The Handbook of Environmental Chemistry 111
Series Editors: Damià Barceló · Andrey G. Kostianoy

Friederike Stock
Georg Reifferscheid
Nicole Brennholt
Evgeniia Kostianaia *Editors*

Plastics in the Aquatic Environment - Part I

Current Status and Challenges

Springer

The Handbook of Environmental Chemistry

Volume 111

Founding Editor: Otto Hutzinger

Series Editors: Damià Barceló • Andrey G. Kostianoy

Editorial Board Members:
Jacob de Boer, Philippe Garrigues, Ji-Dong Gu,
Kevin C. Jones, Thomas P. Knepper, Abdelazim M. Negm,
Alice Newton, Duc Long Nghiem, Sergi Garcia-Segura

In over four decades, *The Handbook of Environmental Chemistry* has established itself as the premier reference source, providing sound and solid knowledge about environmental topics from a chemical perspective. Written by leading experts with practical experience in the field, the series continues to be essential reading for environmental scientists as well as for environmental managers and decision-makers in industry, government, agencies and public-interest groups.

Two distinguished Series Editors, internationally renowned volume editors as well as a prestigious Editorial Board safeguard publication of volumes according to high scientific standards.

Presenting a wide spectrum of viewpoints and approaches in topical volumes, the scope of the series covers topics such as

- local and global changes of natural environment and climate
- anthropogenic impact on the environment
- water, air and soil pollution
- remediation and waste characterization
- environmental contaminants
- biogeochemistry and geoecology
- chemical reactions and processes
- chemical and biological transformations as well as physical transport of chemicals in the environment
- environmental modeling

A particular focus of the series lies on methodological advances in environmental analytical chemistry.

The Handbook of Environmental Chemistry is available both in print and online via http://link.springer.com/bookseries/698. Articles are published online as soon as they have been reviewed and approved for publication.

Meeting the needs of the scientific community, publication of volumes in subseries has been discontinued to achieve a broader scope for the series as a whole.

Plastics in the Aquatic Environment - Part I

Current Status and Challenges

Volume Editors: Friederike Stock · Georg Reifferscheid ·
Nicole Brennholt · Evgeniia Kostianaia

With contributions by

A. Anastasopoulou · N. Brennholt · A. Cabanes · I. P. Chubarenko ·
S. Cieplik · G. Dierkes · H.-J. Endres · T. R. Eremina · A. A. Ershova ·
E. E. Esiukova · G. V. B. Ferreira · C. Földi · T. Fortibuoni ·
A. Fullana · L. Gjyli · D. S. Green · O. Horodytska · C.-S. Hu ·
O. Ilyina · G. Kalčíková · O. Kalinina · J. Kolitari · A. M. Korsunsky ·
E. Kostianaia · A. P. Krelling · T. Kusui · C. Laforsch ·
T. Lauschke · H. Lee · A. R. A. Lima · M. G. J. Löder ·
V. K. B. Narayana · E. D. Osorio · F. E. Possatto · G. Reifferscheid ·
A. Salimon · P. Sapozhnikov · C. Scherer · M. D. Silva ·
A. Snigirova · E. Statnik · F. Stock · M. A. N. Tanchuling ·
B. A. Walther · N. Yen · S. Zandaryaa · A. Žgajnar Gotvajn

Springer

Editors
Friederike Stock
German Federal Institute of Hydrology
Koblenz, Germany

Nicole Brennholt
German Federal Institute of Hydrology
Koblenz, Germany

Georg Reifferscheid
German Federal Institute of Hydrology
Koblenz, Germany

Evgeniia Kostianaia
P.P. Shirshov Institute of Oceanology
Russian Academy of Sciences
Moscow, Russia

ISSN 1867-979X ISSN 1616-864X (electronic)
The Handbook of Environmental Chemistry
ISBN 978-3-030-84117-1 ISBN 978-3-030-84118-8 (eBook)
https://doi.org/10.1007/978-3-030-84118-8

© Springer Nature Switzerland AG 2022
This work is subject to copyright. All rights are reserved by the Publisher, whether the whole or part of the material is concerned, specifically the rights of translation, reprinting, reuse of illustrations, recitation, broadcasting, reproduction on microfilms or in any other physical way, and transmission or information storage and retrieval, electronic adaptation, computer software, or by similar or dissimilar methodology now known or hereafter developed.
The use of general descriptive names, registered names, trademarks, service marks, etc. in this publication does not imply, even in the absence of a specific statement, that such names are exempt from the relevant protective laws and regulations and therefore free for general use.
The publisher, the authors, and the editors are safe to assume that the advice and information in this book are believed to be true and accurate at the date of publication. Neither the publisher nor the authors or the editors give a warranty, expressed or implied, with respect to the material contained herein or for any errors or omissions that may have been made. The publisher remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

This Springer imprint is published by the registered company Springer Nature Switzerland AG.
The registered company address is: Gewerbestrasse 11, 6330 Cham, Switzerland

Series Editors

Prof. Dr. Damià Barceló

Department of Environmental Chemistry
IDAEA-CSIC
C/Jordi Girona 18–26
08034 Barcelona, Spain
and
Catalan Institute for Water Research (ICRA)
H20 Building
Scientific and Technological Park of the University of Girona
Emili Grahit, 101
17003 Girona, Spain
dbcqam@cid.csic.es

Prof. Dr. Andrey G. Kostianoy

Shirshov Institute of Oceanology
Russian Academy of Sciences
36, Nakhimovsky Pr.
117997 Moscow, Russia
and
S.Yu. Witte Moscow University
Moscow, Russia
kostianoy@gmail.com

Editorial Board Members

Prof. Dr. Jacob de Boer

VU University Amsterdam, Amsterdam, The Netherlands

Prof. Dr. Philippe Garrigues

Université de Bordeaux, Talence Cedex, France

Prof. Dr. Ji-Dong Gu

Guangdong Technion-Israel Institute of Technology, Shantou, Guangdong, China

Prof. Dr. Kevin C. Jones

Lancaster University, Lancaster, UK

Prof. Dr. Thomas P. Knepper

Hochschule Fresenius, Idstein, Hessen, Germany

Prof. Dr. Abdelazim M. Negm

Zagazig University, Zagazig, Egypt

Prof. Dr. Alice Newton

University of Algarve, Faro, Portugal

Prof. Dr. Duc Long Nghiem

University of Technology Sydney, Broadway, NSW, Australia

Prof. Dr. Sergi Garcia-Segura

Arizona State University, Tempe, AZ, USA

Series Preface

With remarkable vision, Prof. Otto Hutzinger initiated *The Handbook of Environmental Chemistry* in 1980 and became the founding Editor-in-Chief. At that time, environmental chemistry was an emerging field, aiming at a complete description of the Earth's environment, encompassing the physical, chemical, biological, and geological transformations of chemical substances occurring on a local as well as a global scale. Environmental chemistry was intended to provide an account of the impact of man's activities on the natural environment by describing observed changes.

While a considerable amount of knowledge has been accumulated over the last four decades, as reflected in the more than 150 volumes of *The Handbook of Environmental Chemistry*, there are still many scientific and policy challenges ahead due to the complexity and interdisciplinary nature of the field. The series will therefore continue to provide compilations of current knowledge. Contributions are written by leading experts with practical experience in their fields. *The Handbook of Environmental Chemistry* grows with the increases in our scientific understanding, and provides a valuable source not only for scientists but also for environmental managers and decision-makers. Today, the series covers a broad range of environmental topics from a chemical perspective, including methodological advances in environmental analytical chemistry.

In recent years, there has been a growing tendency to include subject matter of societal relevance in the broad view of environmental chemistry. Topics include life cycle analysis, environmental management, sustainable development, and socio-economic, legal and even political problems, among others. While these topics are of great importance for the development and acceptance of *The Handbook of Environmental Chemistry*, the publisher and Editors-in-Chief have decided to keep the handbook essentially a source of information on "hard sciences" with a particular emphasis on chemistry, but also covering biology, geology, hydrology and engineering as applied to environmental sciences.

The volumes of the series are written at an advanced level, addressing the needs of both researchers and graduate students, as well as of people outside the field of "pure" chemistry, including those in industry, business, government, research establishments, and public interest groups. It would be very satisfying to see these volumes used as a basis for graduate courses in environmental chemistry. With its high standards of scientific quality and clarity, *The Handbook of Environmental Chemistry* provides a solid basis from which scientists can share their knowledge on the different aspects of environmental problems, presenting a wide spectrum of viewpoints and approaches.

The Handbook of Environmental Chemistry is available both in print and online via http://link.springer.com/bookseries/698. Articles are published online as soon as they have been approved for publication. Authors, Volume Editors and Editors-in-Chief are rewarded by the broad acceptance of *The Handbook of Environmental Chemistry* by the scientific community, from whom suggestions for new topics to the Editors-in-Chief are always very welcome.

Damià Barceló
Andrey G. Kostianoy
Series Editors

Contents

Role of Environmental Science in Solving the Plastic Pollution Issue . . . 1
Friederike Stock, Georg Reifferscheid, Nicole Brennholt, and Evgeniia Kostianaia

Pitfalls and Limitations in Microplastic Analyses . . . 13
Friederike Stock, Vinay Kumar B. Narayana, Christian Scherer, Martin G. J. Löder, Nicole Brennholt, Christian Laforsch, and Georg Reifferscheid

Analytical Methods for Plastic (Microplastic) Determination in Environmental Samples . . . 43
Georg Dierkes, Tim Lauschke, and Corinna Földi

Biodegradable Plastics: End of Life Scenarios . . . 69
Hans-Josef Endres

Biological and Ecological Impacts of Plastic Debris in Aquatic Ecosystems . . . 111
Dannielle Senga Green

Impact of Plastic Pollution on Marine Life in the Mediterranean Sea . . . 135
Aikaterini Anastasopoulou and Tomaso Fortibuoni

Plastic in the Aquatic Environment: Interactions with Microorganisms . . . 197
Philipp Sapozhnikov, Alexey Salimon, Alexander M. Korsunsky, Olga Kalinina, Olesya Ilyina, Eugene Statnik, and Anastasiya Snigirova

Freshwater Microplastic Pollution: The State of Knowledge and Research . . . 255
Sarantuyaa Zandaryaa

From Land to Sea: Model for the Documentation of Land-Sourced Plastic Litter . . . 273
Stephanie Cieplik

Plastic Waste Management: Current Status and Weaknesses . . . 289
Oksana Horodytska, Andrea Cabanes, and Andrés Fullana

Plastic Pollution in Slovenia: From Plastic Waste Management to Research on Microplastics . . . 307
Gabriela Kalčíková and Andreja Žgajnar Gotvajn

Marine Litter Assessment on Some Beaches Along the Southeastern Adriatic Coastline (Albania) . . . 323
Jerina Kolitari and Laura Gjyli

Plastic Pollution in East Asia: Macroplastics and Microplastics in the Aquatic Environment and Mitigation Efforts by Various Actors . . . 353
Bruno Andreas Walther, Takashi Kusui, Ning Yen, Chieh-Shen Hu, and Hyemi Lee

The Microplastics in Metro Manila Rivers: Characteristics, Sources, and Abatement . . . 405
Maria Antonia N. Tanchuling and Ezra D. Osorio

Plastic Contamination in Brazilian Freshwater and Coastal Environments: A Source-to-Sea Transboundary Approach . . . 427
André R. A. Lima, Manuela D. Silva, Fernanda E. Possatto, Guilherme V. B. Ferreira, and Allan P. Krelling

Marine Litter in the Russian Gulf of Finland and South-East Baltic: Application of Different Methods of Beach Sand Sampling . . . 461
Alexandra A. Ershova, Tatjana R. Eremina, Irina P. Chubarenko, and Elena E. Esiukova

Role of Environmental Science in Tackling Plastic Pollution . . . 487
Friederike Stock, Georg Reifferscheid, Nicole Brennholt, and Evgeniia Kostianaia

Role of Environmental Science in Solving the Plastic Pollution Issue

Friederike Stock, Georg Reifferscheid, Nicole Brennholt, and Evgeniia Kostianaia

Contents

1 Introduction 2
2 Overview of This Volume 2
3 Discussion 8
References 10

Abstract The first volume of the book "Plastics in the Aquatic Environment" – "Part I: Current Status and Challenges" – gives insights into the role of environmental science and a global perspective. The volume includes 15 chapters dealing with different methods for sampling, sample preparation and analyses of these methods as well as monitoring studies and risks for organisms. Moreover, case studies about the plastic pollution problem from Asia, Latin America, and Europe are presented which gives the reader an integrated overview of the global scope of this issue.

Keywords Global problem aquatic environment, Monitoring, Plastic, Pollution, Risks, Science

---

F. Stock (✉), G. Reifferscheid, and N. Brennholt
Department of Biochemistry, Ecotoxicology, German Federal Institute of Hydrology, Koblenz, Germany
e-mail: stock@bafg.de; reifferscheid@bafg.de; brennholt@bafg.de

E. Kostianaia
P.P. Shirshov Institute of Oceanology, Moscow, Russia
e-mail: evgeniia.kostianaia@ocean.ru

Friederike Stock, Georg Reifferscheid, Nicole Brennholt, and Evgeniia Kostianaia (eds.),
Plastics in the Aquatic Environment - Part I: Current Status and Challenges,
Hdb Env Chem (2022) 111: 1–12, DOI 10.1007/698_2021_780,
© Springer Nature Switzerland AG 2021, Published online: 23 July 2021

1 Introduction

Plastics are emerging contaminants and found in all environments of the world (including the Arctic [1] and remote mountain regions [2]). Plastics in the environment have been investigated since several decades. Researchers all around the world focus on monitoring in different compartments [3–5]. In the last decades, the focus was set on larger plastics (macroplastics) in the world oceans and on beaches as well as impacts on biota, while small plastic particles (<5 mm, the so-called microplastics [6]) in coastal waters were mentioned for the first time in the 1970s [7, 8] and were then discarded for a long time. In the last decade, researchers started to investigate microplastics in detail in marine and freshwaters, biota, sediments, and soil [9–13]. Moreover, many studies have been published about plastic pollution in different countries worldwide.

Some researchers include plastics as part of the "Anthropocene", a new stratigraphically epoch. Humans are a geological factor and strongly influence the environment. Waters et al. [14] argument that the Anthropocene is another stratigraphically epoch which has to be differentiated from the Holocene. Different definitions and starting points are discussed, one among them the Industrial Revolution [15, 16]. The impacts of the Anthropocene can be investigated in different geo-bio-archives such as floodplains, colluvial deposits, beaches, lakes, deltas, soils or ice caps [17].

Besides research on plastics, waste management is one of the main aspects to find solutions to the plastic pollution. Several directives, national frameworks, and bans on certain products have been established in order to deal with the plastic problematic.

The first volume of the book "Plastics in the Aquatic Environment" – "Part I: Current Status and Challenges" – is composed of 15 chapters about environmental science and the state-of-the-art of research on plastic pollution conducted all over the world. The idea to publish a book about science and stakeholders and their role in preventing, reducing, and eliminating plastic pollution emerged at the International Summer School about plastics in marine and freshwater environments which was organized by the German Federal Institute of Hydrology and the International Centre for Water Resources and Global Change (ICWRGC) in July 2017 in Koblenz, Germany. The international researchers and stakeholders discussed the state-of-the-art of research and also presented the diverse efforts and monitoring techniques which are conducted in the countries all over the world [18].

2 Overview of This Volume

The book "Plastics in the Aquatic Environment" consists of two parts: "Part I: Current Status and Challenges" and "Part II: Stakeholders' Role against Pollution" with a total of 27 chapters. The goal of this book is to introduce the current state-of-the-art in science and to show efforts of industry, commerce, and governments to tackle the plastic pollution. Such efforts are manifold and include many directions of action, including but not limited to: formal and informal education, various activities performed by NGOs, development of alternative industrial products, voluntary trade reduction measures, and governmental regulatory actions.

The problem of plastic pollution needs immediate attention. While the existing scientific knowledge is still incomplete and the problem is very complex, there is a pressing demand from the public and policymakers to take prompt measures, including regulatory. This two-volume book aims to provide answers as to how the society, scientists, and policymakers can contribute to solving this problem, what is required of them. Moreover, the book also discusses what data are needed from which stakeholders in order to take the more efficient actions.

Whereas Part II of the book, “Plastics in the Aquatic Environment – Part II: Stakeholders’ Role against Pollution” deals with the society and policy perspectives, Part I gives an overview about the role of environmental science with detailed chapters about sampling, sample preparation and analyses. Moreover, impacts and risks (exposition, hazard, aquatic organisms) for organisms are mentioned and monitoring studies about sources and sinks, transport and fate in the environment for macro- and microplastics are presented. The book also displays case studies from Asia, Latin America, and Europe which gives the reader an overview of the global scope of this issue. Moreover, the last chapters highlight the perception of the plastic issue from the perspective of different countries worldwide.

Stock et al. [19] study pitfalls and limitations in microplastic analyses. The chapter gives an overview about sampling, sample preparation, and analytical methods for analyzing microplastics and discusses the related limitations and challenges of these methods. Although many research papers have been published in the last years about this topic, a comparability of data is still not given. Therefore, the authors describe in detail local and temporal variations and sampling devices in different environmental compartments such as water (plankton and neuston nets, manta trawls, pump-based filter cascades), sediment/soil (shovels, spoons, grabber, corer), or biota. Due to the small size of microplastics (<5 mm) and the complexity of matrices, a thoroughly sample preparation is crucial (size fractionation, density separation, sample purification). Moreover, the authors describe sample loss (adhesion and static forces) and contamination verification via blank samples. The analytical approach covers visual identification and different spectroscopical, spectrometrical, and thermoanalytical methods.

Analytical methods are crucial for detecting microplastics from environmental samples. In their chapter, Dierkes et al. [20] present different spectroscopic and thermoanalytical methods. While spectroscopic methods are, on the one hand, more time-consuming regarding sample preparation, they reveal information about particle numbers, size distribution, color and are non-destructive. Thermoanalytical methods, on the other hand, require less pretreatment of samples and mass concentrations are given. The authors describe the two vibrational spectroscopic methods: Fourier-transform infrared spectroscopy (FT-IR) and Raman spectroscopy which enable reliable microplastic detection when coupled with a particle finding software or a focal plane array (FPA) detector. Identification of particles is conducted with a comparison of recorded spectra to spectra in a library. These methods are widely used by researchers. Different thermoanalytical techniques (pyrolysis, thermogravimetry, and differential scanning calorimetry) have been used for microplastic analyses in the last years. They may serve as an alternative to spectroscopical methods but also complement the existing methods. However, the authors underline that harmonization of these different methods is still missing.

In the "European Strategy for Plastics in a Circular Economy" the European Commission, inter alia, touches the issue of biodegradable plastics. The Commission recognizes the need to establish a clear regulatory framework for plastics with biodegradable properties as some alternative materials claiming biodegradability properties, such as "oxo-degradable plastics," have been found to offer no proven environmental advantage over conventional plastics. In view of this, the chapter by Endres provides insights into the issue of biodegradable plastics from a technical as well as historical perspective [21]. He defines properly the terminology that is used in the whole context, i.e. "biopolymer", "bioplastic", and "biodegradable plastic". In order to better assess the complex issue, Endres focuses on different degradation mechanisms, gives an overview of different standards to characterize degradability, and describes the physico-chemical environmental conditions and main environmental and microbiological parameters for biodegradability.

Biological and ecological impacts of plastic debris (including bioplastics) in aquatic ecosystems are described by Green [22]. Hereby individual organisms as well as biodiversity and ecosystem functioning are considered. Individual organisms suffer, e.g., from entanglement and ingestion. With regard to ingestion, physical (like shape and size), chemical (like polymer type, additives, and persistent organic pollutants), and biological (associated microbes) characteristics of plastics play a decisive role and were intensively looked at. Furthermore, Green also discusses potential spreading of antimicrobial resistance and present research about effects of plastics on ecosystems including changes of benthic assemblages, nutrient cycling, and primary productivity. Moreover, floating debris has always been important for spreading of organisms (e.g., sessile and mobile ones, without pelagic larval stage) all over the world. Man-made litter such as micro- or macroplastics plays an important role for a further dispersal. Depending on the density of polymers, they either float on the water or sink. Biofilms may cover the low-density plastic particles so that these particles with attached organisms sink onto the sea floor and may change the structure and composition.

Anastasopoulou and Fortibuoni [23] describe the impact of marine litter, which is mainly composed of plastic, on life in the Mediterranean Sea. They focus on the such impact categories as ingestion, entanglement, and effects like colonization and rafting. They detail which species have been reported in literature and online sources to ingest plastics and microplastics, while plastic ingestion is the most studied impact in the Mediterranean area. Furthermore, they describe possible consequences of the different impact categories at the individual and population level. Finally, Anastasopoulou and Fortibuoni assess the gaps in the current scientific knowledge in order to evaluate effects at the individual and population level.

Sapozhnikov et al. [24] investigate interactions of plastics with microorganisms. The first part of this chapter gives an overview about many research papers which have been published in the last years. The chapter describes in detail the results of these articles. The authors evaluate the papers by forms of interactions of different polymers and microorganisms and by biofilm communities and biodegradation of polymers. In the second part of this chapter the authors provide in-depth information concerning their own research. For several years, they studied species composition of microorganisms which colonize plastic litter in natural reservoirs and conducted experiments with regard to colonization pattern of synthetic polymers which they put in the natural environment under defined conditions.

An overview of the state of knowledge and research of freshwater microplastic pollution is given by Zandaryaa [25] from the UNESCO Division of Water Sciences. In the UN 2030 Agenda for Sustainable Development, microplastics and their reduction can be associated with several Sustainable Development Goals (SDGs), mainly for reaching a better water quality and for better protecting the water. In her chapter, Zandaryaa presents and discusses the manifold results from scientific publications about sources, pathways, and occurrences of microplastics in different freshwater environments showing that microplastics have been detected in freshwater environments all over the world in lakes, rivers, groundwater, or drinking water. Health and ecological risks of microplastics with special regard to ecotoxicology, human health risks, and microplastics as carriers and sources of other pollutants are also mentioned. Moreover, measures to reduce microplastics such as sustainable consumption, public awareness, responsible production and manufacturing (e.g., reduction, recycling, and reuse), improving wastewater treatment and solid waste collection are discussed in the chapter.

Cieplic from the BKV company in Germany describes the model “From Land to Sea – Model for the documentation of land-sourced plastic litter” [26]. The BKV company provides industry with data and fact base to serve as an aid for decision-making and discussion on topics of resource efficiency and plastics recycling. Cieplic’s model systematically records discharges of improperly disposed-of land-sourced plastic litter, with regard to all of its discharge pathways and discharge sources into the North Sea, the Baltic Sea, and the Black Sea. The goal of this project is systematic recording, structuring, and quantification of the principal discharge sources and pathways for plastics. The model uses several categories of discharge sources and pathways for land-sourced litter, namely: “Coastal regions,” “Rivers,” “Ports,” “Landfills,” and “River shipping.” According to Cieplic, efforts to prevent further litter in the seas can only be effective if discharge sources and pathways along with the corresponding mass flows of plastics into the water are detected and analyzed.

Horodytska et al. [27] have a critical look at plastic waste management and describe the current status and its weaknesses. Global plastic production is still rising and with it the plastic pollution of the natural environment due to the weaknesses of existing plastic waste management strategies. Although approx. 75% of the plastic litter come from developing countries, still 25% remain from western countries owing to the limited efficiency of collection systems and low recycling rates. The authors give an extensive overview of current waste management routes and existing recycling and recovery options with focus on rigid and flexible materials, respectively. These two different types of plastic materials differ regarding their behavior and are therefore usually treated separately. Sources of plastic waste also vary and are mainly grouped into post-industrial (generated during the converting operations, e.g. rejects and offcuts, and this type is usually clean and homogeneous) and post-consumer (mixture of products at the end of their service lives) plastics, whereby post-consumer plastics, not only due to its higher amount, pose a substantial challenge when treating. The post-consumer waste can again be further subdivided into the so-called commercial waste (mainly secondary and tertiary packaging from retail industry areas) and the domestic post-consumer waste (from kerbside collection). Commercial waste is generally homogenous, its composition mostly known, and the amount of physical impurities as well as chemical contaminants is usually low. Domestic post-consumer waste, however, is mostly dirty, often highly contaminated and generally heterogeneous. Waste treatment methods can be divided into mechanical recycling, chemical recycling, and energy recovery. Prior to waste treatment, however, a lot of actions are necessary for managing human produced waste. These include collection, transportation, sorting, and disposal or treatment. The European Waste Framework Directive clearly states that operations for managing waste should not cause any damage to the environment and human health, respectively. For this reason, a waste hierarchy has been implemented for specifying precedence in waste treatment processes: (1) prevention or minimization, (2) preparing for reuse, (3) recycling, (4) recovery as energy, and (5) disposal. The present economy model, which based on extraction, use as well as disposal, is not sustainable from an environmental perspective, particularly with regard to fossil-based non-biodegradable plastics. For this reason, the idea of a new economy model, the Circular Economy, has been developed. The main principles are zero waste, diversity, use of renewable energy, and interaction between systems. After the product's service life the following steps should be addressed: (1) maintenance, (2) reuse, (3) refurbishment, and (4) recycling. But a successful waste management has to start with effective collection and sorting operations. Environmental pollution and waste accumulation can often be traced back to the lack and inefficiency of municipal collection strategies, respectively, especially in developing countries. Inexistent waste collection services as well as limited capacities of landfills play an important role so that inhabitants and authorities feel compelled to throw the waste directly into the environment.

The chapter by Kalčikova and Gotvajn [28] deals with plastic pollution in Slovenia. In the first part the practice of waste management and plastic waste is summarized. Slovenia significantly increased the recycling rate of plastic and packaging waste to one of the highest in Europe (62–69% in 2016) and decreased landfilling in the last years. Social aspects, fines, education, and NGOs considerably contribute to this transition. The second part of the chapter presents research about plastics and microplastics in Slovenia in the last years. In order to prevent plastic waste, researchers studied other materials and investigated biodegradable plastics based on natural products. At the same time, researchers conduct conduct monitoring and impact studies on macro-and microplastics in aquatic environments and biota. The results show that the Slovenian coast is significantly polluted by microplastics although a good waste management in the country exists.

Kolitari and Gjyli [29] studied marine litter and its abundance, composition, and sources along the south-eastern coastline of Albania. The four beaches differed significantly. Two beaches were characterized as semi-urban, one as urban and one as rural. During one winter, the authors monitored 12 cross sections with 12,000 m^2 and an extension of more than 1,000 m. The results revealed a mean litter density of 219 items/100 m and 0.219 items/m^2. The litter was dominated by plastics and other polymer materials (58%) and 37.5% of the items collected mostly originated from tourism and recreational activities. Moreover, poor waste management also contributes to marine litter. At the end of their report they provide options to manage the litter of Albanian beaches as well as mitigation actions that may substantially help to address the problem.

An overview about plastic pollution as a regional problem in East Asia is presented by Walther et al. [30]. The detailed chapter gives insights into the state-of-the-art of macro- and microplastic research in the four countries China, Japan, South Korea, and Taiwan. The authors also summarize the efforts which are made by government policies, society, and the industry/inventors to reduce plastic pollution, to increase recycling, and to create alternative products. The authors used their own research results and conducted a literature review by searching in Web of Science and Google Scholar using specific keywords and their combinations. The results show that intensive research has been conducted in the four abovementioned countries in the last 10 years and that all countries are characterized by high pollution with plastics. However, due to the different socio-economic-political systems, a large difference occurs between the countries regarding mitigation efforts to decrease plastic pollution. The government started to ban some single products and some regional frameworks and collaborations exist between the countries.

The case study of Tanchuling and Osorio [31] deals with monitoring of microplastics in rivers within Metro Manila, Philippines, from 2018 until 2020. The authors studied this area as the Philippines was one of the largest contributors of plastics in the ocean in 2010. The results reveal that up to 60,000 particles/m^3 were detected in the rivers of Metro Manila which is higher than in many studies worldwide. The microplastics mostly degraded from larger plastic pieces and originated from solid waste. The second part of the chapter deals with solid waste management and the regulatory framework. Although a solid waste management infrastructure exists within the country, the waste is disposed on landfills. Moreover, the collection coverage is not very high. The recycling rate has increased up to 33% in 2010; however, many more efforts are still needed to better tackle plastic pollution.

One chapter deals with plastic contamination in Brazilian freshwater and coastal environments. Lima et al. [32] reviewed 37 articles on the basis of Scopus and Web of Science. Although literature about plastics in Brazil exists, a detailed research on plastic pollution in freshwater environments has not been conducted yet. A study of interaction with freshwater fish revealed the source of ingested microplastics from discarded trash. Most of the articles deal with estuarine environments showing that biota ingested microplastics. The results reveal that plastics of all sizes are present in the different compartments (water and sediment). The level of pollution has been described in most of the studies; however, information about transport and pathways are mostly missing. Sources of plastics found in the environment were mostly related to fisheries, urbanization, or improper disposal. On beaches, domestic waste, sewage, and fisheries were mentioned as possible sources. Lima et al. suggest using the source-to-sea approach in order to better understand the plastic problems along the Brazilian coast.

Ershova et al. [33] describe the results of a monitoring study of marine litter on regularly cleaned and wild isolated beaches in the Russian part of the Gulf of Finland (Neva Bay) and the south-eastern Baltic Sea. The authors used different methods (OSPAR, NOAA, IOW beach litter) for monitoring micro- to macrolitter (2 mm to >25 mm) during a four-year period from 2016 to 2020. The monitoring included the wave wrack-line zone and the entire width of the beach from the water line to the first vegetation. The results reveal that no difference can be made between both kind of beaches (wild and regularly cleaned ones) as they are all polluted with marine litter and plastics (domination of microplastics). However, a clear distinction can be made of findings in the Gulf of Finland and the south-eastern Baltic Sea. In the south-eastern Baltic Sea, the beaches are relatively clean and no obvious distinction can be made between high and low populated and visited areas. In the inner part of the estuary of the Neva Bay, the authors detected the largest amount of marine litter. In the region of the Gulf of Finland, plastic pellets, glass, cigarette butts, metal and building plaster, synthetic napkins, and cotton-buds dominate on the beaches. The monitoring results show that different methods complement each other and that they partly depend on the composition of the beaches. The results will be integrated in a database for estuaries and lagoons of the Baltic Sea and will be used for recommendations for the national program of marine litter monitoring along the Russian coasts of the eastern part of the Gulf of Finland.

3 Discussion

World publishing houses pay great attention to the topic of plastic pollution. In Springer, several books have been published about the plastic problem in marine and freshwater environments, especially in the most recent years. These books mainly dealt with research which has been conducted lately.

Looking at the contents of the books published in Springer in the past 5 years, it becomes obvious that the interest towards this problem has been growing significantly. The topics covered have now become more specific, detailed; new aspects of microplastic pollution emerge as subjects for scientific research. For example, in 2015, the book “Marine Anthropogenic Litter” (eds. Melanie Bergmann, Lars Gutow, Michael Klages) [34] analyzed the problem of marine litter, mainly plastics and microplastics, starting from the history of marine litter research, its composition and distribution, moving to discussing effects of marine litter on marine life, sources, pathways, and consequences of the presence of microplastics in the marine environment, as well as methodologies for their detection and identification. The book also reviewed the economics, regulation, and management of marine litter and the role of citizen science in monitoring of marine litter. It is seen both from the title of the book and the titles of the chapters that they tackled the main aspects of the problem. Further books started to focus directly on plastics or microplastics. For instance, in 2018, the book "Freshwater Microplastics. Emerging Environmental Contaminants?" (eds. Martin Wagner, Scott Lambert) [35] discussed the specific problem of microplastics in freshwater systems. In analyzing whether freshwater microplastics were new environmental contaminants, the book reviewed research on microplastics in lakes and rivers, including sources, transport, fate, interactions with biota. Acknowledging the complexity of this problem, the book also examined risk perception of plastic pollution and the role of citizen science and stakeholder involvement, discussed the social-ecological risk perspective, challenges for management and regulation, and the potential solutions.

In 2019, the book "Hazardous Chemicals Associated with Plastics in the Marine Environment" (eds. Hideshige Takada, Hrissi K. Karapanagioti) [36] dug deeper and covered chemicals and additives in plastics, their environmental risks, sorption and desorption of hydrophobic organic compounds. The book also examined ingestion of plastics by marine organisms, factors affecting the amount of plastic in digestive tracts of such organisms, and following on this, transfer of hazardous chemicals from ingested plastics to organisms of the higher trophic level. The book also discussed in detail consequences of the fact that plastics have several chemicals within them. In the same year another book was published, "Bioremediation Technology for Plastic Waste" (eds. Mohd Shahnawaz, Manisha Sangale, Avinash Ade) [37], which focused on bioremediation technologies for plastic degradation, as well as covered other aspects of plastic waste degradation and plastic waste in general, including legislation and social awareness.

The books published in 2020 covered yet even more specific research questions. The book "Microplastics in Terrestrial Environments – Emerging Contaminants and Major Challenges" (2020) (eds. Defu He, Yongming Luo) [38] focuses on microplastics in terrestrial environments, discussing sources, distribution, and environmental fate of microplastics, impacts on ecosystems, health risks, as well as the effect of biodegradable polymers on the environment, and management and legal systems for the control of plastic and microplastic pollution. The book "Mare Plasticum – The Plastic Sea. Combatting Plastic Pollution Through Science and Art" (2020) (eds. Marilena Streit-Bianchi, Margarita Cimadevila, Wolfgang Trettnak) [39] describes the impacts of plastics and microplastics; the "plastisphere", or the new marine ecosystem, the current situation of plastic pollution in various parts of the world. The book provides a brief history of plastics, discusses existing effective and innovative solutions, including the use of circular economy methodologies; outlines the causes of plastic problem in China and Asia and the measures being taken. It further depicts the role of rivers and streams in moving plastic debris from land to the ocean and illustrates the problems associated with small plastic wastes in soils. The book "Proceedings of the 2nd International Conference on Microplastic Pollution in the Mediterranean Sea" (2020) (eds. Maria Cristina Cocca, Emilia Di Pace, Maria Emanuela Errico, Gennaro Gentile, Alessio Montarsolo, Raffaella Mossotti, Maurizio Avella) [40] covers a wide range of topics related to microplastics pollution, which confirms both the complexity and the strong interest towards this research area. It deals with such issues as the sources, fate and impacts of microplastics, methods for their analysis, microfiber pollution, biodegradable plastics, inhalable microplastics, occurrence of nanoplastics in drinking water, innovative solutions to the microplastic problem, transport of pollutants in and on microplastics, and many more other specific issues covering various aspects of microplastic pollution.

We are thankful to Springer for their continuous interest towards the problem of plastic and microplastic pollution and are positive that such books contain comprehensive information on the problem and should be used by various stakeholders, such as scientists, decision- and policymakers, national and international institutions, educational establishments, NGOs, in order to better understand the problem and develop effective solutions. We are also certain that such books can be used by the general public thus raising the awareness of this issue, which will assuredly contribute to the behaviour change.

In this book volume, "Plastics in the Aquatic Environment – Part I: Current Status and Challenges," the chapters provide valuable information on current research areas and challenges that scientists deal with now. We believe that this volume will be useful for scientists in various career stages as well as policymakers to inform themselves of the latest developments in this field and case studies in various countries. We hope that this information will be beneficial and will help solve this problem worldwide in the nearest future.

Acknowledgements Evgeniia Kostianaia was partially supported in the framework of the Russian Science Foundation Project N 21-17-00191 (2021–2023) "Monitoring of water exchange through the Kerch Strait based on state-of-the-art methods of observations and numerical modeling".

References

1. Waller CL et al (2017) Microplastics in the Antarctic marine system: an emerging area of research. Sci Total Environ 598:220–227
2. Allen S et al (2019) Atmospheric transport and deposition of microplastics in a remote mountain catchment. Nat Geosci 12(5):339–344
3. Syakti AD (2017) Microplastics monitoring in marine environment – review. Omni-Akuatika 11(2):1–6
4. Wesch C et al (2016) Towards the suitable monitoring of ingestion of microplastics by marine biota: a review. Environ Pollut 218:1200–1208
5. González-Fernández D, Hanke G (2017) Toward a harmonized approach for monitoring of riverine floating macro litter inputs to the marine environment. Front Mar Sci 4
6. Thompson RC et al (2004) Lost at sea: where is all the plastic? Science 304(5672):838

7. Carpenter EJ, Smith KL (1972) Plastics on the Sargasso Sea surface. Science 175 (4027):1240–1241
8. Carpenter EJ et al (1972) Polystyrene spherules in coastal waters. Science 178(4062):749–750
9. Möller JN, Löder MGJ, Laforsch C (2020) Finding microplastics in soils: a review of analytical methods. Environ Sci Technol 54(4):2078–2090
10. Xu S et al (2020) Microplastics in aquatic environments: occurrence, accumulation, and biological effects. Sci Total Environ 703:134699
11. Triebskorn R et al (2019) Relevance of nano- and microplastics for freshwater ecosystems: a critical review. TrAC Trends Anal Chem 110:375–392
12. Stock F et al (2019) Sampling techniques and preparation methods for microplastic analyses in the aquatic environment – a review. TrAC Trends Anal Chem 113:84–92
13. Schwarz AE et al (2019) Sources, transport, and accumulation of different types of plastic litter in aquatic environments: a review study. Mar Pollut Bull 143:92–100
14. Waters CN et al (2016) The Anthropocene is functionally and stratigraphically distinct from the Holocene. Science 351(6269):aad2622
15. Crutzen PJ (2002) Geology of mankind. Nature 415(6867):23–23
16. Crutzen PJ (2006) The "Anthropocene". In: Ehlers E, Krafft T (eds) Earth system science in the anthropocene. Springer, Berlin
17. Dusar B et al (2011) Holocene environmental change and its impact on sediment dynamics in the eastern Mediterranean. Earth Sci Rev 108(3):137–157
18. Stock F et al (2020) Plastics in aquatic environments – results of an international survey. Fundam Appl Limnol 194(1):67–76
19. Stock F et al (2021) Pitfalls and limitations in microplastic analyses. In: Stock F et al (eds) Plastics in the aquatic environment – part I: current status and challenges. Springer, Berlin. https://doi.org/10.1007/698_2020_654
20. Dierkes G, Lauschke T, Földi C (2021) Analytical methods for plastic (microplastic) determination in environmental samples. In: Stock F et al (eds) Plastics in the aquatic environment – part I: current status and challenges. Springer, Berlin. https://doi.org/10.1007/698_2021_744
21. Endres H-J (2021) Biodegradable plastics – end of life scenarios. In: Stock F et al (eds) Plastics in the aquatic environment – part I: current status and challenges. Springer, Berlin. https://doi.org/10.1007/698_2021_745
22. Green DS (2021) Biological and ecological impacts of plastic debris in aquatic ecosystems. In: Stock F et al (eds) Plastics in the aquatic environment – part I: current status and challenges. Springer, Berlin. https://doi.org/10.1007/698_2020_509
23. Anastasopoulou A, Fortibuoni T (2021) Impact of plastic pollution on marine life in the Mediterranean Sea. In: Stock F et al (eds) Plastics in the aquatic environment – part I: current status and challenges. Springer, Berlin. https://doi.org/10.1007/698_2019_421
24. Sapozhnikov P et al (2021) Plastic in the aquatic environment: interactions with microorganisms. In: Stock F et al (eds) Plastics in the aquatic environment – part I: current status and challenges. Springer, Berlin
25. Zandaryaa S (2021) Freshwater microplastic pollution: the state of knowledge and research. In: Stock F, Reifferscheid G, Brennholt N, Kostianaia E (eds) Plastics in the aquatic environment – part I: current status and challenges. Springer, Berlin. https://doi.org/10.1007/698_2021_748
26. Cieplik S (2021) From land to sea – model for the documentation of land-sourced plastic litter. In: Stock F, Reifferscheid G, Brennholt N, Kostianaia E (eds) Plastics in the aquatic environment – part I: current status and challenges. Springer, Berlin
27. Horodytska O, Cabanes A, Fullana A (2021) Plastic waste management: current status and weaknesses. In: Stock F et al (eds) Plastics in the aquatic environment – part I: current status and challenges. Springer, Berlin. https://doi.org/10.1007/698_2019_408
28. Kalčíková G, Žgajnar Gotvajn A (2021) Plastic pollution in Slovenia: from plastic waste management to research on microplastics. In: Stock F et al (eds) Plastics in the aquatic environment – part I: current status and challenges. Springer, Berlin. https://doi.org/10.1007/698_2019_402

29. Kolitari J, Gjyli L (2021) Marine litter assessment on some beaches along the south-eastern adriatic coastline (Albania). In: Stock F et al (eds) Plastics in the aquatic environment – part I: current status and challenges. Springer, Berlin. https://doi.org/10.1007/698_2020_627
30. Walther BA et al (2021) Plastic pollution in East Asia: macroplastics and microplastics in the aquatic environment and mitigation efforts by various actors. In: Stock F et al (eds) Plastics in the aquatic environment – part I: current status and challenges. Springer, Berlin. https://doi.org/10.1007/698_2020_508
31. Tanchuling MAN, Osorio ED (2021) The microplastics in metro Manila Rivers: characteristics, sources, and abatement. In: Stock F et al (eds) Plastics in the aquatic environment – part I: current status and challenges. Springer, Berlin. https://doi.org/10.1007/698_2020_659
32. Lima ARA et al (2021) Plastic contamination in brazilian freshwater and coastal environments: a source-to-sea transboundary approach. In: Stock F et al (eds) Plastics in the aquatic environment – part I: current status and challenges. Springer, Berlin, pp 1–34. https://doi.org/10.1007/698_2020_514
33. Ershova AA et al (2021) Marine litter in the Russian Gulf of Finland and South-East Baltic: application of different methods of beach sand sampling. In: Stock F et al (eds) Plastics in the aquatic environment – part I: current status and challenges. Springer, Berlin. https://doi.org/10.1007/698_2021_746
34. Bergmann M, Gutow L, Klages M (2015) Marine anthropogenic litter. Springer, Berlin, p 447
35. Wagner M, Lambert S (2018) Freshwater microplastics: emerging environmental contaminants? In: Barceló D, Kostianoy AG (eds) The handbook of environmental chemistry, vol 58. Springer, Cham, p 309
36. Takada H, Karapanagioti HK (2019) Hazardous chemicals associated with plastics in the marine environment. In: The handbook of environmental chemistry, vol 78. Springer, Cham, p 315
37. Shahnawaz M, Sangale M, Ade A (2019) Bioremediation technology for plastic waste. Springer, Singapore, p 130
38. He D, Luo Y (2020) Microplastics in terrestrial environments – emerging contaminants and major challenges. In: The handbook of environmental chemistry. Springer, Berlin, p 469
39. Streit-Bianchi M, Cimadevila M, Trettnak W (2020) Mare plasticum – the plastic sea. Combatting plastic pollution through science and art. Springer, Berlin, p 252
40. Cocca M et al (2020) Proceedings of the 2nd international conference on microplastic pollution in the Mediterranean Sea. Springer water. Springer, Berlin, p 329

Pitfalls and Limitations in Microplastic Analyses

Friederike Stock, Vinay Kumar B. Narayana, Christian Scherer, Martin G. J. Löder, Nicole Brennholt, Christian Laforsch, and Georg Reifferscheid

Contents

1 Introduction 14
2 Sampling 15
  2.1 Spatial and Temporal Variations in the Environment 15
  2.2 Sampling Devices in Different Environmental Compartments 16
  2.3 Summary 20
3 Overview and Comparison of Sample Preparation Techniques 21
  3.1 Complexity of Matrices 21
  3.2 Size Fractionation 22
  3.3 Density Separation 22
  3.4 Alternative MP Extraction Approaches 24
  3.5 Sample Purification 25
  3.6 Summary 26
4 Loss of Analyte and Contamination 27
  4.1 Sample Loss Due to Adhesion and Static Forces 27
  4.2 Blanks 27
  4.3 Mitigation Strategies Against Contamination 28
5 Analytics 28
  5.1 Visual Identification 28
  5.2 Fourier Transform Infrared (FTIR) Spectroscopy 29
  5.3 Raman Spectroscopy 30
  5.4 Pyrolysis Gas Chromatography Mass Spectrometry (Pyr-GC-MS) 32
  5.5 Thermal Extraction and Desorption-Gas Chromatography Mass Spectrometry (TED-GC-MS) 33

F. Stock (✉), C. Scherer, N. Brennholt, and G. Reifferscheid
Department of Biochemistry, Ecotoxicology, German Federal Institute of Hydrology, Koblenz, Germany
e-mail: stock@bafg.de; scherer@bafg.de; brennholt@bafg.de; reifferscheid@bafg.de

V. K. B. Narayana, M. G. J. Löder, and C. Laforsch
Department of Animal Ecology, University Bayreuth, Bayreuth, Germany
e-mail: Vinay-Kumar.Bangalore-Narayana@uni-bayreuth.de; Martin.Loeder@uni-bayreuth.de; Christian.Laforsch@uni-bayreuth.de

Friederike Stock, Georg Reifferscheid, Nicole Brennholt, and Evgeniia Kostianaia (eds.),
Plastics in the Aquatic Environment - Part I: Current Status and Challenges,
Hdb Env Chem (2022) 111: 13–42, DOI 10.1007/698_2020_654,
© Springer Nature Switzerland AG 2020, Published online: 29 August 2020

5.6 Scanning Electron Microscopy Coupled with Energy Dispersive X-Ray Spectroscopy (SEM-EDX) 34
5.7 Quantitative Nuclear Magnetic Resonance Spectroscopy (^{1}H-qNMR) 35
6 Conclusion 36
References 37

Abstract The rising plastic production in the last 70 years led to an increase in plastic waste in the environment. Intensive research activities about macroplastics and microplastics (MPs) started some years ago. Different sampling strategies, sample preparations and analysis methods have been described in the literature for different environmental compartments and biota. Until the present, many papers have been published about the quality and quantity of MPs in different matrices. Pitfalls and limitations in MP analyses are often missing or not discussed. Therefore, this chapter summarizes the present methods for sampling, sample preparation and analysis, discusses the related limitations and outlines the complexity regarding MP loss or contamination during sampling and laboratory work.

Keywords Airborne contamination, Analytics, Environmental compartment, Spatial and temporal variation, Sample preparation

1 Introduction

Our modern daily life would not be possible without plastics – a relatively young group of materials that accompanies mankind since the second part of the twentieth century. Meanwhile, global plastic production rises almost exponentially and reached approximately 360 million tons in 2018 [1]. Associated with this is an increased loss of plastic into environmental compartments. The first description of plastic particles in the sea dates back to 1972 [2, 3]. After four decades of silence, the topic microplastic (MP) was discovered again. Since that time, intensive research has been conducted in marine environments [4–6], whereas a focus on freshwater environments was only set some years ago (e.g. [7, 8]).

Already in 2012, Hidalgo-Ruz et al. [4] pointed out the necessity to establish a common standard operation procedure (SOP) for MP sampling, extraction, purification and analysis to facilitate comparability of data between studies, but still no such standard exists. Thus, many reviews address sampling, sample processing and analyses [9–15] in marine [4, 16], freshwater [7, 17–20] or terrestrial environments and soils [21, 22] or biota [23–25].

These reviews clearly show that different methods are used for sampling, preparation and analysis of MPs from different matrices like water, aquatic sediment, beaches, soil, air or biota. The reviews furthermore reveal that – even if there is a common sense for the need of SOPs – still the report of different units and size classes, categorization of synthetic polymer types, forms or shapes additionally to different sampling and analytical methods often hamper comparability of results for a distinct environmental matrix.

A global suggestion for standardized methods is still missing. However, the ISO/TC 61/SC 14 is currently working on a suggestion for standardization [26]. Meanwhile, first attempts concerning SOPs for MP sampling, sample preparation and detection were made in different national and international research projects [27, 28] and by regional action plans [29]; however, the future will show if they gain a broad acceptance. Until that time, comparability of data suffers from a missing consistent definition of MPs [30] and standardized methods.

Besides these general issues, everybody involved in studies on the contamination of the environment with MP faces various methodological specialties and difficulties. However, even if the careful consideration of these pitfalls and limitations occurring during MP sampling, sample preparation and analysis is inevitable and ultimately determines the quality and reliability of data on MPs, such general obstacles are sparsely mentioned in MP literature.

To fill this gap and raise the awareness for researchers, the goal of this chapter is to present and resume challenges, pitfalls and limitations that can occur when dealing with MP sampling, sample preparation and analysis/identification. For clarity of reading, the chapter is subdivided according to the aforementioned topics.

2 Sampling

Generally, MPs are an extremely heterogeneous group of particles covering different synthetic polymeric materials, sizes, forms and shapes that are furthermore subject to ageing and biofouling in the environment. These characteristics determine also their distribution in the environment, because particles behave differently in comparison to, for example, solvable analytes which may be relatively homogenously distributed in water. Therefore, sampling is the first crucial step when assessing MP abundances. The sampling strategy depends on the aggregate state of the target (solid, liquid, gaseous), the sampled environmental compartment (e.g. aquatic, terrestrial and atmospheric systems) or research item of interest (e.g. tissue, food, drink, sewage sludge, etc.) and of course the respective research question. Different sampling strategies are required for water, sediment/soil, air, biota or other samples. Therefore, a clear and concise strategy concerning location, time, compartment and sampling equipment is necessary.

2.1 Spatial and Temporal Variations in the Environment

Environmental factors lead to site-specific and temporal variations in MP concentrations and compositions. Overall, environmental distribution of MPs can be very heterogeneous which makes representative sampling challenging. For instance, the strong spatial variation in current speed, with backflow areas, whirlpools, etc., strongly influences transport, fast export or temporary retention of MPs in rivers and may create hot spots and areas with low concentrations. The same is true for river banks and beaches where floating MPs usually concentrate at the high-water line. In lakes additionally, prevailing wind direction might also have a strong influence at which lakeshore MPs are found in higher concentration as especially larger MP particles are susceptible to wind drift.

In general, the proximity to anthropogenic activities is supposed to affect MP concentrations and compositions [31, 32]. Secondary MPs are generated via abrasion during usage as well as during building and construction activities, by fragmentation of larger plastic items as a consequence of ageing, or are directly produced as primary MPs and enter the environment via different routes. In this context, transport by wastewater originating from households or industries, wind transport, runoffs from roads, urban regions or agricultural areas with agricultural plastic use, fragmentation of littered plastic waste directly in the environment as well as unintentionally spilled pellets are considered as important pathways and sources, respectively [33, 34].

Potential sources and pathways can be analysed prior to sampling in order to choose sampling sites with informative value. However, spatial and temporal variations hamper the significance of findings derived from single measurements. Therefore, replications as well as repeated measurements are needed to display heterogenic MP distributions in the environment. Up to date, no standard for a representative sampling of different environmental compartments is available, and a definition of minimum replication and number of samples is lacking as standardized procedures are missing [4, 21]. Especially environments that are characterized by a strong heterogeneous contamination of MPs can hardly be sampled representatively with reasonable sampling effort.

Limitations

Sampling with a high spatial and temporal resolution is time-consuming and requires intensive sampling campaigns. In addition, a certain degree of flexibility and mobility is needed to adequately address the influence of environmental factors (e.g. heavy rain falls, storm events, floods or low tides) on the MP abundance. Although anthropogenic factors have been linked to site-specific MP concentrations, the identification of relevant sources is biased by the varieties of materials and pathways.

2.2 Sampling Devices in Different Environmental Compartments

In the following section, currently applied methods and sampling strategies are discussed in detail. The MP research focused for a long time on water samples;

hence, a plenty of publications is available. By contrast, sediment, soil, air and biota are also targets of more recent MP research interest, with less published articles.

2.2.1 Water Sampling

The applied sampling device used for water samples predefines the detectable size range of MPs. Surface waters are frequently sampled, filtered and concentrated with plankton and neuston catamaran nets (optimized for higher wave conditions) or manta trawls. Here, mesh sizes >300 µm facilitate large sample volumes for representative MP counts [35]. However, MPs <300 µm are not addressed quantitatively, and, thus, the amount of MP particles in this size class is underestimated [36]. While sampling with smaller mesh sizes (e.g. 100 µm, 80 µm and below) increases the downward size range of quantitatively sampled MPs, simultaneously rapid clogging at higher seston concentrations limits the flow rate, reduces the sampling volume and leads to errors in sample volume measurements. For preventing the latter, a net cascade with decreasing mesh sizes (e.g. 500 µm, 300 µm, 100 µm) may provide a solution. These methods facilitate large sample volumes by simultaneously considering a higher size range of MPs and may be suitable at different sites under different conditions (e.g. lake, wastewater). A similar approach is the sampling of water via pump-based filter cascades with different mesh sizes. Here, defined volumes of water are either piped through connected filter cartridges [37, 38] or stacked steel sieves [39, 40]. By applying isokinetic sampling, i.e. water is withdrawn at the same speed than the water flow, a non-selective and uniform sampling of particles is guaranteed. Additionally, cascade-based filtration enables the on-site separation of size classes down to 10 µm [39]. A continuous flow centrifuge in which all particles with a density >1 g/cm^3 are retained is another potential alternative. However, it has to be tested if MPs can be sampled quantitatively with this approach [41, 42]. In general, the sampled water volume strongly depends on seston concentrations and, thus, may range from several litres to hundreds of cubic metres.

Limitations

Pumps with attached filter cascades are less mobile than nets. Additionally, the risk of fragmentation, particle loss (e.g. hose system) and contamination (e.g. abrasion of components) during sampling has to be acknowledged and validated [38]. However, the comparison of different methods clearly shows that quantitative and representative sampling with a net is only possible for samples of larger MPs.

Another difficulty occurs with the comparability of results. Sampled volumes differ and so do extrapolated MP concentrations (MPs per litre, m^3, mass units or particle numbers). Moreover, reporting sampling-related limitations (e.g. size range) and appropriate units is mandatory for comparable data. In the future, a combined sampling approach might be probably the most practical solution to sample a wide size range of MPs quantitatively. For instance, MPs >300 μm are sampled by nets, and simultaneously the size fraction <300 μm is sampled by a pump-filter combination.

2.2.2 Sediment and Soil Sampling

Sampling site characteristics and the respective research question necessitate application of different methods. Aquatic sediments include samples from river beds, lake bottoms or from the bottom of the sea and terrestrial (sediment and soil) beaches, alluvial plains, river banks or agricultural areas. Plastic-free utensils like metal shovels or spoons are usually used to sample terrestrial sites and beaches [21, 43]. Sediment/soils are sampled with grabbers (Van Veen grab) and corers for reaching deeper layers [21, 35] or in soil profiles. In contrast to sampling with grabbers that allow for sampling of a disturbed mixture of the sediment/soil in the upper layers only, core sampling can provide undisturbed sediment cores. Thus, information for depth-specific MP deposition and age analysis of the respective layer is potentially preserved.

Reported sample volumes range between 0.5 and 10 kg, and sampling is mostly done randomly at many locations (e.g. with transects perpendicular or parallel to the water or in single squares on beaches) [4]. First proposals for standardized procedures were published by Frias et al. [28] and the MSFD Technical Subgroup of Marine Litter [44] for beach sediments. In these protocols, a monitoring time frame of once per season is suggested within a 30 cm^2 area and a sampling volume of about 4.5 l. Sampling depths may range between 5 and 300 cm. The use of a minimum number of replicates (at least 5) [44] has been recommended for sampling every 100 m on a beach [45].

Limitations

Undisturbed sampling may be achieved with cores, but the overall number of samples collected is limited. Corers often have rods of 1 m length and a diameter of 5 cm. Compaction or loss of sediment by drilling in a higher depth (each 1 m has to be drilled separately with a 1 m extension rod) is likely. In addition, bioturbation may limit the validity of age-dependent MP accumulation. Grab sampling includes larger volumes but disturbed sediments. Besides device-related limitations, different sampling approaches hamper the comparability in terms of different mass or sampling depths as well as reference units. Thereby, standardization of these strategies and a definition of replication and sample amount are urgently needed for the generation of representative and comparable data.

2.2.3 Air Sampling

Air is an important vector for transportation of MPs, e.g. MPs may be transported up to 95 km [46]. Dris et al. [47] investigated indoor and outdoor air in Paris and counted atmospheric fallout of 2 to 355 particles per m^2 and day. Samples were taken with a pump over several hours at a height of 1.2 m indoor or using passive samplers (filters) outside [48]. Filter papers or discs moistened with ultrapure water placed in Petri dishes and exposed to the air are one of the strategies used in passive sampling and monitoring potential airborne contamination in the laboratory. Allen et al. [46] counted more than 350 MPs per m^2 (fragments, fibres, films) in a remote mountain catchment in the French Pyrenees. The authors used two collectors (rain sampler and particulate fallout collector) for a combination of wet and dry sampling. Besides MPs, outdoor sampling may also include sampling pollen or mineral dust. Vianello et al. [49] investigated the exposure of humans to indoor MPs with a breathing thermal manikin and detected 2–16 MPs per m^3.

Further work is needed to focus on the analysis of MPs in the air for the standardization of sampling and analytical methodologies. The environmental topic ‘fine dust’ and related sampling standards provide guiding principles that should be considered when developing sampling strategies for airborne MPs [50–53]. Airborne MPs are also a relevant source of contamination during sampling, preparation and analysis of environmental matrices. Here, lab blanks often reveal high numbers of fibres [54, 55]. The previously mentioned sampling methods may be used to monitor levels of contamination and develop prevention strategies.

Limitations

Pump-based active air sampler systems with a constant flow rate are energy-intensive and expensive to maintain. However, isokinetic sampling has been used a lot for PM 2.5 and PM 10 (particulate matter, particles with a diameter < 2.5 μm (PM 2.5) or 2.5–10 μm (PM 10)) measurements. The performance of these samplers will also be influenced by environmental conditions. The same holds true for passive sampling strategies with moistened filters. Here, the deposition of particles is strongly affected by local air flows and weather events. However, filter-based passive samplers are cheap and can be deployed easily but will require long-time durations of sampling. However, a wet and dry deposition sampler could be a good alternative [46]. Furthermore, the number of polymer-based particles in indoor air samples may be far outweighed by particles of inorganic origin (e.g. glass, ceramic [56]). Thus, plastic-free laboratories or laminar flow boxes are needed. However, a spare room is not available in every laboratory due to limited space.

2.2.4 Biota Sampling

The ubiquitous presence of MPs implies interactions with a diversity of species inhabiting aquatic and terrestrial environments. Thus, in theory, sampling of biota can encompass an enormous spectrum of species. The applied sampling strategies depend on the respective compartment and biocoenosis. For instance, pelagic and benthic invertebrates are commonly sampled with nets and grabbers, respectively. The sampling scale and methodology depend on the size of the organisms. Most studies have focused on several aquatic species such as planktonic invertebrates, mussels and fishes [57–59]. Evidence for MP ingestion in larger organisms is rare due to extensive laborious procedures and ethical issues. Here, stranded carcasses or faeces of larger organisms such as birds, cetaceans and seals are commonly examined for MPs [60–64]. Data for field-collected specimens point to variable levels of contamination (see, for example, compilation by O’Connor et al. [65]). Therefore, sampling of large numbers of especially invertebrates will improve the representativeness of the findings. This indicates that only species with high local densities are appropriate targets for investigation of MP contamination in biota. Considering the dynamic structures of biocenoses, temporal and spatial variations have to be acknowledged. On-site fixation of the collected specimens with preservative chemicals (e.g. ethanol) is recommended to prevent egestion of MPs during transport and storage. Prior to sample preparation (e.g. digestion of biota), specimens should be examined for attached MPs. Overall, sampling of biota complements data on MP abundances in abiotic compartments. Representative easily accessible key species should be chosen and investigated along with other monitoring activities.

Limitations

Given the diversity of the biotic environment, only a small fraction of species has been sampled and analysed for MP contamination. For instance, contamination of bacteria, algae, macrophytes, terrestrial plants and vertebrates especially with nanoplastics (NP) remains largely unknown. As MP ingestion depends on the one hand on the feeding type and size selectivity of biota and on the other hand on the bioavailability of MPs, results from biota reflect only selected (e.g. size, shape) and time-dependent (e.g. egestion) MP abundances in the environment. A direct relation of ingested microplastics to environmental MP contaminations in terms of numbers is difficult to impossible. Characterizing the MP uptake via laboratory studies may help to identify these limitations.

The usage of fixatives can lead to spontaneous evacuation of the gastrointestinal tract due to extreme stress. This should be considered and tested prior to sampling. In general, species-specific potentials to ingest MPs impede the analysis of pooled samples of different species. However, laboratory and environmental results are hard to compare as concentrations used during laboratory experiments with biota often exceed MP concentrations in the field by orders of magnitude.

2.3 Summary

- MPs are heterogeneously distributed in the environment. Particulate properties (e.g. polymer type, size) as well as anthropogenic (e.g. WWTPs) and environmental (e.g. weather) factors lead to spatial and temporal variations in MP abundances. Thus, replication and repeated measurements are needed for representative results.
- The diversity of sampling strategies hampers the comparability of data. For instance, applied devices (e.g. size range) and sampled volumes strongly affect

the results of monitoring studies. Documentation of these limitations is mandatory for cross-study comparisons.

- For water, combined approaches using nets for larger MPs and filter cascades for smaller MPs are recommended. Contamination (e.g. abrasion of sampling equipment) has to be considered and validated.
- For sediment and soils, corers are advisable for deeper profiles (stratified layers) and shovels for surface samples. Here, the volume of the sample should ensure representative MP analysis.
- For air, active and passive samplers might be used for monitoring studies. Monitoring airborne MPs is mandatory during sample preparation.
- For biota, applied sampling devices depend on the targeted species. Here, large numbers are usually needed to compensate for high variabilities.
- Overall, parallel sampling of aquatic, terrestrial and aerial compartments is beneficial for a better understanding of the environmental distribution and fate of MPs. Standardized and harmonized sampling strategies are needed for cross-study comparisons.

3 Overview and Comparison of Sample Preparation Techniques

3.1 Complexity of Matrices

Sample preparation is an important task in MP research. Whereas macroplastics may be easily spotted in the environment, detection of MPs requires an extensive processing of the native sample due to an unfavourable target-nontarget ratio.

The complexity of matrices varies between and within environmental compartments. For instance, concentration and composition of suspended solids differ among water bodies and among locations (e.g. groundwater, lake, stream and ocean). The same is true for the composition of sediments (e.g. marine beach, lakeshore), soils (organic- or mineral-rich), biota samples (biomass, content of protein, fat, carbohydrates, biological structures, etc.) or air. As particulate materials (e.g. minerals, organic matter) interfere with MP identification, these abovementioned variations imply different demands on sample preparation. Here the complexity of sample preparation increases with increasing proportions of particulate matrix matter. However, the first step towards representative counts is achieved by reducing the volume of the native sample. This is done by separating MPs from natural components based on material properties. Several methods have been proposed including sieving, density and oil separation, electroseparation as well as digestion of organic material. Depending on the investigated matrices, single or multiple combined techniques (e.g. sieving, density separation, digestion of organic material) are applied for sample preparation.

Limitations

Processing of native samples most often implies manipulations of the original conditions. For instance, MP sizes, polymer composition, hetero-aggregates, biofilms, MP degradation state as well as sorbed pollutants can be altered by sample preparation [66]. In addition, the diversity of methods limits the comparability between studies. Detailed information on each method is presented in the following section.

3.2 Size Fractionation

Size fractionation via sieving may allow for an easier processing of environmental samples in the following extraction and purification steps and is applied on aqueous and sediment samples.

Variable and wide particle sizes limit the detection of MPs via microscopy and spectroscopic analysis. For instance, multiple particle sizes require different magnifications, and large particles may overlay small particles. For an optimized analysis via microscopy and spectroscopic analysis, stacked sieves with different mesh sizes may be used to fractionate MP size classes. Here, wet or dry sieving is possible. Closed wet sieving units prevent formation of dust (e.g. particle loss) and contamination.

Limitations

Clogging of sieves may result in particle loss and unprecise fractionation. In addition, abrasion or fragmentation during sieving may alter numbers, sizes and shapes of MPs. During wet sieving with a small mesh size, small particles get washed away if the washing fluid is not filtered for a recovery of small fractions.

3.3 Density Separation

Plastics are lightweight materials and may be separated from natural components by utilizing differences in densities. This circumstance is broadly utilized for separating MPs from sediment or soil matrices. For instance, the density of commodity plastic polymers varies between <0.01 and >1.4 g/cm^3 (e.g. foamed PS and PVC), whereas mineral materials are typically denser (e.g. >2.5 g/cm^3 for quartz, feldspar and calcite) [4]. If environmental matrices are suspended in high-density liquids (>1.2 g/cm^3), MPs will float to the top, and minerals sink to the bottom, respectively. Depending on its density, selection of the separation fluid determinates the spectrum of recoverable polymers (Table 1). In addition, monitoring the density of separation fluids is imperative to ensure reproducibility. Density separation is conducted with several devices ranging from simple conical flasks and funnels to custom-built equipment (e.g. [67]). For instance, the Munich Plastic Sediment

Table 1 Density separation

Chemical	Sodium chloride (NaCl)	Zinc chloride (ZnCl)	Sodium iodide (NaI), sodium bromide (NaBr)	Sodium tungstate dihydrate ($Na_2WO_4 \cdot 2$ H_2O)	Sodium polytungstate ($Na_6H_2W_{12}O_{40}$)	Potassium formate K (HCOO)
Density [g/cm^3]	Up to 1.2	Up to 1.8	Up to 1.8 (NaI), medium density 1.4 (NaBr)	Up to max. 3.25	Up to max. 3.1	Up to 1.6
Advantage	Low price, eco-friendly	Low price, high density	High density	Low price, high density	High density, non-toxic	Low price, not hazardous
Disadvantage	No recovery of dense polymers (PVC, PET)	Hazardous and corrosive	Expensive, reacts with cellulose filter, hinders visual analysis		Expensive	Hygroscopic
References	[28, 44]	[28, 69]	[68, 70, 71]	[44, 72]	[73, 74]	[75]

Separator allows processing of large sample volumes in a single run (max. 6 l sediment) [67]. Recovery rates of 95.5% and 100% for small and large MPs, respectively, as well as the closed construction are further benefits. After density separation, the supernatant is filtered (pressure or vacuum) and/or centrifuged and further processed. Large MPs in the filtered supernatant may be individually picked with tweezers while sorting under the stereomicroscope. Another possibility is fluidization. Nuelle et al. [68] decreased the mass of a sediment sample by inducing air-generated turbulent gas bubbles into a density solution and adding the sediment. Lighter particles were transported to the top and transferred to another glass vessel.

Limitations

Although differences in densities represent a promising and frequently applied method to separate plastic materials from natural components, several limitations have to be acknowledged. First, densities of organic matter are similar to those of plastic materials. Thus, organic matter as well as any other material with a similar density will float in dense liquids and will be included in the sample. Second, degradation of MPs and formation of biofilms and hetero-aggregates affect density of plastics. Furthermore, aggregation and disaggregation of particles during density separation might affect the fate of MPs (e.g. sedimentation of MPs attached to larger minerals). Third, method validation is mainly conducted with artificial sediments. Therefore, recovery rates are not directly applicable to complex environmental matrices. Fourth, comparability of results is hampered by the variety of methods (e.g. equipment used, sample volume and separation liquid).

3.4 Alternative MP Extraction Approaches

Centrifugal forces may also be used for separating MPs from other materials. Bauer et al. [76] published a paper on sink-float density separation of polyolefins from waste. Two-stage processing with a centrifugal force separator and a hydro jig for the first separation of larger waste has been conducted. Another possibility for separating MPs from environmental samples is the use of oil extraction due to the oleophilic characteristics of plastics. Crichton et al. [77] mixed a dry sample with water and canola oil and swirled several times so that each particle got into contact with oil. The sample settled; oil was decanted, rinsed and filtered; and the filter incubated in reagent alcohol in order to remove all the remaining oil on MPs. A recovery rate of >90% was achieved for expanded polystyrene (EPS), polyvinyl chloride (PVC), acrylonitrile butadiene styrene (ABS), vinyl, polyamide (PA) fibres and polyester fibres (PES). This method is cost-effective and easy to handle. Whereas sediments and particulate matter are conductive, plastics are nonconductive materials [78]. Thus, electroseparation may be used for separation of sediments and MPs. The method is based on electrostatic behaviour of particles [79]. Dry particles are charged with a corona electrode up to 30 kV and separated into conductive and nonconductive particles. In beach sediments, the recovery rate of pristine microplastic particles >63 μm is close to 100%. This method allows processing of high sample volumes and reduces the remaining matrix to 10%. Due to the reduction of material, only small amounts of chemicals are needed. Moreover, it is a relatively cheap method as only the device is needed and no chemicals are required.

Limitations

The application of centrifugation, oil separation and electroseparation remains to be tested and validated for the separation of a diversity of polymers, size classes and shapes. In addition, further research is necessary in order to verify recovery rates for different matrices and environmental MPs (e.g. hetero-aggregates). For centrifugation, one can envision that this method will include expensive apparatuses and high maintenance expenditures as the rotating parts may be subjected to significant force and sensitivity towards the efficiency of the approach when applied to samples from different environments. For oil separation, high concentrations of organic matter may reduce the overall efficiency. Separating sediment components based on their conductivity does only work for completely dry samples. Thus, particle loss, especially in the smaller-size fractions due to the formation of dust, is possible and has to be considered for mitigation strategies.

3.5 Sample Purification

Synthetic polymers are relatively resistant against selected acids and bases. This is beneficial for MP purification. In theory, incubating environmental samples with acids and/or bases result in digestion of organic matter and, thus, in reduction of the volume. However, whether MPs remain unaffected depends on the applied chemicals and the polymer types (Table 2). For instance, polyesters like PET, PBT, PC, PLA and other synthetic polymers such as cellulose acetate (CA) and PVC might be degraded with 10 M NaOH [80, 81]. Application of HCl results in high digestion efficiency (>95%) albeit with melting of synthetic polymers like PET [82]. Digestion with H_2O_2 and Fenton's reagent has been often used with promising efficiencies. However, a study revealed that only 70% of MPs were recovered after incubation with 30% H_2O_2 for 1 week [83]. In this regard, validation of the digestive protocols is mandatory. Enzymatic digestion with single or sequential incubation of enzymes is another gentle possibility to eliminate organic materials without affecting synthetic polymers [58, 84]. Depending on the study and validated workflows, samples are either treated before or after density separation. In general, the complexity of the sample (water, sediment, biota or air) affects duration and efficiency of digestion.

Limitations

Strong acids and alkaline solutions adversely affect synthetic polymers. Here, the degree of degradation and discoloration is related to the used chemicals and is polymer-specific (Table 2). Acid digestion with chemicals like HCl and HNO_3 is not recommended as synthetic polymers like nylon and PET have low resistance to

Table 2 Organic digestion

Digestion	Alkaline	Acids	Oxidizing	Enzymes
Chemicals	Sodium hydroxide (NaOH), potassium hydroxide (KOH)	Nitric acid (HNO_3) or hydrochloric acid (HCl)	Hydrogen peroxide (H_2O_2)	Cellulase (>30 U/ml), lipase (>15,000 U/ml), chitinase (>40 U/ml), protease (1,100 U/ml)
Advantage	Most organics destroyed, KOH: Most polymers resistant	HNO_3: most organics destroyed	Most organics destroyed	Most organics destroyed, not hazardous
Disadvantage	Some polymers degraded (e.g. PC, CA, PET, PVC)	HNO_3: dissolution of PS and PE possible, HCl Incomplete destruction of organics	Polymers might be affected	Time-consuming, partly expensive, different Enzymes for different samples
References	[5, 58, 85]	[5, 83]	[28, 83, 86]	[5, 15, 80, 84, 87]

these acids. Alkaline digestion may also damage or discolour plastic fragments [10]. However, in order to effectively remove organic matter, certain compromises have to be made. This includes concentration of chemicals, incubation temperature and time. Although enzymatic digestion does not dissolve and degrade synthetic polymers and is very effective in the purification result, the proposed protocols are time-intensive [80, 84]. For instance, several sequentially applied enzymes (e.g. lipase, chitinase, lignase and proteinase) are needed to effectively digest biological structures (e.g. lipids, chitin carapaces, lignin, carbohydrates and proteins). In addition, exposure conditions have to be monitored and modified for optimal reaction rates.

3.6 Summary

- Varying complexities require matrix-specific and standardized procedures for water, sediment, soil, air and biota samples. Here, single or multiple combined techniques may be used.
- Sample preparation increases probability of contamination, particle loss and fragmentation. Thus, an extensive validation of the entire process is essential to prevent underestimations and overestimations of MP abundances.
- Size fractionation and density separation are commonly applied procedures to narrow the sample volume and to separate MPs from natural components, respectively. Alternative methods have been published for separation, but have not been tested in detail and are therefore not advisable at the moment.

- If organics prevail, samples must be purified (with alkaline or oxidizing solutions, acids or enzymes). All have advantages and disadvantages, and treatment with strong chemicals might affect polymers.

4 Loss of Analyte and Contamination

4.1 Sample Loss Due to Adhesion and Static Forces

Synthetic materials are characterized by their surface charge [88]. Plastic polymers are often easily charged statically. The cleaner the surface of MP particles from environmental samples, the more the particles are prone to static charge. Due to static forces, especially when being dried or when samples are processed, MP particles may be lost due to these forces when particles interact with one another or with surfaces such as glass used during laboratory work. High hydrophobicity of synthetic polymers such as nylon, polyester, PVC and PE, for example, results in the accumulation of positive or negative charges depending on the material. This suggests that MPs constituted of such synthetic polymers are at a greater risk of being lost due to the influence of static interactions [79].

4.2 Blanks

Airborne fibres and smaller MP particles represent the largest part of contamination. In order to receive representative results, it is imperative to produce blanks to trace the contamination with plastics during the whole process (from sampling to analysis).

Laboratory protocols include organic digestion, density separation and possible counting/photographing MP particles with a microscope. During all steps conducted in the laboratory, a blank sample with distilled water is treated with the same chemicals/solutions as used for the processing of the real samples. In the standardized protocol for monitoring MPs in sediments [28], one blank per day of laboratory work is suggested. Furthermore, the mean value of at least three blank samples allows for the more assured correction of the real samples.

Blanks collected on substrates (e.g. filters) must be analysed by means of FTIR, Raman spectroscopy, mass spectrometry, etc. commonly used for the detection and characterization of the synthetic polymer constituting MP particles in samples. The average amount of synthetic polymer found in blanks may be subtracted from the real sample values. To be conservative, mean values of the found plastic particles should be round up to the next integer (e.g. 2.3–3) prior to correction of the particle numbers in real samples.

4.3 Mitigation Strategies Against Contamination

In order to avoid MP contamination, an accurate clean workspace is absolutely necessary. Especially, the workspace in the laboratory may lead to contaminations due to several handling steps of samples. It should be cleaned before starting to work. The workspace could be cleaned with 70% ethanol and paper tissues; all apparatuses can also be washed with 100% ethanol. It is also of importance to filter all working solutions such as ethanol, hydrogen peroxide, acids, bases, enzymes, etc. prior to use. During the entire sample processing steps, the use of plastic material should be avoided and replaced by steel or glass devices [89]. Moreover, cotton cloths are recommended while handling samples [44]. Samples must be covered with aluminium foils or materials made from glass in order to avoid contamination from air. Furthermore, samples should be handled and processed under controlled air circulation; the use of a laminar flow box or clean bench is suggested [10].

5 Analytics

5.1 Visual Identification

Many researchers have used visual identification with microscopes for identifying MP particles in earlier studies. Most often, visual, light or digital microscopes are used for the analysis of larger plastic particles with a size of 300 μm to 5 mm (e.g. [90–93]). Smaller particles are not so easily recognizable as plastics; hence, they are first analysed under a stereomicroscope, and synthetic polymers are later identified by means of Raman, pyrolysis GC-MS or FTIR spectroscopy [94, 95]. Aqueous samples can be sorted with the help of sorting chambers like Bogorov counting chambers. Size limits for visual inspection without assistance from other characterization techniques are recommended to be >500 μm [87] or even >1 mm [4]. Norén [96] recommends a standard criterion for visual identification of particles to reduce chances of misidentification, such as absence of organic origin structures on MPs, equal thickness in MP fibres, homogenous coloration of the particle and finally application of fluorescence microscopy to exclude structures of biological origin. Advantages of visual identification are that relatively quick first quantification is possible and that each identified particle larger than 500 μm may be described, photographed and measured [15].

Limitations

Larger particles may be identified by their shape, size, degradation stage and colour as possible plastic fragments [4], but a 100% certainty of identification is not given. Smaller particles, especially <300 μm, are not very well identifiable as these fragments may also be of mineral or organic origin [4, 97]. Handling of particles below a certain size using instruments such as forceps can become unmanageable because of their minute size [96]. Moreover, the results of visual identification are subjective, depend on the sample matrix and are very time-consuming [15]. Therefore, visual identification should only be used as preliminary evaluation of results coupled with another method for a higher accuracy and precision [98]. The approach by itself should not be applied to MP particles <500 μm as the probability of misidentification is very high. It is instead highly recommended to use assistance from spectroscopic approaches for the accurate identification of such MPs [99].

5.2 Fourier Transform Infrared (FTIR) Spectroscopy

FTIR spectroscopy is a vibrational spectroscopy approach which has been used for decades for the analysis and characterization of synthetic organic polymers and their products [15]. This technique involves excitation and detection of molecular vibrations of a sample, which leads to the acquisition of a characteristic spectral fingerprint. FTIR spectroscopy is an absorption technique: the IR radiation absorption by molecule vibration depends on the change of the dipole moment of a chemical bond within a molecule constituting the sample. In this approach, the sample is irradiated with IR light (mid-IR range), a part of the IR radiation is absorbed by the exited molecule vibrations within the sample being probed and detected either by reflection or transmission mode.

Synthetic polymers with their ever-repeating molecule composition possess highly specific IR spectra with distinct signals that make it an ideal technique for the identification of MPs. They can be characterized based on their chemical structure and can be identified by comparing them to reference spectra. As a measuring variation, the surface technique attenuated total reflectance (ATR) FTIR spectroscopy allows for the fast and reliable single analysis of large MP particles >500 μm [15]; however, reliable results need a relatively clean surface, and potential MP particles with biofilm need to be cleaned by, for example, wiping with alcohol.

Micro-FTIR spectroscopy, the combination of FTIR spectroscopy with an IR microscope, facilitates the analysis of particles down to a size of 10 μm. Measurements in transmission mode require the use of IR transparent filters (e.g. aluminium oxide or silica) on which the sample is placed. The thickness of the sample that can be analysed with this mode is limited due to the total absorption of IR radiation through samples above certain thickness. Micro-FTIR mapping with a single element detector and focal plane array (FPA)-based FTIR imaging has been extensively applied for the identification of MPs [100]. The former is very time-consuming, whereas the latter allows for the acquisition of several thousand spectra within a short time frame and at a high spatial resolution. This helps in the sequential imaging of the whole sample filters and is currently the predominant method for the identification and characterization of MPs. The use of reflectance mode in FTIR is also possible. However, this mode results in the generation of complex spectra difficult to interpret as particles with irregular shape lead to refractive error [12, 15, 101].

Overall, the advantage of FTIR analysis is that qualitative and quantitative information about each MP particle can be obtained and the sample is not destroyed and is still present for further downstream analysis. The analysis requires relatively short measurement times, and large sample areas of up to 10 × 10 mm can be efficiently measured within less than 2.5 h. This procedure circumvents extrapolation of measurement results from sample filter subareas and potential uncertainties related to unequal particle distribution on the filter. However, measurement of whole filters results in large datasets with up to more than one million IR spectra which are best analysed via automated approaches [102, 103].

Limitations

Information about the mass of MPs is not accessible with this method. One of the major drawbacks of this technique is that very small particles <10 μm cannot be analysed due to the diffraction limit of light. Especially analysis of fibres having a diameter in that size range is very difficult. Furthermore, there are reports showing significant underestimation of MP particles <20 μm [12]. Thickness of the particles >50–100 μm leads to total absorption as well as black particles which absorb strongly in the IR range. This may lead to difficulty in data analysis and underestimation of MPs if such particles are present. Samples must also be thoroughly dried as water is a strong absorber of IR radiation. There are other limitations which are common for the complementary FTIR and Raman spectroscopy techniques. These will be discussed together in the section after Raman spectroscopy below.

5.3 Raman Spectroscopy

Raman spectroscopy is like FTIR a vibrational spectroscopy technique. It is a surface technique involving inelastic scattering of light to unveil molecular composition of a sample by probing vibrational and rotational frequencies of molecules. A monochromatic light source such as a laser is used to interact with the sample. Most commonly used lasers have wavelengths ranging from 500 to 1,064 nm. Radiation from this source interacts with the sample and one in a billion photons from the source is inelastically scattered revealing information about molecular vibrations of the sample. The differences in the frequency of inelastically scattered photons in comparison to the Rayleigh photons (photons which do not interact with the sample) are known as the Raman shift which forms the basis of the Raman spectrum [104, 105]. Like FTIR spectroscopy, this approach also provides a unique spectral fingerprint of different chemical structures, but the basic difference is that Raman spectroscopy depends on changes in the polarizability of the chemical bond within molecules constituting a sample. Hence, the two approaches are complementary; signals which are strong in IR may be weaker in Raman spectra and vice versa. The Raman spectrum of a synthetic polymer has several unique sharp signals corresponding to the chemical functional groups constituting the sample and can be identified by comparing them to reference spectra. Currently, micro-Raman spectroscopy approach (Raman spectrometer setup coupled with a microscope) is predominantly used. A confocal mode of configuration ensures a lateral and depth resolution that allows the analysis of particles up to ~0.5–1 μm depending on the type of sample being analysed. Thus, with respect to size, a higher percentage of MPs can be detected in comparison to FTIR spectroscopy. Raman spectroscopic imaging mode allows qualitative and quantitative estimation of small size-range MPs albeit with time duration being the principal constraint as Raman images are acquired by stepwise point measurements, the so-called mapping. Raman spectroscopy also allows the chemical analysis and localization of specific components like MPs within complex matrices such as biological cells and tissues. Additionally, it is a significant approach to monitor the alterations in the biochemistry of organisms when stressors like MPs are ingested. Micro-Raman spectroscopy has been applied worldwide for the analysis of samples ranging from single microbial cells to the analysis of MPs in recent years [106, 107]. This approach facilitates detection of even the smallest MP particles in environmental samples, but time-effective integration for MP research is yet to be demonstrated [36, 95].

Limitations

One fundamental drawback of the Raman technique is that it is a very weak phenomenon, which leads to long measurement times with Raman images of whole filters requiring 24–48 h or even longer depending on the resolution and sampling area. Another major drawback is that the Raman signal can be severely affected by the presence of coloured and noncoloured organic or inorganic debris or contaminants which result in fluorescence that swamps the entire Raman spectrum. Hence, environmental samples must be processed thoroughly to have least possible impurities on the samples for a clear identification of MPs from the environment. Lasers with longer wavelengths such as 1,064 nm can also be applied to overcome fluorescence, but the Raman intensities are drastically reduced with the use of longer wavelength excitation sources due to loss in energy. Hence, the focus of the future for the realization of a time-effective and fluorescence suppression-based micro-Raman spectroscopic analysis of MPs depends on testing of the alternate approaches such as the wide-field imaging method and the non-linear Raman spectroscopic approaches like coherent anti-Stokes Raman scattering (CARS), stimulated Raman scattering (SRS), time-gated Raman spectroscopy (TGRS) and shifted excitation Raman difference spectroscopy (SERDS), respectively [108–112].

Limitations Prevalent in FTIR and Raman Spectroscopy

FTIR and Raman spectroscopic MP analysis utilize spectral libraries comprising of data acquired from a large collection of mostly pristine synthetic polymers for comparison to carry out identification of MPs in samples being investigated. But a critical issue is that the spectra of environmental MPs tend to be sometimes different from those included in the spectral library which hinders MP identification rates. FTIR and Raman spectra of environmental MPs are influenced by factors such as the presence of additives, plasticizers, colouring agents, residence times in different environmental conditions, etc. [107]. Hence, it is important to include spectra of environmental MPs in the libraries to improve the MP identification efficiency, and the process of establishing such a comprehensive spectral library inclusive of all this data in laboratories worldwide can be highly time-consuming. Thereby, establishing a comprehensive open-source spectral library which includes FTIR and Raman spectroscopic data acquired from a diverse range of synthetic polymers; biodegradable polymers including those which contain additives, colourants and plasticizers; and aged polymers obtained from exposure to different environmental conditions is necessary. These libraries should also include the parameters used for spectral acquisition. Generation of such an open-source spectral library may be of great help to research groups to avoid creation of repetitive databases and encourage more MP identification studies worldwide especially in groups with limited access to reference polymers [107]. Agglomeration of MPs on filters and substrates used in these approaches is another critical issue which may be overcome by using small aliquots of samples for analysis. The estimated MP quantity from such small aliquots can then be extrapolated to the total investigated sample volume [113]. However, it is important to note that more concrete studies are required to investigate the feasibility of this approach to achieve an accurate estimation of MPs [114]. Furthermore, identification of aged MPs using these techniques is a challenging task as long residence times in the environment expose these particles to factors like sunlight which may alter the chemistry of the polymers. This results in changes in the spectrum of the MPs potentially hampering their identification.

5.4 Pyrolysis Gas Chromatography Mass Spectrometry (Pyr-GC-MS)

In contrast to FTIR and Raman spectroscopy, pyrolysis GC-MS (Pyr-GC-MS) allows quantification of masses of MPs in environmental matrices. This analytical approach involves thermal degradation of large molecules into smaller ones in an inert atmosphere to assess the chemical composition of samples like MPs. Tuning of the pyrolysis temperature leads to the generation of unique volatile degradation products which are similar or can be traced to their precursor synthetic polymers [12]. These pyrolysis products can be separated on a gas chromatography (GC) column and mass spectrometry (MS) measurements then make identification of these products possible. Pyrolysis fingerprint or pyrograms facilitate the identification and characterization of the synthetic polymer constituting the MPs. These pyrograms can be compared with reference pyrograms of known virgin synthetic polymer samples. For synthetic polymers with polar subunits like polyesters, thermochemolysis – which is thermally assisted methylation and hydrolysis – is applied. This procedure is known to improve chromatographic separation, increase sensitivity and give additional structural information. Plastic additives can also be simultaneously determined during Pyr-GC-MS analysis if a thermal desorption step is used before pyrolysis. Pyr-GC-MS has been previously applied for the analysis of MPs from environmental samples like marine sediments, river sediments, sea water surface and marine organisms [12, 115, 116]. Fischer and Scholz-Böttcher [117] recently showed the concentration of a whole environmental MP sample on filters after purification and the subsequent Pyr-GC-MS analysis in one run. This approach is promising as it was carried out without the need for optical or mechanical presorting [118]. Furthermore, mass-related quantification was obtained at trace levels using calibration procedures. Dierkes et al. [119] showed that the mass of the most common polymer types PE, PP and PS might be measured with limits of quantification down to 0.007 mg/g sample.

Limitations

However, there are certain limitations for quantification of MPs using Pyr-GC-MS. Firstly, the analysis is destructive as samples are pyrolized. Secondly, quantification is mass-based meaning that no further information about particle number or shape can be obtained. Analysis of copolymers would result in pyrolysis products of each comonomer instead of a specific pyrolysis product for copolymers. Furthermore, additives, fillers and dyes contribute to the mass of MP particles but are not specifically included in mass-based quantification.

In fact, identification of single MP particles via specific pyrolysis products is relatively fast and simple. However, mass-based quantification of MPs in environmental samples, hence, a large variety of MP particles, is far more complex. Specific pyrolysis products for common synthetic polymers are well described in the literature [120, 121]. Nonetheless, abundance of organic substances such as natural polymers (cellulose, keratin, chitin, lignin) and hydrophobic compounds (fats and waxes) results in similar or identical pyrolysis products possibly leading to an overestimation of MP concentration. For instance, polystyrene (PS) is widely evaluated via its monomer styrene which, however, also results from the pyrolysis of, for instance, lignin. Besides, styrene is also a part of other synthetic polymers such as acrylonitrile butadiene styrene (ABS) or styrene-butadiene rubber (SBR) which results in a cumulative mass concentration based on styrene concentration rather than a polymer-specific concentration. An overestimation due to the presence of natural polymers can in fact be overcome by using the trimer instead of the monomer; however, differentiation of its origin, either from PS, ABS or SBR, is not possible.

5.5 *Thermal Extraction and Desorption-Gas Chromatography Mass Spectrometry (TED-GC-MS)*

TED-GC-MS involves a combination of thermal extraction of samples with thermogravimetric analysis (TGA) on solid-phase adsorbers and subsequent analysis of the adsorbers with thermal desorption-gas chromatography mass spectrometry (TDS-GC-MS) [121, 122]. This combination is known as TED-GC-MS. In this approach, the entire sample is pyrolysed in the TGA at temperatures of up to 1,000°C. The synthetic polymer-specific degradation products, which have to be different from the degradation products of the environmental matrix, are adsorbed on a solid-phase adsorber. Decomposition of most synthetic polymers begins at 350°C, therefore enabling the possibility to eliminate the components of the environmental matrix, which decompose at much lower temperatures and are not trapped on a solid-phase adsorber. These relevant degradation products of plastics are trapped out on a solid-phase adsorber such as polydimethylsiloxane (PDMS). The trapped decomposition products are desorbed by thermal desorption and transferred by an inert gas such as helium into a cooled injection system. Furthermore, only compounds that can be thermally desorbed at temperatures 200–300°C are evaporated for analysis with GC-MS. This acts as a filter as it prevents most long-chain polymer-specific contamination products from reaching the GC-MS [123]. Analysis of these products by GC-MS makes identification and quantification of samples like MPs from different environments possible. Synthetic polymer identity is confirmed based on the occurrence of mass fragments representative of different decomposition products specific to that polymer. Thus, TED-GC-MS can act as a fast tool for identification and quantification of MPs in environmental samples. However, up to date, the range of plastic types that were confirmed to be analysed by TED-GC-MS in environmental samples is very limited (e.g. PE, PP, PS, PA 6 and PET, [121]).

Limitations

Large-scale testing and implementation of this approach for the analysis of MPs from different environmental samples is yet to be realized. However, samples such as in the Pyr-GC-MS approach are not available for downstream analysis due to destruction. It also allows analysis of only 20 mg of dry sample during one TED-GC-MS run; thus, several sequential runs will be needed to analyse many samples. Nevertheless, it is suggested that up to 100 mg of a sample can be analysed in a time span of 2–3 h. MP particle size, shape, colour and related information are not relevant in this approach due to complete homogenization of the sample. First studies have shown a detection limit of around 0.5 up to 1 wt% in a 20 mg sample which seems to be relatively high. Furthermore, calibration for an exhaustive collection of relevant synthetic polymers is yet to be realized and proven.

5.6 Scanning Electron Microscopy Coupled with Energy Dispersive X-Ray Spectroscopy (SEM-EDX)

Scanning electron microscopy coupled with energy dispersive X-ray spectroscopy (SEM-EDX) can be used to obtain high-resolution data of the morphology and qualitative information about the chemical composition of samples such as MPs. SEM provides high-magnification, high-resolution images of the surface of the investigated samples [124]. SEM microscopes apply various detectors which generate images based on emission and detection of secondary electrons. Furthermore, the backscattered electron detector provides information on topography and material contrast based on atomic number (Z). SEM can be coupled to EDX where element-specific radiation is used for chemical characterization of the surface near volume. This approach provides elemental analysis on areas as small as nanometres in diameter. The electron beam incident on the sample produces X-rays that are characteristic of the elements found on the sample. The intensities yield quantitative information on the element composition and distribution. Hence, the approach can be used to obtain detailed information on the elemental composition of MPs, with information also on the inorganic additives present in the sample. Furthermore, intense signals from some elements such as nitrogen can be attributed to the presence of substances such as biomass on MPs. In general, SEM-EDX is a promising approach to obtain high-quality images of MPs in comparison to optical microscopy and to deduce their elemental composition subsequently. This information can help in the analysis of synthetic polymer-based materials and nonplastic materials. It can be used as a complementary technique to fluorescence microscopy for tracking MP localization in biota.

Limitations

SEM-EDX is not suitable for discrimination of different plastic polymers in environmental samples as EDX information is only based on elemental composition. In addition, it is a very expensive technique which requires tremendous time, effort and expertise for sample preparation and analysis. Moreover, samples must be sputtered with conducting materials such as Au, AuPd or carbon on nonconducting MP samples. Thereby, samples cannot be used for downstream analysis. However, working with low kV beam energy such as 0.5–1.5 kV may negate the need for sputtering of samples. This may allow speeding up of sample preparation and making the analysis much easier to perform.

5.7 Quantitative Nuclear Magnetic Resonance Spectroscopy (^{1}H-qNMR)

Quantitative nuclear magnetic resonance (^{1}H-qNMR) is a simple, rapid and nondestructive technique that allows simultaneous characterization and quantification of a multitude of molecules. It has been extensively used in various fields of research and development such as polymers, metabolites, pharmaceuticals, forensic sciences, environmental applications, etc. [125]. ^{1}H-qNMR provides access to a wealth of in-depth information about the structure and dynamics of complex molecules. This approach probes changes in the magnetic field experienced by the nuclei of each atom in a molecule when exposed to an external magnetic field. Nuclei with an odd mass or atomic number have a nuclear spin, this includes ^{1}H and ^{13}C, and the spins of their nuclei are sufficiently different to be probed by NMR measurements. Modern instruments are sensitive to local magnetic variations as small as one part in a billion, from which information about the molecular structure can be determined, and even the complex molecules like proteins can be effectively analysed with high accuracy [125]. Information from ^{1}H-qNMR is based on the proportional relationship of integrated signal area and number of resonant nuclei. Different types of determination methods are possible such as relative determination, absolute determination by use of internal and external standards, standard addition and calibration curve method. ^{1}H-qNMR can be applied for the size-independent qualitative and quantitative characterization and determination of MP particles from different matrices. Preliminary studies used for the analysis of MPs such as PE, PET and PS have shown that the calibration curve method is best suited for MP analysis, since the exact composition of all analytes other than polymers in an environmental sample need not be determined and this method is a preferred choice [125]. Therefore, ^{1}H-qNMR is a precise quantification method which can be applied to achieve a quantification accuracy of >98%. It is an effective and fast approach (about 1 min per sample measurement). The calibration curve method allows for a high-throughput analysis.

Limitations

There is the possibility of significant issues arising from environmental components such as microorganisms and other bioorganic components. The errors arising from these components are signal overlays that are not correctable, and systematic errors are complicated to be detected. Thereby, an effective sample preparation step is necessary to remove all these components before analysis. A major drawback of this approach is the need for dissolution of analytes in a suitable deuterated solvent leading to a loss of size information of the MP particles. Thereby, it is of utmost importance that a suitable solvent is identified for the dissolution of different types of synthetic polymers, and this is challenging due to the inherent chemical and physical properties of polymers in comparison to other organic substances. Furthermore, the solvent signal may overlay or coincide within the signal range of the respective synthetic polymers which can make the analysis highly complicated. ^{1}H-qNMR is a promising approach for the size-independent analysis of MPs; nevertheless, several parameters remain to be optimized in the context of analysis of MPs and are in a very preliminary stage of being applied for this purpose.

6 Conclusion

The chapter summarizes the currently applied methods of sampling, preparation and analysis of environmental MPs. Furthermore, advantages, disadvantages and limitations of these methods are highlighted and discussed in detail.

In general, MPs are heterogeneously distributed in the environment. Thus, replicate samples are needed for valid and reliable data on location and time-dependent MP abundances. Sampling should (ideally) generate data for modelling studies to understand the sources and fate of environmental MPs. The selection of sampling equipment must be given critical importance to cover a broad size range of MPs and to ensure an adequate sample volume for representative counts. Likewise, a stringently regulated, ethically complied and harmonized protocol must be implemented for the sampling of biota.

For sample preparation, the diversity of environmental matrices requires the use of adaptable and combinable processing techniques. Here, validation and recovery experiments as well as laboratory controls are mandatory to prevent underestimations or overestimations of MP abundances (e.g., particle loss, fragmentation, contamination). As the applied methods affect the detectable range of MPs (e.g. polymer type, size), only a thorough documentation allows for cross-study comparisons.

Identification of the plethora of environmental MPs is challenging as it encompasses diverse sizes, shapes and colours of aged polymers. Considering all these parameters, it is not possible to recommend a specific analytical method as the most suitable and efficient for MP identification and quantification. Moreover, every method has its unique advantages and disadvantages. For instance, visual identification is biased, and vibrational spectroscopy techniques such as FTIR and Raman spectroscopy strongly depend on the quality of the sample and the available references (e.g., spectra). Thus, a combination of several analytical approaches may be favourable in order to capture and characterize the diversity of environmental MPs.

Overall, there is currently a lack of harmonized and validated sampling and analytical methodologies due to non-standardized research associated with the analysis of MPs. Furthermore, studies report microplastic concentrations related to different units, size classes, synthetic polymer types, forms and shapes. This hampers comparability between studies from laboratories across the world. Thereby, it is of importance to harmonize the methodologies and to report critical parameters and limitations (e.g. mesh size, sample volume, MP size distribution). Standardization of the methodologies can eventually lead to addressing of key issues such as environmental risk assessment of MPs.

References

1. PlasticsEurope (2019) Plastics – the facts 2019*:* an analysis of European plastics production, demand and waste data
2. Carpenter EJ et al (1972) Polystyrene spherules in coastal waters. Science 178(4062):749–750
3. Carpenter EJ, Smith KL (1972) Plastics on the Sargasso Sea surface. Science 175 (4027):1240–1241
4. Hidalgo-Ruz V et al (2012) Microplastics in the marine environment: a review of the methods used for identification and quantification. Environ Sci Technol 46(6):3060–3075
5. Cole M et al (2011) Microplastics as contaminants in the marine environment: a review. Mar Pollut Bull 62(12):2588–2597
6. Bergmann M, Gutow L, Klages M (2015) Marine anthropogenic litter. Springer, Heidelberg, p 447
7. Eerkes-Medrano D, Thompson RC, Aldridge DC (2015) Microplastics in freshwater systems: a review of the emerging threats, identification of knowledge gaps and prioritisation of research needs. Water Res 75:63–82
8. Wagner M, Lambert S (2018) Barceló D, Kostianoy AG (eds) Freshwater microplastics: emerging environmental contaminants? The handbook of environmental chemistry, vol 58. Springer, Cham, p 309

9. Stock F et al (2019) Sampling techniques and preparation methods for microplastic analyses in the aquatic environment – a review. TrAC Trends Anal Chem 113:84–92
10. Prata JC et al (2019) Methods for sampling and detection of microplastics in water and sediment: a critical review. TrAC Trends Anal Chem 110:150–159
11. Priyanka GM, Udayashankara TH (2018) Methodology for sampling, purification, extraction and identification of microplastic – a review. Int J Sci Res 7(1):1006–1009
12. Käppler A et al (2018) Comparison of μ-ATR-FTIR spectroscopy and py-GCMS as identification tools for microplastic particles and fibres isolated from river sediments. Anal Bioanal Chem 410(21):5313–5327
13. Wang W, Wang J (2018) Investigation of microplastics in aquatic environments: an overview of the methods used, from field sampling to laboratory analysis. TrAC Trends Anal Chem 108:195–202
14. Mai L et al (2018) A review of methods for measuring microplastics in aquatic environments. Environ Sci Pollut Res 25(12):11319–11332
15. Löder MGJ, Gerdts G (2015) Methodology used for the detection and identification of microplastics – a critical appraisal. Marine Anthropogenic Litter. pp 201–227
16. Wright SL, Thompson RC, Galloway TS (2013) The physical impacts of microplastics on marine organisms: a review. Environ Pollut 178:483–492
17. Blettler M et al (2018) Freshwater plastic pollution: recognizing research biases and identifying knowledge gaps. Water Res 143:416–424
18. Horton AA et al (2017) Large microplastic particles in sediments of tributaries of the river Thames, UK – abundance, sources and methods for effective quantification. Mar Pollut Bull 114(1):218–226
19. Triebskorn R et al (2019) Relevance of nano- and microplastics for freshwater ecosystems: a critical review. TrAC Trends Anal Chem 110:375–392
20. Li J, Liu H, Paul Chen J (2018) Microplastics in freshwater systems: a review on occurrence, environmental effects, and methods for microplastics detection. Water Res 137:362–374
21. Möller JN, Löder MGJ, Laforsch C (2020) Finding microplastics in soils: a review of analytical methods. Environ Sci Technol 54(4):2078–2090
22. He D et al (2018) Microplastics in soils: analytical methods, pollution characteristics and ecological risks. TrAC Trends Anal Chem
23. Anbumani S, Kakkar P (2018) Ecotoxicological effects of microplastics on biota: a review. Environ Sci Pollut Res 25(15):14373–14396
24. Hermsen E et al (2018) Quality criteria for the Analysis of microplastic in biota samples: a critical review. Environ Sci Technol 52(18):10230–10240
25. Wesch C et al (2016) Towards the suitable monitoring of ingestion of microplastics by marine biota: a review. Environ Pollut 218:1200–1208
26. ISO/CD 24187. Principles for the development of standards for investigation procedures of plastics in environmental matrices and related materials
27. González-Fernández D, Hanke G (2017) Toward a harmonized approach for monitoring of riverine floating macro litter inputs to the marine environment. Front Mar Sci 4
28. Frias J et al (2018) Standardised protocol for monitoring microplastics in sediments. JPI Oceans BASEMAN project
29. OSPAR (2014) Regional action plan for prevention and Management of Marine Litter in the north-East Atlantic
30. Hartmann NB et al (2019) Are we speaking the same language? Recommendations for a definition and categorization framework for plastic debris. Environ Sci Technol 53 (3):1039–1047
31. Sun J et al (2019) Microplastics in wastewater treatment plants: detection, occurrence and removal. Water Res 152:21–37
32. Hu Y et al (2019) Current research trends on microplastic pollution from wastewater systems: a critical review. Rev Environ Sci Biotechnol 18(2):207–230

33. Windsor FM et al (2019) A catchment-scale perspective of plastic pollution. Glob Chang Biol, vol 25, p 1207
34. Schmidt C, Krauth T, Wagner S (2017) Export of plastic debris by Rivers into the sea. Environ Sci Technol 51(21):12246–12253
35. Klein S et al (2018) Wagner M, Lambert S (eds) Analysis, occurrence, and degradation of microplastics in the aqueous environment, in freshwater microplastics : emerging environmental contaminants? Springer, Cham, pp 51–67
36. Dris R et al (2015) Microplastic contamination in an urban area: a case study in Greater Paris. Environ Chem 12(5):592–599
37. Lenz R, Labrenz M (2018) Small microplastic sampling in water: development of an encapsulated filtration device. Water 10:1055
38. Mintenig SM et al (2017) Identification of microplastic in effluents of waste water treatment plants using focal plane array-based micro-Fourier-transform infrared imaging. Water Res 108:365–372
39. Klinger J (2018) DVGW-Technologiezentrum Wasser. Sonderdruck gwf Gas und Engergie 06/2018 und gwf Wasser/Abwasser 06/2018. 159:43–51
40. Urgert W (2015) Microplastics in the rivers Meuse and Rhine – developing guidance for a possible future monitoring program. Master's thesis Open University of the Netherlands, Heerlen
41. Conn KE et al (2016) Continuous-flow centrifugation to collect suspended sediment for chemical analysis. In: U.S. Geological Survey Techniques and Methods, book 1, chap. D6, 31 p., plus appendixes. US Geological Survey
42. Hildebrandt L et al (2019) Evaluation of continuous flow centrifugation as an alternative technique to sample microplastic from water bodies. Mar Environ Res:104768
43. Pinto da Costa J et al (2019) Microplastics in soils: assessment, analytics and risks. Environ Chem 16(1)
44. MSFD (2013) Technical subgroup on marine litter, guidance on monitoring of marine litter in European Seas – a guidance document within the Common Implementation Strategy for the Marine Strategy Framework Directive, in JRC Scientific and Policy Reports
45. Besley A et al (2017) A standardized method for sampling and extraction methods for quantifying microplastics in beach sand. Mar Pollut Bull 114(1):77–83
46. Allen S et al (2019) Atmospheric transport and deposition of microplastics in a remote mountain catchment. Nat Geosci 12(5):339–344
47. Dris R et al (2016) Synthetic fibres in atmospheric fallout: a source of microplastics in the environment? Mar Pollut Bull 104(1–2):290–293
48. Dris R *First assessment of sources and fate of macro- and micro- plastics in urban hydrosystems: Case of Paris megacity*, in *LEESU – Laboratoire Eau Environnement et Systèmes Urbains* 2016. Environ Sci
49. Vianello A et al (2019) Simulating human exposure to indoor airborne microplastics using a breathing thermal manikin. Sci Rep 9(1):8670
50. John AC et al (1999) Entwicklung eines PM 10/PM 2.5-Kaskadenimpaktors zur Messung der Emission von Feinstäuben. Gefahrstoffe – Reinhalt. Luft, 59(11/12):449–454
51. DIN (2014) Ambient air – standard gravimetric measurement method for the determination of the PM_{10} or PM2,5 mass concentration of suspended particulate matter. In: DIN (ed) DIN EN 12341: 2014-08. Beuth Verlag, Berlin
52. VDI (2004) Messen von Partikeln; Staubmessung in strömenden Gasen; Messung der Emissionen von PM10 und PM2,5 an geführten Quellen nach dem Impaktionsverfahren (Particulate matter measurement; Dust measurement in flowing gases; Measurement of PM10 and PM2,5 emissions at stationary sources by impaction method. In: VDI (ed) 2066 Blatt 10. Beuth Verlag, Berlin
53. VDI (2013–2012) Measurement of indoor air pollution – measurement strategies for determination of airborne particles in indoor environment – PM<(Index)2,5>−fraction. In: VDI (ed) 4300 Blatt 11:2013-2012, Beuth Verlag, Berlin

54. Scherer C et al (in review) Microplastics in the water and sediment phase of the Elbe river, Germany. Sci Total Environ
55. Dris R et al (2017) A first overview of textile fibres, including microplastics, in indoor and outdoor environments. Environ Pollut 221:453–458
56. Stanton T et al (2019) Freshwater and airborne textile fibre populations are dominated by 'natural', not microplastic, fibres. Sci Total Environ 666:377–389
57. Prata JC et al (2019) Effects of microplastics on microalgae populations: a critical review. Sci Total Environ 665:400–405
58. Foekema EM et al (2013) Plastic in North Sea fish. Environ Sci Technol:130711150255009
59. Setälä O, Norkko J, Lehtiniemi M (2016) Feeding type affects microplastic ingestion in a coastal invertebrate community. Mar Pollut Bull 102(1):95–101
60. Bravo Rebolledo EL et al (2013) Plastic ingestion by harbour seals (*Phoca vitulina*) in the Netherlands. Mar Pollut Bull 67(1–2):200–202
61. Besseling E et al (2015) Microplastic in a macro filter feeder: humpback whale *Megaptera novaeangliae*. Mar Pollut Bull 95(1):248–252
62. Nelms SE et al (2018) Investigating microplastic trophic transfer in marine top predators. Environ Pollut 238:999–1007
63. Wright SL et al (2013) Microplastic ingestion decreases energy reserves in marine worms. Curr Biol 23(23):R1031–R1033
64. Bond AL, Lavers JL (2013) Effectiveness of emetics to study plastic ingestion by Leach's storm-petrels (*Oceanodroma leucorhoa*). Mar Pollut Bull 70(1):171–175
65. O'Connor JD et al (2019) Microplastics in freshwater biota: a critical review of isolation, characterization, and assessment methods. Global Chall:1800118
66. Guo X, Wang J (2019) The chemical behaviors of microplastics in marine environment: a review. Mar Pollut Bull 142:1–14
67. Imhof HK et al (2012) A novel, highly efficient method for the separation and quantification of plastic particles in sediments of aquatic environments. Limnol Oceanogr Methods 10:524–537
68. Nuelle MT et al (2014) A new analytical approach for monitoring microplastics in marine sediments. Environ Pollut 184:161–169
69. Coppock RL et al (2017) A small-scale, portable method for extracting microplastics from marine sediments. Environ Pollut 230:829–837
70. Claessens M et al (2013) New techniques for the detection of microplastics in sediments and field collected organisms. Mar Pollut Bull 70(1–2):227–233
71. Quinn B, Murphy F, Ewins C (2017) Validation of density separation for the rapid recovery of microplastics from sediment. Anal Methods 9(9):1491–1498
72. Pagter E, Frias J, Nash R (2018) Microplastics in Galway Bay: a comparison of sampling and separation methods. Mar Pollut Bull 135:932–940
73. Corcoran PL, Biesinger MC, Grifi M (2009) Plastics and beaches: a degrading relationship. Mar Pollut Bull 58(1):80–84
74. Martin J et al (2017) The deposition and accumulation of microplastics in marine sediments and bottom water from the Irish continental shelf. Sci Rep 7(1):10772
75. Zhang K et al (2016) Microplastic pollution of lakeshore sediments from remote lakes in Tibet plateau, China. Environ Pollut 219:450–455
76. Bauer M et al (2018) Sink-float density separation of post-consumer plastics for feedstock recycling. J Mater Cy Waste Manag 20(3):1781–1791
77. Crichton EM et al (2017) A novel, density-independent and FTIR-compatible approach for the rapid extraction of microplastics from aquatic sediments. Anal Methods 9(9):1419–1428
78. Köhnlechner R, Sander S (2009) Praktischer Einsatz elektrostatischer Separatoren in der Sekundärrohstoffindustrie. Berg Hüttenmännische Monatshefte 154:136–137
79. Felsing S et al (2018) A new approach in separating microplastics from environmental samples based on their electrostatic behavior. Environ Pollut 234:20–28
80. Hurley RR et al (2018) Validation of a method for extracting microplastics from complex, organic-rich, environmental matrices. Environ Sci Technol 52(13):7409–7417

81. Dehaut A et al (2016) Microplastics in seafood: benchmark protocol for their extraction and characterization. Environ Pollut 215:223–233
82. Karami A et al (2017) A high-performance protocol for extraction of microplastics in fish. Sci Total Environ 578:485–494
83. Lusher AL, McHugh M, Thompson RC (2013) Occurrence of microplastics in the gastrointestinal tract of pelagic and demersal fish from the English Channel. Mar Pollut Bull 67 (1–2):94–99
84. Löder MGJ et al (2017) Enzymatic purification of microplastics in environmental samples. Environ Sci Technol 51(24):14283–14292
85. Kuhn S et al (2017) The use of potassium hydroxide (KOH) solution as a suitable approach to isolate plastics ingested by marine organisms. Mar Pollut Bull 115(1–2):86–90
86. Avio CG, Gorbi S, Regoli F (2015) Experimental development of a new protocol for extraction and characterization of microplastics in fish tissues: first observations in commercial species from Adriatic Sea. Mar Environ Res 111:18–26
87. Catarino AI et al (2017) Development and optimization of a standard method for extraction of microplastics in mussels by enzyme digestion of soft tissues. Environ Toxicol Chem 36 (4):947–951
88. Köhnlechner R (2000) Triboelectric charging and electrostatic separation of diverse non-conductive mixed waste especially plastic employs Enclosed Vibro-conveyor followed by in-flight separation influenced by non-linear electrostatic field. Patent DEI
89. Woodall LC et al (2015) Using a forensic science approach to minimize environmental contamination and to identify microfibres in marine sediments. Mar Pollut Bull 95(1):40–46
90. Eriksen M et al (2013) Plastic pollution in the South Pacific subtropical gyre. Mar Pollut Bull 68(1–2):71–76
91. Lattin GL et al (2004) A comparison of neustonic plastic and zooplankton at different depths near the southern California shore. Mar Pollut Bull 49(4):291–294
92. Desforges JP et al (2014) Widespread distribution of microplastics in subsurface seawater in the NE Pacific Ocean. Mar Pollut Bull 79(1–2):94–99
93. Goldstein MC, Rosenberg M, Cheng L (2012) Increased oceanic microplastic debris enhances oviposition in an endemic pelagic insect. Biol Lett 8(5):817–820
94. Lenz R et al (2015) A critical assessment of visual identification of marine microplastic using Raman spectroscopy for analysis improvement. Mar Pollut Bull 100(1):82–91
95. Cole M et al (2013) Microplastic ingestion by Zooplankton. Environ Sci Technol:130606145528005
96. Norén F (2007) Small plastic particles in Coastal Swedish waters. Report by N-research on behalf of KIMO Sweden, pp 1–11
97. Lavers JL, Oppel S, Bond AL (2016) Factors influencing the detection of beach plastic debris. Mar Environ Res 119(Supplement C):245–251
98. Vianello A et al (2013) Microplastic particles in sediments of lagoon of Venice, Italy: first observations on occurrence, spatial patterns and identification. Estuar Coast Shelf Sci 130 (3):54–61
99. Dekiff JH et al (2014) Occurrence and spatial distribution of microplastics in sediments from Norderney. Environ Pollut 186
100. Löder MGJ et al (2015) Focal plane array detector-based micro-Fourier-transform infrared imaging for the analysis of microplastics in environmental samples. Environ Chem 12(5):563
101. Harrison JP, Ojeda JJ, Romero-González ME (2012) The applicability of reflectance micro-Fourier-transform infrared spectroscopy for the detection of synthetic microplastics in marine sediments. Sci Total Environ 416(C):455–463
102. Primpke S et al (2017) An automated approach for microplastics analysis using focal plane array (FPA) FTIR microscopy and image analysis. Anal Methods 9(9):1499–1511
103. Hufnagl B et al (2019) A methodology for the fast identification and monitoring of microplastics in environmental samples using random decision forest classifiers. Anal Methods 11(17):2277–2285

104. Kuhar N et al (2018) Challenges in application of Raman spectroscopy to biology and materials. RSC Adv 8:25888–25908
105. McNesby KL, Pesce-Rodriquez RA (2006) Applications of vibrational spectroscopy in the study of explosives. In: Chalmers JM, Griffiths PR (eds) Handbook of vibrational spectroscopy. John Wiley & Sons, Ltd, Chichester
106. Stöckel S et al (2016) The application of Raman spectroscopy for the detection and identification of microorganisms. J Raman Spectrosc 47(1):89–109
107. Araujo CF et al (2018) Identification of microplastics using Raman spectroscopy: latest developments and future prospects. Water Res 142:426–440
108. Krafft C, Dietzek B, Popp J (2009) Raman and CARS spectroscopy of cells and tissues. Analyst 134:1046–1057
109. Matousek P et al (2005) Subsurface probing in diffusely scattering media using spatially offset Raman spectroscopy. Appl Spectrosc 59:393–400
110. Djaker N et al (2007) Coherent anti-stokes Raman scattering microscopy (CARS): instrumentation and applications. Nucl Instrum Methods Phys Res, Sect A 571(1):177–181
111. Li H et al (2018) A practical wide-field Raman imaging method with high spectral and spatial resolution. Rev Sci Instrum 89:083103
112. Shreve AP, Cherepy NJ, Mathies RA (1992) Effective rejection of fluorescence interference in Raman spectroscopy using a shifted excitation difference technique. Appl Spectrosc 46 (4):707–711
113. Thaysen C et al (2020) EXPRESS: toward Raman automation for microplastics: developing strategies for particle adhesion and filter subsampling. Appl Spectrosc:000370282092290
114. Cabernard L et al (2018) Comparison of Raman and Fourier transform infrared spectroscopy for the quantification of microplastics in the aquatic environment. Environ Sci Technol 52
115. Fries E et al (2013) Identification of polymer types and additives in marine microplastic particles using pyrolysis-GC/MS and scanning electron microscopy. Environ Sci: Processes Impacts 15
116. Hermabessiere L et al (2018) Optimization, performance and application of a pyrolysis-GC/MS method for the identification of microplastics. Anal Bioanal Chem 410
117. Fischer M, Scholz-Böttcher BM (2017) Simultaneous trace identification and quantification of common types of microplastics in environmental samples by pyrolysis-gas chromatography–mass spectrometry. Environ Sci Technol 51(9):5052–5060
118. Ter Halle A et al (2017) Nanoplastic in the North Atlantic Subtropical Gyre. Environ Sci Technol 51
119. Dierkes G et al (2019) Quantification of microplastics in environmental samples via pressurized liquid extraction and pyrolysis-gas chromatography. Anal Bioanal Chem 411 (26):6959–6968
120. David J et al (2019) Introducing a soil universal model method (SUMM) and its application for qualitative and quantitative determination of poly(ethylene), poly(styrene), poly(vinyl chloride) and poly(ethylene terephthalate) microplastics in a model soil. Chemosphere 225:810–819
121. Dümichen E et al (2017) Fast identification of microplastics in complex environmental samples by a thermal degradation method. Chemosphere 174:572–584
122. Dümichen E et al (2015) Analysis of polyethylene microplastics in environmental samples, using a thermal decomposition method. Water Res 85:451–457
123. Duemichen E et al (2014) Assessment of a new method for the analysis of decomposition gases of polymers by a combining thermogravimetric solid-phase extraction and thermal desorption gas chromatography mass spectrometry. J Chromatogr A 1354:117–128
124. Gniadek M, Dąbrowska A (2019) The marine nano-and microplastics characterisation by SEM-EDX: the potential of the method in comparison with various physical and chemical approaches. Mar Pollut Bull 148:210
125. Peez N et al (2019) Quantitative analysis of PET microplastics in environmental model samples using quantitative 1H-NMR spectroscopy: validation of an optimized and consistent sample clean-up method. Anal Bioanal Chem 411:7409–7418

Analytical Methods for Plastic (Microplastic) Determination in Environmental Samples

G. Dierkes, T. Lauschke, and C. Földi

Contents

1 Introduction 44
2 Description of Different Methods 45
  2.1 Spectroscopic Methods 45
  2.2 Thermo-Analytical Methods 50
  2.3 Other Methods 56
3 Discussion 57
4 Conclusion 63
References 63

Abstract Beside several studies about the occurrence of microplastic (MP) there is still a huge gap of knowledge regarding the dynamic processes of MP distribution and fate. Consequently, there is a need for reliable, fast, and robust analytical methods for MP monitoring. However, due to the physicochemical attributes of plastic, new analytical approaches fundamentally different from those for most other environmental contaminants are required. Promising strategies include spectroscopic and thermo-analytical methods. The two vibrational spectroscopic methods, Fourier-transform infrared spectroscopy (FT-IR) and Raman spectroscopy, have been implemented for MP detection. Especially in combination with particle finding software or a focal plane array (FPA) detector, they enable reliable determination of MP particle numbers in environmental samples. In recent years, different thermo-analytical techniques, such as pyrolysis (Py), thermogravimetric analysis (TGA), and differential scanning calorimetry (DSC) have been adapted for MP detection. All thermo-analytical methods are based upon measurement of physical or chemical changes of the polymer under thermal treatment. While DSC measures differences in

---

G. Dierkes (✉), T. Lauschke, and C. Földi
German Federal Institute of Hydrology, Koblenz, Germany
e-mail: Dierkes@bafg.de; Lauschke@bafg.de; Foeldi@bafg.de

Friederike Stock, Georg Reifferscheid, Nicole Brennholt, and Evgeniia Kostianaia (eds.),
Plastics in the Aquatic Environment - Part I: Current Status and Challenges,
Hdb Env Chem (2022) 111: 43–68, DOI 10.1007/698_2021_744,

© Springer Nature Switzerland AG 2021, Published online: 30 March 2021

heat flux caused by phase transitions of the polymer, TGA-MS is based upon detection of specific thermal degradation products. By means of a gas chromatographic separation step, an enhanced detection of the marker compounds is possible, enabling a more sensitive MP detection even in complex matrices. The extent of analytical information obtained as well as the complexity and effort of the methods increase by TGA-DSC < TGA-MS < Py-GC-MS/TED-GC-MS. The results are comparable to those of spectroscopic methods (FT-IR, Raman), but both techniques have different benefits and limitations. While thermo-analytical methods require minor sample pretreatment and reveal mass concentrations, spectroscopic methods are non-destructive and yield particle numbers and size distribution by imaging techniques. Whichever is the most suitable method depends on the scientific question and what kind of information is required.

Keywords Microplastic analysis, Spectroscopy, Thermo-analytical methods

1 Introduction

Coming along with the emerging awareness of microplastics (MP) as a meaningful contaminant in aquatic systems, there is a need for reliable, fast, and robust analytical methods for MP monitoring [1]. However, due to the high molecular masses of the polymers, they are not suitable for direct analysis by standard techniques in environmental science such as liquid chromatography (LC) or gas chromatography (GC) coupled to mass spectrometry (MS). The physicochemical attributes of plastics ask for new analytical approaches fundamentally different from those for most other environmental contaminants. In order to solve this issue, intensive scientific development work was done worldwide during the last decade [2–9]. Different strategies were developed, among which thermo-analytical methods and spectroscopic methods are the most promising.

Synthetic polymers consist of one or more types of small organic molecules, the so-called monomers, linked by covalent bonds. During different kinds of polymerization reactions, the monomers are polymerized to macromolecules with molecular weights of several thousand g/mol. The particular polymer types vary in composition and arrangement of a large number of available monomers. For the characterization of a polymer, the building monomers have to be identified and their order in the molecular chain has to be elucidated. Beside spectroscopic methods, thermo-analytical methods are standard applications for the characterization of synthetic polymers in polymer science [10–12].

Microscopy coupled with vibrational spectroscopy for chemical characterization enables visualization and identification of small particles such as MP. Identification is based upon comparison of recorded spectra to those in spectra libraries. Sample imaging techniques can provide morphological parameters such as particle size, shape, and color. By scanning of a representative sample surface, MP particle numbers and a rough estimation of MP mass can be determined.

In recent years, different thermo-analytical techniques (pyrolysis, thermogravimetry, and differential scanning calorimetry) have been adapted for MP detection [3, 9, 13]. These methods can be applied for qualitative and quantitative MP detection in environmental samples. Therefore, thermo-analytical methods are a good alternative or complement to the widely used spectroscopic methods [14]. Today there is no harmonization of the existing analytical methods and only a few comparative studies exist [15, 16]. Consequently, comparison between results of different studies is limited.

In the following sections, the different techniques for the analytical investigation of MP are introduced and benefits and challenges are discussed. Furthermore, a comparison of the benefits and limitations of the different methods is drawn.

2 Description of Different Methods

In contrast to other contaminants, MP do not occur dissolved or sorbed to solids in the environment, but as discrete particles visual for the naked eye or by microscopy. This may lead to the misjudgment that MP determination could be done by visual sorting and counting. However, optical microscopy is not suitable for reliable identification of MP in environmental samples. Classification especially of small particles (<100 μm) by properties such as color, shape, and hardness is prone to errors and leads to false estimation of MP burden [17, 18]. Accuracy can be improved by use of staining with fluorescence dyes such as Nile Red [19–21]. Nile red adsorbs to lipophilic surfaces like those of MP, while hydrophilic and inorganic surfaces are not stained [22]. Under a UV-lamp or a fluorescence microscope MP can be identified by fluorescence. For distinct results and identification of the polymer types, a chemical characterization of the MP particles is necessary [23]. This can be realized by spectroscopic or thermo-analytical methods.

2.1 Spectroscopic Methods

FT-IR and Raman spectroscopy are widespread used methods for MP identification. Both spectroscopic methods are based upon excitation of intramolecular vibrations by electromagnetic radiation. In FT-IR-spectroscopy, the absorption of IR light (wavenumbers between 400 and 4,000 cm^{-1}) is measured. Positions and intensities of the absorption bands depend on the resonance frequencies of vibrations and the magnitude change of the atomic bond's permanent dipole moment. In contrast, Raman spectroscopy is based on measurement of a shift in the wavelength of a scattered laser beam. Interaction of the photons and the sample molecules is related to a change in the polarizability of chemical bonds. Due to these fundamental

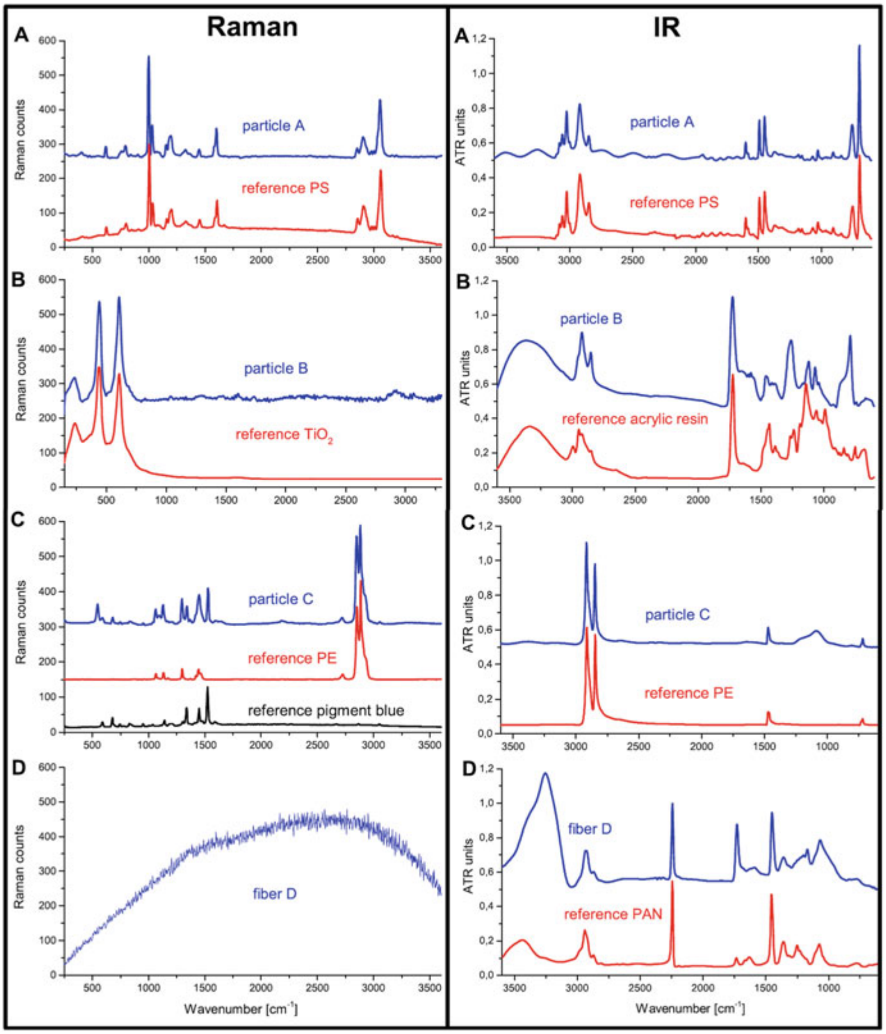


Fig. 1 Raman spectra (left) and ATR-FTIR spectra (right) of MP (blue) each in comparison with a reference (red) (Reprinted by permission from Springer Nature, Anal Bioanal Chem, Käppler et al. [24], 2016)

differences in the physical principles of the absorption processes, both spectroscopic methods are complementary, since some kinds of vibrations are IR active while Raman inactive and vice versa (Fig. 1). In both cases, the resulting spectra consist of characteristic bands for the functional groups in the molecules and are like a chemical fingerprint of the substances. Thus, characterization of the chemical structure and identification of polymers by comparison with reference spectra is possible [24]. By employing spectra libraries and matching software identification can be done by automated routines. The success of matching depends upon the comprehensiveness of the library and the matching algorithms of the software.

For MP identification in environmental samples, a division into two size fractions is recommended: (1) particles >300 μm which can be handled manually and (2) particles <300 μm which are too small for manual picking. For identification of the larger particles, FT-IR in attenuated total reflection (ATR) mode is the best choice [25–27]. The isolated particles are put upon a crystal probe and IR spectra from the near surface region of the sample are recorded. Due to the direct contact between sample and crystal and the poor penetration depth, strongly contaminated particle surfaces may lead to cross contamination and adulterated spectra. Therefore, MP isolated from environmental samples may need cleaning of the polymer surface by acids, bases, or oxidants such as H_2O_2 to remove biofilms or other contaminants. Advanced weathering of polymers leads to chemical modifications of the surface and consequently to an alteration of the IR spectra hampering the identification. Consequently, databases of pristine polymers have to be complemented by spectra of weathered polymers [3].

For analysis of the <300 μm fraction, the particles have to be isolated from the matrix (e.g., by density separation) and purified by chemical or enzymatic digestion of the organic matter (see Stock et al. [28]). Subsequently, they are concentrated onto filters. Coupling a vibrational spectrometer to a microscope enables direct identification of single particles on filters [26, 29]. The filter choice depends on the requirements of the used spectroscopic method. If FT-IR is used in transmission mode, the filters have to be translucent for IR radiation, such as aluminum oxide, silicon, or metal covered polycarbonate [30–32]. For Raman spectroscopy, filters that do not induce background fluorescence are required, such as quartz fiber, polytetrafluoroethylene (PTFE), or silicon filters [31, 33, 34]. The minimum detectable particle size is 10 μm for FT-IR, whereas Raman microscopy can detect particles down to 1 μm. In environmental samples MP are highly outnumbered by natural particles (e.g., clay minerals or organic substances). Such contaminants can hamper the detection of MP by agglomeration and alteration of spectra or strong fluorescence. Therefore, decrease of natural particles and organic contamination by extensive sample pretreatment protocols is essential (see Stock et al. [28]).

There are three different options for particle identification: (1) manual selection of single particles, (2) automated selection by particle finder approaches, and (3) chemical imaging of the filter surface. The manual step-by-step analysis of all particles is very time-consuming. Hence, not the whole filter area is analyzed, but often only less than 10%. The accuracy of the extrapolation depends on the representativeness of the chosen measurement area [3]. More efficient are approaches using particle finding software [33], where high-resolution dark-field images of the whole filter are scanned for particles and a map of all particles is created. Spectra are collected only at those dedicated points [34, 35]. Since numbers of particles in complex environmental samples are very high, particles may be not well separated and agglomerates of small particles may be mistaken for larger particles. Furthermore, analysis of all particles is not possible in a reasonable time. Hence, a diluted sample aliquot or a randomly chosen subset representative for all particles has to be analyzed. The minimal needed number of particles depends on the total number of particles, the ratio of MP to natural particles, and the accepted analysis error. Using a method called "simple random sample of units selected without replacement," Anger et al. [33] calculated a minimum of particle measurements of approx. 6,000 assuming a total particle number of 10^6 containing 0.5% MP and with an accepted error of 20%. Assuming typical measurement times of Raman setups capable of measuring particles between 1 and 5 μm in the range of 10 s per particle, this leads to a total measuring time of approx. 17 h per sample. To reduce the number of the needed particle measurements, Erni-Cassola et al. [19] used the fluorescent dye Nile red for selective staining of MP particles. By fluorescence imaging, the stained particles are mapped and are exclusively considered for chemical identification via spectroscopy. Another approach to reduce measuring time is the use of an IR laser instead of a classical light source in FT-IR. In the so-called Laser Direct Infrared (LDIR) chemical imaging method, very short measuring times of 1 s per spectrum can be realized by focusing a bright laser source onto the particles. The fast spectra acquisition in combination with optical particle finding enables identification of a huge number of particles scattered over a wide area in comparatively short time. Unfortunately, spectral range is limited to 975 to 1,800 cm^{-1} which could be a handicap in polymer identification.

Another option is the automatic chemical imaging of the full area by focal plane array (FPA) detectors [30, 36–38] or Raman imaging [24, 35]. FPA detectors are capable of recording several thousand spectra simultaneously while Raman imaging is realized by time-consuming point-by-point analysis. MP particles are detected by selected bands such as carbon–hydrogen stretch vibration (2,980–2,780 cm^{-1}) in FT-IR. Detected particles are counted and identified automatically by comparison of full spectra. By measuring the full filter area, the bias resulting from extrapolation is circumvented. However, this advantage comes at the costs of long measuring times especially in Raman spectrometry and huge data amounts. Spectral correlations of millions of spectra lead to data analysis times of 4–48 h depending on the used software and hardware [39].

By measuring the dimensions of the identified MP particles, a rough estimation of the mass is possible [38]. More suitable for determination of mass concentrations of MP are near-infrared (NIR) and FT-IR spectroscopic methods in combination with chemometrics [40–42]. By measuring bulk samples mixed spectra of all components are generated. Using multivariate calibration models spectral information about the different polymers can be isolated and used for quantification. So, MP at levels above 0.5 to 1.0 g/100 g depending on the polymer can be detected and classified very rapidly in bulk samples without any chemical pretreatment. Limits of detection are rather high, and applicability of the chemometric model for other matrices and particle sizes than those used for the calibration has yet to be tested. Furthermore, these bulk methods deliver no information about particle sizes and shapes.

Peez, Janiska [43] developed a nuclear magnetic resonance spectroscopy (^{1}H NMR) method for quantitative MP detection. After dissolving the particular polymers in an appropriate solvent, ^{1}H NMR spectra of the polymers are recorded. Quantification is based upon integration of signals showing a polymer specific

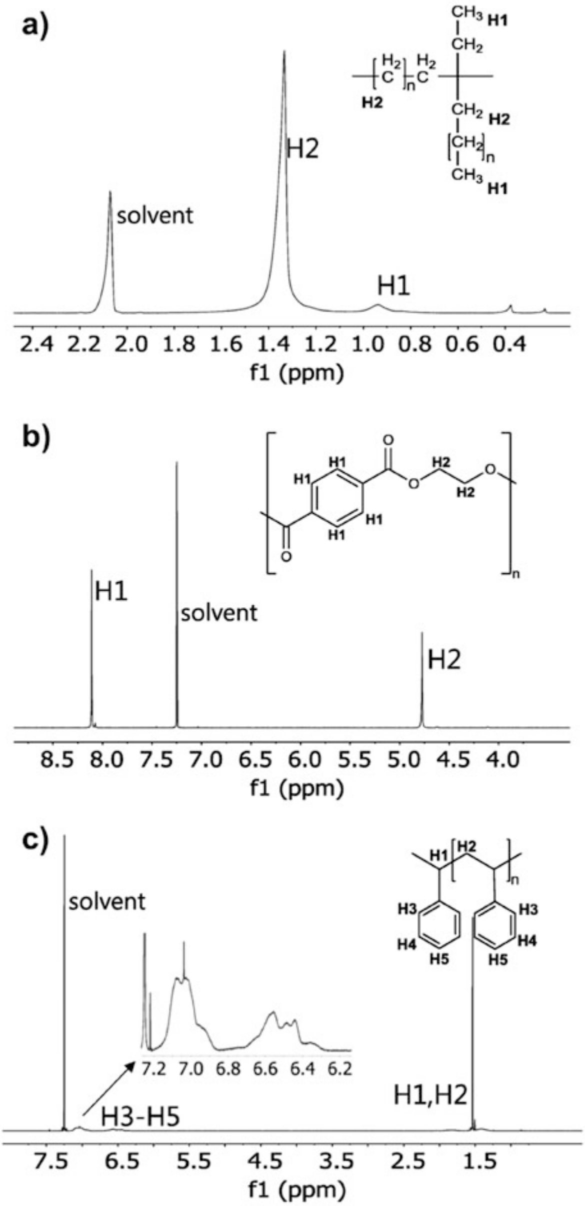


Fig. 2 1H NMR spectra of MP particles and their structural formulas. (**a**) PE granule measured in toluene-d8 at 60°C. (**b**) PET fibers measured in CDCl3/ TFA 4:1 at 25°C. (**c**) PS beads measured in CDCl3 at 25°C. In addition, the enlarged range of 7.20–6.20 ppm is shown (Reprinted by permission from Springer Nature, Anal Bioanal Chem, Peez et al. [43], 2018)

chemical shift (ref. Fig. 2). Limits of detection are in the range of 20–84 μg/mL [43, 44]. Applicability for environmental samples was shown for PET [45]. After chemical digestion, to reduce organic matrix compounds and dissolution in $CDCl_3$ selective determination of PET was realized in water, biofilms, and invertebrates.

The disadvantage of this method is the need of specific solvents for the different polymers and due to the dissolution step information about particle size gets lost.

2.2 Thermo-Analytical Methods

All thermo-analytical methods are based upon measuring physical or chemical changes of the polymer under thermal treatment. At elevated temperatures, polymers undergo phase transitions such as changes in crystal structure and melting. These processes can be observed by differential scanning calorimetry (DSC). During DSC analysis, the difference in heat flow between the sample and a reference is measured over a controlled temperature gradient using a defined heating rate. The result of a DSC experiment is a curve of heat flux versus temperature containing peaks at substance's specific temperatures. These peaks represent the temperature of thermal transitions such as the glass transition temperature, crystallization temperature, and melting temperature. Majewsky et al. [46] determined characteristic endothermic phase transition temperatures for seven polymer types using thermogravimetric analysis (TGA) coupled to DSC (Fig. 3). By integration of the associated peaks, a qualitative and quantitative analysis method for PE and PP was developed. The method was applied for analysis of wastewater samples. However, due to overlapping transition temperatures, the method is not reliable for other polymers [46]. Furthermore, transition temperatures are highly affected by product specific attributes such as fillers, additives, length and branching of the polymer chain. Even

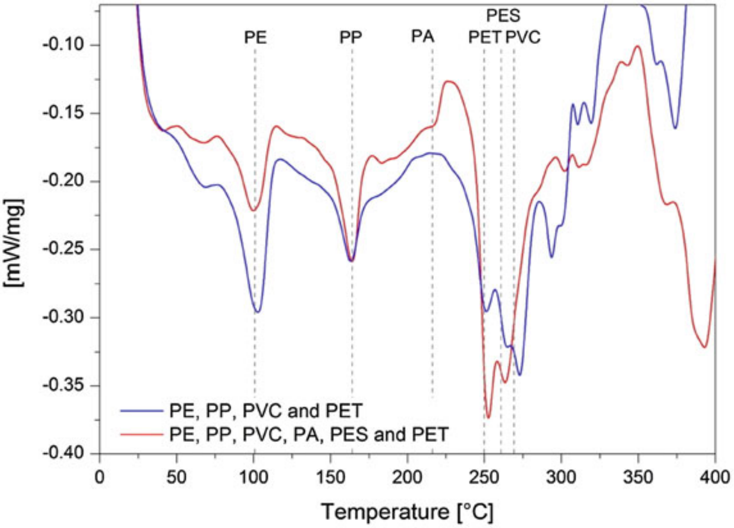


Fig. 3 DSC signals of a mixture of pure polymers. PE and PP show discrete peaks, while the other signals overlap (Reprinted by permission from Elsevier, Sci total Environ, Majewsky et al. [46], 2016)

particle size is a critical parameter, since larger particles can cause interferences by lagged response in terms of an unfavorable mass to surface ratio [3, 47].

Temperatures higher than the decompositions temperature of the polymers lead to fragmentation of the macromolecules and generation of smaller volatile molecules. The resulting mass reduction as a function of the temperature can be monitored by a thermobalance. David et al. [48] developed a thermogravimetric method for MP quantification in soils without sample pre-separations or pre-treatments. The method is based upon mass losses at selected temperatures specific for the particular polymers. To compensate interfering mass losses by the soil matrix they used the so-called soil universal model which was calculated from previously determined relationships between typically observed mass losses and soil properties. The method revealed quantification of PET, PS, and PVC with limits of detection in the range of 0.3–2.2 wt%. For PE determination the method was not suitable, due to overlapping temperature ranges of PE and soil organic matter degradation [48]. Altogether, the method does not enable specific MP detection, and applicability for different kinds of matrices has yet to be tested. More specific detection of MP can be realized by analyzing specific degradation products.

Decomposition of polymers at elevated temperatures is a complex process of different types of reactions, the most common ones being eliminations and rearrangements [49]. Consequently, there is not only one decomposition product, but a pattern of several products. Under standardized conditions (time, temperature, atmosphere), the decomposition process is reproducible and a characteristic product pattern can be obtained for the particular polymers [50, 51]. The fragments of the initial compound are volatile and can be analyzed in the gas phase. FT-IR [52] and mass spectrometry (MS) [53] are the most frequently used detection techniques for analysis of decomposition gases in the so-called evolved gas analysis (EGA-FTIR, EGA-MS) [49]. The fragment pattern is like a fingerprint of the different polymers and can be used for identification, especially in combination with TGA [52–54]. David et al. [54] developed a method for the qualitative and quantitative determination of polyethylene terephthalate (PET) using specific ion fragments of decomposition products resulting from PET (vinyl benzene and benzoic acid). In Fig. 4 typical output for determination of PET in soil is displayed. At the degradation temperature of PET (300–650°C) a clear signal for the PET decomposition compound vinyl benzene can be observed. However, due to interferences by matrix compounds, detection limits of the method are rather high (600 μg/g).

Unfortunately, in most cases the EGA is dominated by small unspecific molecules like H_2O, CO_2, and NH_3, hampering the identification of more complex and specific decomposition products. This disadvantage can be evaded by an additional separation step such as gas chromatography (GC). GC provides separation of the compounds and facilitates identification by provision of retention times and undisturbed mass spectra. Coupling of pyrolysis and GC can be done on-line and off-line. In on-line Py-GC/MS the pyrolysis chamber (filament, micro furnace, or Curie Point pyrolyzer) is directly coupled to the injection system by a heated transfer line or interface and pyrolysis products are directly transferred onto the separation column [10, 55–61]. Major differences between the different techniques lay in

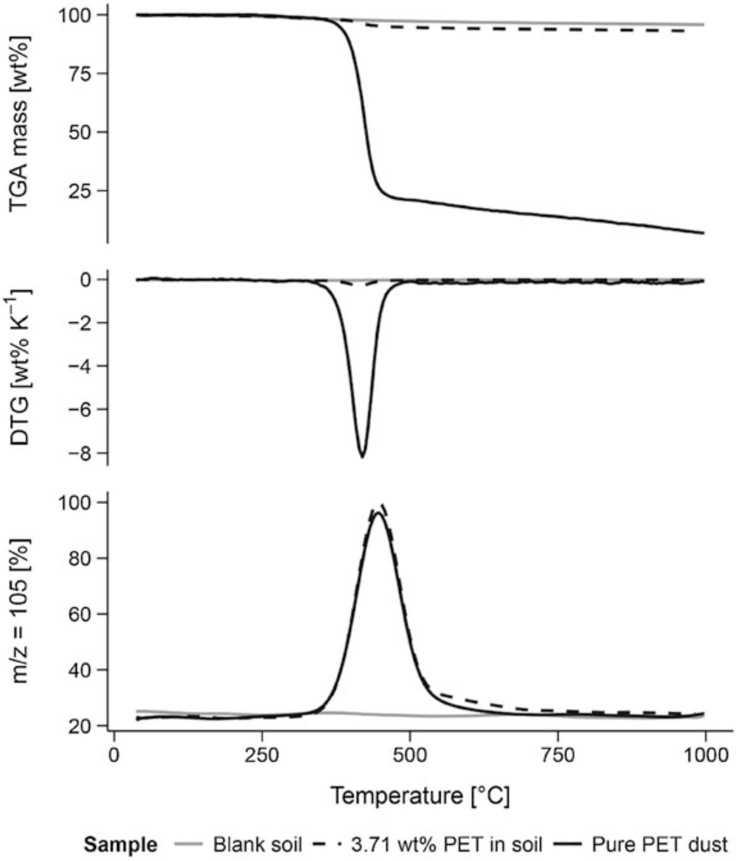


Fig. 4 Output of TGA-MS analysis of PET (Reprinted by permission from ACS Publications, David et al. [54], 2018)

heating rates and sample capacity [62]. Off-line coupling can be realized by trapping the pyrolysate on a sorbent (TED-GC/MS) [63–66] or in a cooled solvent trap [67]. Pyrolysis products are transferred onto separation column by thermal desorption and liquid injection, respectively.

Polymer identification can be based upon specific pyrolysis products or the peak pattern of the pyrogram. Thermal decomposition of some polymers such as polystyrene (PS) results in a small number of specific degradation products, and, therefore, in simple pyrograms with few discrete peaks (ref. Fig. 5a). The pyrogram of polyethylene (PE), for example, is much more complex. It consists of a series of triplets originating from homologous series of alkanes, alkenes, and alkadienes (ref. Fig. 5b). The interpretation of pyrograms of cross-linked polymers like varnishes and resins is very complex and identification of individual decomposition products is very difficult (ref. Fig. 5c). However, identification of the polymers is possible by comparison of the pyrograms with those of reference materials or comprehensive libraries [10, 60, 68, 69].

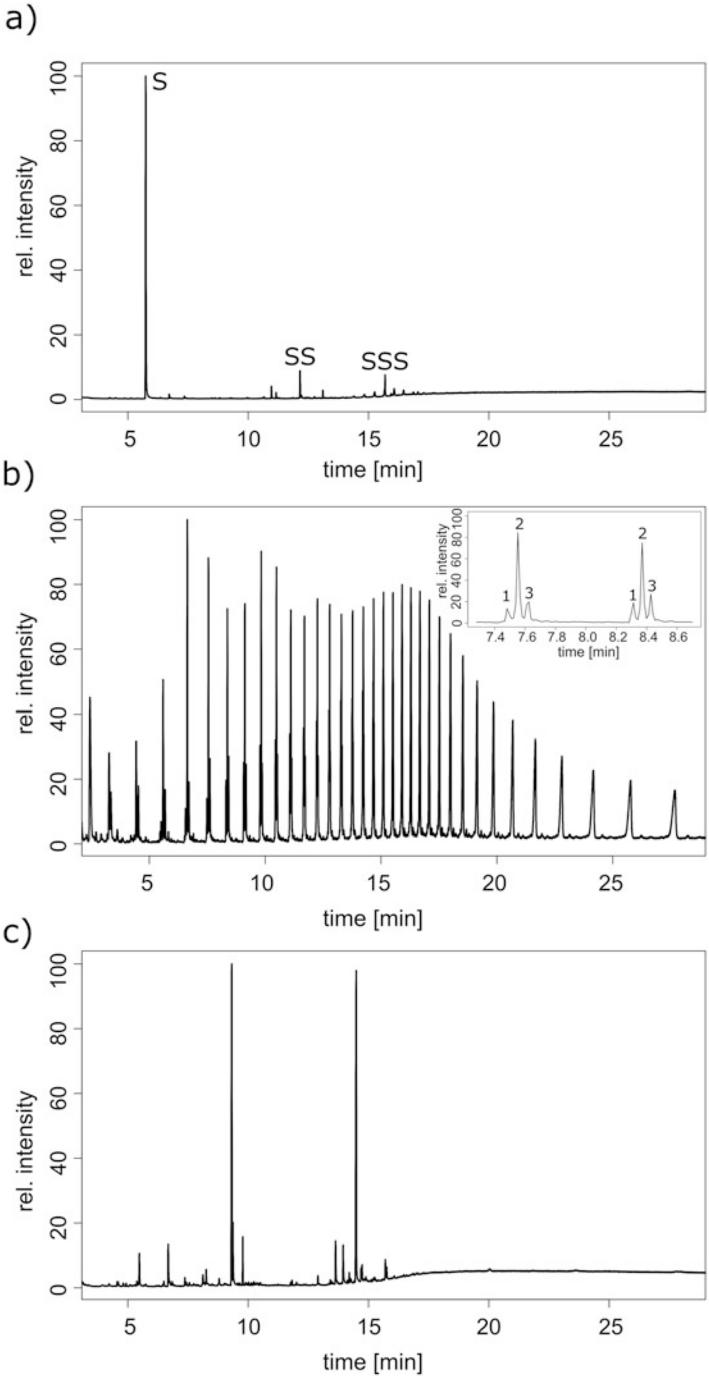


Fig. 5 Pyrograms of different polymers (**a**) Polystyrene: s = styrene, ss = dimer, sss = trimer (**b**) polyethylene: 1 = alkadiene, 2 = alkene, 3 = alkane (**c**) resin

Table 1 Specific indicator compounds for various polymers

Polymer	Indicator compound	Fragment mass	Reference
Polyethylene	α-Alkenes	97, 83	[55, 56, 70]
	α,ω-Alkadienes	95, 82	[55, 56, 70]
Polypropylene	2,4-Dimethylhept-1-ene	126, 70	[55, 56, 70]
Polystyrene	Styrene	104, 78	[55, 56, 70]
	5-Hexene-1,3,5-triyltribenzene	117, 91	[56, 70]
Polyvinylchloride	Benzene	78	[56]
	Naphthalene	128	[67]
Polyethylene terephthalate	Dimethyl terephthalate[a]	194, 163	[56]
	Vinyl benzoate	105, 77	[70]
	Ethyl benzoate	105	[67]
Polyamide	ε-Caprolactam	113, 85	[56, 70]
	N-methyl ε-caprolactam[a]	127, 70	[56]
Poly(methyl methacrylate)	Methyl methacrylate	100, 69	[56]
Styrene butadiene rubber	Vinylcyclohexene	54, 108	[59]
	Styrene	104, 78	[59]
	Cyclohexenylbenzene	104, 158, 129, 115	[66]
Natural rubber	Dipentene	136, 68	[59, 66]

Quantification of MP in environmental samples is still a major challenge for analytical chemists. Thermo-analytical methods using selected degradation products as indicator compounds are promising approaches for MP quantification [2]. These indicator compounds have to be specific for the different polymers and a linear relationship between polymer mass and the amount of indicator compound released during thermal degradation is required. By means of calibration curves, indirect quantification of the polymers is possible using the detector signal of the indicator compounds [62]. However, since some of these degradation products could also be formed from matrix ingredients present in the environmental sample, the selectivity of the indicator compounds has to be evaluated for the respective environmental matrix. Table 1 shows specific indicator compounds for the most important polymers. These compounds have been successfully used for quantification of MP in environmental samples.

Duemischen et al. [65] developed a fully automated two-step method enabling high sample throughput, the so-called thermoextraction-desorption-gas chromatography-mass spectrometry (TED-GC-MS) [65]. In this method, several mg untreated sample (soil, suspended matter) are weighed into a crucible and decomposed in a thermogravimetric analyzer (TGA). Decomposition gases originating from degradation of natural polymers and other matrix compounds in a temperature range of 100–338°C are discarded while degradation products of synthetic polymers emerging in a temperature range of 339–600°C are trapped on a solid-phase absorber. In a second step, the trapped compounds are thermally desorbed and analyzed via GC-MS. By this way PE, PP, PS, PET, PA, and SBR can be directly quantified from environmental matrices without sample pretreatment.

However, high organic contents lead to overloading of the absorber and have to be considered during sample in-weight. Low in-weights result in increased detection limits and representativeness of the analyzed sub-sample may be not given. Due to interferences of inorganic matrix compound with the pyrolysis process matrix matched calibration has to be done [61, 66]. Unice et al. [59] used deuterated internal standards to compensate these effects. However, such standards are not available for all polymers and deuterium exchange during pyrolysis limits the applicability. Therefore, sample pretreatment to reduce the inorganic and organic matrix is recommended for complex matrices. Fischer and Scholz-Böttcher used enzymatic and chemical digestion in combination with density separation for MP analysis in fish [56]. This clean-up is very effective but extreme time-consuming and hard to automate. Alternatively, the polymers can be extracted by organic solvents such as tetrahydrofuran or dichloromethane at elevated temperatures and pressure [55, 57, 58, 71]. In a first step organic matrix compounds are extracted by an organic solvent such as methanol in which most of synthetic polymers are insoluble. In a second step polymers are extracted and separated from the inorganic matrix. The extraction procedure provides high in-weights in the range of 1–10 g, reduction of organic matrix, and enrichment of the polymers. These methods are limited to polymers soluble in the particular solvent under the chosen conditions. Cross-linked polymers such as elastomers and duroplasts are scarcely soluble and currently no extraction methods exist.

Beside the polymer matrix, additives are important and environmentally relevant constituents of plastics. These low molecular substances are added to polymers to improve their attributes such as stability, flexibility, or flame resistance. They can be leached into the environment and harm organisms by toxic effects [72–75]. Pyrolysis/thermal desorption gas chromatography–mass spectrometry (Py/TD-GC–MS) allows solvent-free screening of additives and a subsequent identification of the polymer [60, 76]. In a first step, the additives are thermally desorbed from the polymer matrix at temperatures in the range of 280 and 320°C. After GC-MS analysis of the additives, the remaining polymer matrix is pyrolyzed [77]. Figure 6 shows the thermogram and pyrogram of a plastic sample. The thermogram shows the detected additives: Among others, the UV filter octrizole can be identified. In the pyrogram, however, degradation products specific for polycarbonate such as bisphenol A can be found.

Complete thermal degradation permits determination of the elemental composition (C, H, N, S, O) of polymers which differs distinctly from those of biogenic and inorganic materials. Malow et al. described an approach using elemental analysis combined with Overdetermined Equation Method (EA-OEM) based on different ratios of the elements H, C, O, N, and S in polymers and environmental matrices [78]. The method is limited to relative high MP concentrations (LOD: 33 mg/g) and matrices containing biogenic material with known and stable elemental composition. Biogenic material with high C and H contents such as fats cannot be distinguished from synthetic polymers by this method. Consequently, they have to be removed during sample preparation. The method was validated only for PE and PP, since these two polymers show the strongest differences in elemental composition

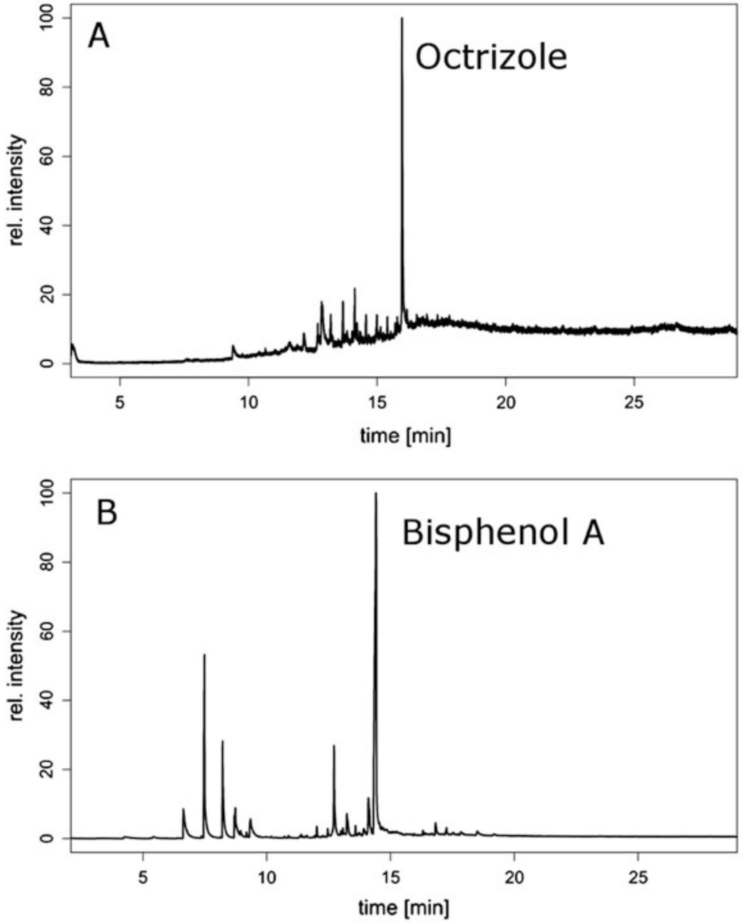


Fig. 6 (**a**) Thermogram and (**b**) pyrogram of a plastic particle

compared to biogenic material. Applicability of the method to other polymers with elemental compositions more like biogenic material such as polyesters has to be tested. However, beside these drawbacks the method reveals reliable results for PE and PP in a consistent matrix like industrial discharge.

2.3 Other Methods

Apart from thermal degradation, polyesters can be depolymerized by chemical treatment. After alkali-assisted thermal hydrolysis in butanol, polycarbonate (PC) and PET can be analyzed by LC-MS [79]. Determination is based upon detection of the emerging building blocks bisphenol A and terephthalic acid. The use of a very sensitive LC-MS-MS method enables very low detection limits in the µg/kg range. However, the suitability of terephthalic acid as specific marker for PET is questionable since it can be released from other polyesters, terephthalate plasticizers, or even natural compounds like humic acids.

3 Discussion

Spectroscopic and thermo-analytical methods are suitable for the identification of MP in environmental samples. The results of both methods are comparable, but both techniques have benefits and limitations [14]. Table 2 summarizes the differences and specifics. Spectroscopic methods are very fast and deliver identification results of isolated particles regardless of the mass. There is only a limitation in particle size (FT-IR >10 µm, Raman >1 µm) [24]. In Py-GC-MS, detection limits depend strongly on the polymer type and lie in the range of ng (e.g., PS) and µg (e.g., PE). Additionally, spectroscopic methods in combination with microscopes reveal information about particle sizes, shapes, and numbers. Another advantage is that spectroscopic methods, in opposition to thermo-analytical methods, are non-destructive and enable additional analyses using the same particle. On the other hand, identification by spectroscopic methods may be disturbed by impurities at the surface of the MP particles, such as biofilms or inorganic contaminants. Such influences are mostly negligible when using thermo-analytical methods.

Furthermore, Py-GC-MS provides a deeper insight into the molecular structure of polymers. In addition to the type of monomers building up the macromolecules, the arrangement of the monomers within the polymer can also be determined by the occurrence of specific degradation products such as oligomers. For example, in Fig. 7 an unknown polymer could be identified as styrene acrylonitrile copolymer with an alternating arrangement of the two monomers acrylonitrile and styrene. The

Table 2 Comparison of thermo-analytical and spectroscopic methods

	DSC-TGA	TGA-MS	Py-GC-MS	NIRS	Spectroscopy (FT-IR/Raman)
Identification of polymer	Limited	Yes	Yes	Yes	Yes
Arrangement of monomers	No	Limited	Yes	No	No
Additives	No	Yes	Yes	No	Severely limited
Disturbance by biofilm and inorganic contaminants	No	No	No	No	Yes
Analysis time	2.7 h	3.2 h	>30 min	5 min	6–24 h (ca. 10% of filter surface)
Non-destructive	No	No	No	Yes	Yes
Particle size distribution	No	No	No	No	Yes
Particle number	No	No	No	No	Yes
Quantification (mass concentration)	Limited	Limited	Yes	Limited	Severely limited

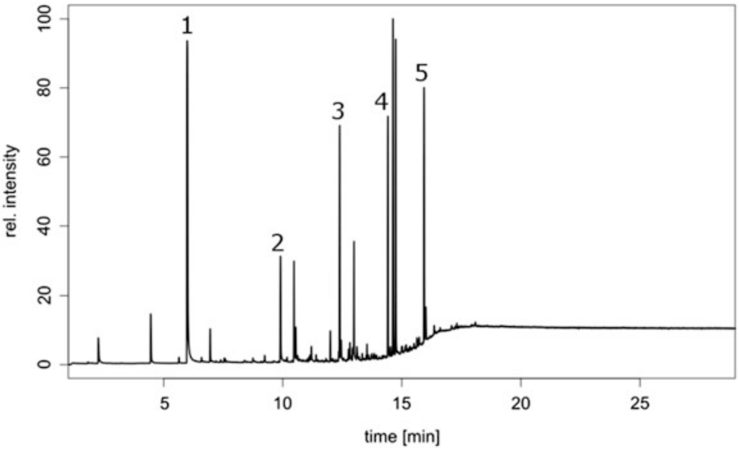


Fig. 7 Pyrogram of styrene acrylonitrile copolymer (SAN). 1: styrene, 2 + 3: S-AN dimer, 4: AN-S-AN trimer, 5: S-AN-S trimer

alternating arrangement, instead of a random or blockwise arrangement is observable by the absence of dimers and trimers consisting of only one kind of monomer and the occurrence of the trimers consisting of two acrylonitrile and one styrene or two styrene and one acrylonitrile units, respectively, in alternating order. Such detailed information can be helpful for the identification of sources of MP emissions and is not accessible by spectroscopic methods.

An interesting example in which such an extensive characterization is indispensable is the differentiation between PS and polystyrene–divinylbenzene resin (PS-DVB) [27, 80]. While PS is a widely used polymer for a multitude of applications (thermal isolation, packaging, electronic cases), PS-DVB is exclusively used for specific applications such as ion exchange materials. Due to their different fields of application, different kinds of emission sources are likely. Both polymers consist of PS chains, but in the case of PS-DVB there are crosslinks between the chains formed by incorporation of the bivalent divinylbenzene. Due to this small difference in molecular structure and the fact that both polymers contain the same chemical groups, distinguishing between both polymers is very difficult when using vibrational spectroscopy (FT-IR, Raman). In contrast, thermo-analytical methods can distinguish between PS and PS-DVB using the thermal degradation product 1,4-divinylbenzene as specific marker for PS-DVB.

Another characteristic hint to the intended use of plastic products and therefore to possible emission sources is provided by the analysis of additives. Typical concentrations of many additives lie in the range of a few mg per kg polymer which is too low for detection by spectroscopic methods. Using thermo-analytical methods, sequential determination of additives and identification of the polymer are possible in one small particle [60, 77].

In summary, both methods provide complementary results. This is especially true for determination of particle numbers and mass concentrations [3, 4, 81].

Determination of particle numbers is possible by microscopy coupled to vibrational spectroscopy. By recording spectra for each particle, reliable identification of the MP particles is provided. However, due to high particle numbers especially for particles <50 μm, measuring times lie in the range of several hours (18 h [34]) per sample. Usage of particle finding software in combination with a reduction of the analyzed particle number by statistical methods leads to a significant decrease of the analysis time. Due to an inhomogeneous distribution of the particles on the filter this approach may lead to high bias especially for minor polymers. Another approach is the analysis of the full filter by chemical mapping technics. To handle the huge amount of data, automatic analysis algorithms have to be established. To avoid a particle overload of the filter the sample volume has to be adapted to the total particle content in the sample [82]. In case of a huge number of natural particles this leads to very small sample volumes and just a few MP particles per filter. Analyzing small aliquots presumes homogeneous samples, which is in case of particle suspensions very challenging. Representativeness of the sub-sample has to be ensured and discrimination of particular particles has to be excluded during aliquoting. Measures for reduction of analysis time often come along with a decrease in spectra quality. Poor spectra characterized by high noise values or missing regions hamper the identification of the particles or lead to false classification. Evaluation of spectra quality and verification of identification by matching software are essential for generation of reliable results and unfortunately neglected in several studies in the past. It is a common practice that library matches with a quality index greater than 700 are accepted, while matches with a ranking between 550 and 700 have to be analyzed again under optimized conditions and interpreted individually [34, 83]. Spectra with an even lower matching are considered as unknown. Despite the additional effort and slowing down of the method each single particle which cannot be identified clearly should be measured under adjusted parameter (e.g., measuring time, laser energy) to render meaningful and distinct spectra for clear identification [30, 34, 83]. Organic matrix or biofilms may disturb the correct identification of the polymers, especially if automated software and libraries based on pristine polymers are relied on exclusively. Spectra in commercial and custom-made libraries usually originate from pristine polymers and may differ significantly from processed or weathered plastics. During processing polymers are blended with additives, fillers, and dyes. Such compounds lead to additional absorption bands in the spectra and may hamper the identification. Environmental stressors such as UV light, heat, or biodegradation result in chemical modifications of the polymers which are summarized in the term weathering [17]. Weathering of polymers lead to formation of polar groups by oxidation (-OH, C=O), double bounds (C=C) by elimination, and free acid groups by hydrolysis. These chemical modifications significantly alter the spectra and may prevent identification or may lead to false classification of polymers. Consequently, integration of spectra recorded from processed and weathered polymers into the libraries is an urgent value for successful identification of MP from the environment. Another aspect is to integrate spectra of widespread non-plastic materials such as inorganic particles, natural polymers (cellulose, keratin, chitin, lignin), and hydrophobic compounds (fats and waxes). Thus, materials can be easily mistaken for MP, due to analogical chemical structure of synthetic polymers and natural compounds. For example, the spectra of the natural compounds waxes and fats are dominated by the C-H vibrations and thus can be mistaken for PE [84]. Differences in the spectra are often marginal and correct identification is only possible by detailed comparison of the fingerprint region (1,500–400 cm^{-1}). To avoid such false classification the organic matrix has to be removed before chemical identification of the particles. This very time-consuming procedure extremely prolongs the analysis time (see Stock et al. [28]).

Due to the immense analytical effort per sample, a preselection of the samples by fast screening methods is advisable for routine analysis or large monitoring campaigns. Promising tools for such a screening are FT-IR or NIR spectroscopic methods for measurement of bulk samples. These methods are very time and cost efficient and reveal semiquantitative MP concentrations [41]. However, these methods are limited by very high detection limits (>1 wt%) and need improvements regarding robustness and reliability before being applied in environmental analysis.

Several methods based on different physical/chemical changes during thermal treatment of polymers have been developed. The extent of the gained analytical information and sensitivity, but also the complexity and effort of the methods increase by TGA-DSC $<$ TGA-MS $<$ Py-GC-MS/TED-GC-MS. TGA-DSC is a user-friendly and cost-effective routine method. It is applicable for qualitative and quantitative analysis of known polymers in a simple sample matrix such as drinking water at high concentrations. However, its viability for trace analysis in complex matrices is severely limited. A more specific detection is given by TGA-MS using selected masses of polymer specific decomposition products. Due to disturbance by matrix compounds forming fragments with the same mass as the analytes, these methods need extensive sample preparation for the analysis of complex samples. Method performance can be improved by implementation of a gas chromatographic separation step. These Py-GC-MS or TED-GC-MS methods enable highly specific and sensitive MP determination by detection of selected marker compounds. These improvements go along with a high complexity of the methods and a weaker robustness. The used analytical instruments are more expensive and well-trained lab staff is needed. Nevertheless, Py-GC-MS and TED-GC-MS are the most promising technics for routine analysis [3]. Table 3 compares the published applications for MP quantification in different environmental matrices. Limits of detection (LOD) vary strongly for different polymers and methods. Py-GC-MS and TED-GC-MS show the lowest LODs in the range of μg/g. Other methods such as TGA-MS or TGA-DSC have LODs two or even three orders higher in magnitude. However, up to date, there is no universal method for all polymers and matrices, and MP quantification is far from standardization. Several improvements have to be done before the methods will be capable for routine analysis. One issue is the strong influence of the sample matrix and the lack of authentic internal standards for most of the polymers [13]. Thus, no universal external calibration can be used, but matrix matched calibrations or standard addition procedures have to be applied [66]. Due to the particulate behavior of the polymers and their inhomogeneous distribution in environmental samples, representativeness of the analyzed sample aliquot is an urgent

Table 3 Comparison of different MP quantification methods

Reference	Method	Polymers	Matrices	Pretreatment	LOD
David et al. [54]	TGA-MS	PET	Soil	None	600 µg/g
David et al. [48]	TGA	PET, PS, PVC	Soil	None	PET: 3.3 mg/g PS: 9.1 mg/g PVC: 22.2 mg/g
Dierkes et al. [55]	Py-GC-MS	PE, PP, PS	Soil, sediment, sewage sludge	Solvent extraction (THF)	LOQs PP: 7 µg/g PS: 8 µg/g PE: 7 µg/g
Eisentraut et al. [66]	TED-GC-MS	PE, PP, PS, SBR, NR	Soil, fish, suspended solids	None	PP: 0.44 µg PS: 0.2 µg PE: 1.6 µg SBR: 0.23 µg NR: 0.22 µg
Fischer and Scholz-Böttcher [56]	Py-GC-MS	PE, PP, PS, PET, PMMA, PA6, PVC	Fish	Enzymatic, chemical digestion	< 0.4 µg
Hahn et al. [41]	FTIRS	PE, PET	Sediment	Milling, mixing with KBr	PE: 10 mg/g PET: 10 mg/g
Majewsky et al. [46]	TGA-DSC	PE, PP	Waste water	None	PE: 25 mg/g PP: 50 mg/g
Mallow et al. [78]	EA-OEM	PE, PP	Water (>250 µm)	Chemical digestion with H2O2	PE + PP: 33 mg/g
Okoffo et al. [57]	Py-GC-MS	PE, PMMA, PS, PET, PC, PP, PVC	Biosolids	Solvent extraction (DCM)	LOQs PE: 0.03 mg/g PMMA: 0.09 mg/g PS: 0.01 mg/g PET: 0.03 mg/g PC: 0.03 mg/g PP: 0.03 mg/g PVC: 0.03 mg/g
Paul et al. [40]	NIRS	PE, PP, PS, PET	Soil	None	5–10 mg/g
Peez et al. [43]	^{1}H NMR	PE, PET, PS		Dissolution	20 µg/mL

(continued)

Table 3 (continued)

Reference	Method	Polymers	Matrices	Pretreatment	LOD
Steinmetz et al. [58]	Py-GC-MS	PE, PP, PS	Soil	Solvent extraction (TCB)	PE: 5 μg/g PP: 86 μg/g PS: 1 μg/g
Unice et al. [59]	Py-GC-MS	SBR, BR, NR	Soil, sediment	None	SBR: 6 μg/g SBR + BR: 5 μg/g NR: 2 μg/g
Wang et al. [79]	LC-MS	PC, PET	Sediment, dust, sludge, mussel, clam	Alkali-assisted thermal hydrolysis in butanol	PC: 2.8 μg/kg PET: 17.7 μg/kg

issue. Frequently used in-weights of 20–50 mg are not representative aliquots, especially if particles >100 μm are analyzed. To overcome these issues, MP has to be isolated from the matrix before analysis. Density separation and chemical or enzymatic digestion of the organic compounds are effective but time-consuming [56]. MP extraction by organic solvents is fast and can be done full automated [55, 57, 58, 71]. Solvent extraction is a widely used procedure in environmental analysis and such machines exist in most routine labs. The existing methods have to be expanded by additional polymers and tested for further matrices.

In contrast to the particle-based methods thermo-analytical methods determine the bulk content of the polymers. These methods deliver mass concentrations of the particular polymers, which is an important indicator for characterization of sources and sinks. Furthermore, for regulatory purposes and environmental thresholds mass concentrations are preferred. However, polymer masses can strongly differ from the actual mass of MPs due to additives and fillers. These plastic ingredients can account for up to 50% of the total mass of plastic objects. Consequently determination of the pure polymer masses may lead to an underestimation of the MP content. Another disadvantage is that a differentiation between homo-polymers and copolymer or polymer composites is not possible. Using styrene as indicator, for example, all styrene containing polymers such as polystyrene (PS), acrylonitrile butadiene styrene (ABS), or styrene butadiene rubber (SBR) will be summed up. On the other hand, this can be utilized for quantification of complex mixtures with known composition such as tire wear particles. Tires consist of a complex mixture of different rubbers (SBR, NR, BR) and fillers. SBR can be quantified by the specific marker compound 3-phenylcyclohexene [66]. Assuming an average SBR-content of 11.3% in the tire material SBR concentration can be transformed into tire material concentrations [85].

4 Conclusion

A realistic risk assessment and the implementation of effective measures to reduce MP emissions require a comprehensive monitoring of sources and sinks. Therefore, there is an urgent need for fast and cost-effective methods for MP determination. Whichever is the best method depends on the scientific question and what kind of information is needed [2]. FT-IR and Raman spectroscopy coupled to microscopy provide a reliable identification of MP particles. Especially in combination with automated particle finding software or an FPA detector, determination of particle numbers in complex environmental samples is possible. If mass concentrations are requested, thermo-analytical methods are the best choice. TGA-DSC and TGA-MS are fast and relatively simple methods. They give good results for MP determination at high concentrations in less complex matrices. For identification of complex polymers or MP determination at low concentrations in environmental matrices, Py-GC-MS or TED-GC-MS is the method of choice. Sample pretreatment is a critical factor regarding cost and time effectiveness. The need of time-consuming pretreatment steps such as density separation and enzymatic/chemical digestion depends on the sample matrix, the investigated polymers, and the aimed detection limits. There is a need for standardized protocols and harmonized quality standards to ensure comparability of the results. However, matrix effects and sample inhomogeneity are still great challenges which have yet to be overcome before the methods will be applied in routine analysis. The overall costs are another factor to be considered [86]. Costs and working time dramatically increase with a decrease in the lower particle size limit and detection limit, respectively. Furthermore, analytical uncertainties and error margin increase. In some cases, the gained knowledge about concentrations of very small particles (<10 μm) may not be worth the immense effort of analyzing them. On the other hand for toxicological studies especially the fraction <10 μm maybe of high interest. So, the request on the analytical methods and the resulting data should be carefully evaluated at the beginning of a study.

References

1. Wagner M et al (2014) Microplastics in freshwater ecosystems: what we know and what we need to know. Environ Sci Eur 26(1):1–9
2. Elert AM et al (2017) Comparison of different methods for MP detection: what can we learn from them, and why asking the right question before measurements matters? Environ Pollut 231 (Part 2):1256–1264
3. Huppertsberg S, Knepper TP (2018) Instrumental analysis of microplastics – benefits and challenges. Anal Bioanal Chem 410:6343–6352
4. Li J, Liu H, Paul Chen J (2018) Microplastics in freshwater systems: a review on occurrence, environmental effects, and methods for microplastics detection. Water Res 137:362–374
5. Renner G, Schmidt TC, Schram J (2018) Analytical methodologies for monitoring micro (nano)plastics: which are fit for purpose? Curr Opin Environ Sci Health 1:55–61

6. Zarfl C (2019) Promising techniques and open challenges for microplastic identification and quantification in environmental matrices. Anal Bioanal Chem 411:3743–3756
7. Silva AB et al (2018) Microplastics in the environment: challenges in analytical chemistry – a review. Anal Chim Acta 1017:1–19
8. Nguyen B et al (2019) Separation and analysis of microplastics and nanoplastics in complex environmental samples. Acc Chem Res 52(4):858–866
9. Peñalver R et al (2020) An overview of microplastics characterization by thermal analysis. Chemosphere 242:125170
10. Tsuge S, Ohtani H, Watanabe C (2011) Pyrolysis-GC/MS data book of synthetic polymers. Elsevier, Oxford
11. Qian K et al (1996) Rapid polymer identification by in-source direct pyrolysis mass spectrometry and library searching techniques. Anal Chem 68(6):1019–1027
12. Everall NJ, Chalmers JM, Griffiths PR (2007) Vibrational spectroscopy of polymers: principles and practice. Wiley-VCH, Weinheim
13. Halle AT (2020) Emerging use thermo-analytical method coupled with mass spectrometry for the quantification of micro(nano)plastics in environmental samples. TrAC Trends Anal Chem:115979
14. Käppler A et al (2018) Comparison of μ-ATR-FTIR spectroscopy and py-GCMS as identification tools for microplastic particles and fibers isolated from river sediments. Anal Bioanal Chem 410(21):5313–5327
15. Müller YK et al (2019) Microplastic analysis – are we measuring the same? Results on the first global comparative study for microplastic analysis in a water sample. Anal Bioanal Chem 412:555–560
16. Becker R et al (2020) Quantification of microplastics in a freshwater suspended organic matter using different thermoanalytical methods – outcome of an interlaboratory comparison. J Anal Appl Pyrolysis 148:104829
17. Lenz R et al (2015) A critical assessment of visual identification of marine microplastic using Raman spectroscopy for analysis improvement. Mar Pollut Bull 100(1):82–91
18. Dekiff JH et al (2014) Occurrence and spatial distribution of microplastics in sediments from Norderney. Environ Pollut 186
19. Erni-Cassola G et al (2017) Lost, but found with Nile red; a novel method to detect and quantify small microplastics (20 μm–1 mm) in environmental samples. Environ Sci Technol 51 (23):13641–13648
20. Shim WJ et al (2016) Identification and quantification of microplastics using Nile Red staining. Mar Pollut Bull 113(1–2):469–476
21. Hengstmann E, E.K.J.E.M. Fischer, and Assessment (2019) Nile red staining in microplastic analysis – proposal for a reliable and fast identification approach for large microplastics. Environ Monit Assess 191(10):612
22. Maes T et al (2017) A rapid-screening approach to detect and quantify microplastics based on fluorescent tagging with Nile Red. Sci Rep 7:44501
23. Hermsen E et al (2018) Quality criteria for the analysis of microplastic in biota samples: a critical review. Environ Sci Technol 52(18):10230–10240
24. Käppler A et al (2016) Analysis of environmental microplastics by vibrational microspectroscopy: FTIR, Raman or both? Anal Bioanal Chem 408(29):8377–8391
25. Klein S, Worch E, Knepper TP (2015) Occurrence and spatial distribution of microplastics in river shore sediments of the Rhine-Main area in Germany. Environ Sci Technol 49 (10):6070–6076
26. Imhof HK et al (2016) Pigments and plastic in limnetic ecosystems: a qualitative and quantitative study on microparticles of different size classes. Water Res 98:64–74
27. Mani T et al (2019) Repeated detection of polystyrene microbeads in the lower Rhine River. Environ Pollut 245:634–641

28. Stock F et al (2020) Pitfalls and limitations in microplastic analyses. Springer, Berlin, pp 1–30
29. Harrison JP, Ojeda JJ, Romero-Gonzalez ME (2012) The applicability of reflectance micro-Fourier-transform infrared spectroscopy for the detection of synthetic microplastics in marine sediments. Sci Total Environ 416:455–463
30. Löder MGJ et al (2015) Focal plane array detector-based micro-Fourier-transform infrared imaging for the analysis of microplastics in environmental samples. J Environ Chem 12 (5):563–581
31. Käppler A et al (2015) Identification of microplastics by FTIR and Raman microscopy: a novel silicon filter substrate opens the important spectral range below 1300 cm^{-1} for FTIR transmission measurements. Anal Bioanal Chem 407(22):6791–6801
32. Cabernard L et al (2018) Comparison of Raman and Fourier transform infrared spectroscopy for the quantification of microplastics in the aquatic environment. Environ Sci Technol 52 (22):13279–13288
33. Anger PM et al (2018) Raman microspectroscopy as a tool for microplastic particle analysis. TrAC Trends Anal Chem 109:214–226
34. Schymanski D et al (2018) Analysis of microplastics in water by micro-Raman spectroscopy: release of plastic particles from different packaging into mineral water. Water Res 129:154–162
35. Araujo CF et al (2018) Identification of microplastics using Raman spectroscopy: latest developments and future prospects. Water Res 142:426–440
36. Tagg AS et al (2015) Identification and quantification of microplastics in wastewater using focal plane Array-based reflectance micro-FT-IR imaging. Anal Chem 87(12):6032–6040
37. Primpke S et al (2017) An automated approach for microplastics analysis using focal plane array (FPA) FTIR microscopy and image analysis. Anal Methods 9(9):1499–1511
38. Simon M, van Alst N, Vollertsen J (2018) Quantification of microplastic mass and removal rates at wastewater treatment plants applying focal plane Array (FPA)-based Fourier transform infrared (FT-IR) imaging. Water Res 142:1–9
39. Primpke S et al (2020) EXPRESS: toward the systematic identification of microplastics in the environment: evaluation of a new independent software tool (siMPle) for spectroscopic analysis. Appl Spectrosc:0003702820917760
40. Paul A et al (2018) High-throughput NIR spectroscopic (NIRS) detection of microplastics in soil. Environ Sci Pollut Res 26:7364–7374
41. Hahn A et al (2019) Using FTIRS as pre-screening method for detection of microplastic in bulk sediment samples. Sci Total Environ 689:341–346
42. Corradini F et al (2019) Predicting soil microplastic concentration using vis-NIR spectroscopy. Sci Total Environ 650:922–932
43. Peez N, Janiska M-C, Imhof W (2018) The first application of quantitative 1H NMR spectroscopy as a simple and fast method of identification and quantification of microplastic particles (PE, PET, and PS). Anal Bioanal Chem
44. Peez N, Imhof W (2020) Quantitative 1H-NMR spectroscopy as an efficient method for identification and quantification of PVC, ABS and PA microparticles. Analyst
45. Peez N et al (2019) Quantitative analysis of PET microplastics in environmental model samples using quantitative 1H-NMR spectroscopy: validation of an optimized and consistent sample clean-up method. Anal Bioanal Chem 411:7603
46. Majewsky M et al (2016) Determination of microplastic polyethylene (PE) and polypropylene (PP) in environmental samples using thermal analysis (TGA-DSC). Sci Total Environ 568:507–511
47. Rodríguez Chialanza M et al (2018) Identification and quantitation of semi-crystalline microplastics using image analysis and differential scanning calorimetry. Environ Sci Pollut Res 25(17):16767–16775
48. David J et al (2019) Introducing a soil universal model method (SUMM) and its application for qualitative and quantitative determination of poly(ethylene), poly(styrene), poly(vinyl chloride) and poly(ethylene terephthalate) microplastics in a model soil. Chemosphere 225:810–819

49. Moldoveanu SC (2005) Moldoveanu SC (ed) Analytical pyrolysis of synthetic organic polymers. Elsevier, Amsterdam
50. Shin T, Hajime O, Chuichi W (2011) Pyrolysis – GC/MS data book of synthetic polymers. In: Pyrolysis – GC/MS data book of synthetic polymers. Elsevier, Amsterdam
51. Wampler TP (1999) Introduction to pyrolysis–capillary gas chromatography. J Chromatogr A 842(1):207–220
52. Schindler A et al (2013) A novel direct coupling of simultaneous thermal analysis (STA) and Fourier transform-infrared (FT-IR) spectroscopy. J Thermal Anal Calorim 113(3):1091–1102
53. Pagacz J et al (2015) Thermal decomposition studies of bio-resourced polyamides by thermogravimetry and evolved gas analysis. Thermochim Acta 612:40–48
54. David J et al (2018) Quantitative analysis of poly(ethylene terephthalate) microplastics in soil via thermogravimetry–mass spectrometry. Anal Chem 90(15):8793–8799
55. Dierkes G et al (2019) Quantification of microplastics in environmental samples via pressurized liquid extraction and pyrolysis-gas chromatography. Anal Bioanal Chem 411(26):6959–6968
56. Fischer M, Scholz-Böttcher BM (2017) Simultaneous trace identification and quantification of common types of microplastics in environmental samples by pyrolysis-gas chromatography–mass spectrometry. Environ Sci Technol 51(9):5052–5060
57. Okoffo ED et al (2020) Identification and quantification of selected plastics in biosolids by pressurized liquid extraction combined with double-shot pyrolysis gas chromatography–mass spectrometry. Sci Total Environ:136924
58. Steinmetz Z et al (2020) A simple method for the selective quantification of polyethylene, polypropylene, and polystyrene plastic debris in soil by pyrolysis-gas chromatography/mass spectrometry. J Anal Appl Pyrolysis:104803
59. Unice KM, Kreider ML, Panko JM (2012) Use of a deuterated internal standard with pyrolysis-GC/MS dimeric marker analysis to quantify tire tread particles in the environment. Int J Environ Res Public Health 9(11):4033–4055
60. Fries E et al (2013) Identification of polymer types and additives in marine microplastic particles using pyrolysis-GC/MS and scanning electron microscopy. Environ Sci Process Impacts 15(10):1949–1956
61. Fabbri D, Trombini C, I.J.J.o.c.s. Vassura (1998) Analysis of polystyrene in polluted sediments by pyrolysis – gas chromatography – mass spectrometry. J Chromatogr Sci 36(12):600–604
62. Fischer M, Scholz-Böttcher BM (2019) Microplastics analysis in environmental samples – recent pyrolysis-gas chromatography-mass spectrometry method improvements to increase the reliability of mass-related data. Anal Methods 11(18):2489–2497
63. Duemichen E et al (2014) Assessment of a new method for the analysis of decomposition gases of polymers by a combining thermogravimetric solid-phase extraction and thermal desorption gas chromatography mass spectrometry. J Chromatogr A 1354:117–128
64. Dümichen E et al (2015) Analysis of polyethylene microplastics in environmental samples, using a thermal decomposition method. Water Res 85:451–457
65. Duemischen E et al (2019) Automated thermal extraction-desorption gas chromatography mass spectrometry: a multifunctional tool for comprehensive characterization of polymers and their degradation products. J Chromatogr A 1592:133–142
66. Eisentraut P et al (2018) Two birds with one stone – fast and simultaneous analysis of microplastics: microparticles derived from thermoplastics and Tire Wear. Environ Sci Technol Lett 5(10):608–613
67. Krauskopf L-M et al (2020) Critical aspects on off-line pyrolysis-based quantification of microplastic in environmental samples. J Anal Appl Pyrolysis:104830
68. Hermabessiere L et al (2018) Optimization, performance, and application of a pyrolysis-GC/MS method for the identification of microplastics. Anal Bioanal Chem 410(25):6663–6676
69. Peters CA et al (2018) Pyr-GC/MS analysis of microplastics extracted from the stomach content of benthivore fish from the Texas Gulf Coast. Mar Pollut Bull 137:91–95
70. Dümichen E et al (2017) Fast identification of microplastics in complex environmental samples by a thermal degradation method. Chemosphere 174:572–584

71. Fuller S, Gautam A (2016) A procedure for measuring microplastics using pressurized fluid extraction. Environ Sci Technol 50(11):5774–5780
72. Lithner D et al (2009) Leachates from plastic consumer products--screening for toxicity with Daphnia magna. Chemosphere 74(9):1195–1200
73. Lithner D, Larsson A, Dave G (2011) Environmental and health hazard ranking and assessment of plastic polymers based on chemical composition. Sci Total Environ 409(18):3309–3324
74. Luft A et al (2017) Nontarget analysis via LC-QTOF-MS to assess the release of organic substances from polyurethane coating. Environ Sci Technol 51(17):9979–9988
75. Zimmermann L et al (2019) Benchmarking the in vitro toxicity and chemical composition of plastic consumer products. Environ Sci Technol 53(19):11467–11477
76. Yanagisawa H et al (2018) Simultaneous screening of major flame retardants and plasticizers in polymer materials using pyrolyzer/thermal desorption gas chromatography mass spectrometry (Py/TD–GC–MS). Molecules 23(4):728
77. Yanagisawa H, Maruyama F, Fujimaki S (2019) Verification of simultaneous screening for major restricted additives in polymer materials using pyrolyzer/thermal desorption gas–chromatography mass spectrometry (Py/TD-GC-MS). J Anal Appl Pyrolysis 137:37–42
78. Mallow O et al (2020) A new thermoanalytical method for the quantification of microplastics in industrial wastewater. Environ Pollut 259:113862
79. Wang L et al (2017) A simple method for quantifying polycarbonate and polyethylene terephthalate microplastics in environmental samples by liquid chromatography–tandem mass spectrometry. Environ Sci Technol Lett 4(12):530–534
80. Scherer C et al (2020) Comparative assessment of microplastics in water and sediment of a large European river. Sci Total Environ 738:139866
81. Mai L et al (2018) A review of methods for measuring microplastics in aquatic environments. Environ Sci Pollut Res 25(12):11319–11332
82. Bergmann M et al (2019) White and wonderful? Microplastics prevail in snow from the Alps to the Arctic. Sci Adv 5(8):eaax1157
83. Woodall LC et al (2014) The deep sea is a major sink for microplastic debris. R Soc Open Sci 1 (4):140317
84. Witzig CS et al (2020) When good intentions go bad-false positive microplastic detection caused by disposable gloves. Environ Sci Technol 54(19):12164–12172
85. Klöckner P et al (2019) Tire and road wear particles in road environment – quantification and assessment of particle dynamics by Zn determination after density separation. Chemosphere 222:714–721
86. Primpke S et al (2020) EXPRESS: critical assessment of analytical methods for the harmonized and cost efficient analysis of microplastics. Appl Spectrosc:0003702820921465

Biodegradable Plastics: End of Life Scenarios

Endres Hans-Josef

Contents

1 Introduction 70
2 Wording 70
  2.1 Degradable Petroleum-Based Biopolymers 72
  2.2 Degradable Bio-Based Biopolymers 73
  2.3 Non-degradable Bio-Based Biopolymers 73
  2.4 Polymerblends and Copolymers 73
  2.5 Old and New Economy Bioplastics 74
  2.6 “Drop-in” Bioplastics 75
3 Degradation 76
  3.1 Primary and Ultimate Degradation 76
  3.2 Oxo-Degradability 77
  3.3 Biological Degradability 78
  3.4 Compostability 80
  3.5 Aquatic Degradability 83
  3.6 Degradability in Soil 90
  3.7 Anaerobic Digestion (Biogas Generation) 94
  3.8 Decomposition/Degradation in Organisms 97
4 Key Factors for Biodegradation 97
  4.1 Microstructure of Material 97
  4.2 Physico-Chemical Environmental Conditions 100
  4.3 Microbiological Conditions 103
5 Conclusions 105
References 105

Abstract When it comes to bioplastics, it is important to differentiate between the biopolymer in its form as a macromolecule and the resulting bioplastic material as a ready-to-use material. Furthermore, a distinction must be made between bio-based and biodegradable plastics. Bio-based refers to the raw material origin of the

---

E. Hans-Josef (✉)
IKK – Institute for Plastics and Circular Economy, Leibniz Universität Hannover, Garbsen, Germany
e-mail: endres@ikk.uni-hannover.de

Friederike Stock, Georg Reifferscheid, Nicole Brennholt, and Evgeniia Kostianaia (eds.),
Plastics in the Aquatic Environment - Part I: Current Status and Challenges,
Hdb Env Chem (2022) 111: 69–110, DOI 10.1007/698_2021_745,
© Springer Nature Switzerland AG 2021, Published online: 31 March 2021

polymer feedstock, while biodegradability describes an end of life option. However, both features are independent of each other. Although biodegradability describes a material property that depends on the microstructure and the chemical structure of the material, in practice biodegradability is a system feature, since there are a variety of environmental conditions, from industrial composting facilities to sewage treatment plants, soils in a variety of climatic regions, the beach and the seabed, or even the human body. It is, therefore, necessary to provide clear information about the environmental conditions and the point in time at which a material or product is biodegradable. In the area of compostability, some test standards for bioplastics and other organic substances cover various environmental conditions well, while test standards and also the understanding of degradation mechanisms in other areas, such as degradability in soil or in marine systems, are only available in small numbers and do not reflect the complex environmental conditions well.

Keywords Anaerobic digestion, Aquatic degradability, Biodegradability, Biodegradable plastics, Bioplastics, Compostability, Environmental conditions, Marine degradability, Material microstructure, Oxo-degradability, Standards for biodegradability

1 Introduction

The number of newly developed bioplastics has increased continuously in recent years but the market volume is still less than 1% of the total plastics market. Bioplastics are not, however, a completely new kind of material, but rather a rediscovered class of materials within the familiar group of materials known as plastics.

The first polymer materials synthesized by man were all based on renewable materials (e.g., caseins, gelatin, shellac, celluloid, cellophane, linoleum, rubber, etc.) because at that time there were simply no petrochemical feedstocks available. Apart from a few exceptions (cellulose- and rubber-based materials), these first bio-based plastics were almost completely replaced by today’s petrochemical plastics.

Bioplastics are now experiencing a renaissance: this is particularly because of ecological aspects as well as limited petrochemical resources and also, in part, innovative property profiles like their biodegradability. This is combined with an increasing awareness amongst the public, politicians, industry and, in particular, research and development. These biopolymers or bioplastics are, however, still very much at the start of their development.

2 Wording

There is still a lot of confusion about the terms “biopolymer”, “bioplastic”, “biodegradable plastic”, “plastics from renewable resources”, etc., because biodegradable plastics can be based on petrochemical as well as on renewable resources and biobased feedstock can lead to degradable as well as durable plastics. The best general definition for biopolymers so far describes a polymer material that fulfills at least one of the two following properties [1]:

- Fully or partly made from bio-based (renewable) raw materials
- In some way biodegradable

Given that, the following three fundamental groups of biopolymers exist:

1. Degradable petro-based biopolymers
2. Degradable (mainly) bio-based biopolymers
3. Non-degradable bio-based biopolymers

Biologically degradable plastics can be based on petrochemical raw materials as well as on renewable raw materials. Degradability in polymeric materials is ultimately influenced only by the chemical and physical microstructure of the polymer, and neither by the origin of the raw materials used nor by the process used for manufacturing these polymers or different products made out of them. This means that biopolymers need not necessarily be made exclusively from renewable materials. Biologically degradable plastics can also be produced from petrochemical ingredients such as polyvinyl alcohols, polycaprolactone, various polyesters, polyesteramides, etc. (Fig. 1, bottom right). Conversely, not all biopolymers based on renewable ingredients are necessarily biologically degradable; for example, highly substituted cellulose acetates, vulcanized rubber, casein plastics, linoleum or bio-based PE, PET, PA, etc. (Fig. 1, top left). Typical examples of the group of

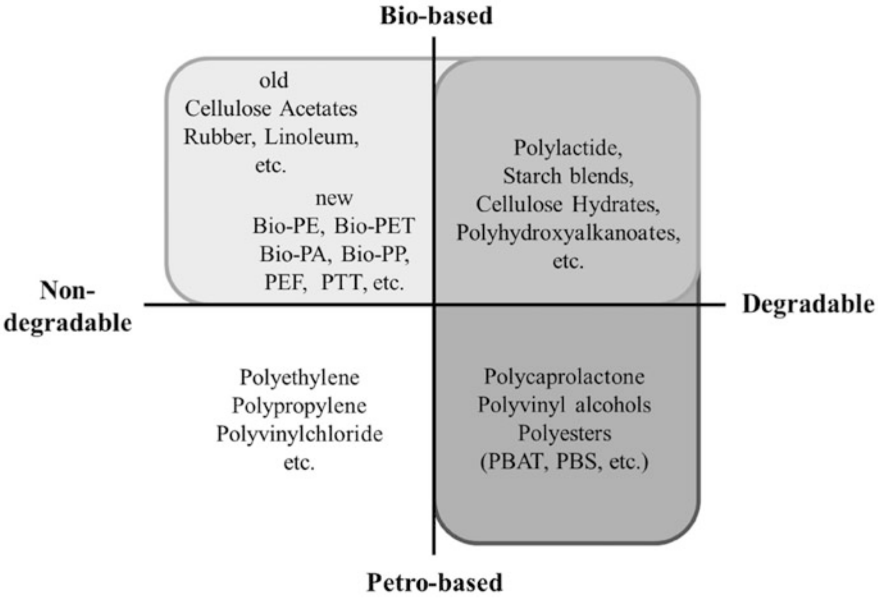


Fig. 1 Bioplastics and the three fundamentally different biopolymer groups ([1], modified)

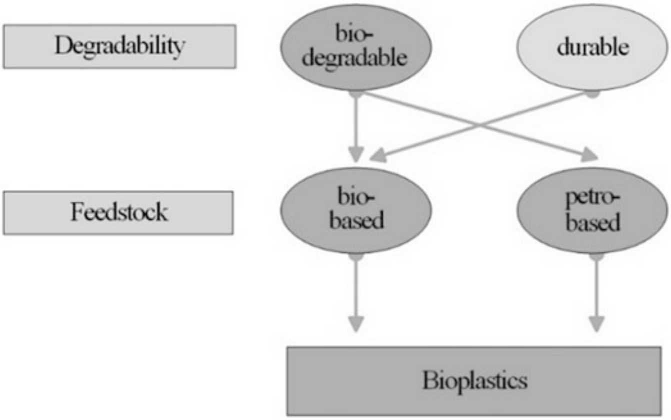


Fig. 2 Raw material basis and degradability of bioplastics ([2], modified)

bio-based and biologically degradable bioplastics are starch-based plastic blends, polyhydroxyalkanoates (PHA), and PLA (polylactic acid).

To avoid misunderstandings when speaking of biopolymers or bioplastics, it is imperative that the most precise nomenclature possible is used. Therefore, it is advisable to speak specifically of biodegradable or bio-based bioplastics. Degradability here means a functional property or disposal option at the end of the materials' life cycle, irrespective of the origin of the raw materials, whilst, conversely, bio-based describes exclusively the origin of the raw ingredients of the polymers at the beginning and provides no statement whatsoever regarding its degradability. These two different approaches are still being pursued and form the technical basis for a variety of bioplastics (Fig. 2).

2.1 Degradable Petroleum-Based Biopolymers

Biopolymers based on petrochemical feedstock, as well as their petro-based secondary products (e.g., polyols, carboxylic acids), are based on hydrocarbon monomers and oligomers gained from crude oil, natural gas, or coal by various methods of fractionated distilling and targeted cracking, as are conventional plastics. The property profile of conventional polymers can be varied by any number of basic resins, polymerization mechanisms, processing parameters, or additives and adapted to any number of applications. Similarly, the property profile of polymer materials can be expanded for degradability by incorporating various heteroatoms (especially oxygen and nitrogen) into their molecules. For conventional plastics, a desirable property was a high level of resistance to chemicals and microbiological or ecologically determined influences. The goal for degradable petrochemical based biopolymers is to design molecules and materials that are not very resistant to environmental influences but rather biodegrade and depolymerize easily under natural influences.

2.2 Degradable Bio-Based Biopolymers

Within the last 25 years the term biopolymers were defined by polymer materials entered the marketplace that are based on renewable resources and which are compostable. Especially cellulose, starch, sugar, vegetable oils and their secondary products like acids or alcohols, as well as some lignins and proteins are renewable resources that can be used as basic components for production of bio-based and biodegradable biopolymers.

2.3 Non-degradable Bio-Based Biopolymers

These biopolymeric materials have been known for a long time. The first engineering polymer materials were based on renewable materials, such as cellulose and natural latex. Raw materials availability was the main feature of these materials. During the materials manufacturing process, these readily available natural resources were modified in such a way that the resulting polymeric materials exhibited property profiles that were utterly new in those times. The main achievement was that for the first time it was possible to turn perishable organic raw materials into durable polymer materials. In the late 1900s, while industrialization continued its advance, petrochemical raw materials were not yet available. Thus, more than 100 years ago, initial, non-degradable biopolymers based on renewable resources were manufactured, although they were not explicitly termed biopolymers. The long-term availability of CO_2-neutral raw materials has recently become the main reason for the development of modern bio-based biopolymers. This is not “only” about the climate-neutral and acute or immediate availability of raw materials, but also about a strategic independence from exhaustible, globally unfavorably distributed petrochemical raw materials for the production of plastics.

2.4 Polymerblends and Copolymers

There are many co- and terpolymers, as well as mixtures, i.e. blends or the so-called polymer alloys, combining various materials from the previously mentioned biopolymer groups.

Besides their main raw materials, biopolymers almost always contain additives suitable for tailoring a particular property profile. These additives are classified by the same method used previously to define biopolymers but often these additives used to upgrade the basic polymers are still petro-based and non degradable.

The increasing use of bio-based, but non-degradable polymers as additives in biopolymer blends tends to impair the biodegradability of these blends. For co- and terpolymers, the increasing use of non-bio-based blend components or petrochemical monomer raw material necessarily leads to a reduction in the amount of bio-based carbon in the final polymer material. Currently, no minimum content levels have been established for bio-based material components in biopolymer blends and co- or terpolymers. Therefore, polypropylene–starch blends, various copolyetsers or even so-called wood plastic composites (WPC) are considered biopolymers, even though they are non-biogradable and their bio-based content is significantly smaller than their petrochemical content.

2.5 Old and New Economy Bioplastics

The first technical, industrially-used polymer materials, the development of which began over 100 years ago, were all bio-based as there were no petrochemical raw materials available at that time. These so-called old economy bioplastics were based on the renewable plant-based raw materials cellulose and natural latex or on animal proteins. With increasing industrialization at the end of the nineteenth century, the availability of the raw materials was of great concern to produce these materials. The natural raw materials available at that time were modified as part of the material manufacturing process in such a way that they resulted in the first really durable polymer materials with a completely new property profile for that time, without being explicitly described as bioplastics. These old economy bioplastics, therefore, belong to the group of bio-based, non-degradable bioplastics. Of the old economy bioplastics, the only ones still of economic significance on the plastics market are natural rubber, regenerated cellulose, and cellulose derivatives (cellophane, viscose, celluloid, cellulose acetate) as well as linoleum in smaller volumes (Fig. 3).

The renewed development of novel bioplastics (new economy bioplastics), which began around 30 years ago, was at first driven by excess supply of food in Europe, such as “butter or tomato mountains” and faced with the waste problem as it existed at the time as well as the unsatisfactory disposal situation with regard to conventional plastics. The objective of these developments was biobased, degradable bioplastics as a solution to the agricultural surpluses and waste problem. There are currently increasing applications in medicine, landscaping and gardening, wastewater treatment, etc., for which degradability permits an additional function under the respective environmental conditions, such as films for use in agriculture which can be plowed in after use, bioresorbable implants or suture materials for use in surgery, or the targeted release of active substances (fertilizers, medical substances).

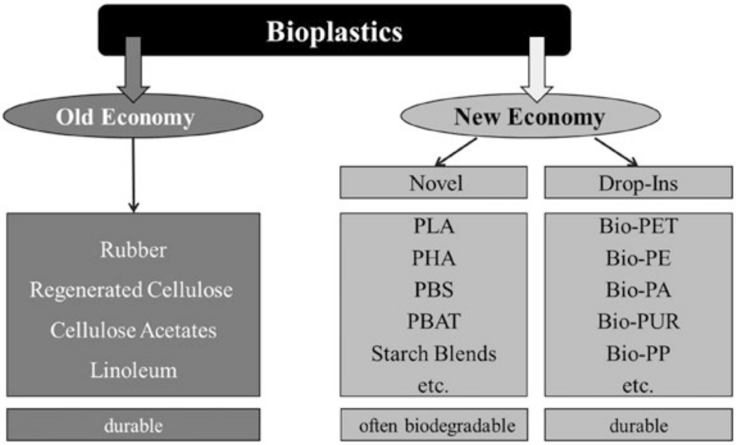


Fig. 3 Old and New Economy Bioplastics ([2], modified)

2.6 "Drop-in" Bioplastics

In the context of the latest developments in bioplastics, the use of sustainable, bio-based raw materials is once again becoming of more concern. Within the group of bio-based and durable bioplastics, the development of the so-called drop-in solutions has made significant technological advances over the past 15 years. The aim is to produce established plastic structures with a maximized bio-based feedstock. To put it simply, the attempt is made to replace the petrochemical raw material with biogenic raw materials, while the established synthesizing processes are retained and ending up in the same chemical structures. That means the goal here is to create bio-based plastics that are similar to familiar conventional plastics. Due to the identical chemical structure, the drop-ins have completely the same property profiles as their petrochemical equivalents. This means that when conventional plastics are replaced by the respective drop-ins, no changes in processing, use and, in particular, recovery and recycling are to be expected. In this context, for example, polyvalent biogenic alcohols or bio-based carboxylic acids are being used to produce a fully bio-based polyethylene (bio-PE) and fully or partly bio-based polyamides (bio-PA), polyurethanes (bio-PUR), acryl butadiene styrene (bio-ABS), polyethylene terephthalates (bio-PET), and various other polyesters. Currently, work is being intensified on these non-degradable bio-based drop-in solutions by large chemical companies, such as BASF, Bayer, Braskem (Brazil), Dow Chemical, DSM, Evonik, or Solvay.

This means at the end that the new economy bioplastics are made up of two basic groups:

1. The chemically-novel biopolymers, that is, unknown in the field of plastics from a chemical point of view until a few years ago (e.g., novel bio-based polyesters such as PLA (polylactic acid), PBAT (polybutylene adipate terephthalate), PEF (polyethylene furanoate), or PTT (polytrimethylene terephthalate)) and
2. Drop-ins which are identical in chemical structure but partially or completely bio-based plastics. Currently, regarding the commercial market share, the most prominent examples of these are bio-PET and bio-PE. Alongside these, work is currently being carried out on further drop-ins, including that in the field of other thermoplastic materials like an also bio-based PP, thermosets (e.g., bio-based EP resins), or elastomer polymer materials (e.g., bio-based EPDM or bio-based polyurethanes).

Depending on the perspective, this means that there are several different types of bioplastics. To avoid misunderstandings, bioplastics should, therefore, generally not be mentioned without further specifying, through additional information, which group is meant.

3 Degradation

3.1 Primary and Ultimate Degradation

As regards material dissociation, it is necessary to differentiate more precisely between a primary degradation (splitting of the macromolecules) and an ultimate degradation of the fission products into water, carbon dioxide, methane, and biomass [3, 4] (Fig. 4).

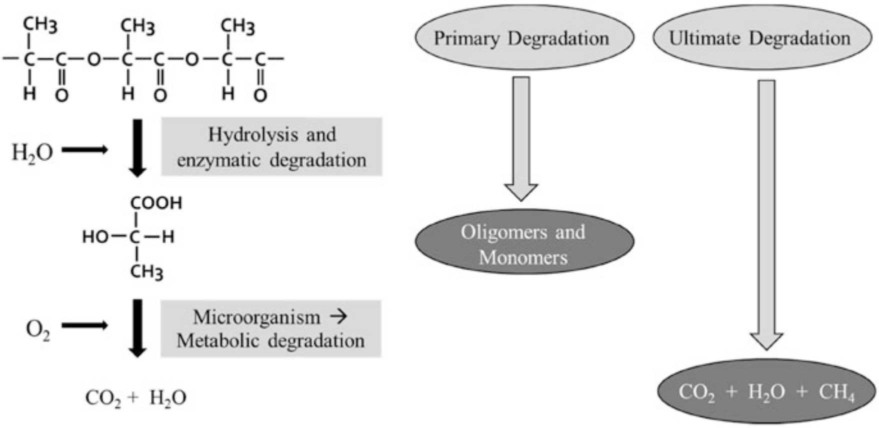


Fig. 4 Primary and ultimate degradation ([1], modified)

The metabolic potential of the (macro-)molecular fission products formed as part of the primary degradation defines here whether the process is simply a macroscopic disintegration process of a component or material or whether it is in fact a complete ultimate degradation [5, 6]. When the ultimate degradation of fission products is not assured, the decomposition products can accumulate, for example, in compost or in groundwater. Therefore, in this case the term "biological degradability" should not be used. One prominent example of a product exclusively coming from primary degradation of (macro-)plastics is microplastic (called as secondary microplastic), which is increasingly accumulating in all the different water systems on earth like the oceans or groundwater and consequently also in living beings. The respective test standards for certifying degradability, therefore, usually include as a significant core element the quantification of the decomposition products formed in the ultimate degradation and/or a record of the oxygen required for this. The oxygen requirement or the amount of CO_2 produced is then compared to the amount theoretically expected in a complete chemical conversion of the material/product to be degraded.

3.2 Oxo-Degradability

During molecular degradation, primary degradation is initiated not only by biologically induced decomposition reactions, but also by other mechanisms. One of these mechanisms is macromolecule scission due to radiation. The most important natural radiation in this respect is the UV content in sunlight. The exposure to sunlight can result in direct polymer chain scission, particularly in polymers with chromophore groups in their molecular structure, e.g. aromatic polyesters or polyamides (photodegradable polymers) [5, 7].

Catalyst residue, contamination, peroxides, as well as other oxygenic components can also absorb sunlight and initiate degradation. Indirect chain scission processes use host molecules, such as aldehydes or conjugated double bond systems; they are excited by radiation and in a second step, the energy required to split the bond is transferred to the actual polymer molecule.

Besides this pure photodegradation, sunlight in combination with oxygen also causes photo-oxidative degradation. Heat or the effect of light can initiate an oxo-degradation sequence by radical formation. Next, alkyl radicals can form and once they react with oxygen, light-sensitive hydroperoxides can form as an intermediate step of photo-oxidative degradation. Continued exposure to light and elevated temperatures together with the previously formed hydroperoxides cause continued radical formation (alkoxy, peroxide, and alkyl radicals), until the polymer chains ultimately degrade. Reaction products such as carboxyl acids or alcohols are subject to further final degradation.

Another method for the initiation of the primary degradation is a chemical dissolving process, for example, in water with subsequent or parallel hydrolysis for water-soluble polymers.

These different reaction mechanisms (Fig. 5) have in common that they can initiate to macroscopic primary degradation, without ensuring final degradation of

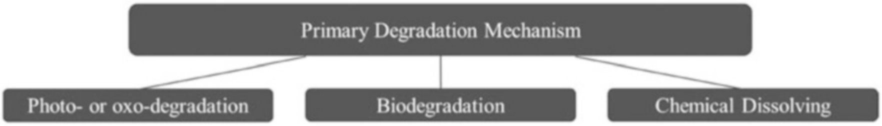


Fig. 5 Degradation mechanisms in degradable polymers ([1], modified)

the products of decomposition. Therefore, total biological degradability or compostability of materials cannot automatically be presumed, even when there is macroscopic disintegration or macrobiological damage and a reduction and/or loss of mechanical properties, surface change, or odor development.

It is, therefore, particularly important in the case of macroscopic disintegration or macrobiological damage, that a complete biological degradability or compostability of the materials is not automatically assumed. The respective additive-enhanced plastics, whose macroscopic decay or primary degradation is initiated solely through oxo-degradation, may therefore not be designated as being degradable or even compostable bioplastics, as ultimate degradation is the crucial process as regards ensuring degradability. From a scientific perspective, there is no further need for discussion as regards oxo-induced or solution-induced primary degradation at all because here, in accordance with the testing standards, the quantification of the resultant final degradation products and the oxygen or possibly hydrogen demand necessary for the metabolization forms the basis for an accurate statement concerning complete degradability.

Current research is concentrating on the oxo-degradability of polyolefins, especially PE, by incorporating special metal ions to initiate a radical oxidation mechanism. However, this oxo-degradation method is very controversial as well as those described before. According to most experts, total microbiological final degradation generally has not been reached in oligomer decomposition products until they are reduced to less than 20 to 25 C atoms (4). The decomposition products of polymer oxo-degradation are usually much larger. In order to obtain smaller, completely, or ultimately degradable oligomers for total degradation, high doping levels are required, which in turn cause a considerable and generally unacceptable decrease in materials properties. The additives for the initiation of the primary degradation once again do not ensure the ultimate degradation. Instead of that, they can, however, lead to a reduction in the stability of the primary materials and also to a contamination of recycled products and thereby, for example, to a reduction in the stability of secondary polyolefins. Additives which solely initiate a primary oxo-degradation present a potential problem for the established recycling of polyolefins.

3.3 Biological Degradability

In most cases, biodegradation of the plastic items occurs on the surface, i.e. at the solid-liquid interface. Microbes and enzymes cannot penetrate the solid plastic, so only the exposed surface is available for biodegradation processes. The physical effect of biodegradation on a solid plastic object is mainly chemical and mechanical erosion as well as light exposure, which leads to thinning and weakening of the object. This process results in the object losing mass, physical properties, such as the reduction of the mechanical values, the visual appearance (surface structure, coloration, etc.), the development of an odor and ultimately the loss of physical integrity through fragmentation into biodegradable particles, the ultimate fate of which is to biodegrade. The term disintegration is used just when the degradation process is prolonged until the original object is completely fragmented into particles below a defined size. This definition does not include the degradation to lower-molecular parts, so that the following definition describes the biological degradability of polymers better: Biological degradation is a process caused by biological activities that leads to a change of the polymer chains to naturally occurring metabolic products. A plastic product is biodegradable, when all its material components end up in microbial metabolic products like water, carbon dioxide, hydrogen or methane and biomass [8]. When microorganisms cause degradation processes, biodegradation, biofragmentation and biological disintegration are the right terms [9].

In the course of total biological degradation, microorganisms in fact require extracellular enzymes to digest plastics and/or their molecular decomposition products. These enzymes essentially use oxidation and hydrolytic processes to break the material down into even smaller components, which can then be absorbed by the cell, where they become metabolized [10–12]. In dependence from the initial starting point of degradation, an exo- and endodegradation mechanism of polymer chain can be distinguished [13]. In case of exodegradation monomers or small oligomers are cleaved from the chain end while endodegradation describes enzymatic chain cleavages which take place statically distributed over the chain. This initial enzymatic degradation step often determinates the degradation rate.

However, the enzymes are too voluminous to penetrate the degrading material efficiently. Therefore, this process can function only as surface erosion, or as a diffusion-controlled sequence in liquid carrier media, especially water. In the other case, the enzymes are adsorptively or covalently bound to the cell wall. The microorganisms have to adhere closely to the polymer surface in order to bring the immobilized enzymes into contact with the substrate so that they can react [14]. Since the enzymes cannot penetrate the plastic due to their size, biodegradation is an interface process, and the material is removed from the surface.

Given that, biodegradation can take place under a wide variety of ambient conditions (ground, water, seawater, clarification plants, compost, human body, etc.) covering a wide range of biological degradation scenarios (Fig. 6).

Finally, biologically degradable plastics consist of natural (renewable) or petrochemical raw materials and, as polymer materials, are amenable to biological degradation reactions – that is, they break down ultimately under the influence of microorganisms and/or enzymes.

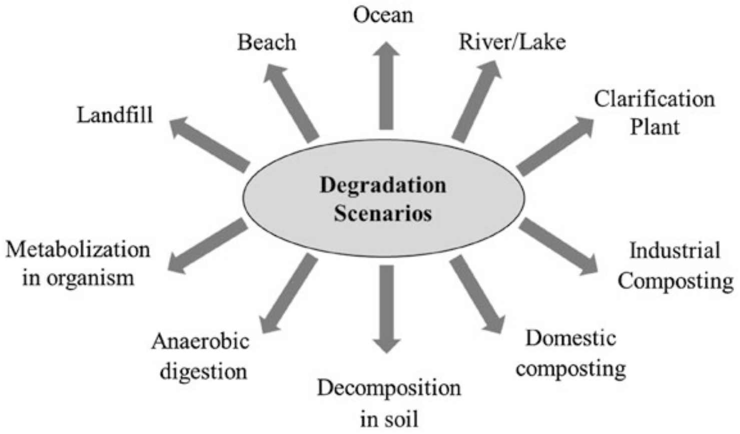


Fig. 6 Various biodegradation scenarios for bioplastics

3.4 Compostability

Even total biodegradation of a material still does not automatically mean that the material or parts produced from it are also compostable. A material is considered biodegradable if all its organic components generally and regardless of any time factor are subject to primary and ultimate degradation by biological activity [15, 16].

A material or material mix and parts produced from it is considered compostable, when, under defined conditions in a composting system, it is entirely transformed into CO_2, H_2O, CH_4, and biomass within a specified length of time, i.e. mostly during a composting cycle ranging from a few weeks to months [17]. In this context, a tree trunk, for example, is biodegradable, but not compostable.

That means, while biodegradability covers a lot of different biological degradation scenarios without any information on the surrounding conditions, temporal course or the duration of the degradation process, compostability describes a degradation process under specified environmental conditions like a domestic compost or industrial composting facility. There are several national, European, and international standards defining compostability of degradable materials and/or products made from these materials (e.g., packaging). At this point it should be noted that the first standard of its kind, German standard DIN V 54900, has been replaced by European standard EN 13432.

Even so, DIN V 54900 represents an important basis for other standards in this field. It consists of five parts and describes succinctly and in detail the individual test/procedures and evaluation criteria (Fig. 7).

Its first part, *DIN V 54900-1* describes the data required regarding the chemical composition of a material. This forces material manufacturers to reveal materials composition. An IR spectrum is created and stored for the precise identification of the particular material.

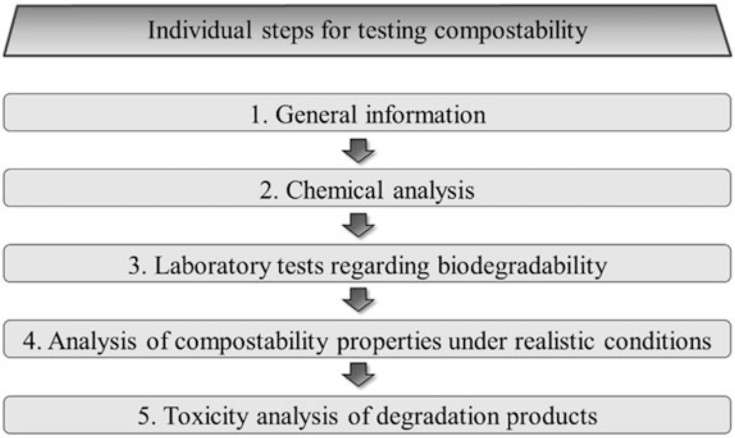


Fig. 7 Steps in the test for compostability according to DIN 54900 ([1], modified)

DIN V 54900-2 describes the test procedures for total biodegradability under clearly defined reproducible laboratory conditions. Two test variations are available, one in aqueous environment and one in compost. During each test, biopolymer metabolization is measured, i.e. the resulting amount of CO_2 or the oxygen consumed during the process. So that a material can be certified as compostable, at least one test version has to indicate total biodegradability of the material, i.e. at least 80% of the theoretical value of total final degradation has to be achieved.

In order to more precisely evaluate degradation behavior of the materials, in the next step screening tests are performed using an aerobically driven, aquatic respirometer testing system. Here, the quantity of oxygen is measured in BOD units (*Biochemical oxygen demand*) that would have to be produced to maintain pressure in the closed system without changing the volume at a constant temperature. *Biochemical oxygen demand* (BOD_m) determines the quantity of oxygen in milligrams consumed in *m* days by the biochemically oxidizable substances contained in 1 L of test water. In order to keep the specific surface of the materials investigated virtually constant, the materials are generally ground to a powder with a specified surface. A fraction of the powder with a particular grain size is sifted out. Subsequently, analogous to DIN 53739, a certain amount of it is added to a potassium phosphate buffered medium (pH value >7) composed as follows (for 1 L):

KH_2PO_4	0.7 g
K_2PO_4	0.7 g
$MgSO_4 \cdot 7H_2O$	0.7 g
NH_4NO_3	1.0 g
NaCl	5.0 mg
$FeSO_4 \cdot 7H_2O$	2.0 mg
$ZnSO_2 \cdot 7H_2O$	2.0 mg
$MnSO_4 \cdot 7H_2O$	1.0 mg

To inoculate the test substance (medium + powdered material) with microbes, an inoculum of conventional fresh compost can be used that represents a wide variety of microorganisms specific to compost.

In a reaction vessel, the samples thus inoculated are thoroughly mixed by a bar magnet throughout the duration of the test so that they can continuously absorb oxygen until saturation. During substrate oxidation, oxygen is consumed and carbon dioxide is formed. The CO_2 generated is absorbed by soda lye. This procedure creates an under-pressure in the reaction vessel that causes an increase in the electrolyte solution (0.5% H_2SO_4) in a precision manometer. Pressure changes cause the contact between the two electrodes to close, triggering the control and regulation unit that generates oxygen electrolytically. The oxygen thus produced causes system pressure to rise again, thereby breaking the contact.

In a second vessel, copper sulfate and sulfuric acid are the electrolyte used to ensure that no further gas can form besides oxygen. The electrolytic current is then kept constant until enough oxygen is produced to recompensate pressure. For the entire duration of the test, current flow is recorded, and the units are added and converted into the corresponding amount of oxygen. The sum of currents serves as a measure for oxygen consumption.

By comparing the measured oxygen requirement ΔO_2 (BOD) with the theoretical, i.e., the chemical amount of oxygen (COD or ThOD) demanded at the start of the test for total oxidation of the test compound, the so-called degree of degradation (DoD) is determined by the following formula:

$$\mathrm{DoD} = \Delta \mathrm{O}_2/\mathrm{COD}$$

The third part of standard *DIN V 54900-3*, in contrast to laboratory tests, describes the test under realistic conditions. It determines the maximum material thickness that can be degraded within a realistic degradation time. One possible measure is the amount of material that can be found after a certain time in compost (sieving).

Subsequently, quality testing regarding recycling properties (*DIN V 54900-4*) and ecotoxicity (*DIN V 54900-5*) is performed on the generated compost [15].

The European standard DIN EN 13432 [16] was conceived specifically for packaging and defines requirements and methods for establishing compostability and anaerobic treatment of packaging and packing materials. DIN EN 13432 has replaced German standard DIN 54900, yet its content is based essentially on the German standard. DIN EN 13432, similar to DIN V 54900, is divided into four technical steps apart from general information about the material from the supplier:

- Characterization of materials composition
- Biological degradability
- Disintegration during biological treatment (no polymer constituent must be visible following composting)
- Effect on the quality of the compost created

In the end, a two-step process is required to certify compostability of a product (Fig. 8), first a positive evaluation for the material(s) used and secondly for the

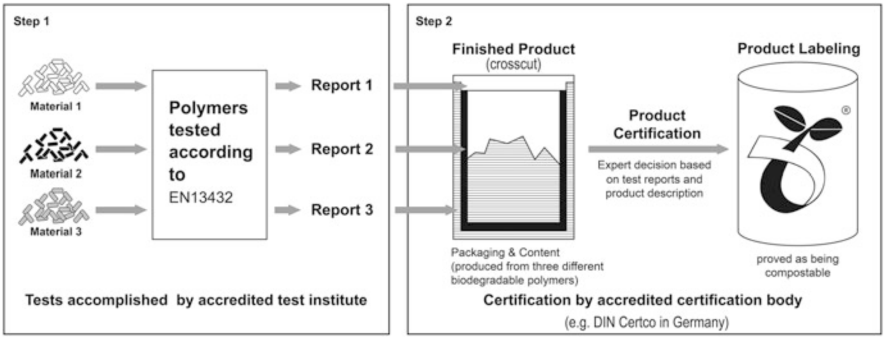


Fig. 8 Two-step evaluation of compostability of a product ([1], modified)

package that was made from the certified material(s). Materials that fulfill the requirements according to DIN EN 13432 can be registered as compostable. The processing industry can use this registration to its advantage when using certified materials. When seeking certification for a product made from certified materials (semi-finished products), the material, but also the articular layer- or wall-thickness, and the specific accessible surface are important characteristics. In Germany DIN CERTCO verifies whether a product with a certain thickness can be certified as being compostable according to the particular standard. If this is the case, the product receives the composting symbol and a certificate stating the maximum permissible layer or wall thickness.

In addition to DIN CERTCO, there are of course further certification organizations worldwide like Vincotte (Belgium) or BPI (USA) employing their own procedures and symbols to certify compostable products. Furthermore, a lot of more international standards are available and used regarding compostability such as DIN EN 14995, ISO 17088, or ASTM 6400. These various standards can be subdivided into two basic groups: Framing standards for product requirements and general descriptions of testing procedures and specific test standards describing in detail how the various investigations are to be performed, including special standards for packaging, aerobic and anaerobic as well as aquatic and terrestrial biodegradation in which the standards for degradability in marine environment and soil are very limited.

3.5 Aquatic Degradability

Pollution of the natural environment, especially the oceans, with conventional plastic becoming more and more a serious environmental issue. Due to the persistence of plastics and their increasing production volume at the same time, plastics in both visible and not directly visible form as micro- and nanoplastics can now be found in almost all ecosystems across the globe. While compostability of plastics has been well studied, there are currently no reliable field test methods and equivalents to these biodegradability standards for unmanaged natural environments. So far, the standards for assessment and certification of biodegradation for aquatic environments, such as wastewater, unmanaged freshwater, or marine habitats have been very incomplete. Freshwater habitats include environments such as rivers, streams, lakes, and wetlands. Marine environments cover a wide variety of habitats, including beaches, ocean surface, open ocean and coastal ecosystems, and deep-sea environments.

In view of the legislative framework and characterization of marine degradability, it is therefore necessary to adapt or adjust existing standards and/or develop new regulations/standards to deliver more results that better match real marine conditions, due to the broad and complex range of physical and chemical conditions as well as microbiological parameters encountered within these natural ecosystems. In addition, the existing standards and test methods for biodegradability in aqueous systems do not contain any toxicity tests and do not take into account the potentially disadvantageous ecological effects of polymer degradation products as well as dissolved additives or small (microscopic) plastic particles that could result from fragmentation of the plastic material [1, 8, 18].

There are various methods for studying aquatic degradability, performed in laboratory systems or in real field experiments. To give an overview, the existing standards and test methods for evaluating biodegradability of plastics in aquatic environments are presented in Table 1 and summarized briefly below, regarding the scale (laboratory or field tests), the inoculum resp. test conditions, the sample, measuring principle, the applied temperatures, and the test duration as well as the way of assessing the results.

The general principle of all laboratory methods is the exact measurement of the end products of degradation under defined environmental conditions, reflecting only a small part of the environment, such as temperature, salinity, nutrition, oxygen availability, exposure, etc. The test conditions on the one hand should represent the various aquatic systems as good as possible, but on the other hand the conditions are selected in a way to achieve a high degradation rate and shorten the examination time. Therefore, the investigations were performed in a laboratory scale with a view preselected inocula such as digested or activated sludge, marine sediments, seawater with indigenous microorganisms and mesophilic temperatures in the range of 15–35°C.

In addition to these laboratory tests, there are also some standards which are carried out under real field conditions to overcome these drawbacks. Given that, the samples are exposed for the degradation studies in the various marine eulittoral or benthic habitats by fixing them in special constructions in order to minimize the physical influence on the material by the experimental design. Afterwards or in the meantime, the remaining material will be evaluated according to specified periods.

The degree of degradation can be assessed either directly by analyzing the remaining material using surfaces' modifications, changes in mechanical or physical properties, weight loss or chemical-molecular polymer analysis such as GC-FID or GC-MS, LC-MS, GPC, HPLC, NMR or FT-IR [37–47] or indirectly by analyzing

Table 1 Overview of different standards to characterize aquatic degradability $CO_2/ThCO_2$: ratio of evolved CO_2 and theoretical amount of carbon dioxide($ThCO_2$) evolved after completely oxidizing the material, calculated from the molecular formula BOD/ThOD: ratio of biochemical oxygen demand (BOD) represents the oxygen consumed under specified conditions by the aerobic biological oxidation and theoretical maximum amount of oxygen (ThOD) required to oxidize the material completely, calculated from the molecular formula, Meso represents tank tests floated by seawater

Standard	Title	Scale	Inoculum/test environment	Sample	Principle	Temp.	Test duration [month]	Assessment
ASTM D 6691 – 2017 [19]	Determining aerobic biodegradation of plastic materials in the marine environment by a defined microbial consortium or Natural Sea water inoculum	Lab	Seawater or microbiological consortium	Powder <25 mm, films, pieces, formed articles	$CO_2/ThCO_2$	30°C	Max. 3	>70% degradation of reference
ASTM D 6692 – 2001 [20]	Determining the biodegradability of radio-labeled polymeric plastic materials in seawater	Lab	Seawater with naturally occurring microbes (adding of marine sediment is possible)	Large grain powder or some shredded form (for example, 1 mm in length)	Amount of (labeled) carbon which is respired to $^{14}CO_2$	n.a.	n.a.	n.a.
ASTM D 7473 – 2012 [21]	Weight attrition of plastic materials in the marine environment by open system aquarium incubations	Meso	Marine sediment and seawater with indigenous microorganisms	Films	Weight loss	Naturally fluctuating	Max. 6	n.a.
ASTM D 7991 – 2015 [22]	Determining aerobic biodegradation of plastics buried in Sandy Marine sediment under controlled laboratory conditions	Lab	Sediment	Films or plates	$CO_2/ThCO_2$	15–25°C	Max. 24	>60% degradation of reference
DIN EN ISO 9408 – 1999 [23]	Evaluation of ultimate aerobic biodegradability of organic compounds in aqueous medium by determination of oxygen demand in a closed respirometer	Lab	Activated sludge, sewage treatment plant, wastewater	Dissolved organic matters	BOD/ThOD	20–25°C	28 days	n.a.

(continued)

Table 1 (continued)

Standard	Title	Scale	Inoculum/test environment	Sample	Principle	Temp.	Test duration [month]	Assessment
EN ISO 9439 – 2000 [24]	Evaluation of ultimate aerobic biodegradability of organic compounds in aqueous medium – carbon dioxide evolution test	Lab	Organic test substance in an inorganic medium and a mixed inoculum from a wastewater treatment plant or from another environmental source	n.a.	$CO_2/ThCO_2$	20–25°C	Max. 1	>60% degradation of reference
EN ISO 11733 – 2004 [25]	Determination of the elimination and biodegradability of organic compounds in an aqueous medium – activated sludge simulation test	Lab	Activated sludge	Dissolved organic matters	Elimination degree of dissolved organic carbon (DOC) or BOD	20–25°C	n.a.	n.a.
DIN EN ISO 14851 – 2019 [26]	Determination of the ultimate aerobic biodegradability of plastic materials in an aqueous medium – method by measuring the oxygen demand in a closed respirometer	Lab	Activated sludge	Powder <250 μm films, shaped parts	BOD/ThOD	20–25°C	Max. 6	>60% degradation of reference
DIN EN ISO 14852 – 2018 [27]	Determination of the ultimate aerobic biodegradability of plastic materials in an aqueous medium – Method by analysis of evolved carbon dioxide	Lab	Activated sludge, eluate from soil or compost	Powder <250 μm films, shaped parts	$CO_2/ThCO_2$	20–25°C	Max. 6	>60% degradation of reference
DIN EN ISO 14853 –	Determination of the ultimate anaerobic biodegradation of plastic materials in an aqueous system –	Lab	Digested sludge	Powder <250 μm films, shaped parts	C in CO_2, methane and dissolved in sludge/C input	35°C	Max. 3	>70% degradation of reference

2018 [28]	method by measurement of biogas production							
DIN EN ISO 15314 – 2018 [29]	Plastics – methods for marine exposure	Field	Salt, fresh, or brackish water	Tensile samples, films, filaments, fibers, ropes, or nettings	Change in phys., chem. or mech. properties	Naturally fluctuating	n.a.	n.a.
DIN EN ISO 18830 – 2018 [30]	Determination of aerobic biodegradation of non-floating plastic materials in a seawater/sandy sediment interface – method by measuring the oxygen demand in closed respirometer	Lab	Marine sediment and synthetic or natural sea water	Parts from films and plates	BOD/ThOD	15–28°C	Max. 24	>70% degradation of reference
DIN EN ISO 19679 – 2020 [31]	Determination of aerobic biodegradation of non-floating plastic materials in a seawater/sediment interface – method by analysis of evolved carbon dioxide	Lab	Marine sediment and synthetic or natural sea water	Parts from films and plates	CO_2/ThCO_2	15–25°C	Max. 24	>60% degradation of reference
ISO 22404 – 2019 [32]	Determination of the aerobic biodegradation of non-floating materials exposed to marine sediment – method by analysis of evolved carbon dioxide	Lab	Sediment (mixed with powdered sample)	Powder	CO_2/ThCO_2	15–25°C	Max. 24	n.a.
ISO 22766 – 2020 [33]	Determination of the degree of disintegration of plastic materials in marine habitats under real field conditions	Field	Marina habitats under real field conditions	Films, fixed in non-degra-dable plastic frames	Weight loss	Naturally fluctuating	Max. 36	>40% disintegration

(continued)

Table 1 (continued)

Standard	Title	Scale	Inoculum/test environment	Sample	Principle	Temp.	Test duration [month]	Assessment
ISO-DIS 23832 – 2020 [34]	Test methods for determination of degradation rate and disintegration degree of plastic materials exposed to marine environmental matrices under laboratory conditions	Lab	(a) wet sandy sediment taken from sandy shoreline (b) interface between a sandy sediment bed and a water column taken from the sea or artificial seawater (c) sea water taken from the sea	Films, sheets or other forms	(1) weight loss (2) erosion rate (decrease of thickness (3) mechanical properties loss (decrease of strength at break) (4) surface area loss	18–24°C	24 (or longer under continuous flow conditions)	
ISO 23977-1 -2020 [35]	Determination of the aerobic biodegradation of plastic materials exposed to seawater – method by analysis of evolved carbon dioxide	Lab	Indigenous population of microorganisms in natural seawater	Powder (<250 μm) or film strips	$CO_2/ThCO_2$	15–28°C, (dark)	Max. 24	n.a.
ISO 23977-2 -2020 [36]	Determination of the aerobic biodegradation of plastic materials exposed to seawater – method by measuring the oxygen demand in closed respirometer	Lab	Indigenous population of microorganisms in natural seawater	Powder (<250 μm) or film strips	BOD/ThOD	15–28°C, (dark)	Max. 24	n.a.
ISO/WD TR 4763	Environmental aspects – analysis of relevant terms used in the sector and need for standardization	Under development						

ISO/WD 5148	Assessment of specific aerobic biodegradation rate of solid plastic materials	Under development
ISO/CD 24187	Principles for the development of standards for investigation procedures of plastic in environmental matrices and related materials	Under development

the biogas evolved (carbon dioxide and methane or stable isotopes) [19, 22, 27, 28, 31, 32, 35] or oxygen demand for the degradation and conversion process of plastics [23, 26, 30, 36] in relationship to the theoretical values, required to degrade the material completely. The CO_2 or biogas generation and the oxygen demand can only be measured by tests which run on a laboratory scale, while direct methods are preferably applied in field tests.

But so far, there are just three standards which are performed under or close to natural conditions [21, 29, 33] whereas all the other standards focus on laboratory methods to characterize biodegradability in aqueous systems. Of course, field tests are much closer to reality. They can represent various marine habitats as well as various freshwater systems if they are carried out within these environments.

However, it is disadvantageous that these field tests only show the material loss as a measured variable. If necessary, the remaining material can also be examined for possible material changes, but no statement can be made about degradation products or the mechanism of the material loss. It is, therefore, not possible to distinguish between a primary and an ultimate degradation. In contrast, laboratory tests provide information about the rate of degradation and, through measurement of the metabolism of the test material to CO_2 in relationship to the theoretical amount of carbon dioxide evolved after completely oxidizing the material, calculated from the molecular formula, also information about primary and final degradability. Further advantages of lab tests are controllable conditions such as temperature, exposure to light or water flow and the additional possibility of analyzing the applied aqueous systems with regard to possible degradation products such as dissolved plastic additives or microplastics.

On the other hand, laboratory tests are closed systems that are not subject to natural fluctuations or a continuous supply of nutrients. The laboratory test systems used can only try to represent a certain environmental condition in the start phase of the test by the applied inocula and therefore deliver results that only reflect the degradation behavior in complex real marine ecosystems to a very limited extent. In addition, they are usually operated at elevated temperatures not occurring in reality to accelerate the degradation behavior and thus reduce the test duration.

3.6 *Degradability in Soil*

Degradation in soil is a disposal option in particular for products in agricultural applications, e.g. mulch films or flowerpots. Degradation in terrestrial systems eliminates the expense of collecting and cleaning products as well as of disposing of the product itself. At the same time, there is the possibility for controlled release of active ingredients such as fertilizer or herbicides. The effects of degradation products on soil quality play a decisive role in degradation behavior. A sufficiently short degradation time is also important. If the degradation is incomplete or if environmentally related, or rather harmful substances are formed, this leads to deterioration of soil quality.

Table 2 Overview of different standards to characterize degradability in soil $CO_2/ThCO_2$: Ratio of evolved CO_2 and theoretical amount of carbon dioxide ($ThCO_2$) evolved after completely oxidizing the material, calculated from the molecular formula, $CH_4/ThCH_4$: Ratio of evolved CH_4 and theoretical amount of methane ($ThCH_4$) evolved after completely digesting the material, calculated from the molecular formula

Standard	Title	Scale	Inoculum/test environment	Sample	Principle	Temp.	Test duration	Assessment
11266 DIN ISO – 1997 [48]	Guidance on laboratory testing for biodegradation of organic chemicals in soil under aerobic conditions	Lab	Soil with knowledge of physical, chemical, and biological properties	Radio-labeled organic chemicals in water, organic solvents, and directly as solid	– Loss of parent compound – Oxygen consumption – CO_2 production – C14-labeled volatile compounds – extractable and non-extractable residues	10–35°C	Max. 120 days	Disappearance time, DT-50 and DT-90
11721-2 DIN EN ISO – 2003 [49]	Determination of the resistance of cellulose containing textiles to microorganisms – soil burial test	Field	Soil, without any specification	Test stripes	Loss of strength	Natural fluctuation	Duration until strength losses 80%	Duration for 80% loss of strength
14239 DIN ISO – 2018 [50]	Soil quality – Laboratory incubation systems for measuring the mineralization of organic chemicals in soil under aerobic conditions	Lab	Soil, without any specification	Soil mixed with (labeled) test substances	Determination of CO_2 production and other volatile (C14-labeled) compounds; $CO_2/ThCO_2$	20°C		No data, standard aims the methods

(continued)

Table 2 (continued)

Standard	Title	Scale	Inoculum/test environment	Sample	Principle	Temp.	Test duration	Assessment
15473 prDIN EN ISO –2019 [51]	Guidance on laboratory testing for biodegradation of organic chemicals in soil under anaerobic conditions	Lab	Soil, with knowledge of physical, chemical, and biological properties, under methanogenic (representing organic marsh (permanently flooded) surface soils, landfill and sludge-amended soils) or water-logged conditions (representing aerobic soils that may be transiently anaerobic)	Test substances (labeled) dissolved in water or organic solvents and directly as solid	Volatile compounds (CO_2, CH_4), water- and solvent-extractable and non-extractable residues	10–35°C	Max. 100 days	Disappearance time, DT-50 and DT-90, additional determination of volatile compounds, formation and persistence of metabolites and non-extractable residues
15985 OENORM EN ISO – 2018 [52]	Determination of the ultimate anaerobic biodegradation under high-solids anaerobic digestion conditions – method by analysis of released biogas	Lab	Methanogenic inoculum (> 20 wt-% total solids content), which is extracted from anaerobic digesters of household waste	Test substances mixed with inoculum	Amount of gaseous carbon released as CO_2 and CH_4	52°C	30 days	Amount of gaseous carbon released as CO_2 and CH_4/ThCO_2 and ThCH_4 release (>70% degradation of reference)

17033 DIN EN – 2018 [53]	Biodegradable mulch films for use in agriculture and horticulture – Requirements and test methods	Lab	Sieved, humidified, natural soil collected from the surface layer of fields and/or forests, ratio of organic carbon in the test material to nitrogen in the soil at least 40:1	Thermoplastic biodegradable films, powder mixed with soil or films, fixed in frames, stacked in the soil	Released CO_2 and ecotoxicity of used soils (reproduction rate of earthworms, microbial nitrification effect, growth of plants)	25°C	24 months	CO_2/Th CO_2 release (>90%)

Focusing on characterizing the terrestrial degradability, just a few test standards are available [48–53]. Table 2 gives an overview on these standards once again regarding the scale (laboratory or field tests), the inoculum resp. test conditions, the sample, measuring principle, the applied temperatures, and the duration as well as the final way of assessing the results. Must standards focus on degradation of organic chemicals and do not consider the degradation of biolastics in soil. Positive exceptions are the OENORM EN ISO 15985 [52], that describes a method for anaerobic degradation of plastics by measuring the released gaseous carbon under high-solids conditions which are typical for digestion facilities and DIN EN 17033 [53], addressing biodegradability of mulch films by measuring the released CO_2 and resulting ecotoxicity of used soils with reproduction rate of earthworms, microbial nitrification effect, and growth of plants as indicators. Other parameters for investigation of degradability in soil are the loss of parent compounds, oxygen consumption, CH_4 production, volatile compounds (C14-labeled), and extractable as well as non-extractable residues. The temperatures vary from 10 to 52°C. Whereas in all the standards the temperatures have a mesophilic level in the range from 10 to 35°C, OENORM EN ISO 15985 recommends a thermophilic temperature of 52°C that is representative for anaerobic digestion. The inoculum respective test environment in all standards consists of soil in a lab scale, partly with knowledge of its physical, chemical, and biological properties. The test duration is between 20 days and 24 months. Just the specific measurement of the resistance of cellulose containing textiles to microorganisms in EN ISO 11721-2 [49] is carried out under real field conditions as soil burial test, whereas all other standards for degradability in soil run on a laboratory scale.

3.7 Anaerobic Digestion (Biogas Generation)

Anaerobic digestion is also known as biogasification. Metabolization to biogas (mainly methane, carbon dioxide, water) is an additional option for degradation of biopolymers that has scarcely been considered so far. The available data are mostly limited to organic waste with high moisture contents, such as mixed green biowaste, kitchen waste, or food waste.

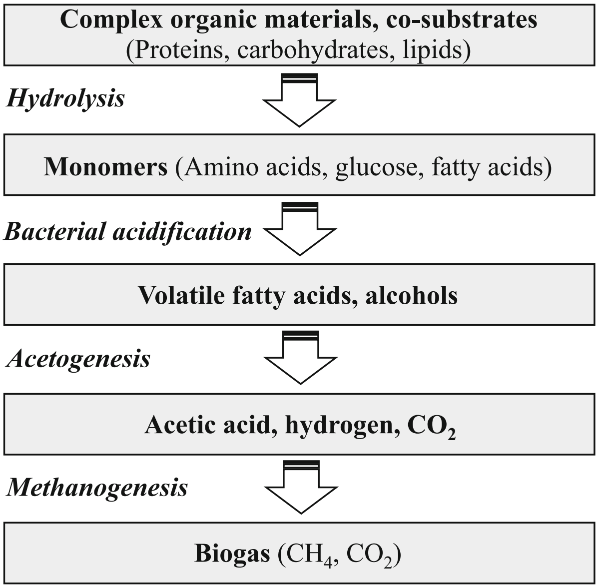
The conversion of organic compounds such as proteins, fats, carbohydrates, or degradable polymers into biogas can generally be subdivided into the following four anaerobic process steps (Fig. 9) [54]:

1. Hydrolysis

Solid substances are broken down (hydrolyzed) by bacterial enzymes into water-soluble monomers (e.g., amino acids, glucose, fatty acids).

2. Bacterial acidification

Fig. 9 Steps of biogas formation ([1], modified)



The dissolved substances are degraded to organic acids (acetic acid, propionic acid, and butyric acid), low alcohols, aldehydes, hydrogen, carbon dioxide, and other gases, such as ammonia and hydrogen sulfide. This process continues until the bacteria are inhibited by their own degradation products (low pH-values).

3. Acetogenesis

In their acetogenic phase, the substances are converted further to acetic acid by acid-forming bacteria.

4. Methanogenesis

Methane bacteria form methane by splitting acetic acid or by reduction of CO_2 with hydrogen in a strongly anaerobic milieu (pH 6.7–8.0) (Fig. 9).

Anaerobic digestion processes can be distinguished as mesophilic and thermophilic processes, 1-phase or 2-phase processes, or as dry and wet processes. In a 2-phase digestion process, hydrolysis and acidification and then subsequently methanogenesis are run in separate tanks. In a 1-phase digestion process, complete digestion is taking place in one unit. Dry digestion processes run at a moisture content $<85\%$, while in wet systems the process is run at a moisture content $>85\%$ [54, 55].

In general, all commercial anaerobic digestion systems consist of a first step of anaerobic fermentation in a wet system, followed by an aerobic composting step. This second step is needed to stabilize the anaerobic sludge. For most biogas facilities, the digestion step runs on a mesophilic level. Whether biodegradation occurs during the first anaerobic phase or during the second aerobic phase impacts only biogas production; however, it does not impact the quality of final compost [56]. Here again, whether or not bioplastics are based on renewable resources is not relevant. The key element is the fact that the material is biodegradable and compatible with the anaerobic digestion process. Correct recovery is assured as long as it is eventually biodegraded and no residues are left after the process is completed.

There are little published data regarding conversion of biopolymers to biogas in a biogas plant, such as temperature, pH-value, microorganisms present, anaerobic/aquatic conditions, etc., or regarding the precise optimum parameters, such as materials flow density, dwell time, gas composition, and gas output. Further research will have to assess potential biogas (energy) production due to bioplastics. Also, the discussion and standardization of requirements for anaerobic biodegradation or anaerobic treatability is still in an early, initial phase [57].

Similar to marine degradability the biogasification rate can be calculated by the ratio of theoretical biogas yield and the real biogas yield. Basically, if the stoichiometric composition is known, the theoretical biogas yield can be calculated approximately according to Buswell:

$$C_cH_hO_oN_nS_s + yH_2O \rightarrow xCH_4 + (c-x)CO_2 + nNH_3 + sH_2S$$

With c, h, o, n and s = molar ratios $x = 1/8\,(4c + h - 2o - 3n - 2\,s)$ and $y = 1/4\,(4c - h - 2o + 3n + 2\,s)$

Examples:

$$\mathrm{PLA}: C_3H_4O_2 + H_2O \rightarrow 1½CH_4 + 1½CO_2 \rightarrow 67.2\ \mathrm{L\ biogas/mol\ PLA}$$

$$\mathrm{PBS}: C_8H_{12}O_4 + 3H_2O \rightarrow 4½CH_4 + 3½CO_2 \rightarrow 179.2\ \mathrm{L\ biogas/mol\ PBS}$$

The investigations on anaerobic digestion of bioplastics often show low conversation rates [1, 58–60]. One underlying reason for these differences may be due to the influence of fungi. Fungi are abundantly available and very active in aerobic composting, while in anaerobic fermentation no fungi are active. Some polymers are mainly (or even only) degraded by fungi and not by bacteria and will therefore biodegrade by aerobic composting and not, or only much slower, by anaerobic digestion. Another reason for the partly insufficient material degradation is the reduced temperature compared to industrial composting processes. The anaerobic mesophilic degradation of PCL, PVAL, PBS, and PLA with its high glass transition temperature is particularly difficult.

First experiments were carried out using a thermal pretreatment of biopolymers [58–60], however, the digestion rate could not be improved. Only PCL with its low melting temperature showed improved digestion rates. For all other biopolymers investigated the digestion rate and the resulting biogas yield decreased. In particular for PLA and other biopolyesters, the thermal pretreatment led to post-processing re-crystallization, which reduced their anaerobic digestibility.

3.8 Decomposition/Degradation in Organisms

These are typically medical applications, either for controlled drug release or to support the healing process following surgery. The best-known applications in this field are absorbable suture materials made from PLA, which can remain inside the body. For the same reason, various temporary implants, such as bone screws for fixating fractures, etc., were developed in recent years using PLA.

Besides these applications, there have also been various attempts to establish biopolymers as edible packaging. However, these attempts have failed mostly, because, among other things, such edible packaging itself requires secondary packaging in order to meet food hygiene and food safety requirements.

4 Key Factors for Biodegradation

Microbiological degradation depends on a couple of parameters. These parameters can be assigned to the three pillars: microbiology/microorganism, the environmental surrounding conditions, and material related factors (Fig. 10). Not only does the variety of these parameters reflect the complexity of this system – the factors are also not necessarily independent of each other.

4.1 Microstructure of Material

While the primary chemical structure determines biodegradability of plastics in principle, the polymer degradation rate is determined by other properties, such as

Fig. 10 Pillars for biodegradation

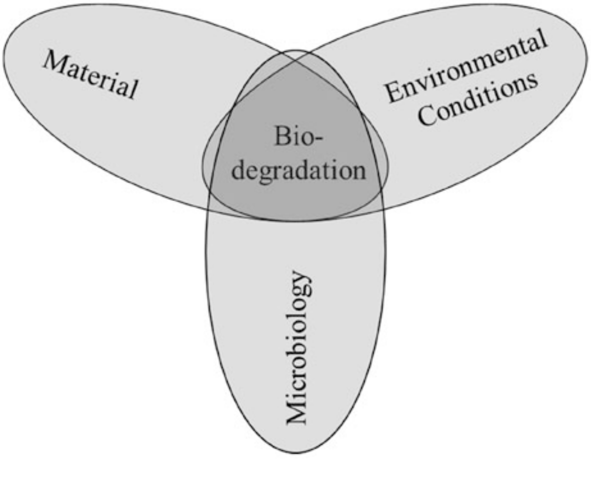
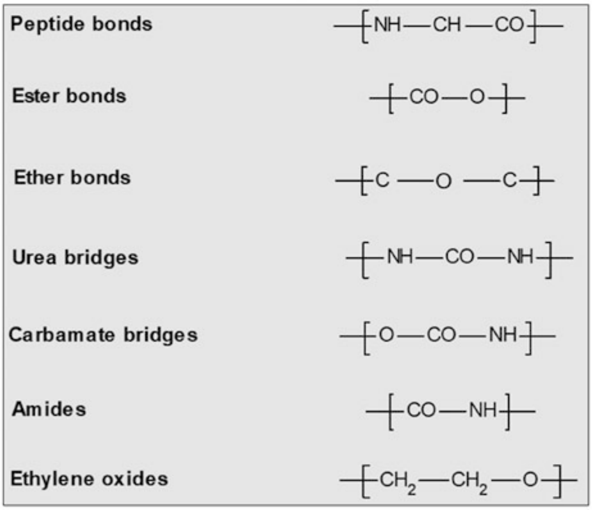


Table 3 Main material parameters for biodegradability [8]

Plastic materials Plastic parts	– Chemical bonds – Branching – Hydrophilic/hydrophobic behavior – Molecular mass – Chain mobility – Melting temperature, glass transition temperature – Crystallinity – Additives – Interaction with copolymers – Morphological and chemical surface structure – Particle size respective surface to volume ratio



Peptide bonds	$-[\mathrm{NH{-}CH{-}CO}]-$
Ester bonds	$-[\mathrm{CO{-}O}]-$
Ether bonds	$-[\mathrm{C{-}O{-}C}]-$
Urea bridges	$-[\mathrm{NH{-}CO{-}NH}]-$
Carbamate bridges	$-[\mathrm{O{-}CO{-}NH}]-$
Amides	$-[\mathrm{CO{-}NH}]-$
Ethylene oxides	$-[\mathrm{CH_2{-}CH_2{-}O}]-$

Fig. 11 Examples of heteroatoms enabling biodegradability ([1], modified)

the melting point and degree of crystallization, which in turn depend on the primary structure. Table 3 gives an overview of significant material related parameters that influence the degradation behavior.

In general, high-molecular engineering polymers whose backbones contain exclusively carbon atoms, such as polyethylene, polypropylene or polystyrene, are inert toward biological degradation [61]. Polyvinyl alcohol is an exception despite its exclusively carbonic backbone [62]. Here, degradation takes place via primary oxidation of the numerous OH-groups with subsequent backbone cleavage – similar to fatty acid degradation [63, 64]. Primarily an enzymatic cleavage of the carbon backbone is caused by either a dehydrogenase or oxidase and subsequently by hydrolase or aldolase reaction [65].

Natural rubber (cis-1,4-polyisoprene) is also biodegradable, although only carbon atoms are present in its polymer backbone [66]. Here, the primary degradation step, backbone cleavage, is initiated by a specific dioxygenase or peroxidase [67].

Other degradable, natural, and synthetic plastic materials usually contain heteroatoms in their backbones, such as oxygen or nitrogen, that represent points of attack for enzymatic catalyzed cleavage products (Fig. 11).

Given that, degradability increases in general as the ratio of heteroatoms to carbon increases, in particular in the main chain, i.e. the biodegradability increases in the following order: PVOH < PCL < PLA < Starch/Cellulose [1].

Chemically unchanged natural polymers, such as cellulose or starch, offer therefore a good biodegradability. However, chemical modifications, e.g. esterification to cellulose triacetate, create plastics that can no longer be cleaved by enzymes.

Polymers with aromatic components or branched structures tend to be more resistant to microbial attack than linear, aliphatic components [56, 61]. For enzymatic hydrolysis, the polymer chain must be flexible enough to fit into the active center of the degrading enzyme. This is the explanation for the easy biological degradation of flexible aliphatic polyester, whereas rigid aromatic polyesters resist biological degradation [68]. An analogous effect can be observed with polyamides. Here, the crystals limit chain flexibility by intermolecular interaction [14].

Crystallinity is an additional factor discussed with regard to the degradation rate in polyesters [68]. In addition to degradation rate dependence on crystallinity, it turns out that the crystalline zones within a plastic are enzymatically hydrolyzed more slowly than amorphous zones. References [69–71] show that crystallinity in PCL films increases during degradation, i.e. the amorphous phases are reduced.

Crosslinking in plastics reduces water concentration via reduced swelling and with it the accessibility of the plastic for the enzyme, also resulting in a reduced degradation rate [67].

A direct connection between the degradation rate and melting temperature was introduced by [72, 73]. They observed that the degradation rate in aliphatic–aromatic copolyester and aliphatic homopolyester decreases with increasing melting temperature [67]. Comprehensive and systematic investigations of the role of chain mobility for polyester degradation show that, for polyesters of similar crystallinity, degradation is controlled exclusively by polymer chain mobility, where mobility is mainly determined by the difference between the ambient temperature and melting temperature [74].

Overall, biodegradability and compostability of biopolymers and/or products made from them increase with certain factors due to the resulting simplified access for microorganisms to the molecules, thus enhancing metabolizability.

Table 4 summarizes the impact of different microstructural material parameters on its degradability.

Another impact factor influencing biodegradability is the finishing process, i.e. the type of modification from biopolymers as virgin polymers to bioplastics as tailor-made “ready to use” materials (Fig. 12). During the finishing process the polymers are modified by some additives such as stabilizers, plasticizer, coloring agents, or reinforcements. Alongside biodegradability of themselves, these additives influence the microstructure and by this biodegradability of the resulting bioplastic.

If additives change the crystallinity or are not themselves degradable, the degradability decreases and if vice versa biodegradable fillers such as starch are used, the

Table 4 Degradability as a function of various micro-structural material parameters (↑ = Increase, ↓ = Decrease), ([1], modified)

Material parameters	Degradability
Intermolecular interaction, crystallinity ↑	↓
Number of unsaturated compounds bonds ↑	↑
Unbranched, flexible molecular structures ↑	↑
Aromatic portion ↑	↓
Molecular weight ↑	↓
Melting and glass transition temperature ↑	↓
Polarity/hydrophilic surfaces ↑	↑
Swelling ↑	↑
Specific surface/surface to volume ratio ↑	↑

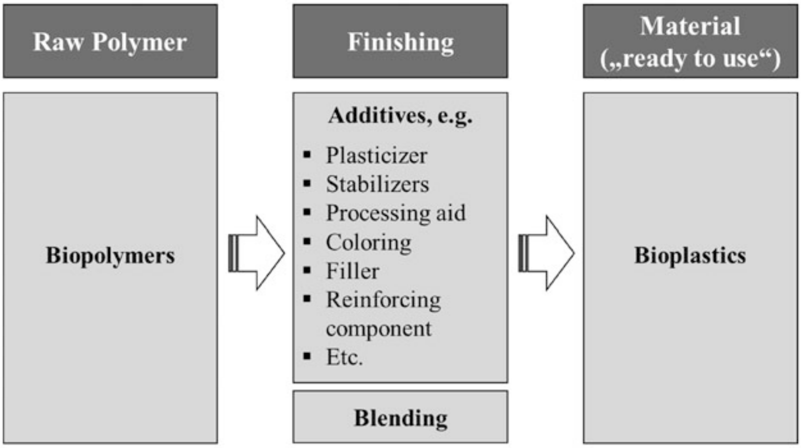


Fig. 12 Finishing process from virgin biopolymers to tailor-made “ready to use” bioplastics ([2], modified)

rate of degradation can be increased significantly. First, the easily biodegradable fillers are broken down. This enlarges the surface area accessible for microorganisms and accelerates the degradation of the remaining matrix.

The most important biodegradable plastics include starch-based polymer materials and various polyesters such as polyhydroxyalkanoates, e.g. PHB (polyhydroxybutyrate), PLA (polylactic acid), PBAT (polybutylene adipate terephthalate), and PBS (Polybutylene succinate).

4.2 *Physico-Chemical Environmental Conditions*

As well as differentiating between macroscopic decomposition of the material (primary degradation) and microscopic ultimate degradation, information about the

Table 5 Main environmental parameters for biodegradability ([8] modified)

Environment	– Temperature – Gas atmosphere – Oxygen availability – Moisture – Salts – Metals – Micronutrients – pH-value – Redox potential – Stability – Light, radiation – Flow conditions – Time

respective environmental conditions and the time span is also essential for a complete description of the degradation process. Biological decomposition can vary enormously under a variety of environmental conditions (soil, water, salt water, compost, human body, etc.) as, apart from the material itself, the degradation process depends on a variety of other (environmental) factors such as microorganisms present, humidity, temperature, available oxygen, pH-value, time, etc. [75]. The following Table 5 summarizes the main factors for biodegradability related to surrounding physico-chemical environmental conditions.

Microorganism development (biodiversity, concentration, activity, adaptation) is also determined by specific environmental influencing factors. They include the presence and absence of oxygen, water content, temperature, pH-value, available nutrients as well as available alternative carbon sources [67]. At the same time, polymer decomposition into small particles or polymer metabolism into water-soluble products, for instance, influences the structure, pH-value, and nutrient content of the environment – not to mention the potential danger that plastics represent to the ecology caused by the accumulation of long-lived, often potentially toxic metabolites. Microorganisms in turn can affect changes in ambient conditions either directly, by excreting metabolism products (e.g., acids) or indirectly, by secreting enzymes that catalyze the formation of reactive reagents in the environment [76].

As an example, biological decomposition of the material in an industrial composting plant with continuous irrigation and turning of the heap takes place more quickly than under a domestic composting process (Fig. 13). Due to the higher temperatures and better oxygen availability, the industrial composting process is much faster than domestic composting and digestion in landfills will take a long time vice versa.

Industrial composting can be defined as “the controlled biological decomposition of organic waste under managed conditions that are predominantly aerobic and that allow the development of thermophilic conditions as a result of biologically produced exothermic heat” [77]. In the course of industrial composting operations, biomass is mixed more frequently, and moisture and oxygen content and temperatures rise up to 50–70°C. When the temperature of the composting pile increases, the microbes adapted to the ambient temperature (mesophiles) stop activity and are

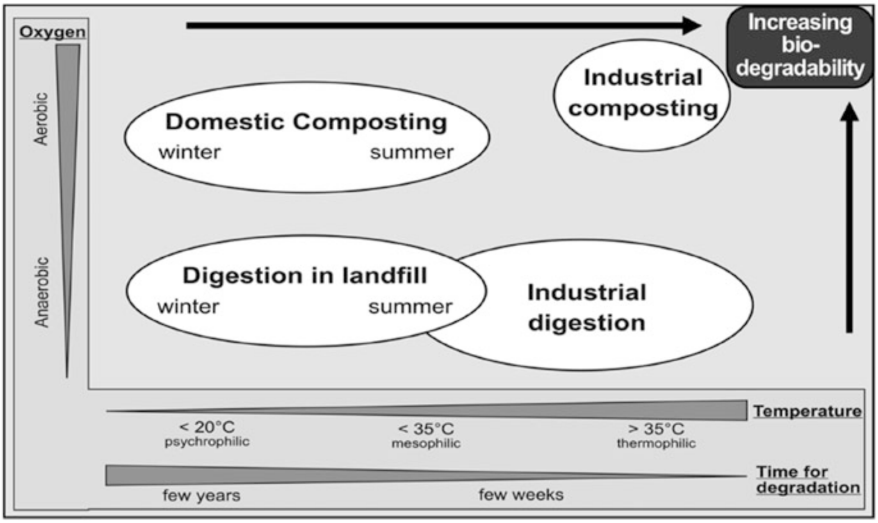


Fig. 13 Conditions in industrial and domestic composting and biogas plants ([1], modified)

replaced by microbes adapted to high temperatures (thermophiles). The rate of activity of microbes is higher at thermophilic temperatures. Due to this shift in microbial populations and the additional treatment of the composting pile, a faster and better degradation of the biopolymers can be ensured. Under these conditions composting is a controlled biotechnological process and therefore the term "industrial" (or municipal) composting is used to distinguish it from "home composting" [78].

Experience has shown that biopolymers certified as compostable under industrial composting conditions are degraded and metabolized well. However, not all biopolymers certified as compostable under industrial composting conditions also degrade under home composting conditions [1, 8, 45].

Municipal composting and home composting share the same designation; however, the conditions for these technologies are quite different [56, 78]. Certified industrial compostability states that products consisting of a certain material and with specified wall thicknesses degrade biologically during a certain time under industrial composting conditions (sufficient oxygen and moisture, regular turning of the pile, temperature development, presence of corresponding microorganisms, pH, carbon/nitrogen ratio, material structure, and size of particles). Industrial compostability of biopolymer materials certified according to legal standards must never be equated with total degradability in domestic compost, also defined as cold composting, i.e. degradation through aerobic biodegradation at ambient temperature (between 21 and 28°C). Certification according to the standards for municipal composting (e.g., EN 13432 or EN 14995) does not imply good in-home composting properties or shortened rotting cycles.

Neither ISO nor ASTM defines home composting rules. The Belgian certification organization AIB Vinçotte issues a specific "home compostability" certification program and an “OK Compost Home” label. Materials degrading to a sufficient level in private composting systems, i.e. home compost, can be labeled additionally or exclusively with this symbol. The certification program for home composting is based only on DIN EN 13432 (see Sect. 3.4). In home composting, 90% biological degradability at ambient temperatures of 20–30°C (in contrast, composting temperatures in industrial plants run approx. 50–70°C) and/or in aquatic surroundings is required (test method according to DIN EN ISO 14851 [26]; cf. Sect. 3.5).

Particularly in Asia, where there is a lack of room and logistics for disposing and industrial composting of biowaste, efforts are being made to support composting in domestic surroundings with heated waste containers to enable domestic composting [79].

However, composting makes sense only when degradability simultaneously offers an additional functional advantage. For example, votive candle holders on graves that can be cleared away with flowers/wreaths etc., films for agriculture that do not have to be collected and disposed of following use, but can be plowed under, laundry bags that dissolve in the washing machine, grocery bags that can be used to collect organic material for composting, or resorbable implants that are metabolized according to the regenerative loop in the human body, etc. In all these applications, degradability and/or compostability results in an additional benefit. By contrast, enforced “composting by decree” requiring separation, collection, and transport to an industrial composting plant represents only additional expense and with it the amount of CO_2 generated by composting equals the amount of CO_2 released by incineration, but composting does not provide an additional energetic benefit.

There are a lot more examples showing the significant influence of the environmental conditions on biodegradability, such as under “normal” ground or anaerobic conditions in a biogas plant, or even on the ocean floor at temperatures of approx. 4°C with no light and a completely different microflora. The impact of the environmental conditions is also very evident for a product such as wood. Under dry conditions, wooden furniture in a house, for example, has an almost unlimited lifespan, whilst in the forest, biological degradation progresses relatively rapidly. Another example is fossilization. Here, too, environmental conditions have prevented a complete degradation of the organic mass.

4.3 Microbiological Conditions

Microbiological degradation is influenced by various factors that can be subdivided into three categories, i.e. the environmental surrounding conditions, material related factors, and microbiological parameters (see Fig. 10). After considering the first two topics in the previous chapters, the following Table 6 gives an overview of the most important microbiological parameters that influence the degradation behavior of plastics.

Biodegradation first requires suitable microorganisms. Certain organisms can usually only degrade a specific group of plastics. For instance, several degrading

Table 6 Main microbiological parameters for biodegradability ([8], modified)

Microorganisms	– Biodiversity – Enzymes and enzyme concentration – Co-metabolism – Competing organisms (protozoa) – Enzyme kinetics – Cell density – Inhibitors/initiators – Intra-/extra cellular mode of action – Aerobic, anaerobic

microorganisms for polyhydroxybutyric acid [67, 80], synthetic aliphatic–aromatic copolyester [14, 81], and synthetic aliphatic homopolyester [82] have been isolated and identified.

Plastics molecules are too large to pass the cell walls of microorganisms. So that bacteria and fungi can use such substances as nutrients, they must produce enzymes that – after being transported through the cell wall – can act outside the cell. The enzymes break down the insoluble macromolecules layer by layer from the surface into short-chain fragments. Here, the organism producing the polymer cleaving enzyme is not necessarily the immediate consumer of these cleavage products. Other organisms in the population can enter the degradation process, absorb low-molecular compounds in the cell and convert them into carbon dioxide, water, and biomass. Under certain circumstances, they can in turn supply other microorganisms with nutrients by excreting metabolites (e.g., acids) that they cannot use. Many degradation sequences take place according to this type of cooperation between different microorganisms. However, the result can also be accumulation of cleavage products which are not further degradable and due to their potentially toxic effects (inhibition, elimination), can pose a potential danger to the microorganisms [8, 14].

In addition, various microorganisms can be responsible for degradation in all types of degradation media. For example, fungi prefer solid surfaces for growth, which is why they are rarely present for degradation in aqueous systems. In contrast, a variety of fungi are involved in degradation in compost [67].

Fungi and other microorganisms, such as bacteria or actinobacteria (actinomycetes), play an important role in the destruction of organic materials [83]. Microorganisms only proliferate in the presence of moisture (relative moisture 63–99%). They will find optimum growth conditions in the temperature range from 10 to 40°C.

For optimum growth, fungi need oxygen and a pH-value of 4.5–5.0. They proliferate over a wide temperature range up to 45°C, with an optimum between 30 and 37°C. Actinobacteria proliferate under aerobic conditions at a pH-value of 5–7 and optimal temperatures between 30 and 37°C. Bacteria can proliferate under both aerobic and anaerobic conditions. Here, too, the optimum is a pH-value of 5–7 and temperatures between 30 and 37°C [84].

In addition to the mesophilic microorganisms, whose optimum growth temperature is between 30 and 37°C, thermophilic microorganisms can proliferate over a much wider temperature range (up to 70°C). These thermophilic microorganisms are mainly applied for the controlled biological degradation of plastics in industrial composting processes.

5 Conclusions

When it comes to bioplastics, it is important to use a clear wording and by this to differentiate between biopolymers as a macromolecules and bioplastics as materials ready-to-use as well as the distinction between bio-based and biodegradable plastics. Biodegradability is a system feature of the material microstructure, physico-chemical and microbiological surrounding conditions. Because in nature are a variety of environmental conditions, from industrial composting facilities to sewage treatment plants, soils in a variety of climatic regions, rivers, the beach, sea surface and the seabed, or even the human body, it is important to provide also clear information about the environmental conditions under which the degradation takes place. On the other side standards are needed to reflect biodegradability under the various conditions. In case of compostability, some test standards cover the environmental conditions well, while test standards in other areas, such as degradability in soil and particularly in marine systems, are only available in small numbers and do not reflect the complex environmental conditions well. For future material development, in addition to the establishment of appropriate standards, an extensive research is required to work out a better understanding of the relationships between the environmental conditions of various habitats and microbiology as well as material parameters on the one hand and the resulting degradation mechanisms and kinetics on the other hand.

References

1. Endres H-J, Siebert-Raths A (2011) Engineering biopolymers, München. 978-3-446-42403-6
2. Endres H-J (2017) Bioplastics. Advances in biochemical engineering/biotechnology. Springer, Cham
3. Haldenwanger HHM, Göttner GH (1970) Biologische Zerstörung der makromolekularen Werkstoffe (Chemie, Physik und Technologie der Kunststoffe in Einzeldarstellungen, Band 15). Springer, Berlin
4. [Online] Empfehlung zur Behandlung von kompostierbaren Verpackungen. http://www.nova-institut.de/news-images/20050425-04/IBAW_VVO_Handlungsempfehlung.pdf [date: 2–24, 2011]
5. N.N. (1989) Mikrobiologische Materialzerstörung und Materialschutz; DECHEMA Arbeitsausschuss. Frankfurt
6. Owen S, Kawamura R, Sakota N (1995) Biodegradation of poly-D,L-lactic acid polyurethanes. J Macromol Sci Pure Appl Chem 32(4):843–850
7. Imam SH, Greene RV, Zaidi BR. Biopolymers. Utilizing nature's advanced materials. ACS symposium series, 723

8. Ehrenstein GW, Pongratz S (2013) Resistance and stability of polymers. Carl Hanser Verlag, München, pp 833–884
9. N.N., ISO/DIS:2020 (E), Plastics – Test methods for determination of degradation rate and disintegration degree of plastic materials exposed to marine environmental matrices under laboratory conditions
10. Hocking P et al (2003) Enzymatic degradability of poly(beta-hydroxybutyrate) as a function of tacticity. Macromol Raprid Commun 15(6):447–452
11. Kawai F (1995) Proposed mechanism for microbial degradation of polyacrylate. J Macromol Sci Pure Appl Chem 32, 4, 835–838
12. N.N. (1987) Degradable plastics. Proceedings of the SPI symposium on degradable plastics. Society of the Plastic Industry, Inc., [Hrsg.], Washington
13. Tänzer W (2000) Biologisch abbaubare Polymere, Deutscher Verlag für die Grundstoffindustrie, Stuttgart
14. Kleeberg I (1999) Untersuchungen zum mikrobiellen Abbau von aliphatisch-aromatischen Copolyestern sowie Isolierung und Charakterisierung eines polyesterspaltenden Enzyms, Dissertation, Technische Universität Braunschweig
15. N.N. (1996) DIN V 54900 (1–5); Prüfung der Kompostierbarkeit von polymeren Werkstoffen, Beuth Verlag, Berlin
16. N.N. (2000) EN 13432, Anforderungen an die Verwertung von Verpackungen durch Kompostierung und biologischen Abbau, Beuth Verlag, Berlin
17. N.N. (2006) DINCERTCO. Zertifizierungsprogramm – Produkte aus kompostierbaren Werkstoffen. Berlin: s.n.
18. Harrison JP, Boardman C, O’Callaghan K, Delort A-M, Song J (2018) Biodegradability standards for carrier bags and plastic films in aquatic environments: a critical review. R Soc Open Sci 5:171792. https://doi.org/10.1098/rsos.171792
19. N.N. ASTM D 6691 – 2017, Standard test method for determining aerobic biodegradation of plastic materials in the marine environment by a defined microbial consortium or natural sea water inoculum
20. N.N., ASTM D 6691:2017, Determining aerobic biodegradation of plastic materials in the marine environment by a defined microbial consortium or natural sea water inoculum
21. N.N. ASTM D 7473 2012, Standard Test Method for Weight Attrition of Plastic Materials in the Marine Environment by Open System Aquarium Incubations
22. N.N. ASTM D 7991 – 2015, Standard test method for determining aerobic biodegradation of plastics buried in sandy marine sediment under controlled laboratory conditions
23. N.N. DIN EN ISO 9408, Bestimmung der vollständigen aeroben biologischen Abbaubarkeit organischer Stoffe im wäßrigen Medium über die Bestimmung des Sauerstoffbedarfs in einem geschlossenen Respirometer
24. N.N., EN ISO 9439:2000, Evaluation of ultimate aerobic biodegradability of organic Compounds in aqueous medium – Carbon dioxide evolution test
25. EN ISO 11733:2004, Determination of the elimination and biodegradability of organic compounds in an aqueous medium – Activated sludge simulation test
26. N.N. DIN EN ISO 14851, Determination of the ultimate aerobic biodegradability of plastic materials in an aqueous medium – method by measuring the oxygen demand in a closed respirometer
27. N.N. DIN EN ISO 14852, Determination of the ultimate aerobic biodegradability of plastic materials in an aqueous medium – method by analysis of evolved carbon dioxide
28. N.N. DIN EN ISO 14853, Plastics – determination of the ultimate anaerobic biodegradation of plastic materials in an aqueous system – method by measurement of biogas production
29. N.N. DIN EN ISO 15314, Plastics – methods for marine exposure
30. N.N. DIN EN ISO 18830, Plastics – determination of aerobic biodegradation of non-floating plastic materials in a seawater/sandy sediment interface – method by measuring the oxygen demand in closed respirometer

31. N.N. DIN EN ISO 19679, Plastics – determination of aerobic biodegradation of non-floating plastic materials in a seawater/sediment interface – method by analysis of evolved carbon dioxide
32. N.N. ISO 22404, Plastics – determination of the aerobic biodegradation of non-floating materials exposed to marine sediment – method by analysis of evolved carbon dioxide
33. N.N. ISO 22766, Plastics – determination of the degree of disintegration of plastic materials in marine habitats under real field conditions
34. N.N., ISO-DIS 23832 – 2020, Test methods for determination of degradation rate and disintegration degree of plastic materials exposed to marine environmental matrices under laboratory conditions
35. N.N. ISO/CDIS 23977-1, Plastics – determination of the aerobic biodegradation of plastic materials exposed to seawater – part 1: method by analysis of evolved carbon dioxide
36. N.N. ISO/CDIS 23977-2, Plastics – determination of the aerobic biodegradation of plastic materials exposed to seawater – part 2: method by measuring the oxygen demand in closed respirometer
37. Nakajima-Kambe T, Shigeno-Akutsu Y, Nomura N, Onuma F, Nakahara T (1999) Microbial degradation of polyurethane, polyester polyurethanes and polyether polyurethanes. Appl Microbiol Biotechnol 51:134–140
38. Molitoris HP, Moss ST, de Koning GJM, Jendrossek D (1996) Scanning electron microscopy of polyhydroxyalkanoate degradation by bacteria. Appl Microbiol Biotechnol 46:570–579
39. Dřímal P, Hrnčiřík J, Hoffmann J (2006) Assessing aerobic biodegradability of plastics in aqueous environment by GC-analyzing composition of equilibrium gaseous phase. J Polym Environ 14:309
40. Dřímal P, Hoffmann J, Družbík M (2007) Evaluating the aerobic biodegradability of plastics in soil environments through GC and IR analysis of gaseous phase. Polym Test 26:729–741
41. Eyheraguibel B et al (2017) Characterization of oxidized oligomers from polyethylene films by mass spectrometry and NMR spectroscopy before and after biodegradation by a Rhodococcus rhodochrous strain. Chemosphere 184:366–374
42. Trimpin S, Eichhorn P, Räder HJ, Müllen K, Knepper TP (2001) Recalcitrance of poly (vinylpyrrolidone): evidence through matrix-assisted laser desorption-ionization time-of-flight mass spectrometry. J Chromatogr A 938:67–77
43. Zgoła-Grześkowiak A, Grześkowiak T, Zembrzuska J, Łukaszewski Z (2006) Comparison of biodegradation of poly(ethylene glycol)s and poly(propylene glycol)s. Chemosphere 64:803–809
44. Fontanella S et al (2010) Comparison of the biodegradability of various polyethylene films containing pro-oxidant additives. Polym Degrad Stab 95:1011–1021
45. Sikorska W, Janeczek H (2014) Techniques useful in biodegradation tracking and biodegradable polymers characterization. See http://www.plastice.org/fileadmin/files/EN_Analyses.pdf. Accessed 19 Oct 2017
46. Koutny M, Lemaire J, Delort A-M (2006) Biodegradation of polyethylene films with prooxidant additives. Chemosphere 64:1243–1252
47. N.N. International Organization for Standardization (ISO), ISO 10640:2011 Plastics – methodology for assessing polymer photoageing by FTIR and UV/visible spectroscopy. ISO, Geneva, Switzerland, pp 1–29
48. N.N. BS EN ISO 11266, Guidance on laboratory testing for biodegradation of organic chemicals in soil under aerobic conditions
49. N.N. EN ISO 11721-2, Determination of the resistance of cellulose containing textiles to microorganisms – soil burial test
50. N.N. DIN ISO 14239, Soil quality – Laboratory incubation systems for measuring the mineralization of organic chemicals in soil under aerobic conditions
51. N.N. DIN EN ISO 15473 – Entwurf, Guidance on laboratory testing for biodegradation of organic chemicals in soil under anaerobic conditions

52. N.N. OENORM EN ISO 15985, Determination of the ultimate anaerobic biodegradation under high-solids anaerobic-digestion conditions – method by analysis of released biogas
53. N.N. DIN EN 17033, Biodegradable mulch films for use in agriculture and horticulture – requirements and test methods
54. Endres H-J, Helmke G (2004) Situation der Biogaserzeugung und Potenzial der Biogasproduktion in Niedersachsen. In: Grabkowsky B (ed) Weiße Reihe, vol 23, Vechtaer Druckerei und Verlag
55. Braun R (1982) Biogas – Methangärung organischer Abfallstoffe. Springer, New York
56. Huang SJ (1989) Biodegradation. Compr Polymer Sci 6:597–606
57. [Online] http://www.european-bioplastics.org/media/files/docs/en-pub/FactSheets/FactSheet_Anaerobic_Digestion.pdf. [date: 5–24, 2011]
58. Kitzler A-S, Endres H-J, Nelles M (2012) Recyclingfähigkeit von Biokunststoffen. In: Tagungsband 15. Dialog Abfallwirtschaft MV Rostock, pp 121–132
59. Kitzler A-S, Endres H-J, Nelles M (2011) Vergärung von Biopolymer-Verpackungen – Eine sinnvolle Verwertungsoption? In: Tagungsband – 8. Biogastagung Dresden, Band 81, pp 117–122
60. Endres H-J, Siebert A, Kitzler A-S, Biopolymers – a discussion on end of life options. In: Bioplastics Magazine 01/08, pp 22–25
61. Shimao M (2001) Biodegradation of plastics. Curr Opin Biotechnol 12(3):242–247
62. Sakazawa C, Shimao M et al (1981) Symbiotic utilization of polyvinyl alcohol by mixed cultures. Appl Environ Microbiol 41(1):261–267
63. Sakai K, Hamada N, Watanabe Y (1986) Degradation mechanism of poly(vinylalcohol) by successive reactions of secondary alcohol oxidase and betadiketone hydrolase from Pseudomonas sp. Agric Biol Chem 50(4):989–996
64. Matsumura S, Tomizawa N et al (1999) Novel poly(vinyl alcohol)-degrading enzyme and the degradation mechanism. Macromolecules 32:7753–7761
65. Arenskötter M, Baumeister D et al (2002) Mikrobieller Abbau von Natur- und Synthesekautschuk. BIOforum 3:124–126
66. Kawai F (1995) Breakdown of plastics and polymers by microorganisms. Adv Biochem Eng 52:151–194
67. Welzel K (2003) Einfluss der chemischen Struktur auf die enzymatische Hydrolyse von Polyester-Nanopartikeln, Dissertation, Technische Universität Braunschweig
68. Huang SJ (1985) Biodegradable polymers. Enzyclopedie Polymer Sci Eng 2:220–241
69. Mochizuki M, Hirami M et al (1995) Hydrolysis of polycaprolactone fibers by lipase: effect of draw ratio on enzymatic degradation. J Appl Polym Sci 55:289–296
70. Gan Z et al (1997) Enzymatic degradation of poly(e-caprolactone) film in phosphate puffer solution containing lipases. Polym Degrad Stab 56:209–213
71. Eldstäter C, Erlandsson B et al (2000) The biodegradation of amorphous and crystalline regions in film-blown poly(e-caprolactone). Polymer 41:1297–1304
72. Witt U, Müllera R-J, Deckwer W-D (1995) New biodegradable polyester-copolymers from commodity chemicals with favorable use properties. J Environ Polymer Degrad 3(4):215–223
73. Tokiwa Y, Suzuki T (1981) Hydrolysis of copolyesters containing aromatic and aliphatic ester blocks by lipase. J Appl Polym Sci 26(2):441–448
74. Marten E (2000) Korrelation zwischen der Struktur und der enzymatischen Hydrolyse von Polyestern, Dissertation, Technische Universität Braunschweig
75. Palmisano AC, Pettigrew CA (1992) Biodegradability of plastics. Bioscience 42(9):680–685
76. Lenz RW (1993) Biodegradable polymers. Adv Polym Sci 107:1–40
77. [Online] Specification for Composted Materials. http://www.wrap.org.uk/downloads/PAS_100.52b60645.6584.pdf [date: 3–31, 2005
78. [Online] http://www.european-bioplastics.org/media/files/docs/en-pub/FactSheets/FactSheet_Industrial_Composting.pdf. [date: 11–29, 2009]

79. Lefebvre F, Daro A, David C (1995) Biodegradation of polycaprolactone by microorganisms from an industrial compost of household refuse. Part II. J Macromol Sci Pure Applchem 32 (4):867–873
80. Jendrossek D (1998) Microbial degradation of polyesters: a review on extracellular poly (hydroxyalkanoic acid)depolymerases. Polym Degrad Stab 59:317–325
81. Kleeberg I, Hetz C et al (1995) Biodegradation of aliphatic-aromatic copolyesters by Thermomonospora fusca and other thermophilic compost isolates. Appl Environ Polymer Degrad A 64(5):1731–1735
82. Tokiwa Y, Suzuki T (1977) Purification and some properties of polyethylene adipatedegradation enzyme produced by Penicillium sp. Strain 14 – 3. Agric Biol Chem 41 (2):265–274
83. Schnabel W (1981) Polymer degradation – principles and practical applications. Carl Hanser Verlag, München
84. Gugumus F, Gächter R, Müller H (Hrsg) (1989) Kapitel antioxidantien. In: Kunststoff-additive, 3. Auflage, Carl Hanser Verlag, München

Biological and Ecological Impacts of Plastic Debris in Aquatic Ecosystems

Dannielle Senga Green

Contents

1 Plastic Debris in Aquatic Ecosystems: From Contamination to Pollution 112
2 Effects of Plastic Debris on Individual Organisms 115
  2.1 Entanglement 115
  2.2 Ingestion of Macro-sized Plastic 116
  2.3 Ingestion of Micro-sized Plastic 117
3 Physical, Chemical and Biological Characteristics of Microplastics 117
  3.1 Physical Characteristics of Microplastics: Shape and Size 118
  3.2 Chemical Composition of Microplastics: Polymer Type, Additives and Persistent Organic Pollutants 119
  3.3 Biological Aspects of Microplastics: As Habitats for Microbes Potentially Increasing the Spread of Antimicrobial Resistance 120
4 Wider Ecological Effects of Plastic Debris on Assemblages and Ecosystem Functioning 122
  4.1 Alterations to Benthic Assemblages Due to Plastic Debris: Evidence from Manipulative Experiments 123
  4.2 Possible Effects of Plastic Debris on Ecosystem Processes: Nutrient Cycling and Primary Productivity 124
5 Closing Remarks and Recommendations 125
References 127

Abstract Plastic debris is now ubiquitous in aquatic ecosystems worldwide and may impact different biological levels of organisation, with effects ranging from individual organisms to ecosystem functioning. Demonstrating these effects is not always straightforward, and there is uncertainty at every level. In particular, understanding of the wider impacts on biodiversity and ecosystem functioning is challenging, and little research has been done in this area. This chapter gives a broad overview of hierarchical impacts of macro- and microplastic pollution on aquatic ecosystems. Topics include the potential for microplastics to spread antimicrobial

---

D. S. Green (✉)
Applied Ecology Research Group, School of Life Sciences, Anglia Ruskin University, Cambridge, UK
e-mail: dannielle.green@aru.ac.uk

Friederike Stock, Georg Reifferscheid, Nicole Brennholt, and Evgeniia Kostianaia (eds.),
Plastics in the Aquatic Environment - Part I: Current Status and Challenges,
Hdb Env Chem (2022) 111: 111–134, DOI 10.1007/698_2020_509,
© Springer Nature Switzerland AG 2020, Published online: 19 June 2020

resistance and a summary of current knowledge concerning wider ecological impacts of macro- and microplastic debris such as changes to assemblage composition and structure and effects on nutrient cycling and primary productivity. The potential impacts of biodegradable plastics are also discussed and, in most cases, have similar effects to plastics made from conventional polymers emphasising that the same precautions need to be taken to ensure that these items do not become the litter of the future.

Keywords Biodiversity, Ecosystem services, Microplastics, Plastic litter, Polylactic acid (PLA)

1 Plastic Debris in Aquatic Ecosystems: From Contamination to Pollution

Natural ecosystems are prone to many stressors resulting from human activities, such as habitat destruction, climate change, invasive species and contamination with anthropogenic materials also known as "anthropogenic debris". Although this includes blown glass, treated wood, metal and galvanised rubber, over the past three decades, plastic has accounted for the majority of anthropogenic debris [1] and is still increasing in abundance [2].

Plastic debris enters aquatic environments in a variety of sizes, from large synthetic fishing nets measuring several hundreds of metres to packaging items such as plastic bags and bottles measuring tens of centimetres down to micro-sized plastics termed "microplastics". The definition of the size of microplastics is under debate by scientists; however, they are commonly defined as being in the range of 5 mm down to 1 nm [3]. Alternatively, others define microplastics as being less than 1 mm [4]. Microplastics are currently the most abundant form of solid waste on Earth [5], and due to their small size, they are bioavailable to organisms throughout the whole food web. Once ingested by an organism, they can elicit a response via their physical presence (possibly altered by shape), their chemical composition (including plasticisers and persistent organic pollutants) or their biological hitchhikers (microbes including pathogens).

Global plastic production is still growing reaching 359 million tonnes in 2018, the majority of which is packaging [6]. Geyer et al. [7] estimated that of all of the plastic produced globally until the year 2015, only 9% has been recycled, 12% incinerated and 79% either sent to landfill or become litter in the environment. They also estimated that of the items produced, more than 40% of them were designed for "single-use" purposes [7]. Mirroring this, almost half (average of 49%) of the plastic debris found in beach clean-ups is composed of single-use items such as food wrappers, plastic bottles and plastic bags [1, 8]. The constant supply of plastic debris to aquatic habitats results in an ever-growing reservoir of plastic waste. Due to its durability, plastic can persist in the environment, possibly for centuries, with fragmentation of larger items providing a constant source of microplastics [2]. Microplastics which are produced intentionally in microscale are referred to as "primary microplastics" (e.g. microbeads, sandblasting particles or pre-production granules). Microplastics produced indirectly from the fragmentation of larger plastic items to smaller pieces are termed as "secondary microplastics" (e.g. synthetic fibres from washing clothing or flaking from packaging).

The growing issue of contamination of aquatic habitats with plastic debris has taken centre stage of many conservation efforts in recent years, especially since the launch of the British Broadcasting Channel's Blue Planet 2 hosted by Sir David Attenborough. This documentary, underpinned by the efforts of many scientists the world over, helped raise awareness of the prevalence of plastic debris in the world's oceans. In order to make scientifically informed policy decisions, however, we must demonstrate whether this contamination is causing harm. If a contaminant has the potential to cause adverse biological effects to organisms in the receiving habitat, then it may be classified as a "pollutant" [9]. In order to classify plastic contamination as pollution, evidence needs to demonstrate that it causes biological and/or ecological harm [10]. Furthermore, in order to demonstrate that an ecological impact has occurred, quantitative data on a population and assemblage or community level is needed [10]. Moreover, an ecological impact could include alterations to ecosystem processes, such as biogeochemical cycles or primary productivity.

A myriad of different types of synthetic plastic polymers are present in the environment, and their density can determine their position in the aquatic environment. For example, low-density polymers such as polyethylene and polypropylene are typically more abundant on the sea surface than denser polymers such as polyester, polyamide and acrylics, which sink quickly and accumulate in sediments [11]. Once low-density polymers are colonised by biofilms, they will also sink into sediments [12, 13]. Plastic debris contaminates aquatic ecosystems worldwide and is present in marine [14] and freshwater [15] water columns, marine beaches [16] and deep sea [17] and in coastal [18] and freshwater lake sediments [19]. It is also found within many marine animals including invertebrates [20], fish [21], reptiles [22] and mammals [23] and freshwater animals including invertebrates [24] and fish [25]. Scientists and the general public are concerned about plastic contamination, and there have been calls to label microplastics as hazardous materials [26], to embed an international agreement to stem the flow of plastic into the oceans and rivers [27] and to facilitate change by holding a Global Convention on Plastic Pollution to enliven collaboration between governments, plastic production industries, scientists and citizens [28]. Despite concerns, regulation to act is sometimes hampered by a lack of knowledge and certainty about the real risks of plastic debris for the environment and specifically about how plastic debris could lead to ecological harm. This is partly a result of a myriad of data arising from experiments on single species, with very few reporting wider ecological effects at higher levels of biological organisation such as populations, communities and ecosystems [29, 30]. Moreover, interpretation of experimental data is often complicated by the fact that exposure experiments use densities of microplastics that far exceed those densities reported in the environment

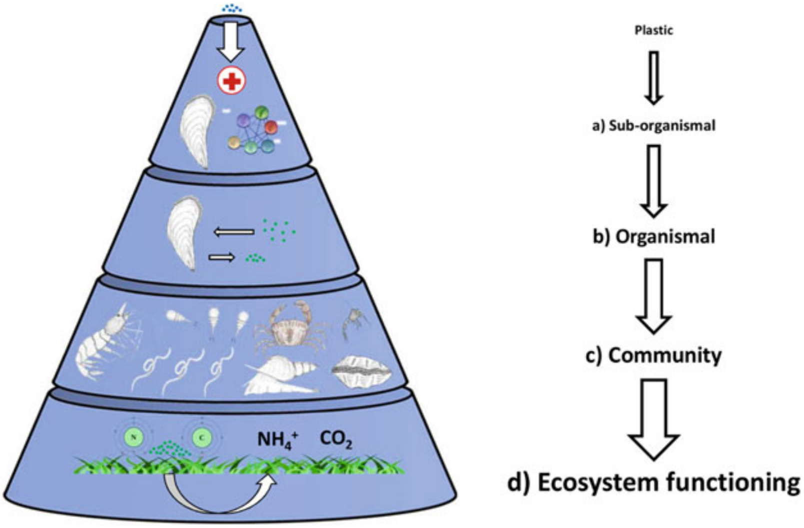


Fig. 1 Hierarchical impacts of plastic debris in aquatic ecosystems from (**a**) sub-organismal effects on the health of individual organisms to (**b**) effects on the biological functioning (e.g. feeding rate, respiration, reproduction rate) of organisms that could lead to wider effects on populations to (**c**) alterations to the structure or composition of assemblages and finally (**d**) alterations to ecosystem processes (e.g. primary productivity, nutrient cycling) that have wider consequences for ecosystem functioning

[31, 32]. Nonetheless, there is a wealth of research attempting to understand the impacts of plastic debris in the aquatic environment. The resulting impacts are, however, context dependent on a number of factors including the size, shape and chemical composition of the plastic debris, on how much of it is present and also on the characteristics of the receiving environment (including both the type of organism and the habitat being contaminated).

In the marine environment, more than 600 species are reported to interact with plastic debris, and of these, at least 17% are on the IUCN Red List and are listed as “threatened” or “near threatened” [33]. In general, effects of marine plastic litter can result from either entanglement or ingestion. For example, larger macroplastic debris such as lost or discarded fishing gear will most likely cause entanglement, whilst smaller items such as bottle caps and microplastics can be ingested.

Impacts of plastic debris can be lethal or sublethal and occur on a hierarchy of scales ranging from effects on the health of individual organisms, to their biological activities and alterations to assemblages and biodiversity and finally to effects on ecosystem processes and ecosystem functioning (Fig. 1). Understanding the wider effects of plastic debris is challenging, and most studies have focused on single species and individual effects. For example, many studies report on the effects of plastic debris on the individual organism (Fig. 1). These often assess cellular or subcellular endpoints associated with health at a sub-organismal level, for example, measuring immune and stress responses using enzyme assays [30]. Other studies assess the impacts of plastic debris on the biological activity of the individual organism (Fig. 1) by measuring, for example, movement, feeding, growth and reproductive rates. These impacts could cascade to wider alterations at the level of whole assemblages whereby the composition and structure of assemblages are changed (Fig. 1). Finally, there is some evidence that plastic debris could alter ecosystem processes such as primary productivity and biogeochemical cycles and, in turn, affect ecosystem functioning (Fig. 1). Very few studies have attempted to assess these wider effects.

This chapter will explore some of the evidence reporting on each tier of hierarchical impacts, from biological effects on individual species to wider ecological impacts on ecosystem functioning, arising from macro- and microplastic litter in aquatic ecosystems.

2 Effects of Plastic Debris on Individual Organisms

2.1 *Entanglement*

The Great Pacific Garbage Patch is still growing in size, and derelict fishing nets account for almost half of the mass of plastic waste in the gyre [34]. Derelict fishing debris, such as nets, ropes, cages and nylon lines, can through tissue abrasion and smothering damage sessile habitat-forming marine invertebrates such as corals and sponges [35, 36]. This can significantly fragment and reduce the extent of coral reefs [37] which provide important habitat for many other species. Abandoned fishing gear can also become colonised by corals, further complicating removal of the settled debris [36]. Regarding mobile animals, "ghostfishing" (the entanglement of marine animals in derelict fishing gear) has been documented for hundreds of species of turtles, seals, whales, seabirds, fishes and invertebrates [38] and poses a risk of mortality. It is the greatest anthropogenic threat to some endangered species, for example, monk seals [39, 40] and turtles [41]. See Laist [42] and Gall and Thompson [33] for a more detailed account of interactions of marine animals with derelict fishing gear. Plastic ropes, nets and lines are the most common causes of entanglement [33] and can lead to lethal or sublethal effects. Entanglement can cause rapid mortality if the animal is asphyxiated (e.g. occlusion of the blowhole in whales [43]), whilst a slower death or reduced fitness can result from injury to the dermal tissue and restricted movement resulting in a decreased ability to capture prey [38] or to evade predation [44]. Pinnipeds, for example, can carry debris wrapped around themselves for sustained periods, and either often die as a result from the chronic wounds as they cut into flesh as animals grow [45] or sometimes this can lead to strangulation [46]. Global estimates indicate that entanglement of pinnipeds with marine debris causes mortality in 16–80% of the cases [47]. The chances of becoming entangled, and the resulting impact, are dependent on several factors including the behaviour of the animal, habitat type, local weather conditions and the type of marine debris involved [47]. Recording and tracking marine debris in the open oceans is difficult but vital in order to prioritise protection of species most at risk. Wilcox et al. [48] combined physical models of oceanic drift with turtle distribution data in order to identify hotspots of potential impact from marine debris. Such models can help guide regulation, enforcement and conservation action regarding ghostfishing. The accumulation patterns of large marine debris can be further ground-truthed and monitored using aerial imagery and spatial analysis [49]. In relative terms, entanglement, especially of large animals, is a visible issue easy to identify. It is more difficult, however, to monitor and quantify the impacts of ingested smaller plastics.

2.2 *Ingestion of Macro-sized Plastic*

A variety of macro-sized plastic debris items, such as plastic bags and straws, but also microplastics, have been found in the digestive system of aquatic organisms including 100% of marine turtle species, ~40% of seabird species and 50% of marine mammal species [38], but it can be difficult to infer a direct causal link of mortality to the ingested plastic. Although the conditions under which death occurs are rarely known, in some cases, ingested plastics can rapidly lead to death, and so causality can be determined. This has been demonstrated in whales where two sperm whales suffered gastric impaction due to ingestion of netting [50] and another had a ruptured stomach containing over 7 kg of plastic [51]. It has also been noted in birds, for instance, a Magellanic penguin with a straw that perforated its internal organs [52] and a great shearwater with plastic debris obstructing the passage from the oesophagus to the proventriculus [53], as well as in several green sea turtles with obstruction of the intestine or oesophagus by plastic debris [54]. For smaller organisms, such as fish and invertebrates, however, there is a lack of direct evidence of mortality resulting from ingestion.

Direct mortality resulting from ingestion of macro-sized plastic debris is less likely than sublethal impacts that compromise fitness and result in long-term health problems. Direct physical damage causing ulcerations, perforations and inflammation of the digestive system can reduce its functionality and affect health. This was noted in a necropsy of a leatherback turtle where severe bacterial gastroenteritis was likely caused by a piece of plastic penetrating the gastric mucosa [55]. Plastic debris can also block off parts of the digestive system, slowing down digestion and preventing the nutrients from food from being properly absorbed and causing the individual to become emaciated. A green sea turtle with plastic debris obstructing the gastrointestinal tract was in an emaciated condition but regained normal behaviour and health after removal of these items, thus indicating a likely cause and effect [56]. Additionally, if the space within an animal stomach is occupied by plastic debris, this can leave less space to accommodate food and can lead to a feeling of satiation, reducing the impulse to feed and subsequently reducing energy intake. This was verified experimentally by Ryan [57] who found that chickens fed with polyethylene pellets had reduced growth rates and ate less food.

2.3 Ingestion of Micro-sized Plastic

The occurrence, abundance and biological effects of microplastics within aquatic organisms have been thoroughly reviewed [32, 58–61]. It is clear that a wide range of marine organisms ingest microplastics. These include cetaceans (whales and dolphins [62, 63]), all seven species of sea turtles [64], sea birds [65], coastal, pelagic and demersal fish [21, 66], deep sea fish [67] and invertebrates including amphipods [68], brittle stars and sea stars [69]. Similarly, freshwater organisms including birds [70] and fish from rivers [25, 71] and lakes [72], together with a range of invertebrates including macroinvertebrates [24], bivalves [73] and insect larvae [74], have been found to contain microplastics in their digestive organs. Despite the overwhelming body of literature reporting ingestion of microplastics, it is possible that some species do not ingest or retain microplastics. For example, around 40% of the species of marine fish tested for microplastics globally showed 0% ingestion rate [75]. There may be a publication bias of results such as these (i.e. not being published in the scientific literature), but reporting 0% plastic ingestion rates needs to be encouraged as it represents important information from a monitoring and risk management perspective.

Once ingested, microplastics can cause impaired health at a molecular or cellular level. These include general stress responses such as oxidative stress (for example, in oysters [76] and sea bass [77]), neurotoxicity in sea bass [77], immunological responses [78], activation of detoxification pathways in coral [79] and alterations to the proteome, for example, genes involved in inflammation and structural development in mussels were altered by microplastics [80].

In addition, microplastics can cause changes to biological processes in some animals, such as respiration in lugworms [81] and oysters [82]; feeding rates in mussels, oysters [83], copepods [84] and water fleas [85]; as well as growth and reproduction in copepods [84], oysters [76] and water fleas [85]. Population level effects arising from reduced fitness and reproduction could lead to economic (loss of stock for aquaculture and fisheries) and ecological impacts (alterations to biodiversity). Microplastics can also alter key biological processes (see Sect. 4), potentially compromising the ability of ecosystem engineers to build reefs or to turn over the sediment. For example, the attachment strength of blue mussels halved after exposure to HDPE microplastics [80], and there was a reduced volume of sand overturned by lugworms exposed to microplastics [81].

3 Physical, Chemical and Biological Characteristics of Microplastics

The term “microplastic” is an umbrella term that encompasses a wide range of different types of particles. In reality, the resulting effect that microplastics have on aquatic organisms can vary depending on their (a) physical characteristics (i.e. the shape and size of the particles), (b) chemical composition (i.e. polymer type, plasticisers and persistent organic pollutants from the environment) and/or (c) biological communities (i.e. the presence of microbes on their surface).

3.1 Physical Characteristics of Microplastics: Shape and Size

Microplastics differ in shape, for example, primary microplastics, such as microbeads, are usually spherical beads, whilst secondary microplastics are fibres, irregular fragments or films. The shape of microplastics could be important in determining their impact as it could affect their bioavailability, the time taken to pass through the gut and the propensity to accumulate within organisms [61, 86]. Of the very few studies to compare the effects of different shapes on aquatic organisms, some have found beads to be more benign than irregular shapes such as fragments or fibres and believe this is due to longer residence times of irregular shapes in the gut or potentially entanglement within the organism. For example, irregular-shaped polyethylene microplastic fragments had a stronger effect than spherical polyethylene microplastics on sheepshead minnows, decreasing their mobility [87]. Similarly, polypropylene fibres had a more toxic effect than polyethylene particles on amphipods [88], although the effects of shape cannot be separated from that of polymer type. Others [89, 90] concluded that size has a greater effect than shape; however, neither of these studies used a balanced design allowing them to separate the effects of shape from that of size or polymer type. Future studies aiming to tease apart the effects of shape need to avoid confounding factors by maintaining equivalent sizes and polymer types for a range of different shapes in their experiments.

There has been considerably more effort from the scientific community to decipher the effects of microplastics of different sizes than those of different shapes. Smaller particles are likely to be more bioavailable than larger particles due to their ability to be ingested by a greater range of smaller organisms, such as zooplankton, and to pass through the food web. Furthermore, nanosized plastic particles are capable of passing through the cell barrier (see review by da Costa et al. [91]) and accumulating in the tissues and organs of both aquatic invertebrates [92] and fishes [93]. There is even evidence that nanoplastics can be transferred maternally to offspring via accumulation in egg sacks as demonstrated for zebra fish [94]. It seems intuitive, due to their greater bioavailability and greater surface area to accumulate chemical toxins, that smaller, nanosized particles would be more likely to lead to toxic effects than larger micro-sized particles, and, indeed, some evidence supports this. For example, 100 nm polystyrene (PS) nanoplastics were more toxic than 5 μm PS microplastics to broad beans as they were able to enter the root tissue likely blocking cell connection or cell wall pores and preventing transport of nutrients [95]. Similarly, in marine copepods, nanosized (0.05 μm) PS beads caused lethal impacts (a greater mortality rate), whilst 0.5 and 6 μm PS beads caused sublethal effects including a decrease in fecundity [96]. Interestingly, however, some organisms respond more strongly to medium or larger microplastics than they do to smaller ones. For instance, Bour et al. [97] found that larger (125–500 μm) microplastics were retained by bivalves and decreased their energy and protein reserves, whilst smaller (4–6 μm) microplastics had negligible effects. Consistent with these results, a comparison of the effects of 0.1, 1 and 5 μm polystyrene particles on aquatic nematodes showed that 1 μm particles accumulated the most often causing the most extensive damage to the reproductive system and greatest mortality [98]. It is possible that toxicity of microplastics is size dependent, altering with their potential to accumulate in organisms and to induce damage via mechanical injury or insufficient nutrition.

3.2 Chemical Composition of Microplastics: Polymer Type, Additives and Persistent Organic Pollutants

Many different polymers are manufactured for use in various applications from single-use packaging to long-lasting items such as toys and electrical goods. Additives such as plasticisers, stabilisers, pigments, fillers and flame retardants are required to improve the physical properties of plastics, such as colour, fire resistance and rigidity. The majority of studies have only tested one polymer type at a time, but given the range of different polymer types found in the environment, it is important to compare their effects in order to prioritise those which may pose more of a risk than others and could be prioritised for regulation. There is some evidence that certain polymers have a stronger effect than others, for example, leachate from polyvinyl chloride (PVC) induced greater mortality of barnacle larvae than leachate from PP [99], whilst PVC microplastics had a marginally stronger effect than PE or PLA microplastics on the metabolism of lugworms [81]. However, a comparison of the effects of five different (PA, PE, PP, PS and PVC) polymer types on zebra fish and nematodes found no polymer-specific effects [98].

It is important to note that biodegradable microplastics may also persist in the environment and could have the same effect as non-biodegradable microplastics. Several experiments in marine mesocosms have revealed that bio-based, biodegradable polylactic acid (PLA) microplastics can have the same effects as non-biodegradable polyethylene microplastics, disrupting the immune system of mussels [80] and respiration in oysters [82] and in lugworms [81], reducing biodiversity and abundance of organisms [82], and decreasing the biomass of primary producers [81, 83]. Similarly, in freshwater biodegradable polyhydroxybutyrate (PHB) and non-biodegradable polymethylmethacrylate (PMMA), microplastics both led to a decrease in biomass of a freshwater amphipod [100]. Although very little is known about the behaviour and breakdown of biodegradable microplastics in aquatic habitats, a recent study found that secondary nanoplastics released from PHB microplastics persist and have negative effects in freshwater environments [101]. Biodegradable plastics will likely become more dominant as packaging in the future, possibly replacing some conventional plastics, and therefore there will be a greater risk of them becoming the litter of the future. We must therefore raise awareness that these polymers are not benign and, as with any other plastic, must be appropriately disposed of.

When present in aquatic habitats, microplastics can also adsorb other persistent organic pollutants (POPs) from the water column, such as phthalates, phenolic endocrine disruptors (e.g. bisphenol A), polycyclic aromatic hydrocarbons (PAHs) and also heavy metals, thereby posing a combined physical and chemical threat to aquatic organisms if ingested [102]. The large surface area to volume ratio of microplastics enables them to readily adsorb many POPs from the surrounding water. Indeed, some of the most commonly occurring microplastics, polyethylene and polypropylene, have a greater propensity to adsorb persistent organic pollutants than naturally occurring particles [103]. Additives and the adsorbed toxins may leach out into organisms that ingest the microplastics [104, 105]. Although there is potential for some of these chemicals, such as phthalates and bisphenol A, to accumulate and biomagnify in aquatic food webs, there is to date little evidence to suggest that microplastics increase the chemical contamination of organisms when compared with other environmental sources (i.e. water, sediments and trophic transfer; see Barboza et al. [102] for review). Even so, it is possible that the combination of microplastics with chemical contaminants could modulate their toxicity, and this has been the focus of many studies [32]. For example, combining microplastics with chromium decreased predatory performance and inhibited neurological enzyme activity (AChE) of the common goby [106]. However, microplastics lessened the effects of bisphenol A on water fleas [107]. Indeed, Horton et al. [108] noted no differences in the effects of organic pesticides on water fleas with and without microplastics. To summarise, our understanding of the combined effects of other environmental chemical contaminants with microplastics is still limited and warrants further investigation.

3.3 Biological Aspects of Microplastics: As Habitats for Microbes Potentially Increasing the Spread of Antimicrobial Resistance

As discussed above, microplastics can present a double-edged sword to organisms, inflicting harm via their physical presence by blocking feeding passages and also by transferring chemical contaminants [86]. There may also be an additional biological effect arising from microbes associated with microplastics. The surfaces of microplastics support taxonomically distinct microbial assemblages [109] with a greater abundance of pathogens, compared to those colonising natural particles or the water column [110, 111].

Antimicrobial-resistant (AMR) microbes have been found extensively in aquatic environments [112] and can occur following the introduction of antimicrobial compounds, such as antibiotics from aquaculture [113], agricultural run-off [114]

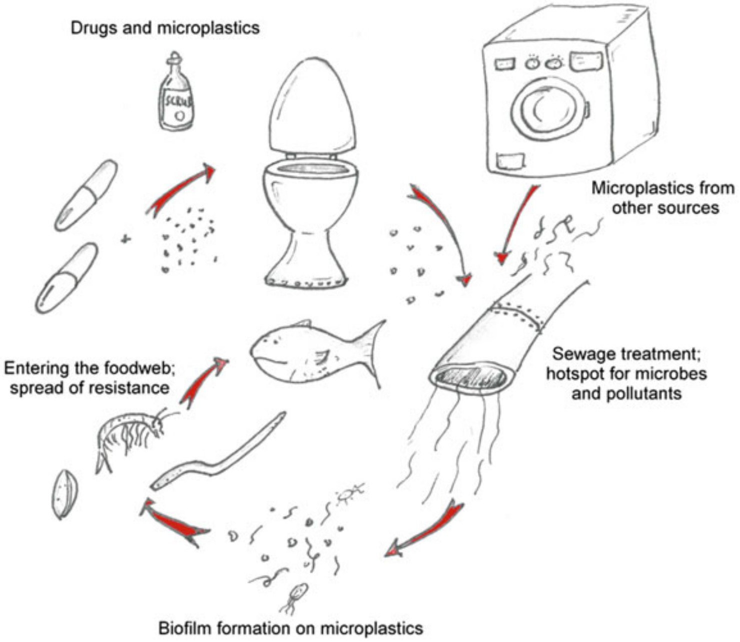


Fig. 2 Potential mechanism for the spread of antimicrobial-resistant bacteria on microplastic particles. Microplastics and antibiotic compounds are simultaneously discharged into wastewater facilities. Here, the microplastics adsorb antibiotics and also become colonised by microbes which become resistant to the adsorbed antibiotics. Microplastics covered in a biofilm of antimicrobially resistant microbes enter the aquatic environment potentially spreading resistance throughout the food web

or wastewater treatment plants [115]. Certain therapeutic antibiotics can pass through the body largely unaltered, for example, during treatment of urinary tract infections. Furthermore, nontherapeutic doses of antibiotics are used as growth promoters in agriculture. The presence of sublethal levels of antimicrobial compounds in wastewater can select for resistant strains giving rise to microbes that are resistant to antimicrobial drugs. Within the environment, microbes can spread their resistance genes among other members of the microbial community via horizontal gene transfer [116, 117] and pose a considerable threat to human health.

Wastewater is a major source of microplastics [118], and antibiotic compounds can adsorb onto microplastics [119] thereby enabling them to play an important role in the transport of water-borne antibiotics [120]. This means that they are likely to play an important role in the evolution and spread of AMR microbes through aquatic food webs (summarised in Fig. 2).

Microbes in biofilms, such as those developed on microplastics, are less susceptible to antibiotics than free-living cells in planktonic culture [121]. Furthermore, horizontal transfer of genes encoding AMR in microbes occurs faster within biofilms [122]. As such, a greater rate of gene transfer was determined in bacteria on microplastics than in those that are free-living in the water column of a laboratory study [123] and within a microcosm sediment experiment [124]. Furthermore, recent sampling in the natural environment in the North Pacific Gyre unveiled that both microplastics and macroplastics are indeed already a reservoir of AMR microbes [125].

Microplastics distributed by rivers and ocean currents persist both in the water column and in sediments, from lakes, rivers and shorelines to the deep ocean floor, and occur in most aquatic habitats worldwide. Given the ubiquitous dispersal of microplastics, AMR genes and microbes carrying them could spread even further than anticipated within freshwater and marine habitats. The global consumption of both antimicrobial compounds and plastic are still increasing; therefore, interaction of these two global stressors could become a major problem in the near future exacerbating the spread of resistant microbial strains also known as “super bugs”.

Although no experiments have explicitly tested the ecotoxicological and/or environmental effects of microplastics laden with AMR microbes, several studies have tested the combined effects of antibiotics and microplastics. For example, Prata et al. [126] conducted a 96-h toxicity experiment to assess the effects of the antibiotic doxycycline with and without plastic microbeads on a marine microalga and found that microbeads increased the toxicity of the antibiotic. Similarly, combined exposure to microplastics and the antibiotic roxithromycin resulted in greater bioaccumulation of roxithromycin in the tissues of the freshwater fish red tilapia than in treatments with roxithromycin alone [127]. Interestingly, the same study found that, despite increasing bioaccumulation, microplastics could mitigate the neurotoxicity and oxidative damage caused by roxithromycin [127]. The effects of combined stressors with microplastics are of growing interest, but more research will be required in order to understand these complex effects.

4 Wider Ecological Effects of Plastic Debris on Assemblages and Ecosystem Functioning

Floating natural debris such as macroalgae, volcanic pumice and timber have played an important role for millions of years, increasing dispersal of sessile and mobile organisms, especially those without a pelagic larval stage. For example, some species of gastropod such as *Littorina saxatilis* are direct developers, and flotsam enhances their range, allowing them to colonise new areas and to increase genetic diversity [128]. Floating anthropogenic marine debris, however, has added a new dimension to the dispersal opportunities of potential rafters. A recorded 387 taxa, including microorganisms, seaweeds and invertebrates, have been found rafting on floating litter in the oceans [129]. For a more detailed account of the types of organisms and the prevalence of rafting on anthropogenic marine debris in different locations, please see Kiessling et al. [129]. Plastic debris often occurs in areas where natural debris is not abundant [130], and, due to being lightweight, it also floats longer and therefore travels further than most other types of natural flotsam [131]. In this way, plastic debris has the potential to act as a vector transporting nonindigenous species into new locations where they could become invasive and could alter assemblages in receiving habitats. The extent to which plastic debris contributes to the spread of invasive species is not well quantified; however, further research is needed [132]. In addition to transportation by rafting, plastic debris, either larger items [129] or microplastics [109], provides a novel substratum for colonisation and can act as habitat. For example, microplastics, in the size range of millimetres, provide habitat for several types of microorganisms including bacteria, diatoms, coccolithophores, dinoflagellates and even some invertebrates [133]. It is possible that they can even sustain these floating communities by providing a food source to those microorganisms capable of biodegrading polymers thereby creating a self-sustaining “plastisphere” with unique communities [109]. Although many plastics have lower density than water and float, once covered in a biofilm, their density increases, and they sink [12, 13]. In this way, benthic habitats are likely the ultimate sink for the majority of macro- and micro-sized plastic debris in marine and freshwater environments.

4.1 Alterations to Benthic Assemblages Due to Plastic Debris: Evidence from Manipulative Experiments

Once accumulated in the benthic environment, plastic debris has the potential to alter the structure and composition of assemblages. This effect has been demonstrated using field experiments with both rigid [134] and flexible [135] plastic debris. The addition of plastic and glass debris to a subtidal sediment habitat resulted in an increase in the total abundance and the number of species of mobile megafauna compared to control sediments after 1 year. The availability of hard substratum also allowed for increased settlement of sessile invertebrates such as ascidians and sponges [134]. Subsequent examination of the macrofaunal assemblages in sediments from the same experiment found that those assemblages associated with experimentally added plastic and glass bottles were altered, driven by an increase in opportunistic macrofaunal species [136]. Another field experiment on an intertidal muddy shore showed that when littered with either conventional (HDPE) or biodegradable (starch-based PLA) plastic bags, assemblage structure of the infaunal macrofauna was altered as the sediment beneath became anoxic and was dominated by opportunistic species [135]. Additionally, a field experiment found little deterioration of biodegradable and oxo-biodegradable plastic bags in marine habitats after 3 years [137], suggesting that they may not offer any advantage over conventional bags in the context of reducing marine litter. Logistical challenges make field experiments testing assemblage level effects of plastic debris rare; however, they provide a level of realism not achievable in laboratory experiments and hence provide important information to policy-makers about the real environmental risks posed by plastic debris.

It is even more challenging to test the effects of microplastics in the field, but a compromise between the realism of field experiments and the feasibility of laboratory experiments can be obtained by using outdoor mesocosms. Outdoor mesocosm experiments using natural flowing seawater and intact sediment cores assessed the impacts of conventional or biodegradable microplastics on invertebrate assemblages from three different habitats [82, 83]. In sandy habitats dominated by flat oysters, the addition of 80 μg L^{-1} of either conventional (HDPE) or biodegradable (PLA) microplastics caused a reduction in the number of species and in the overall abundance of organisms [82]. Similarly, in a follow-up experiment, in muddy sediment dominated by flat oysters, the addition of 25 μg L^{-1} of the same types of microplastics resulted in a shift in community composition whereby opportunistic oligochaetes became dominant and predatory polychaetes declined [83]. Meanwhile muddy sediments dominated by blue mussels showed no change in assemblage structure or composition in response to microplastics [83]. This emphasises that the effects of microplastics can be context dependent and are not easy to predict even when comparing one sedimentary habitat to another.

4.2 Possible Effects of Plastic Debris on Ecosystem Processes: Nutrient Cycling and Primary Productivity

Sedimentary habitats are important sites for nutrient cycling which, in broad terms, is the movement and exchange of organic and inorganic matter back into the production of living matter. Organic matter (e.g. humus) is typically provided to marine sediments by benthic photosynthesis or by the deposition of dissolved and particulate organic matter from the water column. Organic matter in surface sediments is further broken down by microbes and releases inorganic nutrients (e.g. nitrate or ammonium) to the overlying water thereby supporting primary productivity of photosynthetic organisms (e.g. microalgae). Benthic nutrient (re)mineralisation is vitally important and can provide up to two thirds of the nutrients required for primary production in coastal ecosystems [138]. Any alteration to these processes, such as those arising from plastic debris, could lead to cascading effects for benthic and pelagic habitats. As with assemblage level effects, very little research has focused on assessing the impacts of plastic debris on ecosystem processes. Experiments in marine sedimentary habitats, however, found that conventional or biodegradable plastic bags [135] or microplastics [83], decreased the flux of inorganic nutrients (including ammonium and silicate) from the sediment and reduced the biomass of microphytobenthos (microscopic primary producers in sediment). Aquatic primary producers can be macrophytes (seaweed or macroalgae) or microphytes (phytoplankton, microalgae or microphytobenthos), and, like terrestrial primary producers, they form the basis of aquatic food webs and are essential to the functioning of ecosystems. Microalgae are important primary producers and, as well as sequestering carbon, produce the majority of the world's oxygen [139]; therefore, any effects from microplastics will likely have wider impacts on the ecosystem. Even small disruptions of algal populations may lead to cascading impacts on aquatic food webs.

Reviews of the effects of microplastics on aquatic primary producers summarised that microplastics can alter photosynthesis, growth, chlorophyll content, gene expression, colony size and morphology, possibly due to adhesion and/or transfer of adsorbed pollutants from microplastics [140, 141]. Microplastics can adhere onto the surface of micro- [142] and macroalgae [143], acting as a vector for microplastics into aquatic food webs. This adherence can also cause aggregates to be formed altering the sinking rate of microalgae, by (depending on the density of the polymer) increasing or decreasing their buoyancy and causing the microalgae to sink more, or less, rapidly [13, 144, 145].

In marine pelagic ecosystems, zooplankton, such as copepods, contributes to marine nutrient cycling by consuming and subsequently repackaging particulate organic matter into dense faecal pellets with high sinking velocities. These faecal pellets play an important role in the biological pump, transporting organic matter, nutrients, carbon and energy to deeper waters and benthic habitats. Microplastics can reduce the sinking speed of copepod faeces, marine snow and aggregates by increasing the buoyancy, thereby slowing the transport of carbon to the ocean depths via the biological pump [146, 147]. Microplastics also impede feeding in copepods, which over time could lead to sustained reductions in ingested carbon biomass and a reduction in the transport of carbon [84].

5 Closing Remarks and Recommendations

Scaling up from experiments to predict the actual biological and ecological consequences of microplastics is difficult because the vast majority of the studies have used concentrations far exceeding those found in the environment [31]. A handful of recent studies, however, have attempted to use concentrations concomitant with current levels of contamination in relevant habitats and have found mixed results. For example, two infaunal bivalves were exposed to 1, 10 and 25 mg of HDPE microplastics per kg of sediment corresponding to current levels of contamination in some parts of the world [97]. Although there were no effects on mortality, general condition or burrowing, bivalves experienced a decrease in protein content and energy reserves [97]. Similarly, Naidoo and Glassom [148] exposed juvenile glassfish for 95 days to a concentration similar to that found in the local waters (0.17 mg L^{-1}); they also compared virgin microplastics with those extracted from the local harbour. Both types of microplastics reduced growth rates, body weight and probability of survival of glassfish. On the contrary, tests using a range of realistic concentrations of microplastics found a biological effect on only one out of six macroinvertebrates tested [149]. It is clear that more experiments simulating environmentally realistic conditions (i.e. environmentally realistic concentrations, outdoor mesocosm systems and longer-term experimental periods) are required in order to enable understanding of current effects of microplastics.

Despite the mismatch in environmental concentration and experimental conditions, it is worth noting that since Lenz et al. [31], some researchers have further refined sampling and extraction methods to capture the smaller size ranges (<330 μm) of microplastics that were previously omitted by zooplankton sampling methods. Consequently, greater estimates of abundances of microplastics in aquatic habitats have been quantified, for example, up to 25.8 microplastics L^{-1} in surface waters of a Chinese lake [150], 641.3 microplastics L^{-1} in the Los Angeles river [151], up to ~10 microplastics L^{-1} in seawater samples in the Falkland Islands and in the UK [152] and 15.6 microplastics L^{-1} in estuarine waters in South Korea [153].

A recent risk assessment using a modelling approach predicts a 50-fold increase in the total mass of microplastics between 2,010 and 2,100 due to further fragmentation of larger plastic litter and suggests that based on a meta-analysis of the effects of data, ~6 microplastics L^{-1} is the safe limit for floating microplastics beyond which point negative ecological effects will occur [154]. Although the majority of effects outlined in this chapter are described from experiments that used currently unrealistic concentrations of microplastics, these effects could be indicative of future consequences. Given that microplastics are increasing in abundance and that some effects are already evident at current environmental levels, a precautionary approach favouring prevention rather than cure is prudent.

The severity of the environmental impact resulting from plastic debris will depend on a number of variables including the (1) size, (2) shape, (3) chemical composition, (4) abundance of the plastic debris, (5) the type of organism or habitat being contaminated and (6) the presence of other environmental stressors that could exacerbate or ameliorate any impacts. In order to gain a complete picture of the potential impacts of different types of plastic debris, it is important, therefore, to repeat experiments in a range of different environmental contexts and with different mixtures of microplastics.

Finally, as governments and individuals attempt to phase out single-use plastics, alternative materials including bio-based biodegradable plastics are growing in popularity and in global production [155]. It is possible, therefore, that they could be more common as litter items in the future. Research has shown that some biodegradable plastics do not readily biodegrade in natural aquatic or terrestrial environments [137, 156]; and, either as larger consumer items such as plastic bags [135] or as microplastics [81–83, 100], they can have the same effects as non-biodegradable types of plastic if they become litter. We must therefore raise awareness that these are not designed to become litter and emphasise the need for responsible (waste) management of the end-of-life stage of these items to prevent them entering and accumulating in natural habitats.

References

1. International Coastal Cleanup Report (2017) Ocean conservancy. https://oceanconservancy.org/wp-content/uploads/2017/06/International-CoastalCleanup_2017-Report.pdf. Accessed 20 Jan 2020
2. Ostle C, Thompson RC, Broughton D et al (2019) The rise in ocean plastics evidenced from a 60-year time series. Nat Commun 10:1622
3. Oceanic and Atmospheric Administration Marine Debris Program (2014) Report on the entanglement of marine species in marine debris with an emphasis on species in the United States. NOAA, Silver Spring, 28 p
4. Browne MA, Crump P, Niven SJ et al (2011) Accumulation of microplastic on shorelines worldwide: sources and sinks. Environ Sci Technol 45(21):9175
5. Eriksen M, Lebreton LCM, Carson HS et al (2014) Plastic pollution in the world's oceans: more than 5 trillion plastic pieces weighing over 250,000 tons afloat at sea. PLoS One 9: e111913
6. Plastic Europe (2019) Plastics the facts – 2019: an analysis of European plastics production, demand and waste data. Plastic Europe, Brussels
7. Geyer R, Jambeck JR, Law KL (2017) Production, use, and fate of all plastics ever made. Sci Adv 3(7):e1700782
8. Priestland E, Sherrington C, Darrah C et al (2017) Single-use plastics and the marine environment: leverage points for reducing single-use plastics. Report produced for Seas at Risk
9. Chapman PM (2007) Determining when contamination is pollution – weight of evidence determinations for sediments and effluents. Environ Int 33(4):492–501
10. Underwood AJ, Chapman MG, Browne MA (2017) Some problems and practicalities in design and interpretation of samples of microplastic waste. Anal Methods 2017(9):1332–1345
11. Erni-Cassola G, Zadjelovic V, Gibson MI et al (2019) Distribution of plastic polymer types in the marine environment; a meta-analysis. J Hazard Mat 369:691–698
12. Andrady AL (2011) Microplastics in the marine environment. Mar Pollut Bull 62 (8):1596–1605
13. Kaiser D, Kowalski N, Waniek JJ (2017) Effects of biofouling on the sinking behavior of microplastics. Environ Res Lett 12:124003
14. Chae DH, Kim IS, Kim SK et al (2015) Abundance and distribution characteristics of microplastics in surface seawaters of the Incheon/Kyeonggi coastal region. Arch Environ Contam Toxicol 69:269–278
15. Fischer EK, Paglialonga L, Czech E et al (2016) Microplastic pollution in lakes and Lake Shoreline sediments – a case study on Lake Bolsena and Lake Chiusi (Central Italy). Environ Pollut 213:648–657
16. Karthik R, Robin RS, Purvaja R (2018) Microplastics along the beaches of southeast coast of India. Sci Total Environ 645:1388–1399
17. Woodall LC, Sanchez-Vidal A, Canals M et al (2014) The deep sea is a major sink for microplastic debris. R Soc Open Sci 1(4):140317
18. Willis KA, Eriksen R, Wilcox C et al (2017) Microplastic distribution at different sediment depths in an urban estuary. Front Mar Sci 4:419
19. Vaughan R, Turner SD, Rose NL (2017) Microplastics in the sediments of a UK urban lake. Environ Pollut 229:10–18
20. Desforges JP, Galbraith M, Ross PS (2015) Ingestion of microplastics by zooplankton in the Northeast Pacific Ocean. Arch Environ Contam Toxicol 69:320
21. Lusher AL, McHugh M, Thompson RC (2013) Occurrence of microplastics in the gastrointestinal tract of pelagic and demersal fish from the English Channel. Mar Pollut Bull 67:94–99
22. Pham CK, Rodríguez DA et al (2017) Plastic ingestion in oceanic-stage loggerhead sea turtles (*Caretta caretta*) off the North Atlantic subtropical gyre. Mar Pollut Bull 121:222–229

23. Fossi MC, Panti C, Guerranti C (2012) Are baleen whales exposed to the threat of microplastics? A case study of the Mediterranean fin whale (*Balaenoptera physalus*). Mar Pollut Bull 64(11):2374–2379
24. Windsor FM, Tilley RM, Tyler CR et al (2019) Microplastic ingestion by riverine macroinvertebrates. Sci Total Environ 646:68–74
25. Silva-Cavalcanti JS, Diego J, Silva B et al (2017) Microplastics ingestion by a common tropical freshwater fishing resource. Environ Pollut 221:218–226
26. Rochman CM, Browne MA, Halpern BS et al (2013) Policy: classify plastic waste as hazardous. Nature 494:169–171
27. Borrelle SB, Rochman CM, Liboiron M (2017) Opinion: why we need an international agreement on marine plastic pollution. Proc Natl Acad Sci U S A 114(38):9994–9997
28. Worm B, Lotze HK, Jubinville I et al (2017) Plastic as a persistent marine pollutant. Annu Rev Environ Resour 42(1):1–26
29. Rochman CM, Browne MA, Underwood AJ (2016) The ecological impacts of marine debris: unraveling the demonstrated evidence from what is perceived. Ecology 97:302–312
30. Galloway TS, Cole M, Ceri L (2017) Interactions of microplastic debris throughout the marine ecosystem. Nat Ecol Evol 1:0116
31. Lenz R, Enders K, Nielsen TG (2016) Microplastic exposure studies should be environmentally realistic. Proc Natl Acad Sci U S A 113(29):E4121–E4122
32. de Sá LC, Oliveira M, Ribeiro F (2018) Studies of the effects of microplastics on aquatic organisms: what do we know and where should we focus our efforts in the future? Sci Total Environ 645:1029–1039
33. Gall SC, Thompson RC (2015) The impact of debris on marine life. Mar Pollut Bull 92:170–179
34. Lebreton L, Slat B, Ferrari F et al (2018) Evidence that the Great Pacific Garbage Patch is rapidly accumulating plastic. Sci Rep 8:4666
35. Chiappone M, Dienes H, Swanson DW et al (2005) Impacts of lost fishing gear on coral reef sessile invertebrates in the Florida Keys National Marine Sanctuary. Biol Conserv 121 (2):221–230
36. Ballesteros LV, Matthews JL, Hoeksema BW (2018) Pollution and coral damage caused by derelict fishing gear on coral reefs around Koh Tao, Gulf of Thailand. Mar Pollut Bull 135:1107–1116
37. Richards ZT, Beger M (2011) A quantification of the standing stock of macro-debris in Majuro lagoon and its effect on hard coral communities. Mar Pollut Bull 62(8):1693–1701
38. Kühn S, Bravo Rebolledo EL, van Franeker JA (2015) Deleterious effects of litter on marine life. In: Bergmann M, Gutow L, Klages M (eds) Marine anthropogenic litter. Springer, Cham
39. Henderson JR (2001) A pre- and post-MARPOL annex V summary of Hawaiian monk seal entanglements and marine debris accumulation in the northwestern Hawaiian Islands, 1982–1998. Mar Pollut Bull 42:584–589
40. Boland RC, Donohue MJ (2003) Marine debris accumulation in the nearshore marine habitat of the endangered Hawaiian monk seal, *Monachus schauinslandi* 1999–2001. Mar Pollut Bull 46:1385–1394
41. Biddle TM, Limpus CJ (2011) Marine wildlife stranding and mortality database annual reports 2005–2010. Marine Turtles. Conserv Tech Data Rep 2010(1):1–124
42. Laist DW (1997) Impacts of marine debris: entanglement of marine life in marine debris including a comprehensive list of species with entanglement and ingestion records. In: Coe JM, Rogers DB (eds) Marine debris, sources, impacts, and solutions. Springer, New York, pp 99–139
43. Cassoff RM, Moore KM, McLellan WA et al (2011) Lethal entanglement syndrome in baleen whales. Dis Aquat Org 96:175–185
44. Beck CA, Barros NB (1991) The impact of debris on the Florida Manatee. Mar Pollut Bull 22:508–510

45. Byard RW, Machado A (2018) Characteristic "neck collar" injuries in Australian sea lions (*Neophoca cinerea*) caused by marine debris. Forensic Sci Med Pathol 15(4):631–634. https://doi.org/10.1007/s12024-018-0060-1
46. Dau BK, Gilardi KV, Gulland FM et al (2009) Fishing gear-related injury in California marine wildlife. J Wildl Dis 45:355–362
47. Butterworth A, Clegg I, Bass C (2012) Marine debris: a global picture of the impact on animal welfare and of animal-focused solutions. WSPA International, London
48. Wilcox C, Hardesty B, Sharples R et al (2013) Ghostnet impacts on globally threatened turtles, a spatial risk analysis for northern Australia. Conserv Lett 6(4):247–254
49. Moy K, Neilson B, Chung A et al (2018) Mapping coastal marine debris using aerial imagery and spatial analysis. Mar Pollut Bull 132:52–59
50. Jacobsen JK, Massey L, Gulland F (2010) Fatal ingestion of floating net debris by two sperm whales (*Physeter macrocephalus*). Mar Pollut Bull 60(5):765–767
51. De Stephanis R, Gimenez J, Carpinelli E et al (2013) As main meal for sperm whales: plastics debris. Mar Pollut Bull 69(1–2):2016–2214
52. Brandao ML, Braga KM, Luque JL (2011) Marine debris ingestion by magellanic penguins, *Spheniscus magellanicus* (Aves: Sphenisciformes), from the Brazilian Coastal Zone. Mar Pollut Bull 62:2246–2249
53. Colabuono FI, Barquete V, Domingues BS et al (2009) Plastic ingestion by procellariiformes in Southern Brazil. Mar Pollut Bull 58:93–96
54. Tourinho PS, Ivar do Sul JA, Fillmann G (2010) Is marine debris ingestion still a problem for the coastal marine biota of southern Brazil? Mar Pollut Bull 60(3):396–401
55. Poppi L, Zaccaroni A, Pasotto D et al (2012) Post-mortem investigations on a leatherback turtle *Dermochelys coriacea* stranded along the Northern Adriatic coastline. Dis Aquat Org 100:71–76
56. Stamper A, Spicer CW, Neiffer DL et al (2009) Morbidity in a juvenile green sea turtle (Chelonia mydas) due to ocean-borne plastic. J Zoo Wildl Med 40:196–198
57. Ryan PG (1988) Effects of ingested plastic on seabird feeding: evidence from chickens. Mar Pollut Bull l9:125–136
58. Lusher A (2015) Microplastics in the marine environment: distribution, interactions and effects. In: Bergmann M, Gutow L, Klages M (eds) Marine anthropogenic litter. Springer, Berlin, pp 245–308
59. Auta HS, Emenike CU, Fauziah SH (2017) Distribution and importance of microplastics in the marine environment: a review of the sources, fate, effects, and potential solutions. Environ Int 102:165–176
60. Rezania S, Park J, Din MFM et al (2018) Microplastics pollution in different aquatic environments and biota: a review of recent studies. Mar Pollut Bull 133:191–208
61. Botterell ZLR, Beaumont N, Dorrington T et al (2019) Bioavailability and effects of microplastics on marine zooplankton: a review. Environ Pollut 245:98–110
62. Lusher AL, Hernandez-Milian G, Berrow S et al (2018) Incidence of marine debris in cetaceans stranded and bycaught in Ireland: recent findings and a review of historical knowledge. Environ Pollut 232:467–476
63. Nelms SE, Barnett J, Brownlow A et al (2019) Microplastics in marine mammals stranded around the British Coast: ubiquitous but transitory? Sci Rep 9:1075
64. Duncan E, Broderick A, Fuller W et al (2019) Microplastic ingestion ubiquitous in marine turtles. Glob Chang Biol 25:744–752
65. Provencher JF, Bond AL, Mallory ML (2015) Marine birds and plastic debris in Canada: a national synthesis and a way forward. Environ Rev 23:1–13
66. Pozo K, Gomez V, Torres M et al (2019) Presence and characterization of microplastics in fish of commercial importance from the Biobío region in Central Chile. Mar Pollut Bull 140:315–319
67. Wieczorek AM, Morrison L, Croot P et al (2018) Frequency of microplastics in mesopelagic fishes from the Northwest Atlantic. Front Mar Sci 5:39

68. Jamieson AJ, Brooks LSR, Reid WDK et al (2019) Microplastics and synthetic particles ingested by deep-sea amphipods in six of the deepest marine ecosystems on Earth. R Soc Open Sci 6(2):180667. https://doi.org/10.1098/rsos.180667
69. Courtene-Jones W, Quinn B, Ewins C (2019) Consistent microplastic ingestion by deep-sea invertebrates over the last four decades (1976–2015), a study from the North East Atlantic. Environ Pollut 244:503–512
70. Holland ER, Mallory ML, Shutler D (2016) Plastics and other anthropogenic debris in freshwater birds from Canada. Sci Total Environ 571:251–258
71. Sanchez W, Bender C, Porcher J-M (2014) Wild gudgeons (Gobio gobio) from French rivers are contaminated by microplastics: preliminary study and first evidence. Environ Res 128:98–10
72. Biginagwa FJ, Mayoma BS, Shashoua Y et al (2016) First evidence of microplastics in the African Great Lakes: recovery from Lake Victoria Nile perch and Nile tilapia. J Great Lakes Res 42(1):146–149
73. Su L, Cai H, Kolandhasamy P et al (2018) Using the Asian clam as an indicator of microplastic pollution in freshwater ecosystems. Environ Pollut 234:347–355
74. Nel HA, Dalu T, Wasserman RJ (2018) Sinks and sources: assessing microplastic abundance in river sediment and deposit feeders in an Austral temperate urban river system. Sci Total Environ 612:950–956
75. Liboiron F, Ammendolia J, Saturno J et al (2018) A zero percent plastic ingestion rate by silver hake (*Merluccius bilinearis*) from the south coast of Newfoundland, Canada. Mar Pollut Bull 131:267–275
76. Sussarellu R, Suquet M, Thomas Y et al (2016) Oyster reproduction is affected by exposure to polystyrene microplastics. Proc Natl Acad Sci 113(9):2430–2435
77. Barboza LGA, Vieira LR, Branco V et al (2018a) Microplastics cause neurotoxicity, oxidative damage and energy-related changes and interact with the bioaccumulation of mercury in the European seabass, *Dicentrarchus labrax* (Linnaeus, 1758). Aquat Toxicol 195:49–57
78. Greven AC, Merk T, Karagöz F et al (2016) Polycarbonate and polystyrene nanoplastic particles act as stressors to the innate immune system of fathead minnow (*Pimephales promelas*). Environ Toxicol Chem 35(12):3093–3100
79. Tang J, Ni X, Zhou Z et al (2018) Acute microplastic exposure raises stress response and suppresses detoxification and immune capacities in the scleractinian coral *Pocillopora damicornis*. Environ Pollut 243:66–74
80. Green DS, Colgan TJ, Thompson RC et al (2019) Exposure to microplastics reduces attachment strength and alters the haemolymph proteome of blue mussels (*Mytilus edulis*). Environ Pollut 246:423–434
81. Green DS, Boots B, Sigwart J et al (2016) Effects of conventional and biodegradable microplastics on a marine ecosystem engineer (*Arenicola marina*) and sediment nutrient cycling. Environ Pollut 208:426–434
82. Green DS (2016) Effects of microplastics on European flat oysters, *Ostrea edulis* and their associated benthic communities. Environ Pollut 216:95–103
83. Green DS, Boots B, O'Connor NE et al (2017) Microplastics affect the ecological functioning of an important biogenic habitat. Environ Sci Technol 51(1):68e77
84. Cole M, Lindeque P, Fileman E et al (2015) The impact of polystyrene microplastics on feeding, function and fecundity in the marine copepod *Calanus helgolandicus*. Environ Sci Technol 492:1130–1137
85. Ogonowski M, Schür C, Jarsén Å et al (2016) The effects of natural and anthropogenic microparticles on individual fitness in *Daphnia magna*. PLoS One 11(5):e0155063
86. Wright SL, Rowe D, Thompson RC et al (2013) Microplastic ingestion decreases energy reserves in marine worms. Curr Biol 23(23):1031–1033
87. Choi JS, Jung Y-J, Hong N-H et al (2018) Toxicological effects of irregularly shaped and spherical microplastics in a marine teleost, the sheepshead minnow (*Cyprinodon variegatus*). Mar Pollut Bull 129(1):231–240

88. Au SY, Bruce TF, Bridges WC et al (2015) Responses of *Hyalella azteca* to acute and chronic microplastic exposures. Environ Toxicol Chem 34:2564–2572
89. Lehtiniemi M, Hartikainen S, Näkki P et al (2018) Size matters more than shape: ingestion of primary and secondary microplastics by small predators. Food Webs 17:e00097
90. Gray AD, Weinstein JE (2017) Size- and shape-dependent effects of microplastic particles on adult daggerblade grass shrimp (*Palaemonetes pugio*). Environ Toxicol Chem 36:3074–3080
91. da Costa JP, Santos PSM, Duarte AC et al (2016) (Nano)plastics in the environment – sources, fates and effects. Sci Total Environ 566–567:15–26
92. Yu P, Liu Z, Wu D et al (2018) Accumulation of polystyrene microplastics in juvenile *Eriocheir sinensis* and oxidative stress effects in the liver. Aquat Toxicol 200:28–36
93. Pitt JA, Kozal JS, Jayasundara N et al (2018) Uptake, tissue distribution, and toxicity of polystyrene nanoparticles in developing zebrafish (*Danio rerio*). Aquat Toxicol 194:185–194
94. Pitt JA, Trevisan R, Massarsky A et al (2018) Maternal transfer of nanoplastics to offspring in zebrafish (*Danio rerio*): a case study with nanopolystyrene. Sci Total Environ 643:324–334
95. Jiang X, Chen H, Liao Y et al (2019) Ecotoxicity and genotoxicity of polystyrene microplastics on higher plant *Vicia faba*. Environ Pollut 250:831–838
96. Lee K-W, Shim WJ, Kwon OY et al (2013) Size-dependent effects of micro polystyrene particles in the marine copepod *Tigriopus japonicus*. Environ Sci Technol 47 (19):11278–11283
97. Bour A, Haarr A, Keiter S, Hylland K (2018) Environmentally relevant microplastic exposure affects sediment-dwelling bivalves. Environ Pollut 236:652–660
98. Lei L, Wu S, Lu S et al (2018) Microplastic particles cause intestinal damage and other adverse effects in zebrafish *Danio rerio* and nematode *Caenorhabditis elegans*. Sci Total Environ 619–620:1–8
99. Li H-X, Getzinger GJ, Ferguson PL et al (2016) Effects of toxic leachate from commercial plastics on larval survival and settlement of the barnacle *Amphibalanus amphitrite*. Environ Sci Technol 50:924e931
100. Straub S, Hirsch PE, Burkhardt-Holm P (2017) Biodegradable and petroleum-based microplastics do not differ in their ingestion and excretion but in their biological effects in a freshwater invertebrate *Gammarus fossarum*. Int J Environ Res Public Health 14(7):774
101. González-Pleiter M, Tamayo-Belda M, Pulido-Reyes G et al (2019) Secondary nanoplastics released from a biodegradable microplastic severely impact freshwater environments. Environ Sci Nano 6:1382–1392
102. Barboza LGA, Vethaak AD, Lavorante BRBO et al (2018) Marine microplastic debris: an emerging issue for food security, food safety and human health. Mar Pollut Bull 133:336–348
103. Rochman CM, Hoh E, Hentschel BT et al (2013) Long-term field measurement of sorption of organic contaminants to five types of plastic pellets: implications for plastic marine debris. Environ Sci Technol 47(3):1646–1654
104. Bakir A, Rowlans SJ, Thompson RC (2014) Enhanced desorption of persistent organic pollutants from microplastics under simulated physiological conditions. Environ Pollut 185:16–23
105. Chua EM, Shimeta J, Nugegoda D et al (2014) Assimilation of polybrominated diphenyl ethers from microplastics by the marine amphipod, *Allorchestes compressa*. Environ Sci Technol 48(14):8127–8134
106. Luis LG, Ferreira P, Fonte E et al (2015) Does the presence of microplastics influence the acute toxicity of chromium(VI) to early juveniles of the common goby (*Pomatoschistus microps*)? A study with juveniles from two wild estuarine populations. Aquat Toxicol 164:163–174
107. Rehse S, Kloas W, Zarfl C (2018) Microplastics reduce short-term effects of environmental contaminants. Part I: effects of bisphenol a on freshwater zooplankton are lower in presence of polyamide particles. Int J Environ Res Public Health 15(2):200
108. Horton AA, Vijver MG, Lahive E et al (2018) Acute toxicity of organic pesticides to Daphnia magna is unchanged by co-exposure to polystyrene microplastics. Ecotoxicol Environ Saf 166:26–34

109. Zettler ER, Mincer TJ, Amaral-Zettler LA (2013) Life in the “Plastisphere”: microbial communities on plastic marine debris. Environ Sci Technol 47:7137–7146
110. Oberbeckmann S, Loder MGJ, Labrenz M (2015) Marine microplastic associated biofilms – a review. Environ Chem 12(5):551–562
111. Zettler LA, Zettler ER, Boyd GD et al (2015) The biogeography of the Plastisphere: implications for policy. Front Ecol Environ 13(10):541–546
112. Zenker A, Cicero MR, Prestinaci F et al (2014) Bioaccumulation and biomagnification potential of pharmaceuticals with a focus to the aquatic environment. J Environ Manag 133:378–387
113. Burridge L, Weis JS, Cabello F et al (2010) Chemical use in salmon aquaculture: a review of current practices and possible environmental effects. Aquaculture 306(1–4):7–23
114. Van Boeckel TP, Brower C, Gilbert M (2015) Global trends in antimicrobial use in food animals. Proc Natl Acad Sci U S A 112(18):5649–5654
115. Amos CGA, Hawkey PM, Gaze WH et al (2014) Waste water effluent contributes to the dissemination of CTX-M-15 in the natural environment. J Antimicrob Chemother 69:1785–1791
116. Allen HK, Donato J, Wang HH et al (2010) Call of the wild: antibiotic resistance genes in natural environments. Nat Rev Microbiol 8:251–259
117. Wellington EM, Boxall AB, Cross P et al (2013) The role of the natural environment in the emergence of antibiotic resistance in Gram-negative bacteria. Lancet Infect Dis 13(2):155–165
118. Morritta D, Stefanoudis PV, Pearce D et al (2014) Plastic in the Thames: a river runs through it. Mar Pollut Bull 78(1–2):196–200
119. Li J, Zhang K, Zhang H (2018) Adsorption of antibiotics on microplastics. Environ Pollut 237:460–467
120. Wu C, Zhang K, Huang X, Liu J (2016) Sorption of pharmaceuticals and personal care products to polyethylene debris. Environ Sci Pollut Res 23(9):8819–8826
121. Hall-Stoodley L, Costerton JW, Stoodley P (2004) Bacterial biofilms: from the natural environment to infectious diseases. Nat Rev Microbiol 2:95–108
122. Stewart PS, Costerson JW (2001) Antibiotic resistance of bacteria in biofilms. Lancet 358:135–138
123. Arias-Andres M, Klümper U, Rojas-Jimenez K et al (2018) Microplastic pollution increases gene exchange in aquatic ecosystems. Environ Pollut 237:253–261
124. Huang FY, Yang K, Zhang ZX et al (2019) Effects of microplastics on antibiotic resistance genes in estuarine sediments. Huan Jing Ke Xue 40(5):2234–2239
125. Yang Y, Liu G, Song W et al (2019) Plastics in the marine environment are reservoirs for antibiotic and metal resistance genes. Environ Int 123:79–86
126. Prata JC, Lavorante BRBO, da Conceição M et al (2018) Influence of microplastics on the toxicity of the pharmaceuticals procainamide and doxycycline on the marine microalgae *Tetraselmis chuii*. Aquat Toxicol 197:143–152
127. Zhang S, Ding J, Razanajatovo RM et al (2019) Interactive effects of polystyrene microplastics and roxithromycin on bioaccumulation and biochemical status in the freshwater fish red tilapia (*Oreochromis niloticus*). Sci Total Environ 648:1431–1439
128. Wilhelmsen U (1998) Rapid colonization of new habitats in the Wadden Sea by the ovoviviparous *Littorina saxatilis* (Olivi). Helgol Meeresunters 52:325
129. Kiessling T, Gutow L, Thiel M (2015) Marine litter as habitat and dispersal vector. In: Bergmann M, Gutow L, Klages M (eds) Marine anthropogenic litter. Springer, Cham
130. Rothäusler E, Gutow L, Thiel M (2012) Floating seaweeds and their communities. In: Wiencke C, Bischof K (eds) Seaweed biology. Springer, Berlin, pp 359–380
131. Thiel M, Gutow L (2005) The ecology of rafting in the marine environment. I. The floating substrata. Oceanogr Mar Biol Annu Rev 42:181–264
132. Rech S, Borrell Y, García-Vazquez E (2016) Marine litter as a vector for non-native species: what we need to know. Mar Pollut Bull 113(1–2):40–43
133. Reisser J, Shaw J, Hallegraeff G et al (2014) Millimeter-sized marine plastics: a new pelagic habitat for microorganisms and invertebrates. PLoS One 9:e100289

134. Katsanevakis S, Verriopoulos G, Nicolaidou A et al (2007) Effect of marine litter on the benthic megafauna of coastal soft bottoms: a manipulative field experiment. Mar Pollut Bull 54(6):771–778
135. Green DS, Boots B, Blockley DJ et al (2015) Impacts of discarded plastic bags on marine assemblages and ecosystem functioning. Environ Sci Technol 49(9):5380–5389
136. Akoumianaki I, Kontolefas P, Katsanevakis S et al (2008) Subtidal littering: indirect effects on soft substratum macrofauna? Mediterr Mar Sci 9(2):35–52
137. Napper IE, Thompson RC (2019) Environmental deterioration of biodegradable, oxo-biodegradable, compostable, and conventional plastic carrier bags in the sea, soil, and open-air over a 3-year period. Environ Sci Technol 53(9):4775–4783
138. Giles H, Pilditch CA, Bell DG (2006) Sedimentation from mussel (*Perna canaliculus*) culture in the Firth of Thames, New Zealand: impacts on sediment oxygen and nutrient fluxes. Aquaculture 261(1):125–140
139. Barbosa AB (2009) Dynamics of living phytoplankton: implications for paleoenvironmental reconstructions. IOP Conf Ser Earth Environ Sci 5(1):012001. https://doi.org/10.1088/1755-1307/5/1/012001
140. Yokota K, Waterfield H, Hastings C et al (2017) Finding the missing piece of the aquatic plastic pollution puzzle: interaction between primary producers and microplastics. Limnol Oceanogr 2:91–104
141. Prata JC, da Costa JP, Lopes I et al (2019) Effects of microplastics on microalgae populations: a critical review. Sci Total Environ 665:400–405
142. Bhattacharya P, Lin S, Turner JP et al (2010) Physical adsorption of charged plastic nanoparticles affects algal photosynthesis. J Phys Chem C 114(2010):16556–16561
143. Gutow L, Eckerlebe A, Giménez L et al (2016) Experimental evaluation of seaweeds as a vector for microplastics into marine food webs. Environ Sci Technol 50:915–923
144. Long MB, Moriceau M, Gallinari C et al (2015) Interactions between microplastics and phytoplankton aggregates: impact on their respective fates. Mar Chem 175:39–46
145. Lagarde FO, Olivier M, Zanella P et al (2016) Microplastic interactions with freshwater microalgae: hetero-aggregation and changes in plastic density appear strongly dependent on polymer type. Environ Pollut 215:331–339
146. Cole M, Lindeque PK, Fileman E et al (2016) Microplastics Alter the properties and sinking rates of zooplankton faecal pellets. Environ Sci Technol 50(6):3239–3246
147. Porter A, Lyons BP, Galloway TS et al (2018) Role of marine snows in microplastic fate and bioavailability. Environ Sci Technol 52(12):7111–7119
148. Naidoo T, Glassom D (2019) Decreased growth and survival in small juvenile fish, after chronic exposure to environmentally relevant concentrations of microplastic. Mar Pollut Bull 145:254–259
149. Redondo-Hasselerharm PE, Falahudin D, Peeters ETHM et al (2018) Microplastic effect thresholds for freshwater benthic macroinvertebrates. Environ Sci Technol 52(4):2278–2286
150. Su L, Xue Y, Li L et al (2016) Microplastics in Taihu Lake, China. Environ Pollut 216:711–719
151. Wiggin KJ, Holland EB (2019) Validation and application of cost and time effective methods for the detection of 3–500 μm sized microplastics in the urban marine and estuarine environments surrounding Long Beach, California. Mar Pollut Bull 143:152–162
152. Green DS, Kregting L, Boots B et al (2018) A comparison of sampling methods for seawater microplastics and a first report of the microplastic litter in coastal waters of Ascension and Falkland Islands. Mar Pollut Bull 137:695–701
153. Kang J-H, Kwon OY, Lee K-W et al (2015) Marine neustonic microplastics around the southeastern coast of Korea. Mar Pollut Bull 96(1–2):304–312
154. Everaert G, Van Cauwenberghe L, De Rijcke M et al (2018) Risk assessment of microplastics in the ocean: modelling approach and first conclusions. Environ Pollut 242:1930–1938
155. European Bioplastics (2018) www.european-bioplastics.org. Accessed July 2019
156. Narancic T, Verstichel S, Chaganti SR (2018) Biodegradable plastic blends create new possibilities for end-of-life management of plastics but they are not a panacea for plastic pollution. Environ Sci Technol 5218:10441–10452

Impact of Plastic Pollution on Marine Life in the Mediterranean Sea

Aikaterini Anastasopoulou and Tomaso Fortibuoni

Contents

1 Introduction 136
2 Ingestion 138
    2.1 Invertebrates 163
    2.2 Teleosts 164
    2.3 Elasmobranchs 167
    2.4 Marine Mammals 169
    2.5 Sea Turtles 170
    2.6 Seabirds 171
3 Entanglement 172
4 Other Impacts 179
5 Conclusions 181
References 183

Abstract Marine litter is an environmental problem of global concern with well-documented impacts on marine biodiversity and ecosystems. At a global scale, marine litter is mainly composed of plastic. Plastics can affect marine organisms mainly through ingestion and entanglement but also through the facilitation of transport of organisms via rafting or the provision of new habitats for colonization. Impacts vary according to the type and size of the plastics and can occur at different levels of biological organization in a wide variety of habitats. In this chapter, we reviewed and synthesized literature in order to describe the impact of litter on marine

---

Anastasopoulou Aikaterini and Fortibuoni Tomaso contributed equally.

A. Anastasopoulou (✉)
Hellenic Centre for Marine Research, Institute of Marine Biological Resources and Inland Waters, Attica, Greece
e-mail: kanast@hcmr.gr

T. Fortibuoni
Italian National Institute for Environmental Protection and Research, Ozzano dell'Emilia, Italy

National Institute of Oceanography and Applied Geophysics, Trieste, Italy
e-mail: tomaso.fortibuoni@isprambiente.it

Friederike Stock, Georg Reifferscheid, Nicole Brennholt, and Evgeniia Kostianaia (eds.),
Plastics in the Aquatic Environment - Part I: Current Status and Challenges,
Hdb Env Chem (2022) 111: 135–196, DOI 10.1007/698_2019_421,

© Springer Nature Switzerland AG 2019, Published online: 5 December 2019

life in the Mediterranean sensitive ecosystem. The review focused on the following impact categories: ingestion, entanglement and other effects (e.g. colonization and rafting). In the Mediterranean, reports of ingestion were made for more than 49,454 individuals from 116 species, of which the taxonomic group with the greatest number of species impacted was Teleosts (~59%). Forty-four species were found entangled in marine litter (59% were invertebrates, mainly Cnidarians), of which the species with the highest number of entanglement records in the Mediterranean Sea was the loggerhead sea turtle (*Caretta caretta*). One hundred and seventy-eight taxa were found rafting on floating objects or using marine litter as a substratum, including Chromista and Bacteria. The most common phyla rafting on marine litter were Arthropods and Cnidarians.

Keywords Entanglement, Ingestion, Mediterranean, Plastic pollution, Rafting

1 Introduction

Marine litter has been globally recognized as a growing environmental concern, which can significantly affect wildlife, from marine worms [1] to whales [2] and potentially humans [3]. Globally, the proportion of plastic among marine litter ranges from 60 to 80%, although it has reached over 90–95% in some areas [4]. Plastics are produced in huge quantities and have become an essential part of our life mostly due to their low production costs and specific characteristics like durability and flexibility [5]. As reported by PlasticsEurope in 2017 [6], about 335 million tons of plastics were produced in 2016. Up to 5% of plastics produced each year ends up in the sea [7], where it persists, accumulates and fragments into smaller pieces (e.g. microplastics) by UV radiation, waves and mechanical forces [8], increasing the potential for ingestion by organisms [9]. The colour, density, shape, size and abundance of these tiny plastic particles may affect their potential availability to marine organisms [1, 10]. Jovanović [11] reported that, in the relatively near future, the abundance of plastic micro- and nanoparticles will be greater than the count of plankton. Plastics, including microplastics, contaminate habitats from shallow water to the deep sea and from the poles to the equator. They are present on shorelines, in the water column, on the seafloor, in sediments and in organisms (e.g. [12, 13]).

The Mediterranean Sea is the largest and deepest enclosed sea on Earth [14]. It constitutes 1 of the 25 biodiversity centres that are recognized on a planetary scale, and it is considered a biodiversity hotspot by exceptionally high levels of endemism [15]. Moreover, the Mediterranean is a popular touristic destination with high pressures from anthropogenic activities (e.g. intense shipping, fishing, aquaculture, coastal urbanization) that take place in its waters and around its coastlines, factors that increase the input of marine litter into the basin [16, 17]. The Mediterranean is experiencing a deterioration of habitats, related to various human-origin activities, including global warming, uncontrolled urbanization and coastal development, fish farming, pollution and unsustainable fishing [15].

Plastic pollution in the Mediterranean has been well-documented: litter was found from shallow waters down to 4,500 m, as well as in surface waters where plastic concentration in some areas is comparable to that of the subtropical gyres (e.g. [16, 18, 19]). Several studies have demonstrated that plastics are ingested by marine organisms (e.g. [10, 20, 21]) and the number of reports on the impacts of plastics on marine organisms has increased over time. The high amount of plastics in the Mediterranean exposes its marine biodiversity to a direct threat [22].

Several reviews of the impacts of litter on marine life have been published (e.g. [8, 20, 21, 23, 24]). Marine litter impacts on the Mediterranean marine biodiversity were reviewed by Deudero and Alomar [25], who provided a list of 134 species affected by marine litter. More recently, Fossi et al. [26] provided a list of Mediterranean species known to ingest marine litter. This chapter provides the current state of knowledge about the impact of litter on marine organisms in the Mediterranean Sea based on the relevant literature (peer-reviewed scientific literature, grey literature, web sites and reports) published so far. We considered the effects of all kinds of marine litter, even if the great majority of records were referred to plastic litter, in particular as concerns ingestion.

The reviews published by Laist [20], Kühn et al. [23], Kiessling et al. [27], Deudero and Alomar [25] and Fossi et al. [26] were used as the starting point for this review. Web of Science, Scopus and Google Scholar scientific databases were used to search for the keywords: “plastics, marine litter, debris, microplastics, impact, ingestion, entanglement, rafting, hitch-hiking, ghost-fishing, entrapment, bioindicators, Mediterranean Sea, marine organisms, invertebrates, fish, marine mammals, turtles, seabirds, cetaceans, molluscs, crustaceans”, appropriately combined. We further consulted the web sites Litterbase (https://litterbase.awi.de) and “Marine litter and biodiversity interactions in the Mediterranean Sea” (https://panaceacatalogue.adabyron.uma.es/gvsigonline/core/public_project_load/marinelitter/). No laboratory/experimental studies were included in the review. The bibliographic search was closed in August 2019. Our literature search spanned records from 1980 to 2019. Taxonomic information and species names were updated using the World Register of Marine Species (WoRMS) database (www.marinespecies.org). Species habitat information was based on several databases (FishBase, www.fishbase.org; SeaLifeBase, www.sealifebase.org; the Reptile Database, www.reptile-database.org; the World Cetacea Database, www.marinespecies.org/cetacea/). The species’ conservation status was retrieved from the International Union for Conservation of Nature (IUCN) database (www.iucnredlist.org), referring to the Mediterranean subpopulation when specific information was available. Species were arranged into taxonomic groups (bacteria, plankton, marine plants, invertebrates, tunicates, teleosts, elasmobranchs, marine mammals, sea turtles and seabirds), and areas were grouped into Mediterranean subregions, as defined by the Marine Strategy Framework Directive (MSFD) (Western Mediterranean Sea, Ionian Sea and the Central Mediterranean Sea, Adriatic Sea and Aegean-Levantine Sea). It should be noted that a study was considered for all subregions if it covered two or more subregions. This work focuses mainly on ingestion and entanglement since they constitute the most common impacts of anthropogenic litter on marine organisms [23]. However, evidence of other kinds of impacts (e.g. colonization and rafting) on Mediterranean marine life is reported.

A total of 128 documents reporting impacts of marine litter on 329 taxa of the Mediterranean life were collected; among them, 156 taxa were found affected with regard to ingestion and/or entanglement (Tables 1 and 2). Figure 1 summarizes the number of species affected by marine litter either by ingestion or by entanglement according to different subregions of the Mediterranean; Fig. 2 presents the number of documents reporting ingestion/entanglement according to different subregions.

The highest number of species documented for marine litter ingestion has been reported for the Western Mediterranean Sea ($N = 67$), followed by that of the Aegean-Levantine Sea ($N = 37$). The highest number of species affected by entanglement was also reported for the Western Mediterranean Sea ($N = 67$), followed by the Ionian Sea and the Central Mediterranean Sea ($N = 23$).

The subregions with the lowest number of published reports on the effects of marine litter on biota were the Adriatic and the Aegean-Levantine Sea, indicating a gap of knowledge for these areas (Fig. 2).

2 Ingestion

Ingestion has been defined as the main impact of litter on marine organisms according to the MSFD Criterion 10.2 “Impacts of litter on marine life”, which will be used towards the achievement of a Good Environmental Status (GES) that is reached when “Properties and quantities of marine litter do not cause harm to the coastal and marine environment” [152]. Specifically, Indicator 10.2.1 refers to “Trends in the amount and composition of litter ingested by marine animals”.

Plastic is the most common litter category ingested by marine organisms (e.g. [8, 20, 21, 23, 26, 58, 80, 153]). Plastic particles have been found in all trophic levels, from zooplanktonic species such as copepods to invertebrates like polychaetes and bivalves, as well as vertebrates like fish, birds and marine mammals (e.g. [8, 10, 23, 154]). The proportion of species ingesting plastics varies a lot among the several taxonomic groups. Litter ingestion may occur in several ways either intentionally, accidentally, as a result of secondary ingestion or through parental delivery.

Intentional plastic ingestion can occur because of misidentification of litter items as natural prey. For example, Moser and Lee [155] studied the guts of seabirds and reported that some seabirds selected specific plastic shapes and colours and mistook them for potential prey items. Additionally, Ory et al. [156] showed that the planktivorous fish *Decapterus muroadsi* (Carangidae) ingested preferentially blue microplastics resembling their copepod prey, whereas Campani et al. [81] mentioned that turtles frequently ingested plastic bags as they may have mistaken them for jellyfish, which is a common prey in their diet. Moreover, the small size of

Table 1 List of Mediterranean species that have ingested plastics and microplastics according to published literature and online sources (search closed in August 2019)

ML effect	Taxon	Phylum	Species	Habitat	IUCN status	MSFD subregion	Reference
Animalia							
	Invertebrates	Annelida					
I			*Saccocirrus papillocercus*	Benthic	NE	W Med	[28]
		Mollusca					
I			*Mytilus galloprovincialis*	Benthic	NE	Ion & C Med; W Med	[29–32]
I			*Octopus salutii*	Demersal	DD	W Med	[22]
		Cnidaria					
I			*Pelagia noctiluca*	Pelagic	NE	Ion & C Med	[33]
		Echinodermata					
I			*Holothuria (Panningothuria) forskali*	Benthic	NE	W Med	[34]
I			*Holothuria (Holoturia) tubulosa*	Benthic	NE	Ion & C Med	[35]
		Arthropoda					
I		Amphipoda	*Gammarella fucicola*	Benthic	NE	W Med	[36]
I			*Gammarus aequicauda*	Benthic	NE	W Med	[36]
I			*Melita hergensis*	Demersal	NE	W Med	[36]
I			*Nototropis guttatus*	Benthic	NE	W Med	[36]
I		Leptostraca	*Nebalia strausi*	Demersal	NE	W Med	[36]
I		Decapoda	*Aristeus antennatus*	Demersal	NE	W Med	[37]
I			*Athanas nitescens*	Demersal	NE	W Med	[36]
I			*Galathea intermedia*	Demersal	NE	W Med	[36]
I			*Liocarcinus navigator*	Benthic	NE	W Med	[36]

(continued)

Table 1 (continued)

ML effect	Taxon	Phylum	Species	Habitat	IUCN status	MSFD subregion	Reference
I			*Nephrops norvegicus*	Benthic	LC	W Med; AD; Ion & C Med, AG & LEV	[38]
I			*Palaemon xiphias*	Benthic	NE	W Med	[36]
I			*Plesionika narval*	Benthic	NE	AG & LEV	[39]
Vertebrates							
		Chordata					
I	Teleosts		*Argyrosomus regius*	Benthopelagic	LC (Med)	AG & LEV	[40]
I			*Boops boops*	Demersal	LC (Med)	W Med	[41, 42]
I			*Caranx crysos*	Reef-associated	LC	AG & LEV	[40]
I			*Cataetyx laticeps*	Benthopelagic	LC (Med)	W Med	[43]
I			*Centracanthus cirrus*	Benthopelagic	LC (Med)	W Med	[22]
I			*Chelidonichthys cuculus*	Demersal	LC (Med)	W Med	[22]
I			*Chelidonichthys lucerna*	Demersal	LC (Med)	AD; AG & LEV	[40, 44]
I			*Chelon auratus*	Pelagic	LC (Med)	AD; AG & LEV	[40, 45]
I			*Citharus linguatula*	Demersal	LC (Med)	Ion & C Med	[45]
I			*Coryphaena hippurus*	Pelagic	LC (Med)	W Med	[46]
I			*Dentex gibbosus*	Benthopelagic	DD (Med)	AG & LEV	[40]
I			*Diaphus metopoclampus*	Bathypelagic	LC (Med)	Ion & C Med	[47]
I			*Diplodus annularis*	Benthopelagic	LC (Med)	AG & LEV	[40]
I			*Electrona risso*	Bathypelagic	LC (Med)	Ion & C Med	[47]
I			*Engraulis encrasicolus*	Pelagic	LC (Med)	W Med; AD	[41, 48, 49, 60]
I			*Glossanodon leioglossus*	Bathydemersal	LC (Med)	W Med	[22]
I			*Helicolenus dactylopterus*	Bathydemersal	LC (Med)	W Med	[22]

I			*Hoplostethus mediterraneus*	Benthopelagic	LC (Med)	W Med	[22]
I			*Hygophum benoiti*	Bathypelagic	LC (Med)	Ion & C Med	[47]
I			*Lepidion lepidion*	Bathypelagic	LC (Med)	W Med	[22]
I			*Lepidopus caudatus*	Benthopelagic	LC (Med)	W Med	[50]
I			*Lepidotrigla dieuzeidei*	Demersal	LC (Med)	W Med	[22]
I			*Lithognathus mormyrus*	Demersal	LC (Med)	AG & LEV	[40]
I			*Merluccius merluccius*	Demersal	VU (Med)	AD; Ion & C Med	[44, 51, 52]
I			*Mora moro*	Bathypelagic	LC (Med)	W Med	[43]
I			*Mullus barbatus*	Demersal	LC	AD; Ion & C Med; AG & LEV; W Med	[29, 40, 44, 45, 51, 53, 54]
I			*Mullus surmuletus*	Demersal	LC	AD; AG & LEV; W Med	[40, 45, 55]
I			*Myctophum punctatum*	Bathypelagic	LC (Med)	Ion & C Med; W Med	[47, 56]
I			*Naucrates ductor*	Reef-associated	LC (Med)	W Med	[22]
I			*Nemipterus randalli*	Demersal	LC	AG & LEV	[40]
I			*Nettastoma melanurum*	Bathypelagic	LC (Med)	W Med	[43]
I			*Nezumia aequalis*	Benthopelagic	LC (Med)	W Med	[22]
I			*Notacanthus bonaparte*	Bathypelagic	LC (Med)	W Med	[57]
I			*Pagellus acarne*	Benthopelagic	LC (Med)	AG & LEV	[40]
I			*Pagellus bogaraveo*	Benthopelagic	LC (Med)	Ion & C Med	[58, 59]
I			*Pagellus erythrinus*	Benthopelagic	LC (Med)	AD; Ion & C Med; AG & LEV	[29, 40, 45, 59]
I			*Pagrus pagrus*	Benthopelagic	LC (Med)	AG & LEV	[40]
I			*Pelates quadrilineatus*	Reef-associated	NE	AG & LEV	[40]
I			*Phycis phycis*	Benthopelagic	LC (Med)	W Med	[31]
I			*Polyacanthonotus rissoanus*	Bathydemersal	LC (Med)	W Med	[57]

(continued)

Table 1 (continued)

ML effect	Taxon	Phylum	Species	Habitat	IUCN status	MSFD subregion	Reference
I			*Polyprion americanus*	Demersal	DD (Med)	W Med	[22]
I			*Pomadasys incisus*	Demersal	LC (Med)	AG & LEV	[40]
I			*Sardina pilchardus*	Pelagic	LC (Med)	AD; Ion & C Med; AG & LEV; W Med	[29, 40, 41, 44, 45, 48, 60]
I			*Saurida undosquamis*	Reef-associated	LC	AG & LEV	[40]
I			*Schedophilus ovalis*	Benthopelagic	LC (Med)	W Med	[22]
I			*Sciaena umbra*	Demersal	VU (Med)	AG & LEV	[40]
I			*Scomber japonicus*	Pelagic	LC	AD; AG & LEV	[40, 45]
I			*Scorpaena* spp.	Demersal		W Med	[31]
I			*Seriola dumerili*	Reef-associated	LC	W Med	[22]
I			*Serranus cabrilla*	Demersal	LC (Med)	AG & LEV	[40]
I			*Siganus luridus*	Reef-associated	LC	AG & LEV	[40, 61]
I			*Siganus rivulatus*	Reef-associated	LC	AG & LEV	[61]
I			*Solea solea*	Demersal	LC (Med)	AD	[45, 62]
I			*Sparus aurata*	Demersal	LC (Med)	AD; AG & LEV	[40, 45]
I			*Spondyliosoma cantharus*	Benthopelagic	LC (Med)	W Med	[31]
I			*Synchiropus phaeton*	Demersal	LC (Med)	W Med	[22]
I			*Thunnus alalunga*	Pelagic	LC (Med)	Ion & C Med	[63]
I			*Thunnus thynnus*	Pelagic	EN (Med)	Ion & C Med	[63]
I			*Trachinotus ovatus*	Pelagic	LC (Med)	Ion & C Med	[64]
I			*Trachinus draco*	Demersal	LC (Med)	W Med	[22]

I			*Trachurus mediterraneus*	Pelagic	LC	AG & LEV; W Med	[40, 41]
I			*Trachurus picturatus*	Benthopelagic	LC (Med)	W Med	[22]
I			*Trachurus trachurus*	Pelagic	LC (Med)	AD	[45]
I			*Trachyrincus scabrus*	Bathydemersal	LC (Med)	W Med	[43]
I			*Upeneus moluccensis*	Reef-associated	LC	AG & LEV	[40]
I			*Upeneus pori*	Demersal	NE	AG & LEV	[40]
I			*Uranoscopus scaber*	Demersal	LC (Med)	W Med	[31]
I			*Zeus faber*	Benthopelagic	LC (Med)	W Med	[50]
I			*Xiphias gladius*	Pelagic	NT (Med)	Ion & C Med	[63]
I	Elasmobranchs		*Centroscymnus coelolepis*	Bathydemersal	LC (Med)	W Med	[43, 65]
I			*Etmopterus spinax*	Bathydemersal	LC (Med)	Ion & C Med; W Med	[43, 55, 58, 66]
I			*Galeus melastomus*	Demersal	LC (Med)	Ion & C Med; W Med	[43, 55, 58, 65, 66]
I			*Prionace glauca*	Pelagic	CR (Med)	W Med	[67]
I			*Pteroplatytrygon violacea*	Pelagic	LC (Med)	Ion & C Med	[58]
I			*Raja clavata*	Demersal	NT (Med)	W Med	[22]
I			*Scyliorhinus canicula*	Demersal	LC (Med)	W Med	[66]
I			*Squalus acanthias*	Benthopelagic	EN (Med)	AD	[44]
I			*Squalus blainville*	Demersal	DD (Med)	Ion & C Med	[58]
I	Marine mammals		*Grampus griseus*	Pelagic	DD (Med)	AG & LEV; Ion & C Med	[68, 69]
I			*Phocoena phocoena*	Pelagic	LC	AG & LEV	[68]
I			*Physeter macrocephalus*	Pelagic	EN (Med)	AD; AG & LEV; Ion & C Med; W Med	[2, 68, 70–74]
I			*Stenella coeruleoalba*	Pelagic	VU (Med)	AD	[75]
I			*Tursiops truncatus*	Pelagic	VU (Med)	AD; AG & LEV	[76–78]
I			*Ziphius cavirostris*	Pelagic	DD (Med)	AD; Ion & C Med	[68, 79]

(continued)

Table 1 (continued)

ML effect	Taxon	Phylum	Species	Habitat	IUCN status	MSFD subregion	Reference
I	Sea turtles		*Caretta caretta*	Benthopelagic	LC (Med)	AD; Ion & C Med; AG & LEV; W Med	[80–95]
I			*Chelonia mydas*	Benthopelagic	EN	Ion & C Med; AG & LEV	[84, 90, 94, 95, 96]
I			*Dermochelys coriacea*	Benthopelagic	VU	AD; AG & LEV	[73, 97]
I			*Trionyx triunguis*	Benthopelagic	CR (Med)	AG & LEV	[90]
I	Seabirds		*Calonectris diomedea*		LC	W Med	[98]
I			*Stercorarius skua*		LC	W Med	[98]
I			*Falco eleonorae*		LC	AG & LEV	[99]
I			*Ichthyaetus audouinii*		NE	W Med	[98]
I			*Ichthyaetus melanocephalus*		NE	W Med	[98]
I			*Larus michahellis*		LC	W Med	[98]
I			*Morus bassanus*		LC	W Med	[98]
I			*Puffinus yelkouan*		VU	W Med	[98]
I			*Puffinus mauretanicus*		CR	W Med	[98]
I			*Rissa tridactyla*		VU	W Med	[98]

Study locations were grouped according to MSFD subregions (*W Med* the Western Mediterranean Sea, *Ion & C Med* the Ionian Sea and the Central Mediterranean Sea, *AD* the Adriatic Sea, *AG & LEV* the Aegean-Levantine Sea). The species conservation status according to the IUCN Red List is reported (*NE* not evaluated, *LC* least concern, *DD* data deficient, *NT* near threatened, *EN* endangered, *VU* vulnerable, *CR* critically endangered). It is reported when the assessment is specific for the Mediterranean Sea; otherwise, it is the global assessment

Table 2 List of Mediterranean species affected by marine litter (*E* entanglement, *O* other effects, such as rafting, colonization, etc.) according to published literature and online sources (search closed in August 2019)

ML effect	Taxon	Phylum	Species	Habitat	IUCN status	MSFD subregion	Reference
Plantae							
		Tracheophyta					
O			*Cymodocea nodosa*		LC (Med)	W Med	[100]
O			*Posidonia oceanica*		LC (Med)	W Med	[100]
		Rhodophyta					
O			*Hydrolithon farinosum*			W Med	[100]
Chromista							
		Ochrophyta					
O			*Achnanthes* spp.			W Med	[101, 102]
O			*Amphora* spp.			W Med	[101]
O			*Ceratoneis closterium*			W Med	[101]
O			*Cyclotella* spp.			W Med	[101]
O			*Cylindrotheca* spp.			W Med	[102]
O			*Cymbella* spp.			W Med	[101]
O			*Cystoseira* spp.			W Med	[100]
O			*Cocconeis* spp.			W Med	[101]
O			*Entomoneis* spp.			W Med	[101]
O			*Licmophora* spp.			W Med	[101]
O			*Navicula* spp.			W Med	[101, 102]
O			*Pleurosigma* spp.			W Med	[101]
O			*Thalassionema nitzschioides*			W Med	[101]
O			*Thalassionema* spp.			W Med	[102]
O			*Thalassiosira* spp.			W Med	[102]

(continued)

Table 2 (continued)

ML effect	Taxon	Phylum	Species	Habitat	IUCN status	MSFD subregion	Reference
		Myzozoa					
O			*Alexandrium taylori*			W Med	[103]
O			*Coolia monotis*			W Med	[102]
O			*Coolia* spp.			W Med	[101, 103]
O			*Dinophysis* spp.			W Med	[101]
O			*Heterocapsa* spp.			W Med	[101]
O			*Ostreopsis* spp.			W Med	[103]
O			*Prorocentrum lima*			W Med	[101]
O			*Prorocentrum minimum*			W Med	[101]
O			*Prorocentrum* spp.			W Med	[103]
O			*Pentapharsodinium tyrrhenicum*			W Med	[101]
		Haptophyta					
O			*Calcidiscus leptoporus*			W Med	[101]
O			*Coronosphaera mediterranea*			W Med	[101]
O			*Helicosphaera carteri*			W Med	[101]
O			*Syracosphaera halldalii*			W Med	[101]
O			*Syracosphaera molischii*			W Med	[101]
O			*Syracosphaera pulchra*			W Med	[101]
O			*Umbilicosphaera sibogae*			W Med	[101]
O			*Zygosphaera hellenica*			W Med	[101]

		Foraminifera					
O			*Miniacina miniacea*			Ion & C Med	[104]
O			*Tretomphaloides concinnus*			W Med	[105]
Bacteria							
		Proteobacteria					
O			*Acinetobacter junii*			AD	[106]
O			*Acinetobacter lwoffii*			AD	[106]
O			*Aeromonas bestiarum*			AD	[106]
O			*Aeromonas salmonicida*			AD	[106]
O			*Aeromonas sanarellii*			AD	[106]
O			*Aestuariibacter halophilus*			AD	[106]
O			*Aestuariibacter litoralis*			AD	[106]
O			*Alteromonas macleodii*			AD	[106]
O			*Alteromonas marina*			AD	[106]
O			*Alteromonas mediterranea*			AD	[106]
O			*Alteromonas* spp.			W Med	[107]
O			*Croceicoccus naphtovorans*			W Med	[107]
O			*Erythrobacter* spp.			W Med	[107]
O			*Erythrobacter citreus*			AD	[106]
O			*Haemophilus piscium*			AD	[106]
O			*Haliea salexigens*			AD	[106]
O			*Hyphomonas* spp.			W Med	[107]

(continued)

Table 2 (continued)

ML effect	Taxon	Phylum	Species	Habitat	IUCN status	MSFD subregion	Reference
O			*Oceanibaculum pacificum*			AD	[106]
O			*Parasphingopyxis lamellibrachiae*			AD	[106]
O			*Parvularcula oceani*			AD	[106]
O			*Pelagibacter ubique*			W Med	[107]
O			*Proteus mirabilis*			AD	[106]
O			*Pseudoruegeria sabulilitoris*			AD	[106]
O			*Roseobacter* spp.			W Med	[107]
O			*Roseovarius algicolus*			AD	[106]
O			*Shimia marina*			W Med	[107]
O			*Tepidamorphus gemmatus*			AD	[106]
O			*Thioalkalivibrio sulfidophilus*			AD	[106]
O			*Vibrio anguillarum*			W Med	[107]
O			*Vibrio harveyi*			W Med	[107]
O			*Vibrio pectinicida*			W Med	[107]
O			*Vibrio xiamenensis*			W Med	[107]
		Chloroflexi					
O			*Anaerolinea thermophila*			AD	[106]
		Planctomycetes					
O			*Bythopirellula goksoyri*			AD	[106]

O			*Croceitalea dokdonensis*			AD	[106]
O			*Rhodopirellula baltica*			AD	[106]
O			*Rhodopirellula lusitana*			AD	[106]
		Bacteroidetes					
O			*Nonlabens dokdonensis*			AD	[106]
O			*Tenacibaculum* spp.			W Med	[107]
O			*Ulvibacter litoralis*			W Med	[107]
		Cyanobacteria					
O			*Calothrix* spp.			W Med	[107]
O			*Leptolyngbya* spp.			W Med	[107]
O			*Microchaete tenera*			W Med	[107]
O			*Oscillatoria* spp.			W Med	[107]
O			*Pleurocapsa* spp.			W Med	[107]
O			*Phormidium* spp.			W Med	[107]
O			*Prochlorococcus marinus*			AD	[106]
O			*Rivularia* spp.			W Med	[107]
O			*Scytonema* spp.			W Med	[107]
O			*Synechococcus* spp.			W Med	[107]
Animalia							
	Invertebrates	Annelida					
O			*Bonellia viridis*	Benthic	NE	AG & LEV	[108]
O			*Filograna implexa*	Sessile	NE	W Med	[109]
O			*Filograna* spp.	Sessile		Ion & C Med; W Med	[104, 110]
O			*Filogranula gracilis*	Sessile	NE	Ion & C Med	[104]

(continued)

Table 2 (continued)

ML effect	Taxon	Phylum	Species	Habitat	IUCN status	MSFD subregion	Reference
O			*Filogranula* spp.	Sessile		Ion & C Med	[104]
O			*Hydroides* spp.	Sessile		AG & LEV	[111]
O			*Metavermilia multicristata*	Sessile	NE	Ion & C Med	[104]
O			*Nereis splendida*	Benthic	NE	W Med	[100]
O			*Placostegus tridentatus*	Sessile	NE	Ion & C Med	[104]
O			*Sabella pavonina*	Sessile	NE	W Med	
O			*Salmacina* spp.	Sessile		W Med	[110]
O			*Semivermilia* spp.	Sessile		Ion & C Med	[104]
O			*Semivermilia agglutinata*	Sessile	NE	Ion & C Med	[104]
O			*Serpula vermicularis*	Sessile	NE	Ion & C Med; AG & LEV	[104, 111]
O			Serpulidae	Sessile		Ion & C Med; AG & LEV	[104, 111]
O			*Spirobranchus polytrema*	Sessile	NE	W Med	[100]
O			*Spirobranchus triqueter*	Sessile	NE	AG & LEV	[111]
O			*Vermiliopsis* spp.	Sessile		Ion & C Med; W Med	[104, 112]
		Mollusca					
O			*Anomia ephippium*	Benthic	NE	AG & LEV	[111]
O			*Ascidiella aspersa*	Sessile	NE	AG & LEV	[111]
O			*Corbula gibba*	Benthic	NE	AG & LEV	[111]
O			*Diodora* spp.	Benthic		AG & LEV	[111]
O			*Doto* spp.			W Med	[100]

O			*Fiona pinnata*	Pelagic	NE	W Med	[100]
O			*Mytilus* spp.	Benthic		Ion & C Med	[113]
O			*Modiolus barbatus*	Benthic	NE	AG & LEV	[108]
O			*Musculus subpictus*	Benthic	NE	AG & LEV	[111]
O			*Neopycnodonte cochlear*	Benthic	NE	W Med; Ion & C Med; AG & LEV	[104, 111, 112]
O			*Ostrea edulis*	Benthic	NE	Ion & C Med	[113]
O			*Pedicularia sicula*	Benthic	NE	Ion & C Med	[104]
E			*Sepia officinalis*	Benthic	LC	AG & LEV	[114]
O			*Striarca lactea*	Benthic	NE	Ion & C Med	[104]
		Cnidaria					
E			*Acanthogorgia hirsuta*	Benthic	LC (Med)	Ion & C Med; W Med	[109, 110, 115]
O			*Alcyonium coralloides*	Sessile	LC (Med)	Ion & C Med	[104]
E			*Antipathella subpinnata*	Benthic	NT (Med)	Ion & C Med; W Med	[109, 110, 115–117]
E			*Antipathes dichotoma*	Demersal	NT (Med)	Ion & C Med; W Med	[109, 115, 117]
E			*Bebryce mollis*	Benthic	DD (Med)	Ion & C Med; W Med	[115, 118]
O			*Bougainvillia muscus*	Benthic	NE	AG & LEV	[111]
E			*Callogorgia verticillata*	Sessile	NT (Med)	Ion & C Med; W Med	[109, 115–120]
O			*Coenocyathus cylindricus*	Sessile	DD (Med)	Ion & C Med	[104]
E			*Corallium rubrum*	Sessile	EN (Med)	Ion & C Med; W Med	[109, 110, 115, 116]
O			Caryophylliidae	Sessile		Ion & C Med	[104]
O			*Clytia hemisphaerica*	Sessile	NE	W Med	[100]
E			*Dendrophyllia cornigera*	Reef-associated	EN (Med)	W Med	[109, 117]

(continued)

Table 2 (continued)

ML effect	Taxon	Phylum	Species	Habitat	IUCN status	MSFD subregion	Reference
E			*Dendrophyllia ramea*	Reef-associated	VU (Med)	W Med; AG & LEV	[110, 121]
O			*Desmophyllum dianthus*	Reef-associated	EN (Med)	W Med; Ion & C Med	[104, 122]
E, O			*Desmophyllum pertusum*	Sessile	NE	W Med; Ion & C Med	[104, 120, 123]
O			*Errina aspera*	Sessile	NE	Ion & C Med	[104]
O			*Eudendrium* spp.			W Med	[100]
E			*Eunicella cavolini*	Benthic	NT (Med)	Ion & C Med; W Med	[109, 110, 115, 116, 118, 124, 125]
E			*Eunicella singularis*	Benthic	NT (Med)	Ion & C Med; W Med	[110, 115]
E			*Eunicella verrucosa*	Sessile	NT (Med)	Ion & C Med; W Med	[110, 115]
O			*Gonothyraea loveni*	Pelagic	NE	W Med	[100]
O			Hydrozoa	Sessile		Ion & C Med	[104]
O			*Laomedea angulata*	Pelagic	NE	W Med	[100]
E			*Leiopathes glaberrima*	Reef-associated	EN (Med)	Ion & C Med; W Med	[109, 115, 117, 119, 126]
E, O			*Madrepora oculata*	Sessile	EN (Med)	Ion & C Med; W Med	[104, 109, 112, 120, 123, 127, 128]
O			*Obelia dichotoma*	Sessile	NE	W Med	[100]
O			*Oculina patagonica*	Sessile	LC (Med)	W Med	[129]
E			*Paramuricea clavata*	Sessile	VU (Med)	Ion & C Med; W Med; AD	[109, 110, 115–118, 130, 131]
E			*Paramuricea macrospina*	Benthic	DD (Med)	Ion & C Med	[115]
E			*Parantipathes larix*	Benthic	NT (Med)	Ion & C Med; W Med	[109, 115, 124]
E			*Savalia savaglia*	Benthic	NT (Med)	Ion & C Med; W Med	[115, 116]

O			*Sertularella* spp.	Sessile		Ion & C Med	[104]
O			Sertulariidae	Sessile		Ion & C Med	[104]
E			*Swiftia dubia*	Sessile	NE	Ion & C Med	[132]
E			*Viminella flagellum*	Benthic	NT (Med)	Ion & C Med; W Med	[109, 115, 116]
		Echinodermata					
O			*Arbacia lixula*	Benthic	NE	W Med	[100]
E			*Cidaris cidaris*	Benthic	NE	Ion & C Med	[132]
		Porifera					
O			*Haliclona* (*Reniera*) spp.	Sessile		W Med	[112]
E			*Geodia cydonium*	Sessile	NE	AD	[133]
E			*Pachastrella monilifera*	Sessile	NE	W Med	[134]
E			*Poecillastra compressa*	Sessile	NE	W Med	[134]
E			*Raspailia* (*Raspailia*) *viminalis*	Sessile	NE	Ion & C Med	[115]
O			*Sycon raphanus*	Sessile	NE	Ion & C Med	[104]
		Bryozoa					
O			*Amathia gracilis*	Sessile	NE	W Med	[100]
O			*Callopora lineata*	Sessile	NE	W Med	[100]
O			*Cellaria salicornioides*	Sessile	NE	Ion & C Med	[104]
O			*Cellepora* spp.	Benthic		Ion & C Med	[104]
O			*Celleporina* spp.	Sessile		Ion & C Med	[104]
O			Cheilostomatida	Sessile		Ion & C Med	[104]
O			Cyclostomatida	Sessile		Ion & C Med	[104]
O			*Electra posidoniae*	Benthic	NE	W Med	[100]
O			*Haplopoma* spp.	Sessile		Ion & C Med	[104]

(continued)

Table 2 (continued)

ML effect	Taxon	Phylum	Species	Habitat	IUCN status	MSFD subregion	Reference
O			*Membranipora membranacea*	Sessile	NE	W Med	[100]
O			*Puellina gattyae*	Sessile	NE	Ion & C Med	[104]
O			*Puellina* spp.	Sessile		Ion & C Med	[104]
		Brachiopoda					
O			*Gryphus vitreus*	Benthic	NE	W Med	[135]
		Arthropoda					
O		Amphipoda	*Caprella andreae*	Benthic	NE	W Med	[136]
O			*Caprella hirsuta*	Benthic	NE	W Med	[136]
O			*Elasmopus brasiliensis*	Benthic	NE	W Med	[136]
O			*Hyale grimaldii*	Benthic	NE	W Med	[136]
O			*Jassa cadetta*	Benthic	NE	W Med	[136]
O			*Phtisica marina*	Pelagic	NE	W Med	[100]
O		Lepadiformes	*Lepas* (*Anatifa*) *anatifera*	Sessile	NE	W Med; AG & LEV	[111, 136]
O			*Lepas* (*Anatifa*) *pectinata*	Sessile	NE	W Med	[100]
O			*Lepas* spp.	Sessile		Ion & C Med	[113]
O		Isopoda	*Idotea balthica*	Benthic	NE	Mediterranean	[137]
O			*Idotea metallica*	Pelagic	NE	Mediterranean; W Med	[100, 136–139]
O		Sessilia	*Adna anglica*	Sessile	NE	Ion & C Med	[104]
O			*Austrominius modestus*	Sessile	NE	W Med	[140]
O			*Chelonibia testudinaria*	Sessile	NE	Ion & C Med	[113]
O			*Hesperibalanus fallax*	Sessile	NE	W Med	[140]

O			*Megabalanus tulipiformis*	Sessile	NE	W Med; Ion & C Med	[104, 140]
O			*Octolasmis* spp.	Sessile		Ion & C Med	[104]
O			*Perforatus perforatus*	Sessile	NE	W Med; AG & LEV	[111, 140]
O			*Pachylasma giganteum*	Sessile	NE	Ion & C Med	[104]
E		Decapoda	*Geryon trispinosus*	Benthic	NE	W Med	[141]
O			*Liocarcinus navigator*	Benthic	NE	AD	[142]
E			*Maja squinado*	Benthic	NE	W Med	[143]
E, O			*Paromola cuvieri*	Demersal	NE	W Med	[122, 134]
O			*Planes minutus*	Pelagic	NE	AD	[142]
Vertebrates							
		Chordata					
O	Tunicates		*Phallusia mammillata*	Sessile	NE	AG & LEV	[111]
O			*Ciona intestinalis*	Sessile	NE	AG & LEV	[111]
O			*Styela* spp.	Sessile	NE	AG & LEV	[111]
E	Teleosts		*Conger conger*	Demersal	LC	W Med	[143]
E			*Epinephelus aeneus*	Demersal	NT (Med)	AG & LEV	[114]
E			*Scorpaena notata*	Demersal	LC (Med)	W Med	[143]
E			*Scorpaena porcus*	Demersal	LC (Med)	W Med	[143]
E			*Scorpaena scrofa*	Demersal	LC (Med)	W Med; AG & LEV	[114, 143]
E	Elasmobranchs		*Mobula mobular*	Pelagic	EN (Med)	W Med	[143]
E			*Prionace glauca*	Pelagic	CR (Med)	W Med	[144]
E			*Scyliorhinus* spp.	Demersal		AD	[145]
E	Marine mammals		*Monachus monachus*	Bathydemersal	CR (Med)	Mediterranean	[20]
E	Sea turtles		*Caretta caretta*	Benthopelagic	LC (Med)	AD; Ion & C Med; AG & LEV; W Med	[84, 90, 93, 94, 146–148]

(continued)

Table 2 (continued)

ML effect	Taxon	Phylum	Species	Habitat	IUCN status	MSFD subregion	Reference
E			*Chelonia mydas*	Benthopelagic	EN	AG & LEV	[90, 149]
E	Seabirds		*Charadrius alexandrinus*		LC	W Med	[150]
E			*Charadrius hiaticula*		LC	W Med	[150]
E			*Larus michahellis*		LC	W Med	[150]
O			*Morus bassanus*		LC	W Med	[151]
E			*Phoenicurus ochruros*		LC	W Med	[150]

Study locations were grouped according to MSFD subregions (*W Med* the Western Mediterranean Sea, *Ion & C Med* the Ionian Sea and the Central Mediterranean Sea, *AD* the Adriatic Sea, *AG & LEV* the Aegean-Levantine Sea). The species conservation status according to the IUCN Red List is reported (*NE* not evaluated, *LC* least concern, *DD* data deficient, *NT* near threatened, *EN* endangered, *VU* vulnerable, *CR* critically endangered). It is reported when the assessment is specific for the Mediterranean Sea; otherwise, it is the global assessment

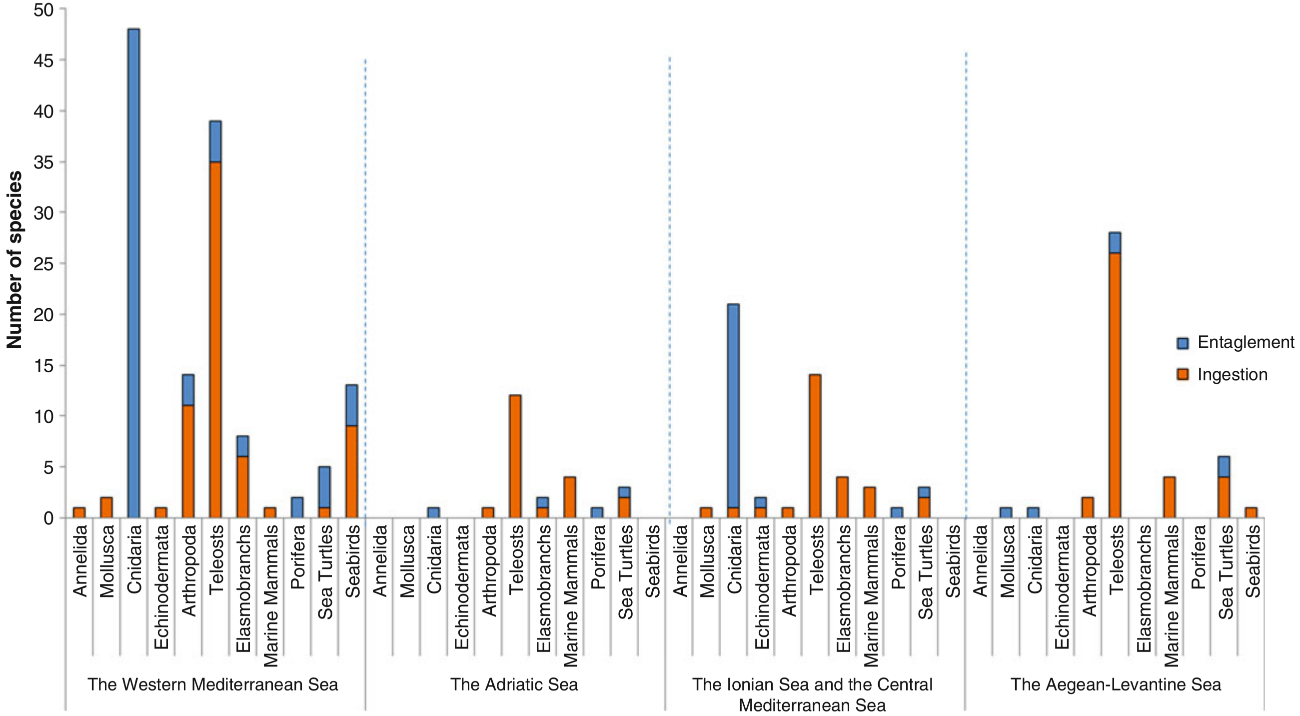


Fig. 1 Number of species affected by marine litter ingestion and entanglement in the Mediterranean subregions

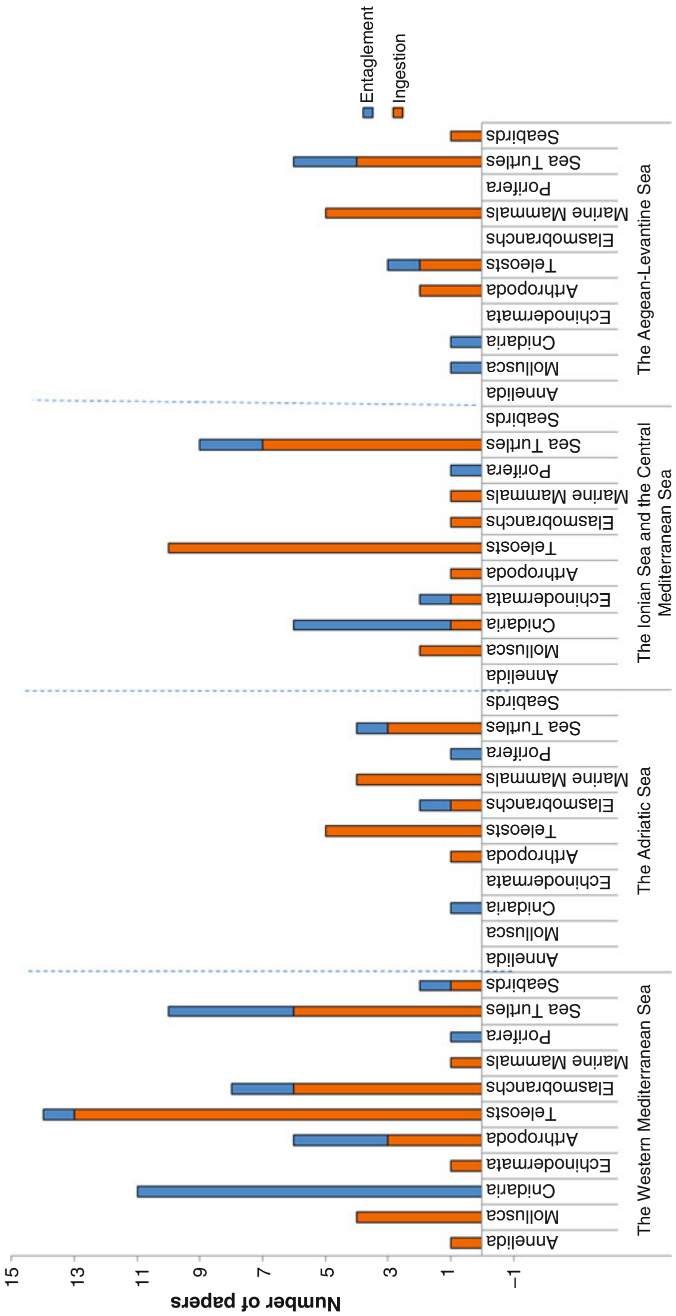


Fig. 2 Number of reports on marine litter ingestion and entanglement in the Mediterranean subregions

microplastics can make them indistinguishable from natural prey items (same size fraction of some planktonic organisms), increasing their availability to a wide range of planktivorous feeders ([10] and references therein). McCord and Campana [157] supposed that the consumption of anthropogenic material by blue sharks is related to the species opportunistic predator behaviour. Anastasopoulou et al. [58] reported that the sharks' ability to consume virtually anything of appropriate size during their feeding activity might be the reason of the plastic consumption by the species *Etmopterus spinax*, *Galeus melastomus* and *Squalus blainvillei* in the deep waters of the Eastern Ionian Sea. Finally, considering plastics as curious/attractive objects ([23] and references therein; [158]), Moss [159] reported that elasmobranchs might consume metal objects because they are attracted by their electric field.

Accidental ingestion usually occurs through passive predation or by filter-feeding activity. Filter-feeding organisms ingest plastics and microplastics by filtering large volumes of water containing them. Uptake of plastics and microplastics may also occur accidentally by passive ingestion of sediments (if plastics are of the same size fraction as sediments) when deposit or detritivores organisms are feeding (e.g. [10] and references therein; [1, 29, 39]).

Litter uptake can also occur as a result of secondary ingestion (through prey that has already ingested plastic) (e.g. [160, 161]). Predatory organisms may indirectly accumulate plastics during the ingestion of contaminated prey, which may lead to bioaccumulation at upper trophic levels. Possatto et al. [161] mentioned that some fish species (e.g. *Cynoscion acoupa*, *Centropomus undecimalis* and *Dasyatis guttata*) prey on smaller fish that have been previously contaminated by plastics. Eriksson and Burton [160] found small plastic particles in the faeces of fur seals, which were attributed to secondary ingestion through the consumption of myctophid species contaminated with plastics. The fact that many small plastics have been found in myctophid species, a common prey for large predators as tunas, supports the hypothesis of secondary ingestion ([23] and references therein). There is evidence also from laboratory studies that microplastics can be transferred from prey to predator [162] and this is therefore very likely in the environment when contaminated organisms are ingested as a whole.

Another way of litter uptake can occur through parental delivery in seabirds [164]. Adult seabirds collect plastic pieces at sea, together with food items, and bring them to feed the fledgeling chicks. Acampora et al. [165] found more plastic in the chicks than adult birds and mentioned that young birds are prone to be fed with plastic particles by their parents before fledging.

Ingested plastics are believed to have a variety of consequences for the consuming organism. The most serious effect is the direct mortality that can occur when stomachs or intestines become completely blocked or internally injured by sharp plastic objects ([8] and references therein). Other harmful effects include the blockage of the digestive tract, false feeling of satiation, reduced fitness, diminished predator avoidance, blockage of gastric enzyme production, diminished feeding stimulus, nutrient dilution, reduced growth rates, lowered steroid hormone levels, delayed ovulation and reproductive failure and absorption of toxins (e.g. [8, 10, 23, 97]). Furthermore, microplastic ingestion may cause several biochemical responses and impacts at the cellular level as cellular necrosis, oxidative stress, neurotoxic effects, liver toxicity, cancer, impaired reproductive activity, decreased immune response and malformation in animals and humans [166, 167]. Large amounts of ingested plastic might also affect an animal's buoyancy, either directly through its low density or by impairing digestive function leading to gas buildup. This may be a serious problem for turtles, potentially impairing their ability to dive and consequently leading them to starvation [168]. Additionally, plastics are liable to carry chemicals of a small molecular size as hydrophobic persistent organic pollutants (POPs) that are absorbed from the surrounding seawater (e.g. PCBs and DDT) and additives/plasticisers monomers or oligomers (e.g. flame retardants and antimicrobial agents, phthalates, bisphenol A, nonylphenols, polybrominated diphenyl ethers) of their component molecules, which were added during the manufacturing of plastics. All the above chemicals are known for their biological consequences (e.g. oestrogenic effects, testosterone reduction) on the organisms [169]. Plastics, when ingested, may also serve as indirect vehicles for the transport of pathogens via the trophic web in wild marine organisms [10]. However, further research is needed in order to understand if and to what extent microplastics pose an actual risk to wildlife and consequently to human health [170].

The extent of the harm posed by plastics on marine animals varies among species [8], and little is known about the factors influencing litter ingestion. Marine organisms feeding in different marine habitats would be exposed to different plastic abundances and consequently to different plastic availability. Few studies have examined this hypothesis. Van Franeker et al. [171, 172] provided evidence that plastic abundance in fulmars' stomachs reflected local or regional pollution levels. According to these authors, fulmars in Arctic Canada experienced lower debris ingestion rates than those in the North Sea, a difference that may result from relatively cleaner seas in Arctic Canada [171]. Anastasopoulou et al. [45], analysing several fish species from the Adriatic-Ionian macroregion, reported that the higher number of macrolitter found in the guts of the pelagic species sampled in the Adriatic Sea may be related with the higher average density of floating macrolitter found in the coastal Adriatic waters than that of the NE Ionian Sea [173]. Nevertheless, this hypothesis is not easy to be verified as many other parameters (e.g. atmospheric and oceanographic conditions) that may alter the plastic abundance at different temporal scales are involved. Differences in plastic ingestion among different locations may also reflect differences in the sampling and processing methodologies followed by each work team and not represent actual local differences in the plastic density. Moreover, different species inhabiting the same geographic area may utilize different feeding strategies, exploit differing habitats, and target different preys and may, therefore, vary in the amount of plastic ingestion. Some authors (e.g. [29, 40]) showed that pelagic fish ingest more microplastics than fish living in other habitats and exhibit higher frequencies of microplastic ingestion than demersal fish. Conversely, there are studies that reported no differences in the frequency of microplastic ingestion between pelagic and demersal fishes (e.g. [174, 175]). Anastasopoulou et al. [45] found higher macroplastic ingestion by pelagic than demersal and mesopelagic fish species, but no difference in microplastic ingestion among them.

Additionally, some organisms are subjected to ontogenetic habitat and feeding strategy shifts, and consequently, the type and number of plastics ingested by a species may vary during its different life stages. For example, juvenile birds have been reported to ingest significantly more pieces of litter than adults [165, 176]. The higher number of ingested plastics in young dolphins found by Denuncio et al. [177] was attributed to the juvenile inexperience to eat the appropriate prey. Bessa et al. [178] found a high amount of microplastics in young specimens (about 120 mm size) of *Diplodus vulgaris*; the young specimens of this species usually find shelter in estuarine waters, which are under strong influence of tidal movements and more sensitive to plastic availability. Their presence in these environments along with their opportunistic behaviour may be linked to a higher likelihood of microplastic ingestion.

First reports of plastic ingestion by marine animals date back to the early 1960s [179, 180], and successively both the number of individuals and the number of species known to ingest litter increased. However, until the 1980s the number of documents reporting ingestion did not significantly change [181]. Laist [20] reported 177 species to have been impacted by plastic ingestion worldwide, whereas Kühn et al. [23] increased this number to 331 species worldwide.

In the Mediterranean, Fossi et al. [26] documented 91 marine species having ingested marine litter, as reported in 48 papers. Our bibliographic research resulted in 76 papers documenting litter ingestion by 116 species, belonging to the taxa annelids, molluscs, cnidarians, echinoderms, arthropods, teleosts, elasmobranchs, marine mammals, sea turtles and seabirds, in a period of 31 years. Thus, a 58% increase in the number of papers was observed after Fossi et al. [26] publication. Despite the high increase in the number of published papers, the number of species increased only by 27.7%. Most of the studies were conducted in the Western Mediterranean Sea and the Ionian Sea and the Central Mediterranean Sea subregions, whereas the Adriatic and Aegean-Levantine Sea subregions were less investigated. With regard to the number of species, the higher number was observed in the Western Mediterranean Sea subregion, while the lower in the Adriatic Sea subregion.

The first study of litter ingestion in the Mediterranean was published in 1988 by Gramentz [82] for the species *Caretta caretta* and was carried out in 1986. In the last decade, the number of publications documenting litter ingestion in marine wildlife increased at an accelerating rate (79% of the collected papers were published in the last decade, from 2010 till August 2019). The low number of published works reporting litter ingestion in the past can be explained by the fact that they were mainly part of diet studies ([58] and references therein). Moreover, Carson [182] reported that this might be because many fishes that consume plastic are able to pass it through their digestive system or that past studies on stomach contents did not note/report any plastic that was encountered. In the last years, the growing recognition and concern of the marine litter problem by the media, by regional and global organizations and by the European Union that have launched several initiatives and legislative tools (e.g. MSFD) to protect the marine environment, have pushed the scientific community to pay more attention in recording and investigating litter ingestion by marine organisms and the potential risk to human consumption.

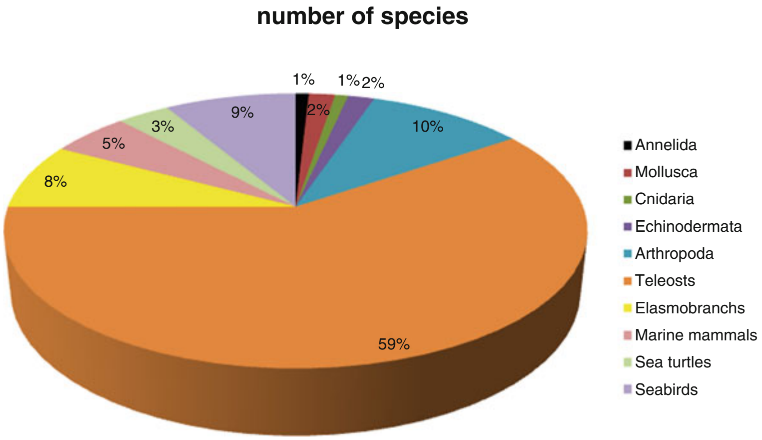


Fig. 3 Percentage of species known to have ingested litter by the taxonomic group from 1988 to August 2019 in the Mediterranean Sea

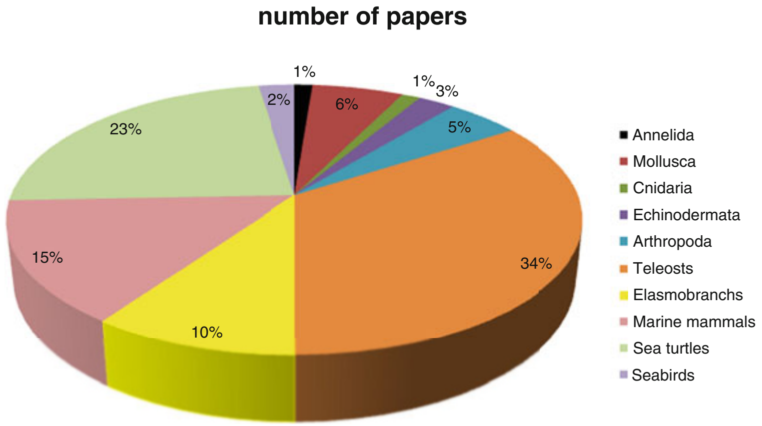


Fig. 4 Percentage of papers documenting the ingestion of marine litter by the taxonomic group from 1988 to August 2019 in the Mediterranean Sea

Teleosts were the taxonomic group with the higher number of species (59%) known to ingest plastics, followed by arthropods (12%), seabirds (10%) and elasmobranchs (9%) (Fig. 3). The number of species belonging to the other taxa was very low (<4%). Accordingly, the majority (34%) of papers reporting litter ingestion by Mediterranean marine life regarded Teleosts (Fig. 4). This could derive from the fact firstly that fish constitutes an important food source for humans and secondly that they are easy to be collected and subsequently be monitored during experimental trawl fishery surveys.

2.1 Invertebrates

In the Mediterranean, ingestion of plastics was verified in annelids (1 species), molluscs (2 species), cnidarians (1 species), echinoderms (2 species) and arthropods (12 species). The small size of microplastics makes them available for ingestion to a variety of invertebrates. Information for annelids was only available from Gusmão et al. [28]. The microfibres identified in the guts of the annelid *Saccocirrus pussicus* collected in Sardinia (Italy) were probably related to the feeding behaviour (nonselective particle feeders) of the species, which may increase the probability of accidental capture and ingestion of microfibres. No indication of physical damage due to the passage of microfibres through their gut and no mortality were detected.

Microplastic ingestion by *Mytilus galloprovincialis* was evidenced in the Mediterranean by [29–32]. Mussels are filter feeders and thus process relatively large amounts of water during feeding. This maximizes their exposure to any harmful material within the water column and can result in the accumulation of chemical pollutants and microplastics available in the environment. According to Digka et al. [29], the majority of ingested microplastics in *M. galloprovincialis* examined were fragments, in accordance with microplastics in seawater in the study area (Northern Ionian Sea). Polyethene was the most frequent polymer type found. The presence of microplastics in wild or cultured mussels [32, 183] sold for human consumption has raised a concern about whether the ingestion of litter by these organisms can cause impact on human health. The risk for human health may derive from the fact that mussels are consumed as a whole, including the gut [3]. However, recent work [184] suggests that microplastics in shellfish pose negligible risk to human health when compared to fibre exposure during a meal via dust fallout in a household. Although it is evident that humans are exposed to microplastics through their diet, there are still a lot of uncertainties around the direct connection between the ingested microplastics and the food targeted for human consumption [3, 183]. Litter ingestion has also been reported for another mollusc, the spider octopus (*Octopus salutii*), in the Western Mediterranean Sea [22].

Marine litter was found in the gastrovascular cavity of *Pelagia noctiluca* specimens in the Tyrrhenian Sea [33]. The authors mentioned that these plastic fragments were probably actively ingested by the jellyfish that wrongly recognized them as food.

Microplastic ingestion was found to occur in two Holothurian species [34, 35]. Renzi et al. [35] found consistency between the types of ingested plastics and those observed in the sediments in the studied areas, supporting the hypothesis of direct ingestion of the plastic litter from the sediment. Holothurians are benthic organisms, which ingest a large volume of sediments. Two opposite hypotheses have been proposed for plastic ingestion by Holothurians related to their feeding strategies: random (the animals had to forage on contaminated sediment to encounter plastic particles) and selective (once encountered, plastic is separated from the sediment and eaten) feeding methods [10].

Litter was also detected in crustaceans and more specifically in amphipods, leptostraca and decapods. Remy et al. [36] investigated litter in four amphipod, one leptostraca and four decapod species in the Western Mediterranean Sea and found many artificial fibres like cellulose or viscose fibres by the textile industry. Information on the presence of plastics in the guts of decapods intended for human consumption is still limited. Evidence of plastic material was reported in the stomachs of *Nephrops norvegicus* by Cristo and Cartes [38], which was attributed to the low selectivity of the species in its feeding activities. Carreras-Colom et al. [37] found that 39.2% of the individuals of blue and red shrimp *Aristeus antennatus*, a species of high commercial interest, sampled in the deep waters of the Western Mediterranean Sea, contained microplastics in their stomachs. Microplastics were mainly fibres but also tightly tangled balls of plastic strands. The uptake of plastics may occur accidentally during species normal feeding activity that occurs in a close relationship with the substrate when the species preys on endobenthic and epibenthic invertebrates, exposing shrimps to the microplastics accumulated in the seabed. Carreras-Colom et al. [37] mentioned that it is also possible that *A. antennatus* actively preys upon microplastic fibres (as balls) because they can be similar in size to small polychaetes, an important diet prey of the species. However, no clear effects on the condition or diet of the species were observed. Finally, plastics were found in another high-value commercial shrimp species, Narwal shrimp (*Plesionika narval*) in the Aegean Sea [39]. Plastics were found in approximately 6% of the stomachs of this species and were identified as nylon related to fishing activities. The plastics were mainly in the form of fibres or tangled together (balls of plastics) as a loose knot occupying the entire stomach content. Approximately 15% of the examined stomachs with ingested plastics from both sexes were empty, especially those with ball plastics, indicating a possible blockage of their digestive system. The low values of the repletion index found in shrimps with ingested plastics further support this hypothesis [39]. The presence of plastics in the stomachs of *P. narval* may be related to the feeding behaviour of the species, which is a scavenger and detritivorous feeder. The simultaneous presence of plant remains is another evidence of the passive ingestion of plastics with sediments [39].

2.2 Teleosts

Plastic ingestion has been observed in a relatively large number of fish species (69) belonging to 13 orders. The most affected species were found to be the demersal ones (30.4%), followed by benthopelagic (24.6%), pelagic (15.9%), bathypelagic and reef-associated (11.6% for each category) and bathydemersal (5.8%) fish.

Most of the demersal species are used for human consumption (e.g. *Mullus surmuletus*, *M. barbatus*, *Merluccius merluccius*, *Polyprion americanus*, *Solea solea* and *Sparus aurata*). Among demersal species, the incidence of plastics in *S. solea* was found to be very high in the Adriatic Sea, where 95% of the specimens examined had microplastics in their gastrointestinal tract [62]. Plastics in the guts of

M. barbatus, a species of high commercial value, were studied by Avio et al. [185], Bellas et al. [53], Güven et al. [40], Anastasopoulou et al. [45], Digka et al. [29], Piccardo et al. [54] and Giani et al. [51]; plastic percentage ingestion ranged from 8 to 92%. Similarly, the percentage of plastic ingestion for *M. surmuletus* ranged between 27.3 and 70% [45, 186] for the Western Mediterranean and the Adriatic Sea, respectively. Anastasopoulou et al. [45] evidenced for the first time the occurrence of litter in the stomach of spotted flounder (*Citharus linguatula*). The high variability (from 8.3 to 100%) in the presence of plastics in the guts of *M. merluccius* among the different studies conducted in the Mediterranean [51, 52, 185] could be explained by the different environmental characteristics of the areas studied and by the different number of individuals examined (in some cases the number of individuals was very low). Moreover, regurgitation of the stomach content during the fishing due to the expansion of gas in the swim bladder, which is common for this species, may result in an underestimation of the presence of plastics [51]. Demersal species live in close association with the seafloor; thus, they are more exposed to settled plastics than other species. Only one single specimen of *Sciaena umbra*, a species characterized as vulnerable in the Mediterranean by IUCN Red List, was found with microplastics in its gut in the Levantine Sea [40].

Eleven pelagic fish species were found to be affected by plastic ingestion in the Mediterranean. The highest proportion of individuals with microplastics in their guts belonged to the pelagic species *Sardina pilchardus*, which showed a highly variable incidence (0.09–96%) in different studies [29, 40, 41, 45, 48, 60, 185]. The highest value (96%) was reported in the Adriatic Sea [48]. Renzi et al. [48] found also high microplastic ingestion (91% of analysed specimens) in the pelagic species *Engraulis encrasicolus*. The authors mentioned that this high plastic incidence may be due to “net feeding”, as has been reported in another study in the North Pacific zone [187]. Renzi et al. [48] suggested that the uptake of microplastics by *S. pilchardus* derived from fish’s prey and not directly from the water column filtration. This was not the case for *E. encrasicolus*, which seemed to actively select microplastics through their feeding mechanisms. Female anchovies ingested on average more items (8 items/ind) than males (5 items/ind) especially during the spawning period [48]. The higher litter occurrence in guts of *S. pilchardus* compared to those of *E. encrasicolus* has been attributed to their filtration system; the larger filtration area and the smaller gap between gill rakers may allow the first species to ingest litter more likely ([41] and references therein) than the latter. Romeo et al. [63] reported that about 18% of large pelagic fishes in the Mediterranean (*Xiphias gladius*, *Thunnus thynnus* and *T. alalunga*) had plastic litter in their stomachs. The uptake of plastics by *T. thynnus*, a species characterized as endangered according to the IUCN Red List for threatened species, could be explained by the opportunistic feeding strategy of this species. Conversely, *T. alalunga* is a specialist feeder. Romeo et al. [63] provided several possible explanations for these high rates of litter ingestion. Firstly, tuna often chase prey schools to shallow water where they are more easily caught, but where plastic fragments are more abundant (given their buoyancy); secondly, feeding on aggregated preys may increase the ingestion of unwanted particles, as the predator is not focusing on a single large prey, but on several small ones; thirdly, secondary ingestion cannot be excluded. The same hypothesis has also been suggested for swordfish *X. gladius* [63]. The fact that the majority of preys in the diet of swordfish [188] are fish species known to ingest plastics, e.g. *S. pilchardus*, *E. encrasicolus* and *Boops boops* [29, 40–42, 45, 48, 49, 60, 185], enhances the hypothesis of secondary ingestion. Massutí et al. [46] found 12% of *Coryphaena hippurus* specimens, a top predator species in the Western Mediterranean pelagic ecosystem, with nonfood material, grouped as miscellaneous items, in their stomachs. The authors suggested that this phenomenon might indicate a nonselective predator activity, although unusual for the species. However, some additional reasons could be the secondary ingestion (the diet of the species is based on other fishes, crustaceans, cnidarians and cephalopods that may ingest plastics) or the fact that the species has been reported to be associated with floating debris ([46] and references therein). Moreover, the adult specimens analysed were caught with longlines around Majorca (Spain, Western Mediterranean Sea) [46], and they remained alive in the gear for many hours, which can increase the bias from "net feeding". Other pelagic fish species have also been investigated for plastics in the Mediterranean. *Scomber japonicus* has been studied from two locations: 43% of individuals from South Adriatic and 57% from the Levantine Sea were found to contain macro- and microplastics, respectively [40, 45]. *Trachurus mediterraneus* has been studied in the Aegean-Levantine Sea and the West Mediterranean Sea: 48% and 42.5% of the individuals respectively were found with ingested litter [40, 41]. Rios-Fuster et al. [41] reported that the species showed the highest level of ingested anthropogenic particles compared to the rest of the examined species. Pelagic planktivorous/filter feeder fish plays a key role in pelagic food webs, representing a central link for larger predators [26]. Moreover, their diel vertical migrations may act as a biological pump, transferring plastic litter from surface waters to mesopelagic waters and from low trophic levels of the food web to top predators [174].

The occurrence of macroplastics and microplastics was studied in several benthopelagic species in the Mediterranean. Macroplastic occurrence in *Pagellus erythrinus* from the Ionian and the Central Mediterranean Sea and the Adriatic Sea was found to be 2% and 3.3%, respectively [45]. Macroplastic occurrence in the species *Pagellus bogaraveo* and *Cataetyx laticeps* was 1.7% and 10% for the Ionian and the Central Mediterranean and the Western Mediterranean subregions, respectively [43, 58]. The microplastic occurrence in benthopelagic fishes showed a high variability ranging from 6.7 to 100%. Avio et al. [31] reported that all specimens of the benthopelagic species *Spondyliosoma cantharus* and *Phycis phycis* studied in Giglio Island (Tyrrhenian Sea) had microplastics in their stomachs. According to the authors, these values may reflect the elevated levels of microplastics in the Tyrrhenian basin, as well as the close proximity to the shoreline, which is an important sink compartment for microplastics, particularly in highly touristic or anthropized areas. Presumably, another cause for these values could be the low number of specimens analysed for both species (five and eight individuals, respectively). The commercial species *P. erythrinus* was studied in several areas in the Mediterranean [29, 40, 45, 59]. The proportion of *P. erythrinus* with microplastics in the Tyrrhenian Sea was 6.7% [59], a value higher than those reported by Anastasopoulou et al. [45] for the presence of macroplastics in the North Adriatic and North-East Ionian Sea (3.3% and 2%, respectively) and lower than those reported for the coastal waters of Turkey (22%) [40]. Significantly higher frequency of microplastic ingestion (42.1% for the Ionian and the Central Mediterranean Sea and 50% for the Adriatic Sea subregions) by the same species was mentioned by Digka et al. [29] and Anastasopoulou et al. [45]. Other benthopelagic species investigated in the Mediterranean were *Argyrosomus regius*, *Diplodus annularis*, *Dentex gibbosus*, *Centracanthus cirrus*, *Hoplostethus mediterraneus*, *Lepidopus caudatus*, *Nezumia aequalis*, *Schedophilus ovalis*, *Trachurus picturatus*, *Zeus faber*, *Pagrus pagrus* and *Trisopterus luscus* (Table 1), with the latter presenting the lowest incidence of litter occurrence (0.03%).

The occurrence of plastics in bathypelagic species ranged from 0.3 to 11.8%. The lowest occurrence of plastics was observed in *Diaphus metopoclampus* (0.3%) [47], whereas the highest one (11.8%) was observed in *Notacanthus bonaparte*, which seems consistent with the species diet, which is based on benthos [57].

Reef-associated (*Caranx crysos*, *Pelates quadrilineatus*, *Saurida undosquamis*, *Siganus luridus*, *Siganus rivulatus* and *Upeneus moluccensis*) species were studied in the Aegean-Levantine Sea by Güven et al. [40] and van der Hal et al. [61] whereas the species (*Naucrates ductor*, *Seriola dumerili*) in the Western Mediterranean Sea by Compa et al. [22]. The authors did not find any correlation between the number of ingested litter and the trophic level of species. Individuals with plastics in their guts ranged between 28 and 60% depending on the species, and fish that ingested a higher number of microplastic particles originated from the sites that also had a higher particle count in the seawater and sediment.

The last category is bathydemersal species. Cartes et al. [43] found that 33.3% of the specimens of *Trachyrincus scabrus* analysed, the largest macrourid fish inhabiting the deep waters of the Western Mediterranean Sea, have ingested macroplastics. This species preys upon other fishes, and the uptake of plastics could be a result of secondary ingestion. Plastic threads occurred in 3.7% of *Polyacanthonotus rissoanus* specimens in the Western Mediterranean Sea [57]. Litter ingestion has also been reported for the species *Glossanodon leioglossus* and *Helicolenus dactylopterus* in the Western Mediterranean Sea [22].

2.3 Elasmobranchs

In the Mediterranean, plastic ingestion has been reported in nine elasmobranch species (Table 1).

Anastasopoulou et al. [58] found that 5 out of 26 examined species from the deep waters of the Eastern Ionian Sea ingested litter. They examined 1,502 individuals, and 28 of them contained debris (~2%). The incidence of plastic in the stomachs of elasmobranchs was much greater than that of Teleosts: 4 (*Galeus melastomus*, *Pteroplatytrygon violacea*, *Squalus blainville*, *Etmopterus spinax*) out of 9 elasmobranch species were found with plastics in their stomachs, while only 1 out of

17 Teleost species examined contained plastics. The authors suggested that the types of litter ingested were related to the feeding behaviour of elasmobranch species. For instance, *G. melastomus*, a nektobenthic opportunistic feeder, swallowed all types of litter, the pelagic and bathypelagic/demersal feeders *P. violacea* and *S. blainville* ingested only pieces of plastic bags, whereas *E. spinax*, which has bathybenthic/ bathydemersal feeding habits, mainly ingested hard plastics ([58] and references therein).

Similarly, Cartes et al. [43] studied three elasmobranch species (*G. melastomus*, *E. spinax* and *Centroscymnus coelolepis*) in the deep waters of the Western Mediterranean Sea. The authors mentioned a variety of allochthonous items in the diets of deep-sea elasmobranchs, including threads/fibres, remains of bags or sacks, cartoon remains, coal fragments of unknown origin and organic remains originating from human activities.

Microplastics were identified in 21 of 125 individuals of *G. melastomus* in the Western Mediterranean Sea [55]. The authors suggested that the higher values of microplastic ingestion in Western Mediterranean compared to that reported for the Eastern Ionian Sea [58] might reflect the general patterns of availability and large-scale distribution of marine litter in the Mediterranean. Indeed, litter densities from trawl surveys in continental slopes were higher in the Western Mediterranean (4.0 ± 1.8 kg/ha) compared to the central (0.6 ± 0.4 kg/ha) and eastern (1.1 ± 0.3 kg/ha) region [19]. Also in deep areas, higher values have been reported in western areas (1.8 ± 1.5 kg/ha) than in central (1.7 ± 0.6 kg/ha) and eastern areas (1.2 ± 0.3 kg/ha) [19]. Valente et al. [66] found very high litter frequency of occurrence values in three deepwater elasmobranch species (*G. melastomus*, *E. spinax* and *Scyliorhinus canicula*) studied in the Tyrrhenian Sea, which may coincide with the high litter densities recorded for the continental slopes in the Western Mediterranean and the wide anthropogenic pressure insisting on the area.

The first record of marine plastic litter in the stomachs of blue sharks (*Prionace glauca*) in the Mediterranean Sea was reported by Bernardini et al. [67]. According to these authors, 24 out of 95 specimens analysed were found with plastic litter in their stomachs, and juvenile blue sharks showed a significantly higher occurrence of ingested plastics than adults did. They proposed several explanations for the high uptake of plastics by the blue sharks. Firstly, blue sharks are pelagic species and feed on the surface to >600 m depth following the prey distribution in mesopelagic waters. Secondly, they are opportunistic feeders, playing the role of scavengers. Thirdly, their position at the top of the Mediterranean food web could increase the probability of exposure to secondary plastic ingestion. Fourthly, the colour of plastics could trick the predators ([67] and references therein). It is worth mentioning that blue shark Mediterranean subpopulation is characterized as critically endangered according to the IUCN Red List for threatened species, and the population is decreasing mainly due to fishing. Plastic ingestion may represent a further threat for the species in the Mediterranean.

Avio et al. [185] documented the presence of plastics in spiny dogfish *Squalus acanthias* in the Adriatic Sea. The Mediterranean subpopulation of *S. acanthias* is considered endangered by IUCN, and plastic can result in a further threat to the species in the area.

2.4 Marine Mammals

Litter ingestion by marine mammals in the Mediterranean Sea is less documented in the scientific literature than for other organisms (e.g. fish, marine birds and sea turtles) [68], although several cases have been reported worldwide [70]. So far, the existing information regarding interactions between large marine mammals and marine litter is related more to entanglement rather than ingestion [70]. Most of the available record of litter ingestion by mammals collected from small sample sizes provided by stranded animals, presenting only a snapshot of the impacts occurring unseen at sea [189]. Litter ingestion in singly stranded animals, although offers important information to researchers [71], may not be representative of mammals' populations and therefore may not provide accurate results [97].

One individual of a sperm whale (*Physeter macrocephalus*), the largest of the toothed whales, was found dead in a Spanish coastal area (Western Mediterranean Sea subregion) in 2012, without evidence of entanglement scars or other injuries. De Stephanis et al. [70] reported ingestion of various plastic items, such as plastic cover material, burlap plastic bags, flower pots, hosepipes and ropes, which originated from the local greenhouse agriculture. Another individual of *P. macrocephalus* that had ingested plastic was reported in the southwest of Crete Island (Aegean-Levantine Sea subregion). A small square piece of rigid plastic mesh 10 × 10 cm was found inside the stomach of the whale, which was most likely to be litter disposed of at sea [2]. In the south Adriatic Sea, seven sperm whales were found stranded in 2009. Pieces of fishing gears and hooks, ropes and plastic objects were found in 74% of the individuals examined [72]. Marine litter was found in the stomach of 10 out of 13 sperm whales (77%) stranded along the Italian coast (Western Mediterranean Sea subregion) between the period from 2009 to 2013, and it was composed mostly of plastic [71]. Similarly, marine litter found in the stomachs of six out of ten sperm whales stranded along the Ionian and Aegean Seas from 1993 to 2014; the majority of litter was plastics except only one metal wire tied at the top of a plastic bag [68]. According to the authors, the high percentage of plastic sheets in the stomachs of sperm whales is probably linked with the high abundance of plastic bags and packaging in both the water column and the seafloor. The sperm whale Mediterranean subpopulation is classified as endangered according to the IUCN Red List, and plastic ingestion could result in additional mortality for the species.

Two Cuvier's beaked whales (*Ziphius cavirostris*), which were found stranded along the Croatian coast (the Adriatic Sea subregion), contained four plastic bags (two of them were shopping bags from soft plastic, and the other two were made of more solid plastics). According to the authors, the cause of death was probably the direct result of ingesting plastic bags [79]. Plastics were found in the stomach of one Cuvier's beaked whale stranded along the Greek Ionian Sea coasts (the Ionian Sea and the Central Mediterranean Sea subregion), and its death may be caused by gastric blockage [68].

Dolphins have been reported to ingest marine litter quite often as well. Shoham-Frider [69] documented ingestion of pieces of plastic bags by a stranded Risso's dolphin (*Grampus griseus*) along the Mediterranean coast of Israel (Aegean-Levantine subregion). The pieces of plastic bags found in its stomach contributed to the dolphin's poor physical condition. Alexiadou et al. [68] mentioned plastic ingestion by a Risso's dolphin stranded along the Greek Ionian Sea coasts (the Ionian Sea and the Central Mediterranean Sea subregion), the death of which may have been caused by gastric blockage. Levy et al. [76] reported ingestion of nylon filaments and nets by a common bottlenose dolphin (*Tursiops truncatus*) in 2002 in the Port of Haifa, Israel. Gomerčić et al. [77] examined 120 stranded bottlenose dolphins along the Croatian coasts of the Adriatic Sea in the period from 1990 to 2008. Almost 10% of these dolphins had from 1 to 13 pieces of gill-net parts in their forestomachs depending on the specimen. Two of these animals had heavy forestomach and oesophagal ulcerations. Probably, these dolphins had torn off a part of the gill nets while feeding on fish entangled in the fishing nets [77]. The authors mentioned that this hypothesis was supported by the fact that dolphins with gill-net parts in their stomachs were found without larynx strangulation. Interaction of marine mammals with fishing nets as part of their feeding strategy is well-known [77]. An individual of striped dolphin (*Stenella coeruleoalba*) was found dead near the SW coast of the island Krk, in the North Adriatic Sea, in 1998. The cause of death was that the entire volume of the stomach was occluded by different kinds of plastic material (garbage bags, rubber gloves, cellophane wrappings). The blubber layer was extraordinarily thin, indicating starvation [75].

Finally, Alexiadou et al. [68] reported the first evidence of plastic ingestion for harbour porpoise *Phocoena phocoena* in the Mediterranean. Ingestion of plastic by harbour porpoises is well-known in the North Sea [190].

2.5 Sea Turtles

The Mediterranean loggerhead sea turtle (*Caretta caretta*), green sea turtle (*Chelonia mydas*), African softshell turtle (*Trionyx triunguis*) and leatherback turtle (*Dermochelys coriacea*) have been found to be affected by litter ingestion (Table 1). The threat from plastic ingestion is well-documented for *C. caretta* (e.g. [80–92, 96]). The species was indeed selected in the framework of the MSFD as the target species for monitoring the amount and composition of litter ingested by marine animals in the Mediterranean. The loggerhead sea turtle has also been recommended by the expert group of the Barcelona Convention LBS protocol as the main target species regarding "Common Indicator 18E: Trends in the amount of litter ingested by or entangling marine organisms" [89]. Conversely, only three papers [84, 90, 96] reported the presence of marine litter in *C. mydas* and just one [90] in *T. triunguis* and *D. coriacea* [97].

Gramentz [82] examined sea turtles *C. caretta* incidentally caught by Maltese fishermen while fishing for *Xiphias gladius* and *Coryphaena hippurus*. Most plastics found in the guts of these specimens were transparent, milky (translucent) or white pieces of polystyrol, styrofoam and PVC, which were considered by the author as strongly indicative that these materials were ingested, being mistaken for jellyfish. Moreover, Gramentz [81] suggested that the pieces of aluminium foil found in the gut of another individual were indicative that the animal was attracted by the silvery reflections of metals, probably mistaking them for fish. Different types of litter appeared in the gastrointestinal tract of 43 loggerhead sea turtles (79.6%) caught illegally by fishermen in Spanish Mediterranean waters, with plastics being the most frequent type (75.9%). Similar results of high plastic ingestion by loggerhead were reported by [80, 85, 87, 89, 91, 93], for other areas of the Western Mediterranean Sea; by Casale et al. [92] for the Ionian Sea and the Central Mediterranean Sea; by Lazar and Gračan [80] for the Adriatic Sea; and by Sönmez [90] for the Aegean-Levantine Sea. The high occurrence of ingested plastics might be explained by the ubiquity of soft floating debris in the marine ecosystem and by the high attraction of loggerheads for this litter type [80, 81, 92]. Tomás et al. [83] mentioned that no lethal effect and neither clear evidence of digestive tract blockage were observed during the necropsies of *C. caretta* captured in the Western Mediterranean Sea. Conversely, this was not the case for seven loggerhead turtles caught in Sicily, in which intestinal occlusions, caused by the ingestion of foreign bodies of various nature, such as pumice stones, pieces of wood and plastics, fragments of electrical wires, candy wrappings, newspaper bits, tar and cellophane, were observed [84]. Campani et al. [81] reported that the presence of plastics principally in the last sections of intestines indicates that probably most of the plastics pass through the gastrointestinal tract of the *C. caretta* are excreted. Matiddi et al. [89] found the marine litter mainly in the intestine of the *C. caretta* specimens examined in the Western Mediterranean, followed by the stomach, while the oesophagus was the least affected part of the gastrointestinal tract. Moreover, Camedda et al. [87] reported that in studies of dead specimens, 70% of the litter was found in the intestines and only 30% in the stomachs.

Similar to the results reported for loggerhead sea turtles, plastics were also the most commonly ingested type of litter in *C. mydas*, *T. triunguis* and *D. coriacea* [84, 90, 96, 97]. These species have been listed as endangered (global population) and critically endangered (Mediterranean subpopulation), respectively, by the IUCN Red List mainly due to fisheries by-catch, and plastic ingestion may represent a significant further threat to these species [96].

2.6 Seabirds

On a global scale, reports of seabird plastic ingestion have been increasing since the 1960s, and they have stabilized over time [23, 153]. However, in the Mediterranean Sea, there is a lack of information on marine litter ingestion by seabirds [99]. Only two papers [98, 99] provided some information on litter ingestion by different seabird species.

Codina-García et al. [98] examined nine seabird species accidentally caught by longliners in the Western Mediterranean Sea from 2003 to 2010. Among them, Scopoli's shearwater (*Calonectris diomedea*), yelkouan shearwater (*Puffinus yelkouan*) and Balearic shearwater (*P. mauretanicus*) presented the highest occurrence of litter ingestion (96%, 71% and 70%, respectively). According to these authors, this result is of conservation concern, since *P. yelkouan* is considered vulnerable by IUCN, while *P. mauretanicus* is considered critically endangered. *C. diomedea*, although characterized as least concern at a global scale by IUCN, in the Spanish national catalogue is listed as vulnerable. The other species (*Ichthyaetus audouinii*, *I. melanocephalus*, *Larus michahellis*, *Rissa tridactyla*, *Stercorarius skua* and *Morus bassanus*) were less affected (13–50%), although the number of specimens examined was low. The highest mean number of items/ind (49.3 ± 77.7) was observed in Audouin's gull (*L. audouinii*). Plastic items found in this study could be classified mostly as microlitter, that is, most of them were smaller than 5 mm [98].

Steen et al. [99] observed, through a camera trap placed in a nest situated in the eastern region of the Aegean Sea, a parental female Eleonora's falcon (*Falco eleonorae*) arriving at the nest with plastic waste (a snack wrapper) and feeding the nestlings. The female bird probably mistook the plastic waste for a small bird or large insect during hunting [99].

3 Entanglement

Entanglement of seabirds, marine mammals, sea turtles and fish in marine debris is the most known and visible effect of plastic pollution on marine organisms [23]. However, many species vulnerable to entanglement are scattered across wide ocean areas and individuals that become entangled and die may quickly sink or be consumed by predators at sea without being detected [20]. Thus, the estimated mortality rates and the effects of entanglement on the population dynamics of many species are probably underestimated [97]. Many marine species interact with marine debris as a result of their normal behaviour patterns: drifting debris attracts fish and invertebrates, and thus marine mammals, seabirds and turtles could be attracted to debris by its associated prey species [20].

Entanglement is the second main impact (following ingestion) to be considered when dealing with criteria 10.2. "Impacts of litter on marine life" of the MSFD [191]. However, no indicator related to entanglement has been defined to date for long-term monitoring programmes due to the difficulties in detecting it (see [192] for a thorough discussion on this issue).

Entanglement of marine life occurs in all ecosystems around the world and affects a wide range of species, including whales, sea turtles, fur seals, seabirds, octopuses, corals, crabs, fish, etc. (for a global review, see [23]). Entanglement can cause wounds and entrapment, hindering animals ability to move, reproduce, feed and escape from predators, and potentially lead to death from starvation, suffocation, strangulation or drowning [20, 23]. Moreover, entanglement could cause lacerations and infections from the abrasive or cutting action of attached debris, and entangled animals may exhibit altered behaviour patterns potentially hampering their survival [97]. Whales and dolphins are usually entangled around their neck, flippers and flukes by fishing gear (e.g. [193]). Seals can become entangled in fishing gear, packing straps or other loop-shaped items that encircle the neck at a young age and create problems during growth [194]. As summarized by Kühn et al. [23], seabirds may become entangled around the bill, wings and feet with rope-like materials, which constrains their ability to fly or forage; marine turtles are prone to entanglement by floating debris, while hatchlings may be entangled in beach debris on their way to the sea; crabs, octopuses, fishes and a wide range of smaller marine biota can be caught in derelict traps and nets on the seafloor where they could die from starvation and serve as bait, attracting new victims. Derelict fishing lines and nets may cause direct physical damage to benthic sessile organisms such as sponges and corals, breaking them or causing a progressive removal of their tissues, making them more vulnerable to parasites or bacterial infections [134].

Most records of entanglement around the world involve fishing gears, six-pack plastic rings and packing strapping bands [97]. As regards fishing gear, generally it is not straightforward determining if the animal became entangled in an active gear (by-catch) or in an abandoned, lost or otherwise discarded fishing gear (ghost fishing) [20, 26]. Indeed, "ghost fishing" refers to derelict fishing gear that continues to catch marine animals, inducing mortality without human control [195]. Moreover, larger vertebrates may continue to travel after becoming entangled in nets, hence transforming active fishing gear into marine debris [196]. For these reasons, the MSFD monitoring criteria for biota considered entanglement as a secondary criterion, and each member state has to decide for its implementation [197]. In this review, we considered as entanglement by marine debris only those cases where the authors explicitly refer to it. Reports in which the gear was identified as likely having been operational at the time of entanglement were excluded (e.g. [78]).

In the Mediterranean Sea, the first documented record of a marine animal entangled in anthropogenic debris dates back to 1979, when a small turtle with a large piece of a plastic sheet wrapped around its shell was observed attempting to swim in the Eastern Mediterranean [146]. The author did not specify the species, but it could probably be the loggerhead sea turtle (*C. caretta*) since it is the most common sea turtle in the Mediterranean [147].

According to the present review, *C. caretta* is the species with the highest number of entanglement records in the Mediterranean Sea. The species resulted impacted both by land-based sources, such as plastic bags and sheets, and by sea-based sources, like fishing aggregating devices (FADs) and fishing lines. During their juvenile pelagic phase, sea turtles are dependent on driftlines for their food supply and shelter [97]. The currents that form driftlines and transport hatchlings to oceanic convergence zones also concentrate floating anthropogenic debris, resulting in a trap for these young turtles, whether it be through ingestion or entanglement [148]. We identified four papers reporting loggerhead entanglement in marine debris in Italian waters (mainly in the Tyrrhenian Sea) and one in Turkey (Aegean-Levantine Sea) (Table 2). Casale et al. [147] reported entangled loggerhead in the South Tyrrhenian

Sea, Ionian Sea, Strait of Sicily and North Adriatic Sea between 2000 and 2008. Even though the authors reported that the entanglement of analysed specimens was due to anthropogenic material that could not be ascribed with certainty to operating or abandoned fishing gear, they concluded that entanglement in ghost gear or in other anthropogenic debris affects high numbers of turtles in the Mediterranean. In 1994, a juvenile loggerhead turtle was found close to the Island of Panarea (South Tyrrhenian Sea, Sicily; Western Mediterranean Sea subregion) trapped in a bundle of polyethene packaging twine; a piece of cord had been swallowed and extended out of the animal's mouth for a length of 20 cm, and it was removed through an endoscopy, as reported in the Marine Turtle Newsletter 71:5, http://www.seaturtle.org/mtn/archives/mtn71/mtn71p5.shtml. Blasi et al. [94] reported loggerhead individuals entangled in anchored illegal FADs and in floating debris (nylon and debris from FADs or land-based sources) near the Aeolian Archipelago (South Tyrrhenian Sea, Sicily; Western Mediterranean Sea subregion) in a study conducted between 2011 and 2014. Turtles became entangled or injured in the anchoring lines and debris of FADs at the neck, flippers and posterior limbs. These entanglements produced injuries and hampered the ability to swim and dive and were responsible for a general state of undernutrition probably due to the inability of turtles to successfully capture preys.

Marine pollution, intended, for example, as entanglement in rope and net, cloth sack or nylon bag, as well as the presence of these substances in the digestive system, was identified as the main cause of death of stranded green turtles (*C. mydas*) found on Samandağ beach (Turkey, Aegean-Levantine Sea subregion) between 2009 and 2017 [90]. Rope entanglement represented the main problem for green turtles at the oceanic stage. *C. mydas* is an endangered species because of the extensive subpopulation declines in all the major ocean basins over the last three generations. The main causes for this decline are the overexploitation of eggs and adult females at nesting beaches, juveniles and adults in foraging areas and, to a lesser extent, incidental mortality relating to marine fisheries and degradation of marine and nesting habitats. For this reason, green turtles are subjected to legislative protection under a number of treaties and laws (e.g. Annex II of the SPAW Protocol to the Cartagena Convention; Appendix I of the Convention on International Trade in Endangered Species of Wild Fauna and Flora; Appendices I and II of the Convention on Migratory Species (CMS)). On the same beach, marine pollution was identified as the third cause of death for loggerhead, following fishing activities and intentional killing [90]. Özdilek et al. [149] found that solid waste accumulations on beaches along the Samandağ coast negatively affect green turtle hatchlings trying to reach the sea. The authors observed that litter represented an obstacle for the rushing hatchlings, and when they hang on the litter, they were easily hunted by ghost crabs living there.

Through in situ experiments using some of the most common items found on beaches (plastic bottles, styrofoam cups, plastic canisters and fishing nets), Triessnig et al. [198] showed that most sea turtle hatchlings were permanently entrapped in cups and canisters or entangled in nets. The study was conducted in the Gulf of Fethiye (southwest Turkey) in the Aegean-Levantine Sea subregion. Turkey is one of the main contributors to marine plastic debris in the Mediterranean [7], and at the same time, it hosts the main nesting concentrations of loggerhead [199], resulting in a high risk for the species since nesting beaches are extremely important habitats for marine turtles [200].

In the first global review ever published on the entanglement of marine life, Laist [20] reported only one species from the Mediterranean, i.e. the Mediterranean monk seal (*Monachus monachus*). The author reported that the animal was entangled in derelict fishing gear and a rubber hoop. Further evidence of entanglement of Mediterranean monk seals was reported in the Western Mediterranean Sea, the Ionian Sea and the Central Mediterranean Sea and the Aegean-Levantine Sea [201], but it is not clear if these were cases of by-catch or ghost fishing, and thus they were not considered in this review. Caution is needed when attributing entanglement to marine debris. Kühn et al. [23], for instance, attributed to plastic litter the entanglement of an individual of monk seal reported in [201]. However, in the article, it is clearly described that it was a case of by-catch.[1]

Other marine mammals threatened by marine litter (and in particular fishing gear), in terms of entanglement, are cetaceans [189]. However, we did not find in the literature any reference for the Mediterranean Sea clearly distinguishing cases of cetaceans' entanglement due to by-catch from cases due to ghost fishing. Even if some marine mammals, especially juveniles, may attempt to rest on the debris, may want to investigate it by curiosity or even play with it, risking their entanglement, most cases of entanglement records of cetaceans are related to active fishing gear [97]. Entanglement in fishing gear was observed for the endangered sperm whale (*P. macrocephalus*) in the Southern Tyrrhenian Sea [203] and in the Greek Seas [204]; for the vulnerable common bottlenose dolphin (*T. truncatus*) in the Adriatic Sea [77] and Greek Seas [204]; and for Risso's dolphin (*G. griseus*) in the Greek Seas [204].

As regards seabirds, the only entanglement records found for the Mediterranean were anecdotal evidence on accidental entrapments by fishing lines on a beach along the Tyrrhenian coast of central Italy: one adult of *Charadrius hiaticula* (Fig. 5), an adult male of *Phoenicurus ochruros*, an adult female *Charadrius alexandrinus* and an adult of *Larus michahellis* [150]. It is worth noting that *C. alexandrinus* is an endangered species in Italy [205] that breeds locally and is included in Annex 1 of 147/2009 "Birds" EU Directive as threatened species.

Three elasmobranch species subjected to entanglement in marine debris in the Mediterranean Sea were identified in the literature. However, the number of sharks and rays that become entangled and die undetected could be much greater than those reported since they will invariably die at sea and very likely be rapidly consumed

---

[1] "A hungry seal raids a fishing net but suddenly finds itself ensnared. In its desperation, it bites and tears and struggles until the net is reduced to shreds. The harder it struggles to escape, the tighter the surviving rope and mesh entwines it. In the end it lies at the water's surface, gasping, exhausted, the remnants of the net wound tight around its throat. The following morning, the unfortunate owner of the tattered nets set out from the little fishing village of Komi on Chios to bring in his night's catch, only to discover the seal still struggling to liberate itself" [202].

Fig. 5 An adult individual of *Charadrius hiaticula* entrapped by fishing line with the hook [150]

[145]. An individual of giant devil ray (*Mobula mobular*) was found dead entrapped in a ghost net in 2011 in the Port-Cros National Park (Western Mediterranean Sea, France) [143]. According to IUCN, it is an endangered species, and its geographic range is limited to the Mediterranean Sea and possibly adjoining North Atlantic waters. *M. mobular* is also included in Annex II "List of endangered or threatened species" to the Protocol concerning Special Protected Areas and Biological Diversity in the Mediterranean of the Barcelona Convention and in Annex II "Strictly protected fauna species" to the Bern Convention. In 2016, a juvenile female of blue shark *Prionace glauca*, with a yellow plastic polyolefin strapping band collar surrounding its gill area, was captured by a commercial longline boat in the North-Western Mediterranean Sea (Fig. 6). The ring encircled the gill region causing damage to the tissue of this area and in the front part of the right pectoral fin, and the fifth-gill slit was obstructed, which could cause breathing problems [144]. The investigative behaviour of sharks in relation to inanimate objects is probably the main cause of such encircling by debris [97]. Entanglement is more frequent in juveniles [20] since they are attracted to floating debris by curiosity and thus plastic loops can easily slip onto their body. A plastic collar around a shark's gill slits or body can cause traumatic cutting into tissue and chronic infection; it may compromise its ability to feed and grow and cause its death by strangulation [145]. Finally, in a report by Butterworth et al. [145] a picture of a deceased catshark (*Scyliorhinus* spp.) entangled in a fishing net in Croatia (Adriatic Sea) is shown, but it is not possible to identify the species.

Fig. 6 Lateral view of the head of the *Prionace glauca* specimen showing damage on the gill region and the pectoral fin by a plastic debris collar [144]

Only two documented cases of teleost species entanglement in marine debris were found in the literature for the Mediterranean Sea. Houard et al. [143] reported ghost fishing for red scorpionfish (*Scorpaena scrofa*), small red scorpionfish (*Scorpaena notata*), black scorpionfish (*Scorpaena porcus*) and European conger (*Conger conger*) in the Port-Cros National Park (Western Mediterranean Sea, France). Furthermore, Ayaz et al. [114] reported ghost fishing for red scorpionfish (*S. scrofa*), as well as for white grouper (*Epinephelus aeneus*) in the Gökova Special Environmental Protection Area (Aegean Sea, Turkey); all specimens were found dead. However, entanglement for teleosts is probably underreported because of the difficulties in its observation at sea, due, for instance, to considerable scavenging pressure on entrapped fish [206], and because information about the number of fishing gears lost or for how long such gears continue to fish is limited [207]. Potentially, a wide range of teleost species may be affected by ghost fishing. Ayaz et al. [208], for instance, performed an experimental study of ghost fishing in southwest Izmir Bay, Turkey (Aegean-Levantine Sea subregion): 29 species (22 fish, 5 crustacea, 1 cephalopod and 1 gastropod) were captured by the ghost gillnets.

Entanglement can also occur in invertebrate benthic/sessile species, such as cnidarians and sponges, which can suffer broken parts, necrosis, progressive removal of the tissues and wounds susceptible to infections [116]. Marine debris may act as a significant stressor for coral reefs, causing suffocation, shading, tissue abrasion and mortality of corals [209]. Rock habitats support dense aggregations of tridimensional complex sessile fauna called “marine animal forests”, which have been proposed as indicators to monitor the temporal and spatial trends of entanglement by marine litter. Indeed, they are vulnerable to damage due to their slow growth rate, they are widely distributed from shallow waters to the deep sea, and they are immobile enabling the precise location of the entanglement event and reducing the risk of misinterpretation due to possible interaction with active fishing gears [209].

In the Mediterranean Sea, 20 cnidarian species were found to be affected by entanglement in marine litter (Table 2), representing the taxon with the highest number of species showing this kind of impact. Most cases of entanglement were due to fishing lines and nets. The largest number of cnidarian species subjected to entanglement was found in the Ionian Sea and Central Mediterranean Sea and

Western Mediterranean Sea subregions (19 species). Only one species (*Paramuricea clavata*) was found in the Adriatic Sea subregion and one species (*Dendrophyllia ramea*) in the Aegean-Levantine Sea subregion. Endangered species in the Mediterranean *Dendrophyllia cornigera*, *Leiopathes glaberrima*, *Corallium rubrum* and *Madrepora oculata* were included in the list of species entangled by marine litter. The latter is considered critically endangered in Italian Seas, where it was found in the Ionian Sea stuck by plastic litter [127]. Moreover, nine near threatened species and two vulnerable species were found to be impacted by entanglement in the Mediterranean. The likelihood of diseases increased 20-fold once a coral is entangled in plastic [210]. For instance, Bavestrello et al. [130] found along the Portofino Promontory (Ligurian Sea, Italy) that severe damage to gorgonians (*P. clavata*) was caused by lost fishing gear (mostly monofilament lines) affecting tens of colonies through a continuing abrasive action. The stretched lines, under the action of sea currents, mechanically excoriated the coenenchyme. The authors concluded that the major cause of mortality in *P. clavata* facies along the Portofino Promontory was due to damage by fishing lines, followed by the attachment of several epibionts in the damaged surface [130]. Indeed, if injuries are of minor intensity, gorgonians are able to rapidly healing the wound by coenenchyme regeneration. Conversely, pioneering species such as hydroids may settle on the damaged surface and are soon replaced by stronger competitors like bryozoans, macroalgae, serpulids or sponges, which can no longer be removed by the newly growing coenenchyme.

Four species belonging to the taxon Porifera were found entangled in marine litter in the Mediterranean Sea, i.e. *Geodia cydonium* in the North Adriatic Sea [133], *Pachastrella monilifera* and *Poecillastra compressa* in the Ligurian Sea (Western Mediterranean Sea) [134] and *Raspailia* (*Raspailia*) *viminalis* in the Strait of Sicily. Most entanglement records involved fishing nets and lines.

Finally, two crustaceans (spinous spider crab *Maja squinado* and three-spined Geryon *Geryon trispinosus*; Fig. 7), one mollusc (common cuttlefish *Sepia officinalis*) and one echinoderm (pencil urchin *Cidaris cidaris*) were found entangled in marine litter (mainly fishing nets) in the Mediterranean Sea (Table 2). Ramirez-

Fig. 7 Ghost fishing of *Geryon trispinosus* by a discarded/lost net recovered from 1,200 m depth in the Western Mediterranean Sea [135]

Llodra et al. [135] reported evidence of ghost fishing in one sample from 1,200 m in the Western Mediterranean, where several Geryon crabs were observed dead or moribund in a broken fishing net (Fig. 7). Indeed, benthic organisms entangled in derelict fishing gear or other litter items on the seafloor can eventually die because of starvation [209].

4 Other Impacts

Marine litter may have an impact on marine life indirectly by offering available substrates for rafting species and facilitating species dispersal [27]. A wide range of sessile and motile marine organisms colonize floating litter (also called hitch-hikers, [211]), including bivalves, barnacles, algae, foraminifera and a rich microbial community that forms biofilms known as the "Plastisphere" [212]. The composition of the microbial community has been found to significantly differ from the surrounding seawater suggesting that plastic litter forms a novel habitat for microbiota [212]. Many species may extend their distribution range through transport by floating rafts: several taxa, including potential invaders, were already found on marine litter far beyond their natural dispersal range [27]. Passive transport can last for years, with marine current-driven journeys covering vast distances across the oceans [213]. Floating natural debris (e.g. plants, trunks, pumice) has always acted as a dispersal vector for marine organisms, but since the quantities of synthetic and non-biodegradable materials in marine debris have increased manifold over the last decades in the Mediterranean Sea [214], the dispersal has probably been accelerated [211]. Abundant floating marine litter may facilitate the spread of invasive and pathogenic species [27].

We found 29 papers describing the effects of marine litter on the biota in the Mediterranean Sea other than ingestion and entanglement, impacting bacteria, algae, marine plants, invertebrates, tunicates and seabirds (Table 2). Most of these species use marine litter as a substratum, but marine litter can also be used instead of sponges by the crab *Paromola cuvieri* to cover its carapace [122, 134] (Fig. 8). Overall, the most common phyla rafting on or encrusting marine litter were Bacteria, Algae, Arthropods and Cnidarians. Most taxa were associated with plastic litter.

The most comprehensive study on hitch-hikers in the Mediterranean Sea was published in 2003 by Aliani and Molcard [100], who found 21 species rafting on plastic floating litter (mainly plastic bags, bottles and styrofoam) in the Ligurian and Tyrrhenian Sea (Western Mediterranean Sea) (Table 2). The most common species found were the barnacle *Lepas* (*Anatifa*) *pectinata* and the isopod *Idotea metallica*, but many other species were identified, including coralline algae (e.g. *Hydrolithon farinosum*), seagrass species (e.g. *Cymodocea nodosa*), hydrozoans (e.g. *Obelia dichotoma*), polychaetes (e.g. *Nereis splendida*), nudibranchs (e.g. *Doto* spp.), etc. No alien species was found. *I. metallica* is an obligate rafter without benthic populations and was found hitch-hiking on marine litter also along the Catalan coast [138] and in the Alboran Sea [136]. This species is adapted to the rafting life-style and has low food requirements compared to its congener *I. baltica* that

Fig. 8 The crustacean *Paramola cuvieri* carries plastic on its exoskeleton, instead of usual sponges (470 m depth) [134]

predominantly colonizes algal rafts, which are rapidly consumed by this voracious herbivore [27]. Gutow and Franke [139] observed that specimens of *I. metallica* found rafting on floating litter in the Mediterranean represented not just ephemeral assemblages but persistent local populations.

Two papers analysed the bacterial films in the Mediterranean Sea [106, 107]. The authors identified 50 bacterial species on the microplastics sampled in the Northern Adriatic Sea and Western Mediterranean Sea (Table 2). Some of them were hydrocarbon-degrading bacterial species, able to degrade various types of plastics through the secretion of specific extracellular enzymes. The bacteria *Aeromonas salmonicida* was also identified for the first time on microplastics, which is responsible for bacterial diseases in fishes. The risk associated with microplastic pollution for the spreading of diseases should be evaluated taking into account the occurrence of pathogenic bacteria in other environmental matrices, like water and other floating objects. Dussud et al. [107] found that some putative pathogens were particularly abundant on plastic marine debris and rather rare in free-living and organic particle-attached bacteria, such as the fish pathogen *Tenacibaculum* spp. or the crustacean and invertebrate pathogens *Phormidium* spp. and *Leptolyngbya* spp. This result suggests that microplastics may serve as a vector of pathogenic bacterial species in the marine environment, although this issue has been little studied [103].

Another notable example of hitch-hiker in the Mediterranean Sea includes the Arch-fronted swimming crab (*Liocarcinus navigator*), reported for the first time ever rafting on floating marine litter in the Adriatic Sea [142]. The authors also observed two specimens of Columbus crab (*Planes minutus*) jumping from one to another piece of floating plastic marine debris (a plastic sandal and a sports shoe).

Marine litter can also serve as a substratum for cnidarians, including the alien species *Oculina patagonica* found in the Alicante harbour (Spain) inhabiting plastic bags and cans, and the potentially harmful dinoflagellates *Coolia* spp. and *Ostreopsis* spp. found on plastic that littered the waterfront of La Fosca beach (Costa Brava, Catalan coast). Plastic is used as a substratum also by sessile annelids like *Filograna implexa* [109], *Placostegus tridentatus* [104] and *Spirobranchus triqueter* [111].

Fig. 9 The northern gannets plastic nest of Porto Venere (La Spezia Gulf), built over the deck of a boat [151]

Battaglia et al. [104] reported evidence of colonization of rafting floats in expanded PVC from abandoned, lost or derelict fishing gears by 3,014 deepwater organisms belonging to 38 taxa of macro-invertebrates (Arthropoda, Cnidaria, Mollusca, Porifera, Bryozoa and Foraminifera) in the Strait of Messina (Ionian Sea). Four of these species are protected: the three deepwater corals *Errina aspera*, *Desmophyllum pertusum* and *Madrepora oculata* as well as the deep-sea cirripede crustacean *Pachylasma giganteum*. The authors speculated that the hydrodynamism of the Strait of Messina produces a continuous and strong frictional stress on the abandoned, lost or derelict fishing gears laying on the bottom. This causes the breakage of ropes and nets releasing the fishing floats, which may emerge and strand on the shore or follow the course of currents and disperse far from the point of release.

Finally, marine litter can be used by seabirds to build their nests. Indeed, plastic lightness and flexibility make it attracting to birds for the construction of nests, but it can easily twist around the body of new-borns or even of adults, damaging them and possibly causing their death [215]. Merlino et al. [151] found that northern gannets (*Morus bassanus*) in the La Spezia Gulf (Ligurian Sea, Italy; Western Mediterranean Sea subregion) build their nests using more anthropogenic objects than natural ones, in particular fragments of nets made with polypropylene and polyethene that are used for mussel farming (Fig. 9). The study concluded that the prevalent productive activities of the area (mussel farming) are responsible for the problem of marine and coastal pollution and pose a danger for the local fauna.

5 Conclusions

Our review demonstrates that the impact of plastic pollution on Mediterranean marine life, including a number of commercially important species, is a widespread and pervasive phenomenon. This is not surprising, since the Mediterranean Sea is both a crucial biodiversity hotspot and a critically polluted area, and it has been described as one of the areas most affected by marine litter in the world [26]. Evidence of plastic impact on marine life is accumulating fast because ocean plastic is a contemporary focal point of concern for the marine environment. Consequently, even if this is the most up-to-date collection of information on the impact of plastic pollution on marine life in the Mediterranean Sea, the number of species known to be impacted by plastic is deemed to increase along with growing scientific literature. A variety of direct and indirect effects of plastic pollution on marine biota has been described, including ingestion; entanglement; substrate for the dispersal of some organisms, e.g. bacteria; vectors for exposure to potential pathogens; and much more.

Plastic ingestion is the most studied impact in the region. There is scientific evidence that a large number of Mediterranean species, across multiple habitats and trophic levels, ingest plastics, mainly microplastics. It is documented that macroplastics are also regularly uptaken and retained by a variety of marine animals including fish, birds, turtles and cetaceans. The consequences of microplastic ingestion for marine organisms are still largely unknown, but generally considered as a lower risk than ingestion of larger plastic items. For instance, direct mortality in wild fish caused by ingested microplastics has not yet been described in the published literature [24]. Conversely, ingestion of and entanglement in the plastic waste can cause the suffering and death of seabirds, turtles and cetaceans. For instance, gastric blockage from plastic can be lethal for marine mammals [68]. Nevertheless, the population effects of these deaths are still largely unknown [216].

From this review, it emerges that the literature regarding seabirds for the Mediterranean is scarce. Seabirds are among the animals the most impacted by marine litter worldwide, with 165 species of the 367 species (45%, but limiting the analysis to species actually checked for plastic, the proportion is 78%) that have been recorded to ingest plastics [168]. Since seabird ingestion rates scale with plastic exposure [217], the high densities of plastic litter in the Mediterranean allow expecting ingestion rates by seabirds to increase proportionately. Thus, much research effort is needed in order to evaluate the impact of marine litter on Mediterranean seabirds.

Commercially important fish species contaminated with microplastics represent a potential source of these particles and the chemicals they contain for human consumers [218]. Many commercially important species for Mediterranean fisheries, such as European anchovy, sardine and common sole, have been found with microplastics in their guts. However, recently different scientists have denounced that the risk deriving from microplastics is probably overstated since their quantity in the marine environment is generally so low that they do not represent an environmental risk [219, 220]. It is worth noting that Katsanevakis [97] affirmed in 2008 that "The impact of microscopic plastic particles on marine fauna and the marine food web is largely unknown", and 10 years later Markic et al. [24] still recognize that "Plastic ingestion is of special concern, as its magnitude and consequences for marine organisms and potentially humans are still largely unknown". Also, a recent report by the Food and Agriculture Organization (FAO) stated that "Despite the increasing literature on marine plastic contamination, there is very little information on its effects at ecosystem, habitat, population or even individual level" [154]. Thus, further research is needed to investigate the effects of plastic particles on marine biota in the wild as well as those of their additives and adhering contaminants.

The presence of microplastics in lower trophic levels raises the possibility that microplastics and/or their contaminants may be transferred through the food web. However, the number of studies reporting trophic transfer remains limited. To date there is only initial evidence that suggests the potential of trophic transfer of microplastic in wild-caught organisms [221]. For this reason, further studies are needed in order to better understand the effects of plastics on the Mediterranean food webs and ecosystem.

Concluding, according to the reviewed literature, it seems that ingestion and entanglement can have dramatic consequences on marine life at the individual level. Conversely, it is unlikely to occur frequently enough to have adverse demographic impacts in the Mediterranean, with the possible exception of some marine turtles [168]. However, it must be noted that the available literature generally pertains to individuals rather than on a population level. Studies at the population level are indeed hindered by multiple environmental and human-induced stressors to which wild animals are subjected, which may mask the possible role played by microplastics [154].

References

1. Van Cauwenberghe L, Claessens M, Vandegehuchte MB, Janssen CR (2015) Microplastics are taken up by mussels (*Mytilus edulis*) and lugworms (*Arenicola marina*) living in natural habitats. Environ Pollut 199:10–17. https://doi.org/10.1016/j.envpol.2015.01.008.
2. Roberts SM (2003) Examination of the stomach contents from a Mediterranean sperm whale found south of Crete, Greece. J Mar Biol Assoc UK 83:667–670. https://doi.org/10.1017/S0025315403007628h
3. Rochman CM, Tahir A, Williams SL, Baxa DV, Lam R, Miller JT, Teh F-C, Werorilangi S, Teh SJ (2015) Anthropogenic debris in seafood: plastic debris and fibers from textiles in fish and bivalves sold for human consumption. Sci Rep 5:14340. https://doi.org/10.1038/srep14340
4. Moore CJ (2008) Synthetic polymers in the marine environment: a rapidly increasing, long-term threat. Environ Res 108:131–139. https://doi.org/10.1016/j.envres.2008.07.025
5. Worm B, Lotze HK, Jubinville I, Wilcox C, Jambeck J (2017) Plastic as a persistent marine pollutant. Annu Rev Environ Resour 42:1–26. https://doi.org/10.1146/annurev-environ-102016-060700
6. PlasticsEurope, Plastics – the Facts (2017) An analysis of European plastics production, demand and waste data, 2017. https://doi.org/10.1016/j.marpolbul.2013.01.015
7. Jambeck JR, Geyer R, Wilcox C, Siegler TR, Perryman M, Andrady A, Narayan R, Law KL (2015) Plastic waste inputs from land into the ocean. Science 347:768–771
8. Derraik JGB (2002) The pollution of the marine environment by plastic debris: a review. Mar Pollut Bull 44:842–852. https://doi.org/10.1016/S0025-326X(02)00220-5
9. Boerger CM, Lattin GL, Moore SL, Moore CJ (2010) Plastic ingestion by planktivorous fishes in the North Pacific Central Gyre. Mar Pollut Bull 60:2275–2278. https://doi.org/10.1016/j.marpolbul.2010.08.007

10. Wright SL, Thompson RC, Galloway TS (2013) The physical impacts of microplastics on marine organisms: a review. Environ Pollut 178:483–492. https://doi.org/10.1016/j.envpol.2013.02.031
11. Jovanović B (2017) Ingestion of microplastics by fish and its potential consequences from a physical perspective. Int Environ Assess Manag 13:510–515. https://doi.org/10.1002/ieam.1913
12. Barnes DKA, Galgani F, Thompson RC, Barlaz M (2009) Accumulation and fragmentation of plastic debris in global environments. Philos Trans R Soc B Biol Sci 364:1985–1998. https://doi.org/10.1098/rstb.2008.0205
13. Law KL, Thompson RC (2014) Microplastics in the seas. Nature 345:144–145. https://doi.org/10.1002/2014EF000240/polymer
14. Coll M, Piroddi C, Steenbeek J, Kaschner K, Lasram FBR, Aguzzi J, Ballesteros E, Bianchi CN, Corbera J, Dailianis T, Danovaro R, Estrada M, Froglia C, Galil BS, Gasol JM, Gertwage R, Gil J, Guilhaumon F, Kesner-Reyes K, Kitsos MS, Koukouras A, Lampadariou N, Laxamana E, de la Cuadra CMLF, Lotze HK, Martin D, Mouillot D, Oro D, Raicevich S, Rius-Barile J, Saiz-Salinas JI, Vicente CS, Somot S, Templado J, Turon X, Vafidis D, Villanueva R, Voultsiadou E (2010) The biodiversity of the Mediterranean Sea: estimates, patterns, and threats. PLoS One 5:e11842. https://doi.org/10.1371/journal.pone.0011842
15. UNEP-MAP RAC/SPA (2010) The Mediterranean Sea biodiversity: state of the ecosystems, pressures, impacts and future priorities
16. Cózar A, Sanz-Martín M, Martí E, González-Gordillo JI, Ubeda B, Gálvez JÁ, Irigoien X, Duarte CM (2015) Plastic accumulation in the Mediterranean Sea. PLoS One 10:1–12. https://doi.org/10.1371/journal.pone.0121762
17. Liubartseva S, Coppini G, Lecci R, Clementi E (2018) Tracking plastics in the Mediterranean: 2D Lagrangian model. Mar Pollut Bull 129:151–162. https://doi.org/10.1016/j.marpolbul.2018.02.019
18. Pham CK, Ramirez-Llodra E, Alt CHS, Amaro T, Bergmann M, Canals M, Company JB, Davies J, Duineveld G, Galgani F, Howell KL, Huvenne VAI, Isidro E, Jones DOB, Lastras G, Morato T, Gomes-Pereira JN, Purser A, Stewart H, Tojeira I, Tubau X, van Rooij D, Tyler PA (2014) Marine litter distribution and density in European seas, from the shelves to deep basins. PLoS One 9:e95839. https://doi.org/10.1371/journal.pone.0095839
19. Suaria G, Avio CG, Mineo A, Lattin GL, Magaldi MG, Belmonte G, Moore CJ, Regoli F, Aliani S (2016) The Mediterranean plastic soup: synthetic polymers in Mediterranean surface waters. Sci Rep 6:1–10. https://doi.org/10.1038/srep37551
20. Laist DW (1997) Impacts of marine debris: entanglement of marine life in marine debris including a comprehensive list of species with entanglement and ingestion records. In: Coe JM, Rogers DB (eds) Marine debris. Sources, impacts solution. Springer, New York, pp 99–413. https://doi.org/10.1007/978-1-4613-8486-1
21. Gall SC, Thompson RC (2015) The impact of debris on marine life. Mar Pollut Bull 92:170–179. https://doi.org/10.1016/j.marpolbul.2014.12.041
22. Compa M, Alomar C, Wilcox C, van Sebille E, Lebreton L, Hardesty BD, Deudero S (2019) Risk assessment of plastic pollution on marine diversity in the Mediterranean Sea. Sci Total Environ 678:188–196. https://doi.org/10.1016/j.scitotenv.2019.04.355
23. Kühn S, Bravo Rebolledo EL, van Franeker JA (2015) Deleterious effects of litter on marine life. In: Bergmann M, Gutow L, Klages M (eds) Marine anthropogenic litter. Springer, Cham, pp 75–116. https://doi.org/10.1007/978-3-319-16510-3_4
24. Markic A, Gaertner J-C, Gaertner-Mazouni N, Koelmans AA (2019) Plastic ingestion by marine fish in the wild. Crit Rev Environ Sci Technol:1–41. https://doi.org/10.1080/10643389.2019.1631990
25. Deudero S, Alomar C (2015) Mediterranean marine biodiversity under threat: reviewing influence of marine litter on species. Mar Pollut Bull 98:58–68. https://doi.org/10.1016/j.marpolbul.2015.07.012

26. Fossi MC, Pedà C, Compa M, Tsangaris C, Alomar C, Claro F, Ioakeimidis C, Galgani F, Hema T, Deudero S, Romeo T, Battaglia P, Andaloro F, Caliani I, Casini S, Panti C, Baini M (2018) Bioindicators for monitoring marine litter ingestion and its impacts on Mediterranean biodiversity. Environ Pollut 237:1023–1040. https://doi.org/10.1016/j.envpol.2017.11.019
27. Kiessling T, Gutow L, Thiel M (2015) Marine litter as habitat and dispersal vector. In: Marine anthropogenic litter, pp 141–181. https://doi.org/10.1007/978-3-319-16510-3
28. Gusmão F, Di Domenico M, Amaral ACZ, Martínez A, Gonzalez BC, Worsaae K, Ivar do Sul JA, da Cunha Lana P (2016) In situ ingestion of microfibres by meiofauna from sandy beaches. Environ Pollut 216:584–590. https://doi.org/10.1016/j.envpol.2016.06.015
29. Digka N, Tsangaris C, Torre M, Anastasopoulou A, Zeri C (2018) Microplastics in mussels and fish from the Northern Ionian Sea. Mar Pollut Bull 135:30–40. https://doi.org/10.1016/j.marpolbul.2018.06.063
30. Vandermeersch G, van Cauwenberghe L, Janssen CR, Marques A, Granby K, Fait G, Kotterman MJJ, Diogène J, Bekaert K, Robbens J, Devriese L (2015) A critical view on microplastic quantification in aquatic organisms. Environ Res 143:46–55. https://doi.org/10.1016/j.envres.2015.07.016
31. Avio CG, Cardelli LR, Gorbi S, Pellegrini D, Regoli F (2017) Microplastics pollution after the removal of the Costa Concordia wreck: first evidences from a biomonitoring case study. Environ Pollut 227:207–214. https://doi.org/10.1016/j.envpol.2017.04.066
32. Renzi M, Guerranti C, Blašković A (2018) Microplastic contents from maricultured and natural mussels. Mar Pollut Bull 131:248–251. https://doi.org/10.1016/j.marpolbul.2018.04.035
33. Macali A, Semenov A, Venuti V, Crupi V, D'Amico F, Rossi B, Corsi I, Bergami E (2018) Episodic records of jellyfish ingestion of plastic items reveal a novel pathway for trophic transference of marine litter. Sci Rep 8:6105. https://doi.org/10.1038/s41598-018-24427-7
34. Alomar C, Estarellas F, Deudero S (2016) Microplastics in the Mediterranean Sea: deposition in coastal shallow sediments, spatial variation and preferential grain size. Mar Environ Res 115:1–10. https://doi.org/10.1016/j.marenvres.2016.01.005
35. Renzi M, Blašković A, Bernardi G, Russo GF (2018) Plastic litter transfer from sediments towards marine trophic webs: a case study on holothurians. Mar Pollut Bull 135:376–385. https://doi.org/10.1016/j.marpolbul.2018.07.038
36. Remy F, Collard F, Gilbert B, Compère P, Eppe G, Lepoint G (2015) When microplastic is not plastic: the ingestion of artificial cellulose fibers by macrofauna living in seagrass macrophytodetritus. Environ Sci Technol 49:11158–11166. https://doi.org/10.1021/acs.est.5b02005
37. Carreras-Colom E, Constenla M, Soler-Membrives A, Cartes JE, Baeza M, Padrós F, Carrassón M (2018) Spatial occurrence and effects of microplastic ingestion on the deep-water shrimp *Aristeus antennatus*. Mar Pollut Bull 133:44–52. https://doi.org/10.1016/j.marpolbul.2018.05.012
38. Cristo M, Cartes JE (1998) A comparative study of the feeding ecology of *Nephrops norvegicus* (L.), (Decapoda: Nephropidae) in the bathyal Mediterranean and adjacent Atlantic. Sci Mar 62:81–90. https://doi.org/10.3989/scimar.1998.62s181
39. Bordbar L, Kapiris K, Kalogirou S, Anastasopoulou A (2018) First evidence of ingested plastics by a high commercial shrimp species (*Plesionika narval*) in the eastern Mediterranean. Mar Pollut Bull 136:472–476. https://doi.org/10.1016/j.marpolbul.2018.09.030
40. Güven O, Gökdağ K, Jovanović B, Kıdeyş AE (2017) Microplastic litter composition of the Turkish territorial waters of the Mediterranean Sea, and its occurrence in the gastrointestinal tract of fish. Environ Pollut 223:286–294. https://doi.org/10.1016/j.envpol.2017.01.025
41. Rios-Fuster B, Alomar C, Compa M, Guijarro B, Deudero S (2019) Anthropogenic particles ingestion in fish species from two areas of the western Mediterranean Sea. Mar Pollut Bull 144:325–333. https://doi.org/10.1016/j.marpolbul.2019.04.064
42. Nadal MA, Alomar C, Deudero S (2016) High levels of microplastic ingestion by the semipelagic fish Bogue *Boops boops* (L.) around the Balearic Islands. Environ Pollut 214:517–523. https://doi.org/10.1016/j.envpol.2016.04.054

43. Cartes JE, Soler-Membrives A, Stefanescu C, Lombarte A, Carrassón M (2016) Contributions of allochthonous inputs of food to the diets of benthopelagic fish over the Northwest Mediterranean slope (to 2300 m). Deep-Sea Res I Oceanogr Res Pap 109:123–136. https://doi.org/10.1016/j.dsr.2015.11.001
44. Avio CG, Gorbi S, Regoli F (2015) Experimental development of a new protocol for extraction and characterization of microplastics in fish tissues: first observations in commercial species from Adriatic Sea. Mar Environ Res 111:18–26. https://doi.org/10.1016/j.marenvres.2015.06.014
45. Anastasopoulou A, Kova M, Bojani D, Digka N, Fortibuoni T, Koren Š, Mandi M, Mytilineou C, Pe A, Ronchi F, Jasna Š, Torre M, Tsangaris C, Tutman P (2018) Assessment on marine litter ingested by fish in the Adriatic and NE Ionian Sea macro-region (Mediterranean). Mar Pollut Bull 133:841–851. https://doi.org/10.1016/j.marpolbul.2018.06.050
46. Massutí E, Deudero S, Sánchez P, Morales-Nin B (1998) Diet and feeding of dolphin (*Coryphaena hippurus*) in Western Mediterranean waters. Bull Mar Sci 63:329–341
47. Romeo T, Pedà C, Battaglia P, Fossi MC, Andaloro F (2016) First record of plastic debris in the stomach of Mediterranean lanternfishes. Acta Adriat 57:115–124
48. Renzi M, Specchiulli A, Blašković A, Manzo C, Mancinelli G, Cilenti L (2019) Marine litter in stomach content of small pelagic fishes from the Adriatic Sea: sardines (*Sardina pilchardus*) and anchovies (*Engraulis encrasicolus*). Environ Sci Pollut Res 26:2771–2781. https://doi.org/10.1007/s11356-018-3762-8
49. Collard F, Gilbert B, Eppe G, Parmentier E, Das K (2015) Detection of anthropogenic particles in fish stomachs: an isolation method adapted to identification by Raman spectroscopy. Arch Environ Contam Toxicol 69:331–339. https://doi.org/10.1007/s00244-015-0221-0
50. Bottari T, Savoca S, Mancuso M, Capillo G, Panarello G, Bonsignore M, Crupi R, Sanfilippo M, D'Urso L, Compagnini G, Neri F, Romeo T, Luna GM, Spanò N, Fazio E (2019) Plastics occurrence in the gastrointestinal tract of *Zeus faber* and *Lepidopus caudatus* from the Tyrrhenian Sea. Mar Pollut Bull 146:408–416. https://doi.org/10.1016/j.marpolbul.2019.07.003
51. Giani D, Baini M, Galli M, Casini S, Fossi MC (2019) Microplastics occurrence in edible fish species (*Mullus barbatus* and *Merluccius merluccius*) collected in three different geographical sub-areas of the Mediterranean Sea. Mar Pollut Bull 140:129–137. https://doi.org/10.1016/j.marpolbul.2019.01.005
52. Mancuso M, Savoca S, Bottari T (2019) First record of microplastics ingestion by European hake (*Merluccius merluccius*) from the Tyrrhenian Sicilian coast (Central Mediterranean Sea). J Fish Biol 94:517–519. https://doi.org/10.1111/jfb.13920
53. Bellas J, Martínez-Armental J, Martínez-Cámara A, Besada V, Martínez-Gómez C (2016) Ingestion of microplastics by demersal fish from the Spanish Atlantic and Mediterranean coasts. Mar Pollut Bull 109:55–60. https://doi.org/10.1016/j.marpolbul.2016.06.026
54. Piccardo M, Felline S, Terlizzi A (2018) Preliminary assessment of microplastic accumulation in wild Mediterranean species. In: Cocca M et al (eds) Proceedings of the international conference on microplastic pollution in the Mediterranean Sea. Springer, Cham, pp 115–120. https://doi.org/10.1007/978-3-319-71279-6_16
55. Alomar C, Deudero S (2017) Evidence of microplastic ingestion in the shark *Galeus melastomus* Rafinesque, 1810 in the continental shelf off the western Mediterranean Sea. Environ Pollut 223:223–229. https://doi.org/10.1016/j.envpol.2017.01.015
56. Collignon A, Hecq J-H, Galgani F, Voisin P, Collard F, Goffart A (2012) Neustonic microplastic and zooplankton in the North Western Mediterranean Sea. Mar Pollut Bull 64:861–864. https://doi.org/10.1016/j.marpolbul.2012.01.011
57. Romeu OR, Cartes JE, Solé M, Carrassón M (2016) To what extent can specialized species succeed in the deep sea? The biology and trophic ecology of deep-sea spiny eels (Notacanthidae) in the Mediterranean Sea. Deep-Sea Res I Oceanogr Res Pap 115:74–90. https://doi.org/10.1016/j.dsr.2016.05.006

58. Anastasopoulou A, Mytilineou C, Smith CJ, Papadopoulou KN (2013) Plastic debris ingested by deep-water fish of the Ionian Sea (Eastern Mediterranean). Deep-Sea Res I Oceanogr Res Pap 74:11–13
59. Savoca S, Capillo G, Mancuso M, Bottari T, Crupi R, Branca C, Romano V, Faggio C, D'Angelo G, Spanò N (2019) Microplastics occurrence in the tyrrhenian waters and in the gastrointestinal tract of two congener species of seabreams. Environ Toxicol Pharmacol 67:35–41. https://doi.org/10.1016/j.etap.2019.01.011
60. Compa M, Ventero A, Iglesias M, Deudero S (2018) Ingestion of microplastics and natural fibres in *Sardina pilchardus* (Walbaum, 1792) and *Engraulis encrasicolus* (Linnaeus, 1758) along the Spanish Mediterranean coast. Mar Pollut Bull 128:89–96. https://doi.org/10.1016/j.marpolbul.2018.01.009
61. van der Hal N, Yeruham E, Angel DL (2018) Dynamics in microplastic ingestion during the past six decades in herbivorous fish on the Mediterranean Israeli coast. In: Cocca M et al (eds) Proceedings of the international conference on microplastic pollution in the Mediterranean Sea. Springer, Cham. https://doi.org/10.1007/978-3-319-71279-6_21
62. Pellini G, Gomiero A, Fortibuoni T, Ferrà C, Grati F, Tassetti AN, Polidori P, Fabi G, Scarcella G (2018) Characterization of microplastic litter in the gastrointestinal tract of *Solea solea* from the Adriatic Sea. Environ Pollut 234:943–952. https://doi.org/10.1016/j.envpol.2017.12.038
63. Romeo T, Battaglia P, Pedà C, Consoli P, Andaloro F, Fossi MC (2015) First evidence of presence of plastic debris in stomach of large pelagic fish in the Mediterranean Sea. Mar Pollut Bull 95:358–361. https://doi.org/10.1016/j.marpolbul.2015.04.048
64. Battaglia P, Pedà C, Musolino S, Esposito V, Andaloro F, Romeo T (2016) Diet and first documented data on plastic ingestion of Trachinotus ovatus L. 1758 (Pisces: Carangidae) from the strait of Messina (Central Mediterranean Sea). Ital J Zool 83:121–129. https://doi.org/10.1080/11250003.2015.1114157
65. Carrassón M, Stefanescu C, Cartes JE (1992) Diets and bathymetric distributions of two bathyal sharks of the Catalan deep sea (Western Mediterranean). Mar Ecol Prog Ser 82:21–30
66. Valente T, Sbrana A, Scacco U, Jacomini C, Bianchi J, Palazzo L, de Lucia GA, Silvestri C, Matiddi M (2019) Exploring microplastic ingestion by three deep-water elasmobranch species: a case study from the Tyrrhenian Sea. Environ Pollut 253:342–350. https://doi.org/10.1016/j.envpol.2019.07.001
67. Bernardini I, Garibaldi F, Canesi L, Fossi MC, Baini M (2018) First data on plastic ingestion by blue sharks (*Prionace glauca*) from the Ligurian Sea (North-Western Mediterranean Sea). Mar Pollut Bull 135:303–310. https://doi.org/10.1016/j.marpolbul.2018.07.022
68. Alexiadou P, Foskolos I, Frantzis A (2019) Ingestion of macroplastics by odontocetes of the Greek Seas, Eastern Mediterranean: often deadly! Mar Pollut Bull 146:67–75. https://doi.org/10.1016/j.marpolbul.2019.05.055
69. Shoham-Frider E, Amiel S, Roditi-Elasar M, Kress N (2002) Risso's dolphin (*Grampus griseus*) stranding on the coast of Israel (eastern Mediterranean). Autopsy results and trace metal concentrations. Sci Total Environ 295:157–166. https://doi.org/10.1016/S0048-9697(02)00089-X
70. De Stephanis R, Giménez J, Carpinelli E, Gutierrez-Exposito C, Cañadas A (2013) As main meal for sperm whales: plastics debris. Mar Pollut Bull 69:206–214. https://doi.org/10.1016/j.marpolbul.2013.01.033
71. Panti C, Baini M, Lusher A, Hernandez-Milan G, Bravo Rebolledo EL, Unger B, Syberg K, Simmonds MP, Fossi MC (2019) Marine litter: one of the major threats for marine mammals. Outcomes from the European Cetacean Society workshop. Environ Pollut 247:72–79. https://doi.org/10.1016/j.envpol.2019.01.029
72. Mazzariol S, di Guardo G, Petrella A, Marsili L, Fossi CM, Leonzio C, Zizzo N, Vizzini S, Gaspari S, Pavan G, Podestà M, Garibaldi F, Ferrante M, Copat C, Traversa D, Marcer F, Airoldi S, Frantzis A, de Beraldo Quirós Y, Cozzi B, Fernández A (2011) Sometimes sperm whales (*Physeter macrocephalus*) cannot find their way back to the high seas: a multidisciplinary study on a mass stranding. PLoS One 6:e19417. https://doi.org/10.1371/journal.pone.0019417

73. Poppi L, Zaccaroni A, Pasotto D, Dotto G, Marcer F, Scaravelli D, Mazzariol S (2012) Post-mortem investigations on a leatherback turtle *Dermochelys coriacea* stranded along the northern Adriatic coastline. Dis Aquat Org 100:71–76. https://doi.org/10.3354/dao02479
74. Viale D, Verneau N, Tison Y (1992) Stomach obstruction in a sperm whale beached on the Lavezzi islands: macropollution in the Mediterranean. J Rech Oceanogr 16:100–102
75. Pribanic S, Holcer D, Miokovic D (1998) First report of plastic ingestion by striped dolphin (Stenella coeruleoalba) in the Croatian part of the Adriatic Sea. Eur Res Cetac 13:443–446
76. Levy AM, Brenner O, Scheinin A, Morick D, Ratner E, Goffman O, Kerem D (2009) Laryngeal snaring by ingested fishing net in a common bottlenose dolphin (*Tursiops truncatus*) off the Israeli shoreline. J Wildl Dis 45:834–838. https://doi.org/10.7589/0090-3558-45.3.834
77. Gomerčić MD, Galov A, Gomerčić T, Škrtić D, Ćurković S, Lucić H, Vuković S, Arbanasić H, Gomerčić H (2009) Bottlenose dolphin (*Tursiops truncatus*) depredation resulting in larynx strangulation with gill-net parts. Mar Mamm Sci 25:392–401. https://doi.org/10.1111/j.1748-7692.2008.00259.x
78. Milani CB, Vella A, Vidoris P, Christidis A, Koutrakis E, Frantzis A, Miliou A, Kallianiotis A (2018) Cetacean stranding and diet analyses in the North Aegean Sea (Greece). J Mar Biol Assoc UK 98:1011–1028. https://doi.org/10.1017/S0025315417000339
79. Gomerčić H, Duras Gomerčić M, Gomerčić T, Lucić H, Dalebout M, Galov A, Škrtić D, Ćurković S, Vuković S, Huber D (2006) Biological aspects of Cuvier's beaked whale (*Ziphius cavirostris*) recorded in the Croatian part of the Adriatic Sea. Eur J Wildl Res 52:182–187. https://doi.org/10.1007/s10344-006-0032-8
80. Lazar B, Gračan R (2011) Ingestion of marine debris by loggerhead sea turtles, *Caretta caretta*, in the Adriatic Sea. Mar Pollut Bull 62:43–47. https://doi.org/10.1016/j.marpolbul.2010.09.013
81. Campani T, Baini M, Giannetti M, Cancelli F, Mancusi C, Serena F, Marsili L, Casini S, Fossi MC (2013) Presence of plastic debris in loggerhead turtle stranded along the Tuscany coasts of the Pelagos Sanctuary for Mediterranean Marine Mammals (Italy). Mar Pollut Bull 74:225–230. https://doi.org/10.1016/j.marpolbul.2013.06.053
82. Gramentz D (1988) Involvement of loggerhead turtle with the plastic, metal, and hydrocarbon pollution in the Central Mediterranean. Mar Pollut Bull 19:11–13. https://doi.org/10.1016/0025-326X(88)90746-1
83. Tomás J, Guitart R, Mateo R, Raga J (2002) Marine debris ingestion in loggerhead sea turtles, *Caretta caretta*, from the Western Mediterranean. Mar Pollut Bull 44:211–216. https://doi.org/10.1016/S0025-326X(01)00236-3
84. Russo G, di Bella C, Insacco G, Palazzo P, Violani C, Zava B (2003) Notes on the influence of human activities on sea chelonians in Sicilian waters. J Mt Ecol 7:37–41
85. Revelles M, Cardona L, Aguilar A, Fernández G (2007) The diet of pelagic loggerhead sea turtles (*Caretta caretta*) off the Balearic archipelago (western Mediterranean): relevance of long-line baits. J Mar Biol Assoc UK 87:805–813. https://doi.org/10.1017/S0025315407054707
86. Casale P, Freggi D, Paduano V, Oliverio M (2016) Biases and best approaches for assessing debris ingestion in sea turtles, with a case study in the Mediterranean. Mar Pollut Bull 110:238–249. https://doi.org/10.1016/j.marpolbul.2016.06.057.
87. Camedda A, Marra S, Matiddi M, Massaro G, Coppa S, Perilli A, Ruiu A, Briguglio P, de Lucia GA (2014) Interaction between loggerhead sea turtles (*Caretta caretta*) and marine litter in Sardinia (Western Mediterranean Sea). Mar Environ Res 100:25–32. https://doi.org/10.1016/j.marenvres.2013.12.004
88. Mecozzi M, Pietroletti M, Monakhova YB (2016) FTIR spectroscopy supported by statistical techniques for the structural characterization of plastic debris in the marine environment: application to monitoring studies. Mar Pollut Bull 106:155–161. https://doi.org/10.1016/j.marpolbul.2016.03.012
89. Matiddi M, Hochsheid S, Camedda A, Baini M, Cocumelli C, Serena F, Tomassetti P, Travaglini A, Marra S, Campani T, Scholl F, Mancusi C, Amato E, Briguglio P, Maffucci F, Fossi MC, Bentivegna F, de Lucia GA (2017) Loggerhead Sea turtles (*Caretta caretta*): a target

species for monitoring litter ingested by marine organisms in the Mediterranean Sea. Environ Pollut 230:199–209. https://doi.org/10.1016/j.envpol.2017.06.054

90. Sönmez B (2018) Sixteen year (2002–2017) record of sea turtle strandings on Samandağ Beach, the Eastern Mediterranean coast of Turkey Bektaş Sönmez. Zool Stud 57:53. https://doi.org/10.6620/ZS.2018.57-53
91. Domènech F, Aznar FJ, Raga JA, Tomás J (2019) Two decades of monitoring in marine debris ingestion in loggerhead sea turtle, *Caretta caretta*, from the western Mediterranean. Environ Pollut 244:367–378. https://doi.org/10.1016/j.envpol.2018.10.047
92. Casale P, Abbate G, Freggi D, Conte N, Oliverio M, Argano R (2008) Foraging ecology of loggerhead sea turtles *Caretta caretta* in the Central Mediterranean Sea: evidence for a relaxed life history model. Mar Ecol Prog Ser 372:265–276. https://doi.org/10.3354/meps07702
93. Blasi MF, Mattei D (2017) Seasonal encounter rate, life stages and main threats to the loggerhead sea turtle (*Caretta caretta*) in the Aeolian Archipelago (southern Tyrrhenian Sea). Aquat Conserv Mar Freshwat Ecosyst 27:617–630. https://doi.org/10.1002/aqc.2723
94. Blasi MF, Roscioni F, Mattei D (2016) Interaction of loggerhead turtles (*Caretta caretta*) with traditional fish aggregating devices (FADs) in the Mediterranean Sea. Herpetol Conserv Biol 11:386–401
95. Duncan EM, Broderick AC, Fuller WJ, Galloway TS, Godfrey MH, Hamann M, Limpus CJ, Lindeque PK, Mayes AG, Omeyer LCM, Santillo D, Snape RTE, Godley B (2019) Microplastic ingestion ubiquitous in marine turtles. Glob Change Biol 25(2):744–752. https://doi.org/10.1111/gcb.14519
96. Duncan EM, Arrowsmith JA, Bain CE, Bowdery H, Broderick AC, Chalmers T, Fuller WJ, Galloway TS, Lee JH, Lindeque PK, Omeyer LCM, Snape RTE, Godley BJ (2019) Diet-related selectivity of macroplastic ingestion in green turtles (*Chelonia mydas*) in the eastern Mediterranean. Sci Rep 9:11581. https://doi.org/10.1038/s41598-019-48086-4
97. Katsanevakis S (2008) Marine debris, a growing problem: sources, distribution, composition, and impacts. In: Hofer TN (ed) Marine pollution: new research. Nova Science, New York, pp 53–100
98. Codina-García M, Militão T, Moreno J, González-Solís J (2013) Plastic debris in Mediterranean seabirds. Mar Pollut Bull 77:220–226. https://doi.org/10.1016/j.marpolbul.2013.10.002
99. Steen R, Torjussen CS, Jones DW, Tsimpidis T, Miliou A (2016) Plastic mistaken for prey by a colony-breeding Eleonora's falcon (*Falco eleonorae*) in the Mediterranean Sea, revealed by camera-trap. Mar Pollut Bull 106:200–201. https://doi.org/10.1016/j.marpolbul.2016.02.069
100. Aliani S, Molcard A (2003) Hitch-hiking on floating marine debris: macrobenthic species in the Western Mediterranean Sea. Hydrobiologia 503:59–63
101. Masó M, Fortuño JM, de Juan S, Demestre M (2016) Microfouling communities from pelagic and benthic marine plastic debris sampled across Mediterranean coastal waters. Sci Mar 80:117–127. https://doi.org/10.3989/scimar.04281.10A
102. Fortuño J, Masó M, Sáez R, de Juan S, Demestre M (2010) SEM microphotographs of biofouling organisms on floating and benthic plastic debris. In: Proceedings of CIESM congress, Venice, 10–14 May 2010, pp 7947
103. Masó M, Garcés E, Pagès F, Camp J (2003) Drifting plastic debris as a potential vector for dispersing Harmful Algal Bloom (HAB) species. Sci Mar 67:107–111. https://doi.org/10.3989/scimar.2003.67n1107
104. Battaglia P, Consoli P, Ammendolia G, D'Alessandro M, Bo M, Vicchio TM, Pedà C, Cavallaro M, Andaloro F, Romeo T (2019) Colonization of floats from submerged derelict fishing gears by four protected species of deep-sea corals and barnacles in the strait of Messina (Central Mediterranean Sea). Mar Pollut Bull 148:61–65. https://doi.org/10.1016/j.marpolbul.2019.07.073
105. Jorissen FJ (2014) Colonization by the benthic foraminifer Rosalina (Tretomphalus) concinna of Mediterranean drifting plastics. In: CIESM workshop monographs, pp 18–21

106. Viršek MK, Lovšin MN, Koren Š, Kržan A, Peterlin M (2017) Microplastics as a vector for the transport of the bacterial fish pathogen species *Aeromonas salmonicida*. Mar Pollut Bull 125:301–309. https://doi.org/10.1016/j.marpolbul.2017.08.024
107. Dussud C, Meistertzheim AL, Conan P, Pujo-Pay M, George M, Fabre P, Coudane J, Higgs P, Elineau A, Pedrotti ML, Gorsky G, Ghiglione JF (2018) Evidence of niche partitioning among bacteria living on plastics, organic particles and surrounding seawaters. Environ Pollut 236:807–816. https://doi.org/10.1016/j.envpol.2017.12.027
108. Ioakeimidis C, Papatheodorou G, Fermeli G, Streftaris N, Papathanassiou E (2015) Use of ROV for assessing marine litter on the seafloor of Saronikos Gulf (Greece): a way to fill data gaps and deliver environmental education. Springerplus 4(1):1–9. https://doi.org/10.1186/s40064-015-1248-4
109. Cau A, Alvito A, Moccia D, Canese S, Pusceddu A, Rita C, Angiolillo M, Follesa MC (2017) Submarine canyons along the upper Sardinian slope (Central Western Mediterranean) as repositories for derelict fishing gears. Mar Pollut Bull 123:357–364. https://doi.org/10.1016/j.marpolbul.2017.09.010
110. Consoli P, Romeo T, Angiolillo M, Canese S, Esposito V, Salvati E, Scotti G, Andaloro F, Tunesi L (2019) Marine litter from fishery activities in the Western Mediterranean Sea: the impact of entanglement on marine animal forests. Environ Pollut 249:472–481. https://doi.org/10.1016/j.envpol.2019.03.072
111. Gündoğdu S, Çevik C, Karaca S (2017) Fouling assemblage of benthic plastic debris collected from Mersin Bay, NE Levantine coast of Turkey. Mar Pollut Bull 124:147–154. https://doi.org/10.1016/j.marpolbul.2017.07.023.
112. Lastras G, Canals M, Ballesteros E, Gili J-M, Sanchez-Vidal A (2016) Cold-water corals and anthropogenic impacts in La Fonera submarine canyon head, northwestern Mediterranean Sea. PLoS One 11(5):e0155729. https://doi.org/10.1371/journal.pone.0155729
113. Frazier JG, Margaritoulis D (1990) The occurrence of the barnacle, *Chelonibia patula* (Ranzani, 1818), on an inanimate substratum (Cirripedia, Thoracica). Crustaceana 59 (2):213–218
114. Ayaz A, Ünal V, Acarli D, Altinagac U (2010) Fishing gear losses in the Gökova Special Environmental Protection Area (SEPA), eastern Mediterranean, Turkey. J Appl Ichthyol 26:416–419. https://doi.org/10.1111/j.1439-0426.2009.01386.x
115. Consoli P, Andaloro F, Altobelli C, Battaglia P, Campagnuolo S, Canese S, Castriota L, Cillari T, Falautano M, Pedà C, Perzia P, Sinopoli M, Vivona P, Scotti G, Esposito V, Galgani F, Romeo T (2018) Marine litter in an EBSA (Ecologically or Biologically Significant Area) of the Central Mediterranean Sea: abundance, composition, impact on benthic species and basis for monitoring entanglement. Environ Pollut 236:405–415. https://doi.org/10.1016/j.envpol.2018.01.097
116. Angiolillo M, di Lorenzo B, Farcomeni A, Bo M, Bavestrello G, Santangelo G, Cau A, Mastascusa V, Cau A, Sacco F, Canese S (2015) Distribution and assessment of marine debris in the deep Tyrrhenian Sea (NW Mediterranean Sea, Italy). Mar Pollut Bull 92:149–159. https://doi.org/10.1016/j.marpolbul.2014.12.044
117. Bo M, Bava S, Canese S, Angiolillo M, Cattaneo-Vietti R, Bavestrello G (2014) Fishing impact on deep Mediterranean rocky habitats as revealed by ROV investigation. Biol Conserv 171:167–176. https://doi.org/10.1016/j.biocon.2014.01.011
118. Bo M, Canese S, Spaggiari C, Pusceddu A, Bertolino M, Angiolillo M, Giusti M, Loreto MF, Salvati E, Greco S, Bavestrello G (2012) Deep coral oases in the South Tyrrhenian Sea. PLoS One 7(11):e49870. https://doi.org/10.1371/journal.pone.0049870
119. Bo M, Cerrano C, Canese S, Salvati E, Angiolillo M, Santangelo G, Bavestrello G (2014) The coral assemblages of an off-shore deep Mediterranean rocky bank (NW Sicily, Italy). Mar Ecol 35:332–342. https://doi.org/10.1111/maec.12089
120. Fabri MC, Pedel L, Beuck L, Galgani F, Hebbeln D, Freiwald A (2014) Megafauna of vulnerable marine ecosystems in French Mediterranean submarine canyons: spatial distribution and anthropogenic impacts. Deep-Sea Res PT II 104:184–207. https://doi.org/10.1016/j.dsr2.2013.06.016

121. Orejas C, Gori A, Jiménez C, Rivera J, Kamidis N, Abu Alhaija R, Iacono C (2019) Occurrence and distribution of the coral *Dendrophyllia ramea* in Cyprus insular shelf: environmental setting and anthropogenic impacts. Deep-Sea Res PT II 164:190–205. https://doi.org/10.1016/j.dsr2.2019.04.006
122. Taviani M, Angeletti L, Canese S, Cannas R, Cardone F, Cau AB, Cau AB, Follesa MC, Marchese F, Montagna P, Tessarolo C (2017) The "Sardinian cold-water coral province" in the context of the Mediterranean coral ecosystems. Deep Sea Res Part II Top Stud Oceanogr 145:61–78. https://doi.org/10.1016/j.dsr2.2015.12.008.
123. D'Onghia G, Calculli C, Capezzuto F, Carlucci R, Carluccio A, Grehan A, Indennidate A, Maiorano P, Mastrototaro F, Pollice A, Russo T, Savini A, Sion L, Tursi A (2017) Anthropogenic impact in the Santa Maria di Leuca cold-water coral province (Mediterranean Sea): observations and conservation straits. Deep-Sea Res PT II 145:87–101. https://doi.org/10.1016/j.dsr2.2016.02.012
124. Bo M, Canese S, Bavestrello G (2014) Discovering Mediterranean black coral forests: *Parantipathes larix* (Anthozoa: Hexacorallia) in the Tuscan Archipelago, Italy. Ital J Zool 81(1):112–125. https://doi.org/10.1080/11250003.2013.859750
125. Cánovas-Molina A, Montefalcone M, Bavestrello G, Cau A, Bianchi C, Morri N, Canese S, Bo M (2016) A new ecological index for the status of mesophotic megabenthic assemblages in the Mediterranean based on ROV photography and video footage. Cont Shelf Res 121:13–20. https://doi.org/10.1016/j.csr.2016.01.008
126. Deidun A, Andaloro F, Bavestrello G, Canese S, Consoli P, Micallef A, Romeo T, Bo M (2014) First characterisation of a *Leiopathes glaberrima* (Cnidaria: Anthozoa: Antipatharia) forest in Maltese exploited fishing grounds. Ital J Zool 82(2):1–10. https://doi.org/10.1080/11250003.2014.986544
127. Freiwald A, Beuck L, Ruggeberg A, Taviani M, Hebbeln D (2009) The white coral community in the Central Mediterranean Sea revealed by ROV surveys. Oceanography 22:27. https://www.jstor.org/stable/24860923
128. Orejas C, Gori A, Lo Iacono C, Puig P, Pili JM, Dale MRT (2009) Cold-water corals in the Cap de Creus canyon, northwestern Mediterranean: spatial distribution, density and anthropogenic impact. Mar Ecol-Prog Ser 397:37–51. https://doi.org/10.3354/meps08314
129. Fine M, Zibrowius H, Loya Y (2001) *Oculina patagonica*: a non-lessepsian scleractinian coral invading the Mediterranean Sea. Mar Biol 138(6):1195–1203. https://doi.org/10.1007/s002270100539
130. Bavestrello G, Cerrano C, Zanzi D, Cattaneo-Vietti R (1997) Damage by fishing activities to the Gorgonian coral *Paramuricea clavata* in the Ligurian Sea. Aquat Conserv Mar Freshwat Ecosyst 7:253–262. https://doi.org/10.1002/(SICI)1099-0755(199709)7:3<253::AID-AQC243>3.0.CO;2-1
131. Kipson S, Linares C, Čižmek H, Cebrián E, Ballesteros E, Bakran-Petricioli T, Garrabou J (2015) Population structure and conservation status of the red gorgonian *Paramuricea clavata* (Risso, 1826) in the Eastern Adriatic Sea. Mar Ecol 36(4):982–993. https://doi.org/10.1111/maec.12195
132. Pierdomenico M, Cardone F, Carluccio A, Casalbore D, Chiocci F, Maiorano P, D'Onghia G (2019) Megafauna distribution along active submarine canyons of the Central Mediterranean: relationships with environmental variables. Prog Oceanogr 171:49–69. https://doi.org/10.1016/j.pocean.2018.12.015
133. Melli V, Angiolillo M, Ronchi F, Canese S, Giovanardi O, Querin S, Fortibuoni T (2017) The first assessment of marine debris in a site of community importance in the North-Western Adriatic Sea (Mediterranean Sea). Mar Pollut Bull 114:821–830. https://doi.org/10.1016/j.marpolbul.2016.11.012
134. Angiolillo M (2019) Debris in deep water. In: Sheppard C (ed) World seas: an environmental evaluation. Elsevier, Amsterdam, pp 251–268. https://doi.org/10.1016/B978-0-12-805052-1.00015-2
135. Ramirez-Llodra E, de Mol B, Company JB, Coll M, Sardà F (2013) Effects of natural and anthropogenic processes in the distribution of marine litter in the deep Mediterranean Sea. Prog Oceanogr 118:273–287. https://doi.org/10.1016/j.pocean.2013.07.027

136. Cabezas MP, Navarro-Barranco C, Ros M, Guerra-García JM (2013) Long-distance dispersal, low connectivity and molecular evidence of a new cryptic species in the obligate rafter *Caprella andreae* Mayer, 1890 (Crustacea: Amphipoda: Caprellidae). Helgol Mar Res 67:483–497. https://doi.org/10.1007/s10152-012-0337-9
137. Holdway P, Maddock L (1983) A comparative survey of neuston: geographical and temporal distribution patterns. Mar Biol 76(3):263–270
138. Abello P, Guerao G, Codina M (2004) Distribution of the neustonic isopod *Idotea metallica* in relation to shelf-slope frontal structures. J Crustac Biol 24:558–566
139. Gutow L, Franke HD (2003) Metapopulation structure of the marine isopod *Idotea metallica*, a species associated with drifting habitat patches. Helgol Mar Res 56:259–264. https://doi.org/10.1007/s10152-002-0126-y
140. Southward AJ, Hiscock K, Kerckhof FO, Moyse JP, El AS (2004) Habitat and distribution of the warm-water barnacle, *Solidobalanus fallax* (Crustacea: Cirripedia). J Mar Biol Assoc UK 84:1169–1177. https://doi.org/10.1017/S0025315404010616h
141. Ramirez-Llodra E, Tyler PA, Baker MC, Bergstad OA, Clark MR, Escobar E, Levin LA, Menot L, Rowden AA, Smith CR, van Dover CL (2011) Man and the last great wilderness: human impact on the deep sea. PLoS One 6(7):e22588. https://doi.org/10.1371/journal.pone.0022588
142. Tutman P, Kapiris K, Kirinčić M, Pallaoro A (2017) Floating marine litter as a raft for drifting voyages for *Planes minutus* (Crustacea: Decapoda: Grapsidae) and *Liocarcinus navigator* (Crustacea: Decapoda: Polybiidae). Mar Pollut Bull 120:217–221. https://doi.org/10.1016/j.marpolbul.2017.04.063
143. Houard T, Boudouresque CF, Barcelo A, Cottalorda J, Formentin J, Jullian E, Kerlidou B, Pironneau E (2012) Occurrence of a lost fishing net within the marine area of the Port-Cros national park (Provence, northwestern Mediterranean Sea). Sci Rep Port-Cros Natl Park 118:109–118
144. Colmenero AI, Barría C, Broglio E, García-Barcelona S (2017) Plastic debris straps on threatened blue shark *Prionace glauca*. Mar Pollut Bull 115:436–438. https://doi.org/10.1016/j.marpolbul.2017.01.011
145. Butterworth A, Clegg I, Bass C (2012) Untangled. Marine debris: a global picture of the impact on animal welfare and of animal-focused solutions. World Society for the Protection of Animals, London
146. Morris RJ (1980) Floating plastic debris in the Mediterranean. Mar Pollut Bull 11:125. https://doi.org/10.1016/0025-326X(80)90073-9
147. Casale P, Affronte M, Insacco G, Freggi D, Vallini C, d'Astore PP, Basso R, Paolillo G, Abbate G, Argano R (2010) Sea turtle strandings reveal high anthropogenic mortality in Italian waters. Aquat Conserv Mar Freshwat Ecosyst 20:611–620. https://doi.org/10.1002/aqc.1133
148. Duncan EM, Botterell ZLR, Broderick AC, Galloway TS, Lindeque PK, Nuno A, Godley BJ (2017) A global review of marine turtle entanglement in anthropogenic debris: a baseline for further action. Endanger Species Res 34:431–448. https://doi.org/10.3354/esr00865
149. Özdilek Ş, Yalçın-Özdilek F, Ozaner S, Sönmez B (2006) Impact of accumulated beach litter on *Chelonia mydas* L. 1758 (green turtle) hatchlings of the Samandag coast, Hatay, Turkey. Fresenius Environ Bull 15:95–103
150. Battisti C, Kroha S, Kozhuharova E, De Michelis S, Fanelli G, Poeta G, Pietrelli L, Cerfolli F (2019) Fishing lines and fish hooks as neglected marine litter: first data on chemical composition, densities, and biological entrapment from a Mediterranean beach. Environ Sci Pollut Res 26:1000–1007. https://doi.org/10.1007/s11356-018-3753-9
151. Merlino S, Abbate M, Pietrelli L, Canepa P, Varella P (2018) Marine litter detection and correlation with the seabird nest content. Rend Lincei Sci Fis e Nat 29:867–875. https://doi.org/10.1007/s12210-018-0750-3
152. Galgani F, Hanke G, Werner S, De Vrees L (2013) Marine litter within the European marine strategy framework directive. ICES J Mar Sci 70:1055–1064. https://doi.org/10.1093/icesjms/fst122

153. Ryan PG, Moore CJ, Van Franeker JA, Moloney CL (2009) Monitoring the abundance of plastic debris in the marine environment. Philos Trans R Soc B Biol Sci 364:1999–2012. https://doi.org/10.1098/rstb.2008.0207
154. Lusher AL, Hollman PCH, Mendoza-Hill JJ (2017) Microplastics in fisheries and aquaculture: status of knowledge on their occurrence and implications for aquatic organisms and food safety. Food and Agriculture Organization of the United Nations, Rome
155. Moser ML, Lee DS (1992) A fourteen-year survey of plastic ingestion by Western North Atlantic Seabirds. Colon Waterbirds 15:83. https://doi.org/10.2307/1521357
156. Ory NC, Sobral P, Ferreira JL, Thiel M (2017) Amberstripe scad *Decapterus muroadsi* (Carangidae) fish ingest blue microplastics resembling their copepod prey along the coast of Rapa Nui (Easter Island) in the South Pacific subtropical gyre. Sci Total Environ 586:430–437. https://doi.org/10.1016/j.scitotenv.2017.01.175
157. McCord ME, Campana SE (2003) A quantitative assessment of the diet of the Blue Shark (*Prionace glauca*) off Nova Scotia, Canada. J Northwest Atl Fish Sci 32:57–63. https://doi.org/10.1080/01436597.2011.581058
158. Laist DW (1987) Overview of the biological effects of lost and discarded plastic debris in the marine environment. Mar Pollut Bull 18(6):319–326. https://doi.org/10.1016/S0025-326X(87)80019-X
159. Moss SA (1984) Sharks. Prentice-Hall, Englewood Cliffs
160. Eriksson C, Burton H (2003) Origins and biological accumulation of small plastic particles in fur seals from Macquarie Island. Ambio 32:380–384. https://doi.org/10.1579/0044-7447-32.6.380
161. Possatto FE, Barletta M, Costa MF, Ivar do Sul JA, Dantas DV (2011) Plastic debris ingestion by marine catfish: an unexpected fisheries impact. Mar Pollut Bull 62:1098–1102. https://doi.org/10.1016/j.marpolbul.2011.01.036.
162. Watts AJR, Urbina MA, Corr S, Lewis C, Galloway TS (2015) Ingestion of plastic microfibers by the crab *Carcinus maenas* and its effect on food consumption and energy balance. Environ Sci Technol 49:14597–14604. https://doi.org/10.1021/acs.est.5b04026
163. Carey MJ (2011) Intergenerational transfer of plastic debris by short-tailed shearwaters (*Ardenna tenuirostris*). Emu Austral Ornithol 111:229–234. https://doi.org/10.1071/MU10085
164. Rodríguez A, Rodríguez B, Nazaret Carrasco M (2012) High prevalence of parental delivery of plastic debris in Cory's shearwaters (*Calonectris diomedea*). Mar Pollut Bull 64:2219–2223. https://doi.org/10.1016/j.marpolbul.2012.06.011
165. Acampora H, Schuyler QA, Townsend KA, Hardesty BD (2014) Comparing plastic ingestion in juvenile and adult stranded short-tailed shearwaters (*Puffinus tenuirostris*) in eastern Australia. Mar Pollut Bull 78:63–68. https://doi.org/10.1016/j.marpolbul.2013.11.009
166. Law KL (2017) Plastics in the marine environment. Annu Rev Mar Sci 9:205–229. https://doi.org/10.1146/annurev-marine-010816-060409
167. Auta HS, Emenike CU, Fauziah SH (2017) Distribution and importance of microplastics in the marine environment a review of the sources, fate, effects, and potential solutions. Environ Int 102:165–176. https://doi.org/10.1016/j.envint.2017.02.013
168. Ryan PG (2016) Ingestion of plastics by marine organisms. In: Takada HKKH (ed) Hazardous chemicals associated with plastics in the marine environment. Springer, Cham
169. Teuten EL, Saquing JM, Knappe DRU, Barlaz MA, Jonsson S, Björn A, Rowland SJ, Thompson RC, Galloway TS, Yamashita R, Ochi D, Watanuki Y, Moore C, Viet PH, Tana TS, Prudente M, Boonyatumanond R, Zakaria MP, Akkhavong K, Ogata Y, Hirai H, Iwasa S, Mizukawa K, Hagino Y, Imamura A, Saha M, Takada H (2009) Transport and release of chemicals from plastics to the environment and to wildlife. Philos Trans R Soc B Biol Sci 364:2027–2045. https://doi.org/10.1098/rstb.2008.0284
170. Koelmans AA, Bakir A, Burton GA, Janssen CR (2016) Microplastic as a vector for chemicals in the aquatic environment: critical review and model-supported reinterpretation of empirical studies. Environ Sci Technol 50:3315–3326. https://doi.org/10.1021/acs.est.5b06069
171. van Franeker JA, Blaize C, Danielsen J, Fairclough K, Gollan J, Guse N, Hansen PL, Heubeck M, Jensen JK, Le Guillou G, Olsen B, Olsen KO, Pedersen J, Stienen EWM, Turner

DM (2011) Monitoring plastic ingestion by the northern fulmar *Fulmarus glacialis* in the North Sea. Environ Pollut 159:2609–2615. https://doi.org/10.1016/j.envpol.2011.06.008.

172. van Franeker JA, Law KL (2015) Seabirds, gyres and global trends in plastic pollution. Environ Pollut 203:89–96. https://doi.org/10.1016/j.envpol.2015.02.034
173. Zeri C, Adamopoulou A, Bojanić Varezić D, Fortibuoni T, Kovač Viršek M, Kržan A, Mandic M, Mazziotti C, Palatinus A, Peterlin M, Prvan M, Ronchi F, Siljic J, Tutman P, Vlachogianni T (2018) Floating plastics in Adriatic waters (Mediterranean Sea): from the macro- to the micro-scale. Mar Pollut Bull 136:341–350. https://doi.org/10.1016/j.marpolbul.2018.09.016
174. Lusher AL, O'Donnell C, Officer R, O'Connor I (2016) Microplastic interactions with North Atlantic mesopelagic fish. ICES J Mar Sci 73:1214–1225. https://doi.org/10.1093/icesjms/fsv241
175. Neves D, Sobral P, Ferreira JL, Pereira T (2015) Ingestion of microplastics by commercial fish off the Portuguese coast. Mar Pollut Bull 101:119–126. https://doi.org/10.1016/j.marpolbul.2015.11.008
176. Donnely-Greenan E, Hyrenbach D, Beck J, Fitzgerald S, Nevins H, Hester M (2018) First quantification of plastic ingestion by short-tailed albatross *Phoebastria albatrus*. Mar Ornithol 46:79–84
177. Denuncio P, Bastida R, Dassis M, Giardino G, Gerpe M, Rodríguez D (2011) Plastic ingestion in Franciscana dolphins, *Pontoporia blainvillei* (Gervais and d'Orbigny, 1844), from Argentina. Mar Pollut Bull 62:1836–1841. https://doi.org/10.1016/j.marpolbul.2011.05.003
178. Bessa F, Barr P, Neto JM, Frias PGL, Otero V, Sobral P, Marques JC (2018) Occurrence of microplastics in juvenile fish from an estuarine environment. Mar Pollut Bull 128:131–135. https://doi.org/10.1007/978-3-319-71279-6
179. Brongersma LD (1968) Notes upon some turtles from the Canary Islands and from Madeira. Proc K Ned Akad van Wet 71:128–136
180. Caldwell MC, Caldwell DK, Siebenaler JB (1965) Observations on captive and wild Atlantic bottlenosed dolphins, Tursiops truncatus, in the northeastern Gulf of Mexico. Contributions in science. Los Angeles City Museum, Los Angeles, pp 1–10
181. Secretariat of the Convention on Biological Diversity and the Scientific and Technical Advisory Panel – GEF (2012) Impacts of marine debris on biodiversity – current status and possible solutions. Technical Series No. 67
182. Carson HS (2013) The incidence of plastic ingestion by fishes: from the prey's perspective. Mar Pollut Bull 74:170–174. https://doi.org/10.1016/j.marpolbul.2013.07.008
183. van Cauwenberghe L, Janssen CR (2014) Microplastics in bivalves cultured for human consumption. Environ Pollut 193:65–70. https://doi.org/10.1016/j.envpol.2014.06.010
184. Catarino AI, Macchia V, Sanderson WG, Thompson RC, Henry TB (2018) Low levels of microplastics (MP) in wild mussels indicate that MP ingestion by humans is minimal compared to exposure via household fibres fallout during a meal. Environ Pollut 237:675–684. https://doi.org/10.1016/j.envpol.2018.02.069
185. Avio CG, Gorbi S, Milan M, Benedetti M, Fattorini D, D'Errico G, Pauletto M, Bargelloni L, Regoli F (2015) Pollutants bioavailability and toxicological risk from microplastics to marine mussels. Environ Pollut 198:211–222. https://doi.org/10.1016/j.envpol.2014.12.021
186. Alomar C, Sureda A, Capó X, Guijarro B, Tejada S, Deudero S (2017) Microplastic ingestion by *Mullus surmuletus* Linnaeus, 1758 fish and its potential for causing oxidative stress. Environ Res 159:135–142. https://doi.org/10.1016/j.envres.2017.07.043
187. Davison P, Asch RG (2011) Plastic ingestion by mesopelagic fishes in the North Pacific subtropical gyre. Mar Ecol Prog Ser 432:173–180. https://doi.org/10.3354/meps09142
188. Salman A (2004) The role of cephalopods in the diet of swordfish (*Xiphias gladius* Linnaeus, 1758) in the Aegean Sea (Eastern Mediterranean). Bull Mar Sci 74:21–29
189. Baulch S, Perry C (2014) Evaluating the impacts of marine debris on cetaceans. Mar Pollut Bull 80:210–221. https://doi.org/10.1016/j.marpolbul.2013.12.050
190. van Franeker JA, Bravo Rebolledo EL, Hesse E, Ijsseldijk LL, Kühn S, Leopold M, Mielke L (2018) Plastic ingestion by harbour porpoises *Phocoena phocoena* in the Netherlands:

establishing a standardised method. Ambio 47:387–397. https://doi.org/10.1007/s13280-017-1002-y

191. MSFD Technical Subgroup on Marine Litter (2013) Guidance on Monitoring of Marine Litter in European Seas. https://doi.org/10.2788/99475
192. Claro F, Fossi MC, Ioakeimidis C, Baini M, Lusher AL, Mc Fee W, McIntosh RR, Pelamatti T, Sorce M, Galgani F, Hardesty BD (2019) Tools and constraints in monitoring interactions between marine litter and megafauna: insights from case studies around the world. Mar Pollut Bull 141:147–160. https://doi.org/10.1016/j.marpolbul.2019.01.018
193. van der Hoop J, Moore M, Fahlman A, Bocconcelli A, George C, Jackson K, Miller C, Morin D, Pitchford T, Rowles T, Smith J, Zoodsma B (2014) Behavioral impacts of disentanglement of a right whale under sedation and the energetic cost of entanglement. Mar Mamm Sci 30:282–307. https://doi.org/10.1111/mms.12042
194. Allen R, Jarvis D, Sayer S, Mills C (2012) Entanglement of grey seals, *Halichoerus grypus*, at a haul out site in Cornwall, UK. Mar Pollut Bull 64:2815–2819
195. Matsuoka T, Nakashima T, Nagasawa N (2005) A review of ghost fishing: scientific approaches to evaluation and solutions. Fish Sci 71:691–702
196. Galgani F, Claro F, Depledge M, Fossi C (2014) Monitoring the impact of litter in large vertebrates in the Mediterranean Sea within the European Marine Strategy Framework Directive (MSFD): constraints, specificities and recommendations. Mar Environ Res 100:3–9. https://doi.org/10.1016/j.marenvres.2014.02.003
197. European Commission, Commission Decision (EU) 2017/848 (2017). http://eur-lex.europa.eu/pri/en/oj/dat/2003/l_285/l_28520031101en00330037.pdf
198. Triessnig P, Roetzer A, Stachowitsch M (2012) Beach condition and marine debris: new hurdles for sea turtle hatchling survival. Chelonian Conserv Biol 11:68–77. https://doi.org/10.2744/CCB-0899.1
199. Broderick AC, Glen F, Godley BJ, Hays GC (2002) Estimating the number of green and loggerhead turtles nesting annually in the Mediterranean. Oryx 36:227–235. https://doi.org/10.1017/S0030605302000431
200. Nelms SE, Duncan EM, Broderick AC, Galloway TS, Godfrey MH, Hamann M, Lindeque PK, Godley BJ (2016) Plastic and marine turtles: a review and call for research. ICES J Mar Sci 73:165–181. https://doi.org/10.1093/icesjms/fsv165
201. Karamanlidis AA, Androukaki E, Adamantopoulou S, Chatzispyrou A, Johnson WM, Kotomatas S, Papadopoulos A, Paravas V, Paximadis G, Pires R, Tounta E, Dendrinos P (2008) Assessing accidental entanglement as a threat to the Mediterranean monk seal *Monachus monachus*. Endanger Species Res 5:205–213. https://doi.org/10.3354/esr00092
202. Johnson WM, Karamanlidis AA (2000) When fishermen save seals. Monachus Guard 3:18–22
203. Pace DS, Miragliuolo A, Mussi B (2008) Behaviour of a social unit of sperm whales (*Physeter macrocephalus*) entangled in a driftnet off Capo Palinuro (Southern Tyrrhenian Sea, Italy). J Cetacean Res Manag 10:131–135. https://doi.org/10.1016/j.soilbio.2013.12.006
204. Frantzis A (2007) Fisheries interactions with cetacean species in Hellas. In: State of Hellenic fisheries. Hellenic Centre for Marine Research, Athens, pp 274–278
205. Peronace V, Cecere JG, Gustin M, Rondinini C (2012) Lista rossa 2011 degli uccelli nidificanti in Italia. Avocetta 36:11–58
206. Brown J, Macfadyen G, Huntington T, Magnus J, Tumilty J, Ghost Fishing by Lost Fishing Gear (2005) Final report to DG fisheries and maritime affairs of the European Commission. Fish/2004/20. Institute for European Environmental Policy/Poseidon Aquatic Resource Management Ltd joint report
207. Baeta F, Costa MJ, Cabral H (2009) Trammel nets' ghost fishing off the Portuguese central coast. Fish Res 98:33–39. https://doi.org/10.1016/j.fishres.2009.03.009
208. Ayaz A, Acarli D, Altinagac U, Ozekinci U, Kara A, Ozen O (2006) Ghost fishing by monofilament and multifilament gillnets in Izmir Bay, Turkey. Fish Res 79:267–271. https://doi.org/10.1016/j.fishres.2006.03.029.

209. Galgani F, Pham CK, Claro F, Consoli P (2018) Marine animal forests as useful indicators of entanglement by marine litter. Mar Pollut Bull 135:735–738. https://doi.org/10.1016/j.marpolbul.2018.08.004
210. Lamb JB, Willis BL, Fiorenza EA, Couch CS, Howard R, Rader DN, True JD, Kelly LA, Ahmad A, Jompa J, Harvell CD (2018) Plastic waste associated with disease on coral reefs. Science 80(359):460–462
211. Gregory MR (2009) Environmental implications of plastic debris in marine settings – entanglement, ingestion, smothering, hangers-on, hitch-hiking and alien invasions. Philos Trans R Soc B Biol Sci 364:2013–2025. https://doi.org/10.1098/rstb.2008.0265
212. Zettler ER, Mincer TJ, Amaral-Zettler LA (2013) Life in the "plastisphere": microbial communities on plastic marine debris. Environ Sci Technol 47:7137–7146. https://doi.org/10.1021/es401288x
213. Thiel M, Gutow L (2004) Oceanography and marine biology. CRC Press, Boca Raton. https://doi.org/10.1201/9780203507810
214. Suaria G, Aliani S (2014) Floating debris in the Mediterranean Sea. Mar Pollut Bull 86:494–504. https://doi.org/10.1016/j.marpolbul.2014.06.025
215. Deideri J, Deideri M, Beyney MC, Rouger MH (2014) Dix ans de suivi des Fous de Bassan sur la Côte Bleue. Faune-PACA Publ 43:21
216. Stafford R, Jones PJS (2019) Viewpoint – ocean plastic pollution: a convenient but distracting truth? Mar Policy 103:187–191. https://doi.org/10.1016/j.marpol.2019.02.003
217. Wilcox C, van Sebille E, Hardesty BD (2015) Threat of plastic pollution to seabirds is global, pervasive, and increasing. Proc Natl Acad Sci 112:11899–11904. https://doi.org/10.1073/pnas.1502108112
218. Barboza LGA, Dick Vethaak A, Lavorante BRBO, Lundebye A-K, Guilhermino L (2018) Marine microplastic debris: an emerging issue for food security, food safety and human health. Mar Pollut Bull 133:336–348. https://doi.org/10.1016/j.marpolbul.2018.05.047
219. Burton GA (2017) Stressor exposures determine risk: so, why do fellow scientists continue to focus on superficial microplastics risk? Environ Sci Technol 51:13515–13516. https://doi.org/10.1021/acs.est.7b05463
220. Backhaus T, Wagner M (2019) Microplastics in the environment: much ado about nothing? A debate. Global Chall 6:1900022. https://doi.org/10.1002/gch2.201900022
221. Lusher A (2015) Microplastics in the marine environment: distribution, interactions and effects. In: Marine anthropogenic litter. Springer, Cham, pp 245–307. https://doi.org/10.1007/978-3-319-16510-3_10

Plastic in the Aquatic Environment: Interactions with Microorganisms

Philipp Sapozhnikov, Alexey Salimon, Alexander M. Korsunsky, Olga Kalinina, Olesya Ilyina, Eugene Statnik, and Anastasiya Snigirova

Contents

1 Introduction 198
2 Plastic in Water and Microorganisms: State-of-the-Art Knowledge 199
  2.1 Plastic and Microorganisms in Water Bodies: Forms of Interaction 199
  2.2 Microorganisms and Plastics: Biofilm Communities and Biodegradation of Synthetic Polymers 215
3 The Results of Studies of Plastic Colonization by Microorganisms, Obtained by Our Group 221
  3.1 Microplaston on Plastic Waste in Natural Reservoirs 221

P. Sapozhnikov (✉)
P.P. Shirshov Institute of Oceanology, Moscow, Russia

A. Salimon
Center for Energy Science and Technology, Skolkovo Institute of Science and Technology, Moscow, Russia
e-mail: a.salimon@skoltech.ru

A. M. Korsunsky
Department of Engineering Science, University of Oxford, Oxford, UK

Skoltech Institute of Science and Technology, Moscow, Russia
e-mail: alexander.korsunsky@eng.ox.ac.uk

O. Kalinina
Department of Geography, Lomonosov Moscow State University Moscow, Moscow, Russia

O. Ilyina
Department of Biology, Lomonosov Moscow State University Moscow, Moscow, Russia

E. Statnik
Skoltech Institute of Science and Technology, Moscow, Russia

A. Snigirova
Institute of Marine Biology, Odessa, Ukraine

Friederike Stock, Georg Reifferscheid, Nicole Brennholt, and Evgeniia Kostianaia (eds.),
Plastics in the Aquatic Environment - Part I: Current Status and Challenges,
Hdb Env Chem (2022) 111: 197–254, DOI 10.1007/698_2021_747,

© Springer Nature Switzerland AG 2021, Published online: 6 March 2021

3.2 Microplaston in Experiments: Colonization of Synthetic Polymers in the Natural Environment 241
4 Conclusions 247
References 248

Abstract Plastic debris is gradually filling the seas, oceans, and freshwater bodies of the planet. Since the 1950s, a huge amount of plastic has entered into various bodies of water. All these objects decompose at different rates, and aquatic organisms take part in these processes. This book chapter provides an overview of studies carried out in recent decades on the interaction between microorganisms and plastic debris in the aquatic environment. Both prokaryotic communities and algo-bacterial cenoses are considered. A separate section is devoted to the research results of the authors of this book chapter, obtained in the natural environment, contaminated with plastic, and in field experiments in the sea.

Keywords Colonial settlements, Cyanoprokaryotes, Diatoms, Marine plastic, Microplaston, Plastic litter, Synthetic polymers

1 Introduction

Any object that enters any of the planet's water bodies for one reason or another will sooner or later be "assimilated" by the natural environment of these water bodies. Aquatic organisms take part in this process. First of all, these are microorganisms that play the role of pioneers, mastering various elements of the microrelief of the surface of any object that finds itself in the water: leaves, pieces of wood, metal objects, etc. Plastic waste, gradually filling the seas and oceans, lakes, rivers, and streams, is no exception to this simple rule.

However, plastic, unlike traditional materials used by mankind such as wood, metal, or natural textiles, is a material that is relatively new to the aquatic environment. Due to this circumstance, microorganisms from the communities inhabiting water bodies are still only "learning" to interact with synthetic polymers. Nevertheless, since the processes of their interaction take place practically all over the world, this phenomenon – on a global scale – cannot be overlooked in the field of attention of scientific observations.

Such processes, as can be seen from the studies of our colleagues, lead, first, to the formation of fouling on different types of plastics [1, 2]. Secondly, micro-fouling turns out to be able to participate in the biodegradation of polymers, the mechanical strength and integrity of the structure of which changes in water bodies under the influence of ultraviolet radiation from the sun, water movement, and salt aggression occur [3–5].

In this book chapter, we review the published research and our own findings from studies of plastic micro-fouling in a wide variety of water bodies around the world.

2 Plastic in Water and Microorganisms: State-of-the-Art Knowledge

2.1 Plastic and Microorganisms in Water Bodies: Forms of Interaction

In this section let us consider the results of scientific investigations of the last decade devoted to the interaction between the micro-world of water bodies and synthetic polymers – different types of plastics. In doing so, we will consistently focus on the most interesting scientific work performed in this area by researchers from around the world.

Owing to its physical characteristics, plastic debris that is present in the marine environment does form or create the unique environment for placement on its surfaces and for transfer through rivers, lakes, seas, and boundless oceans of a wide variety of different microbial "hitchhikers" (sitters) being attached to its surface and creating a so-called plastisphere, according to the designation given to communities on the surface of plastic [6, 7].

Most plastic waste consists of polyethylene (PE), followed by polypropylene (PP) and polystyrene (PS) [8]. In recent decades, increasing attention is therefore being given to microplastics; the term has been assigned to any plastics or polymers, among which the most prevalent are PE, PP, PET, PVC, PU, PS, acrylic, polyamide, and polyester particles of <5 mm [8, 9], which can accumulate in the depths of the oceans, enter food chains, and cause significant damage to aquatic ecosystems [10].

Past decades have clearly demonstrated that plastic pollution is a global anthropo-ecological concern for the oceans, in particular in the context of continuous growth of plastic concentrations in the aquatic environment. Hence, there is a growing number of publications about the subject, but mostly they highlight that microbial life develops on plastic debris [7, 11, 12] – in the context of the new term "plastisphere," describing the multitude of these communities. In this case, microbial ecotoxicology is rarely integrated into the general canvas of articles. However, microbial ecotoxicology is first following the impacts of contaminants on microbial communities. And, secondly, it determines how many and which microbes are able to take part in the biodegradation of these substrates.

The review presented by Jacquin et al. [11] aimed at introducing the published literature over the previous 15 years. This involves publications related to microbial toxicology, in particular about those microorganisms that turned out to be associated with plastic pollution of the oceans. First, the authors paid great attention to the effects of plastic on marine microbial life as well as to the different functions it realizes in ecosystems. The factors that stimulate the development of biofilms on surfaces of different plastics and the potential role of plastic litter as a vector of pathogenic species of microorganisms are also discussed here. Second, the authors gave critical insights into the extent to which marine microorganisms are able to participate in plastic decomposition. This aspect remains a challenge, in particular with regard to modern biodegradation tests for plastics in the marine environment.

The review highlighted several examples of metabolic pathways of polymer biodegradation. As the conclusion, several questions are formulated concerning gaps in our present knowledge about biodegradation of plastic in the marine ecosystems, as well as on possible directions of new research.

The amount of plastics transported from land into the sea and oceans is extremely large – by 2015 it was estimated as 4.8–12.7 million tons annually [13]. Plastics in the world's oceans can no longer be considered as a limited environmental issue because small pieces of plastics called "microplastic" (<5 mm) were found in the sea and can cover 4.2 million km^2 of the sea surface [14–16]. In fact, a new geologic epoch – the Anthropocene can be designated to mark the ways in which the planet has suffered from the damage caused by plastic pollution [17, 18]. More and more investigations today are focused on the distribution of diverse plastic debris [19, 20] and its toxic effects on the marine fauna [21, 22]. Against this background, a relatively small but growing literature was devoted to microbial ecotoxicology of the marine plastics pollution [23, 24].

Investigations of microbial communities, colonizing plastic surfaces using modern techniques of massive DNA sequencing [7] were introduced only recently. A concept of "plastisphere" with high global significance has been presented to describe the microbial life growing on these surfaces. Zettler et al. also detected that the potentially pathogenic bacteria genus Vibrio can migrate over long distances by floating persistent plastics. Since then, several investigations focused on various marine environments, such as the North Pacific Gyre [25] and the Mediterranean Sea [26]. In the meantime, the scientists described the first steps of colonization of clean plastic until the formation of a mature biofilm [27–29]. All those results offer an interesting insight into the impact of plastic on marine microbial life and ecosystem functions. In view of this fact, a study by Bryant et al. [30] so far using metagenomics tools, showing that plastic-inhabiting microbes present an "enriched gene repertoire" compared to microbes living in the surrounding environment, is of the utmost importance.

For example, irrespective of the polymer type, recent studies emphasized the difference between the bacteria living on plastics and the bacteria in free-living state [25] or on organic particles in the surrounding seawater [29, 31]. Similar observations have been made for fungal communities [32]. Nevertheless, the plastisphere being accumulated at the base of the water column other than the sea surface layer received very little attention until now.

Because of the methodological restrictive guidelines, the majority of studies have been limited to sampling seawater surface with manta trawls, which represents less than 1% of the global load of plastic in the open ocean [33].

Only certain types of plastics made of PE and PP with their higher surface to volume ratio such as rigid plastic articles and bundled fishing nets and ropes have the capability to remain for a very long time at the surface of the oceans [34]. Most other floatable plastic such as films or smaller pieces, fouled by organisms, tend to sink to the bottom and accumulate in sediments [35, 36]. Very limited information is available concerning the composition of microbial communities on plastic items sampled from the seafloor [37]. The photoautotrophic bacteria such as the cyanobacteria of the genera *Phormidium* and *Rivularia* [7, 29, 30] dominate the subsurface plastisphere communities. At the same time, the main microbiome of the seabed and the subsurface plastosphere seems to include several common taxonomic groups: Bacteroidetes (Flavobacteriaceae) and Proteobacteria (Rhodobacteraceae and Alcanivoracaceae) [7, 29, 30, 38].

In parallel to studies on plastics directly sampled at sea, other studies focused on the successive colonization steps of new plastics incubated in marine conditions. At sea, plastics are rapidly covered by the "conditioning film" made of inorganic and organic matter which is then rapidly colonized by bacteria mainly represented by members of Gammaproteobacteria and Alphaproteobacteria [28]. With time, members of Bacteroidetes appear to be more abundant [39].

Such substratum properties as hydrophobicity, crystal structure, glass transition temperature, modulus of elasticity, roughness and melting temperature may play a role in the selection of a bacterial community in the early stages of colonization [40], but they become probably less influential when the biofilm becomes mature [29]. The successive growing and maturation phases of biofilm formation, already described for other surfaces and substances (such as rocks, steel, acryl, glass, and algae), were also observed for plastics of different compositions [28].

Biofilm developments were observed during several weeks in seawater on PE-based plastic bags [27], polyvinyl chloride (PVC) [41], polyethylene terephthalate (PET)-based plastic bottles [42], or polystyrene (PS) coupons [43]. Plastics were also rapidly colonized by microorganisms in marine sediments [44]. It is essential to note that clear differences in bacterial abundance, diversity, and activity were found between non-biodegradable and biodegradable plastics [45, 46].

Faster colonization by active and specific bacteria were found after six weeks on Poly (3-hydroxybutyrate-co-3-hydroxyvalerate – PHBV) and pre-oxidized PE-based oxodegradable polymers (OXO) in comparison with non-biodegradable PE polymers [45, 46]. Longer-term studies (carried out over a 6-month to 1-year period) also showed differences in biofilm formation and maturation according to the polymer type, i.e. PE, PP, PET, PC [38, 47]. Studies have revealed that fungi often form biofilms on plastic surfaces [48]. The structure of these films was mainly dominated by Chytridiomycota, Cryptomycota [32], and Ascomycota [32, 38, 49].

At the time the review was published, it was already becoming clear that marine microorganisms that compose the plastisphere are known to play a key role in the biogeochemical cycles in the oceans [50]. Approximately one-half of oceanic primary production is channeled via heterotrophic bacterioplankton into the microbial loop [51]. Investigation by Dussud et al. [29] has shown that the activity of heterotrophic bacteria living on plastics was 43–48 times higher than that of the free-living fraction. These results were obtained in the framework of a study on colonization of new plastics incubated at sea for a relatively short period (45 days). Similar methodologies applied to plastics that had spent several years at sea would be necessary to evaluate how much large amount of plastic and the accompanying plastisphere influence the biogeochemical carbon cycle in the oceans.

Most of the studies aiming to characterize the plastisphere mentioned that Cyanobacteria were overrepresented on plastics compared to the surrounding free-living and organic particle-attached fractions. The relative importance of photosynthetic activities that Cyanobacteria living on plastic have on global pelagic primary production is still unknown.

Coupling primary production and measurement of the heterotrophic production data obtained from plastics over large temporal and spatial scales will be necessary to obtain a better view of the role of the “plastisphere” on carbon cycling in the oceans. Microorganisms are also involved in the carbon cycle. Cycles include nitrogen, sulfur, iron, manganese, chromium, phosphorus, calcium, and silicate cycles, which may also be impacted by the presence of plastic at sea [52].

Interest has been raised about opportunist pathogen dispersed on plastics, such as animal or human pathogenic *Vibrio* sp. [7]. Putative pathogens of fish (*Tenacibaculum* sp.) and of invertebrates (cyanobacteria *Phormidium* sp. and *Leptolyngbya* sp.) were found to be more common on plastic compared to surrounding seawater [29]. Infusoria *Halofolliculina* spp. that is supposed to be pathogenic for coral was found to be abundant on some western Pacific plastic debris [53].

Some toxic eukaryotic species were also mentioned by Debroas et al. [25] at low abundance (<0.04%), but might be regarded as hitchhiker organisms.

Nevertheless, caution should be taken since the 16S rRNA metabarcoding approach used in all these studies was not an appropriate method for describing bacterial virulence. The recent coupling of the 16S rRNA metabarcoding technique with the detection of virulence-associated genes may be an interesting option to address this question [54]. Though in the meantime, pathogenicity evidence on marine animals in relation to the plastisphere has never been proven, further research will be required before publicizing alarmist conclusions on the possible responsibility of plastic debris as a vector for the spread of disease-causing organisms. However, apart from these potential threats, microplastics colonized by pathogens may also pose threats to humans who are exposed to contaminated beach and bathing environments [55]. Evidence is still missing to determine whether plastic debris could lead to the spread and prolonged persistence of pathogenic species in the oceans.

As a rule, factors driving the plastisphere composition are complex. Most of them are spatial and seasonal, but are also influenced by the polymer type, surface properties, and size. For example, plastisphere communities studied in different polymer types floating in the North Pacific and North Atlantic reflected first their biogeographic origins and to a lesser extent the plastic type [56]. Communities colonizing plastics along an environmental gradient are formed firstly by the water salinity conditions and secondly by the plastic type (PS and PE) [31]. Inversely, another study based on a large number of microplastics sampled in the western Mediterranean Sea showed no effect of geographical location (including coastal and open ocean samples) or plastic type (mainly PE, PP, and PS) on the prokaryotic cenoses community composition [28].

The physical properties of plastic offer a unique habitat that contributes to the long-distance transport of diverse microbial hitchhikers attached to its surface [6, 7]. A vast range of other phyla, including Arthropoda, Annelida, Mollusca, Bryozoa, and Cnidaria have conferred on plastics the role of vector for the transfer of organisms. Some of them are cataloged as invasive alien species [28]. For instance, plastic debris with tropical biota including corals was detected in the Netherlands [57], and Southern Ocean Bryozoans were observed in Antarctica [58]. Interactions between micro- and macro-organisms, their substratum and their surroundings are needed to better predict the ecological consequences of microplastics transported through the global oceans.

In their review, Jacquin et al. [11] lead us to the understanding that the knowledge available is insufficient to formulate clear ideas about the impact of plastic on marine microbial life and ecosystem functions. The growing number of plastisphere studies represents new knowledge about microbial biofilms on plastic in the seas and oceans. However, a complex network of interactions between films, plastic and the external environment is still a subject of discussion. However, the authors of the review express the hope that a clearer understanding will come from more extensive research covering vast amounts of material from different locations, as well as with a more detailed description of the physical and chemical properties of polymers themselves in the marine environment.

Another emerging major challenge for scientists is the possible role of microorganisms in the biodegradation of plastic in the sea. Very recently, an excellent comprehensive review concluded that “current international standards and regional test methods are insufficient in their ability to realistically predict the biodegradability of carrier bags in marine environment, due to several shortcomings in experimental procedures and a paucity of information in the scientific literature” [59].

The capability of microorganisms to biodegrade plastic was reported frequently in past years for numerous bacterial strains [60] and fungi [61]. However, most of the studies were conducted in terrestrial conditions [62–64] whereas very few studies so far provide new insights into the marine conditions [65, 66]. Moreover, most of these studies were based on the selection and testing of single strains in laboratory conditions, which is very far from environmental conditions. Jacquin et al. [11] identify knowledge gaps about the potential for biodegradation of plastics by marine microbes. They are also trying to determine possible ways and directions of further research in this field.

Biodegradation of plastic is a process that results in total or partial conversion of organic carbon into biogas and biomass associated with the activity of a community of microorganisms (bacteria, fungi, and actinomycetes) capable of using plastic as a carbon source [67]. Depending on the respiratory conditions (aerobic/anaerobic) and the microorganisms involved, the biogas will be different (CO2, CH4, H2S, NH4, and H2) [68].

Some microorganisms, including bacteria and fungi, have the ability to destroy or degrade the structure of plastic polymers. Several review papers have presented the updated list of plastic degraders [66, 68–70]. For example, species *Arthrobacter, Corynebacterium, Micrococcus, Pseudomonas, Rhodococcus,* and *Streptomyces* have been identified as significant bacterial taxa capable of using plastic as their sole source of carbon and energy in laboratory experiments under controlled conditions. It is now proposed to update the current list of microorganisms proven to have the abilities to degrade different types of synthetic polymers under laboratory conditions. For PE it is *Brevibacillus borstelensis, Bacillus Weihenstephanensis, Comamonas* sp., *Delftia* sp., *Stenotrophomonas* sp., *Achromobacter xylosoxidans, Bacillus* sp. YP1, *Enterobacter asburiae* YT1, *Bacillus amyloliquefaciens, Bacillus pumilus* M27, *Kocuria palustris* M16, *Lysinibacillus xylanilyticus, Bacillus mycoides, Bacillus subtilis, Pseudomonas aeruginosa* PAO1 (ATCC 15729), *P. aeruginosa* (ATCC 15729) 692), *Pseudomonas putida* (KT2440 ATCC47054), *Pseudomonas syringae* (DC3000 ATCC10862), *Brevibacillus parabrevis, Acinetobacter baumannii, Pseudomonas citronellolis, Bacillus sphaericus, Rhodococcus rubannii, Aspergillus versicolor, Aspergillus* sp., *Chaetomium* sp., *Aspergillus flavus, A. niger*, *Penicillium simplicissimum, Lasiodiplodia theobromae, Paecilomyces lilacinus, P. pinophilum*, *Gliocladium virens, P. chrysosporium, Aspergillus glaucus*. For PET: *Bacillus amyloliquefaciens, Nocardia* sp., *Ideonella sakaiensis, Humilicas insolens, Pseudomonas mendocina, Thermobifida fusca* DSM 43793, *Penicillium citrinum, Thermomonospora fusca, Fusarium oxysporum, Fusarium solani*. In turn, for PHB: *Crupriavidus* sp., *Marinobacter algicola, Schlegella thermodepolymerans, Caenibacterium thermophilum, Acidovorax* sp. Strain TP4, *Pseudomonas stutzeri, Leptothrix discophora, Alcaligenes faecalis, Comamonas acidovorans* YM1609, *Comamonas tetsteroni, Pseudomonas lemoignei, Ralstonia pickettii, Pseudomonas fluoresonas cens* (YM1415), *Aspergillus niger*. For PHBV, the list of possible biodegraders is much shorter: *Clostridium botulinum, Clostridium acetobutylicum, Streptomyces* sp. SNG9, *Pseudomonas lemoignei* and *Paecilomyces lilacinus*. Finally, for PS, the list of strains as agents for biodegradation includes: Strain TM1 and ZM1, *Bacillus subtilis, Staphylococcus aureus, Streptococcus pyogenes, Exiguobacterium* sp., *Bacillus* sp. NB6, *Pseudomonas aeruginosa* NB26, *Exiguobacterium* sp., *Microbacterium* sp. NA23, *Paenibacillus urinalis* NA26, *Rhodococcus ruber, Pseudomonas putida* CA-3 (NCIMB 41162), *Bacillus* sp. STR-Y-O, as well as Mixed microbial communities (*Bacillus, Pseudomonas, Micrococcus,* and *Nocardia*). All strains are given according to the materials of Jacquin et al. [11].

Biodegradation is considered to occur after or concomitant with physical and chemical degradation (abiotic degradation), which weakens the structure of polymers as revealed by roughness, cracks, and molecular changes [71]. The change in plastic properties due to abiotic degradation is called “aging” and depends on several factors such as ultraviolet exposure, water mobility, changes in temperature, and mechanical abrasion, as well as chemicals that increase the rate of degradation by oxidation or destruction of the length of the polymer chain.

Biodegradation processes can be generalized to four main stages [72]:

- Bio-deterioration relates to the biofilm growing on the surface and inside the plastic, which increases the pore size and provokes cracks that weaken the physical properties of the plastic (physical deterioration) or releases acid compounds that modify the pH inside the pores and results in changes in the microstructure of the plastic matrix (chemical deterioration).
- Bio-fragmentation corresponds to the action of extracellular enzymes (oxygenases, lipases, esterases, depolymerases, and other enzymes that may be

as diverse as the large spectrum of polymer types) released by bacteria colonizing the polymer surface. These enzymes will reduce the molecular weight of polymers and release oligomers and then monomers that can be assimilated by cells. These enzymes reduce the molecular weight of polymers and release oligomers, followed by monomers, which can be digested by cells.

- Assimilation allows oligomers of less than 600 Da to be integrated inside the cells to be used as a carbon source, thus increasing the microbial biomass.
- Mineralization is the ultimate step in the biodegradation of a plastic polymer and results in the excretion of completely oxidized metabolites (CO2, N2, CH4, and H2O).

In early 2020 Caruso [73] presented an extensive overview devoted to microbial colonization of various substrates in the marine environment (an overview of current knowledge and new research topics). Among the substrates considered by the author, much attention is paid to synthetic polymers as well. Because of its physical properties, plastic litter represents a unique habitat – both for placement on its surfaces and for transfer through vast spaces of oceans of a wide variety of different hitchhikers from among microorganisms. Most of the plastic litter accumulated in the world's oceans is by now represented by PE, followed by PP and PS [8]. The nature and history of the distribution of plastics particles in the aquatic environment are determined by hydrodynamics (circulations and turbulence of water body) [74–76]. Also, the variability of plastic density contributes to the deluge of aquatic plastic and its sedimentation. The particles are also discovered to be deluged depending on the polymer nature of plastics. Mostly plastic has a higher density than seawater [77]. However, PE and PP are plastics with low density, and therefore show or have generally positive buoyancy. This distinguishes them from PVC, PS, polyester, and polyamide – as plastics of higher density, they proved, in a logical way, to be more prone to deluge [9, 76, 78]. The colonization of organisms on the plastic surface increases the total weight of particles. It also results in accelerating their sinking into deeper pelagic waters and to bottom sediments [9, 27, 76, 79, 80]. In contrast, plastic particles initially having high density can be redistributed within layers of the water column due to turbulence of water masses [74–76].

Cai et al. [81] investigated the effects of bacterial adhesion to plastics associated with substrate properties such as surface charge, hydrophobicity, hydrophilicity, roughness, and hardness. They concluded that it is the hardness of the substrate that is the key factor for the colonization of plastic.

In the structure of microbial biofilms developing on plastic, diatoms are often found which is one of the most common types of unicellular eukaryotes [43, 49, 82, 83]. Diatoms are considered among the first colonizers of plastic surfaces in the sea, and as the most probable pioneers for microbial colonizers of heterotrophic nature coming after them [41, 84, 85]. Diatom species are required for aggregation of the related prokaryotic epibiont communities [86]. Such bacterial communities, also attached to plastic, are often dominated by *Roseobacter, Alteromonas*, and *Pseudoalteromonas* species.

In turn, flavobacteria from the genera *Tenacibaculum* and *Polaribacter* are the main colonizers of diatoms within detritus [87].

Bacteria living on plastic are characterized by a high concentration of alpha- and gamma-proteobacteria, whereas seawater is mostly domain of alpha-proteobacteria (largely *Pelagibacter* sp.) [26].

The current knowledge of the microbial biofilm formation on marine plastic debris is mainly limited by its composition and structure. Prospective research will focus – to a greater extent – on the study of the interactions of microbes and plastic. Plastisphere communities in marine environments are potential microbial degraders of plastics [88]. In this light, biofilms can be an environment for finding new microbes involved in the biodegradation of polymers and genes involved in this enzymatic process.

Review by Caruso [73] provides information on biofilms formed on different polymers: PVC, PE and PET, PS, PU and acrylic.

2.1.1 Polyvinylchloride (PVC)

Plates from PVC have been used as a substrate to study the resumption of the *Balanus amphitrite* shell population in the subtropical waters of Hong Kong [89]. To avoid deposition on plates of invertebrate larvae, these plates were placed in a mesh nylon sac. *B. amphitrite* larvae preferred plates covered with biofilms that formed under littoral area conditions. The crayfish of this species inhabit the coastal cliffs. Probably their larvae reacted to specific biochemical signals produced by the microbial biofilms developed on the littoral. The results of the research strongly support the site-specific variation in marine biofilms developed under different biotopic conditions on the same substrate from PVC.

On PVC sheets arranged in wooden holders, a study was performed concerning the constitution of the biofilm formed on the basis of colonies of heterotrophic bacteria based on the heterotrophic bacteria in the coastal waters of the Laccadive Sea (port of Thoothukudi (former name Tuticorin), India). Sampling for fouling analysis was carried out for 7 days: 0.5, 1, 2, 4, 24, 48, 72, 96, 120, and 144 h from the beginning of the experiment [90]. Marine bacteria belonging to genera *Pseudomonas, Enterobacter, Aeromonas, Cytophaga*, and *Flavobacterium* were the first microbes to colonize the surface of PVC – this was discovered after the first 30 min. After 48 h, Gram-positive bacteria from the genera *Micrococcus* and *Bacillus* were also found in the biofilm structure. Then, between 48 and 96 h, the films developed both as gram-negative and gram-positive groups. After 96 h, the biofilm consisted only of gram-positive bacteria.

Briand et al. [91] studied factors that influenced the microbial community of biofilms developed on PVC and 4 different antifouling coatings. Experiments were carried out on two sites, on the coast of France: in one case the site was under eutrophic conditions (Lorient, Atlantic coast), in the other – under mesotrophic conditions, but strongly contaminated (Toulon, north-west of the Mediterranean Sea). The change of seasons did not have a significant impact on the composition of biofilms. However, significant factors were surface type, as well as habitat features: high temperatures, lead ions and salinity at the Toulon site, and at the Lorient site – nutritious substances and dissolved organic carbon. HTS (high throughput sequencing), using 454 pyrosequencing, showed that gamma- and alpha-proteobacteria were predominant in the prokaryotic communities, as well as Bacteroidetes. The percentage of Bacteroidetes decreased as whole due to the presence of pyrithione as an antifouling coating. It is important to emphasize that all surfaces were dominated by the small *Amphora* spp. and *Navicula* spp. diatoms.

2.1.2 Polyethylene (PE) and Polyethylene Terephthalate (PET)

To verify the effects of habitat factors on a bacterial community capable of developing on the PET surface in the North Sea, bottles made of this polymer were exposed there; in particular near the coastline of Britain, the effect that yearly seasons, geographical location and type of substrate have on the fouling of plastic substrates was monitored during experiments [42]. After 6 weeks of exposure, bacterial biofilms included representatives of Bacteroidetes, Proteobacteria, and Cyanobacteria, along with eukaryotes from among Bacillariophyceae and Phaeophyceae. At the same time, the similarity of biofilms and communities of surrounding water did not exceed 10%. The most significant differences between prokaryote biofilm communities were observed during seasonal changes, and giving consideration to the geographical location of the stations, which could reflect the influence of local physical and chemical conditions.

In continuation of these studies in the North Sea [49], the structure of microbial communities on the surface of PET-bottles has been compared to that of seawater communities (separately for free-living (0.22–3 μm) and particle-related (>3 μm) fractions), as well as with the cenoses of biofilms developing on glass, whereas the 16S rRNA gene sequence analysis has been applied. The prokaryotic biofilms cenoses on the surface of plastic showed species-specificity and high dependency on the season and substrate type. Once again, a large difference in structure was shown between prokaryotic cenoses on PET and in the sea water, but not between biofilms on PET and those communities of prokaryotes that developed on glass or particles suspended in the water. Microbial cenoses developed on plastic, glass, and suspended particles turned out to be very similar to each other. This suggests that PET does not play a decisive role (as a specific substrate) in the formation of the biofilm structure on its surface.

In the biofilm communities on PET, the most widespread and diverse bacteria belonged to the genus Bacteroidetes with the families Flavobacteriaceae, Cryomorphaceae, and Saprospiraceae. In turn, the genus *Tenacibaculum* (Bacteroidetes, Flavobacteriaceae), *Crocinitomix*, and *Owenweeksia* have always prevailed from among the Rhodobacteraceae. In addition, microbes belonging to Sphingobacteriales (notably the Saprospiraceae) and Myxococcales have been found, along with members of the genus *Verrucomicrobia* (subdivision 1 of Verrucomicrobia) and the genus *Phormidium*. The order Sphingobacteriales are successful members of the biofilm's community, probably because of their ability to produce exopolysaccharides and to collect biofilms to generate energy and carbon. The Myxobacteria are known to produce a polymer substance enhancing their gliding and swarming properties, as well as complex bioactive secondary metabolites and hydrolytic enzymes that give them a competitive advantage in resource limitation, for example, in a biofilm environment. Members of the Sphingobacteriales and Myxococcales have also been identified as biofilm components of plastic debris collected from the North Atlantic [7].

De Tender et al. [38] identified a core group of 25-unit OTUs belonging to the Proteobacteria, Bacteriodetes, and *Verrucomicrobia* types on PE-debris fragments collected in the North Sea. However, it has not yet been proven whether these "core organisms" are specific to the habitat on polyethylene, or whether they may develop en masse on other types of synthetic polymers, present at sea.

Misic and Harriague [92] conducted an experiment with fouling the PET-bottles by microbial communities over 34 days. In doing so, they studied how changes in the physical, chemical, and biochemical properties of seawater, as well as temperature and lighting influenced the colonization. They showed that an increase in the temperature of the medium and limitation of lighting were able to change the biofilm communities and enhancing the role of prokaryotes. At the same time, their experiment showed that during summer conditions the role of phototrophic microbes is increasing.

2.1.3 Polystyrene (PS)

To test the hypothesis about the possible effect of bacterial supernatates on the attachment of filter-feeding mollusks larvae onto substrate, PS plates were used. It was observed that attachment of the crustacean larvae (balanoids) is effectively hindered by films of Deleya marina (during the stationary phase of growth), but the culture *Alteromonas macleodii* and *Pseudomonas fluorescens* did not show similar effects [93]. The process of active attachment of the Marine Bacterium *Pseudoalteromonas* sp. D41 to multicell PS-plates in sterile natural seawater was observed during an experiment by Leroy et al. [94]. They recommended this method for primary screening of the efficacy of antifouling agents in the early stages of marine biofilms formation.

During experiments by Chiu et al. [95], PS Petri dishes were also chosen as a substrate for colonization. The authors studied the composition and biomass of mixed algo-bacterial biofilms according to two seasons of the year (winter and summer), focusing on differences in temperature and salinity. For over 20 days biofilms were formed in the laboratory using natural seawater. The largest biomass (10–46 μg of dry weight per cm^2) was formed in summer, with salinity of 34 ppt. This was significantly higher than in winter, at 20 ppt. During summer, the composition of communities of both prokaryotes and diatoms varied significantly depending on salinity. In winter, the composition of cenoses was significantly influenced by temperature.

2.1.4 Polyurethane (PU)

Another plastic material tested as an artificial substrate in the study of microbial colonization is the polyurethane. Xu et al. [96] investigated the colonization of polyurethane foam shells with periphytonic infusoria (developing as part of fouling) in marine ecosystems. At the same time, they identified 27 species using the modified observation method on glass slides, as well as the silver impregnation method. Despite the similarity in the species composition of colonies formed by infusoria, they showed significant differences in terms of structural and functional parameters – between polyurethane foam and ordinary glass slides. Species diversity, uniformity, and colonization rate were distinctly higher, but the time to reach 90% of the equilibrium number of species was less on the surface of PU-foam than on bare glasses. However, the results of these experiments showed that PU foam is a substrate on which periphyton infusoria form colonies, in general, faster, more uniform, and more diverse than on ordinary glasses.

Jeong et al. [97] used PU sponges or sponges made of PU to extract lithium from seawater. At the same time, they studied microbial communities detected on polymer products (adsorbents) of three different configurations: spheroidal (2 μm), discoidal (2 mm), and rachis-like (2 mm). All adsorbents were in the seawater for 30 days. The primary colonization of the substrate was produced by gamma proteobacteria (gamma-Proteobacteria) belonging to the genera *Vibrio, Alteromonas,* and *Pseudoalteromonas*. Observations using confocal microscopy and scanning electron microscopy (SEM) showed that the distribution of bacteria on PU surfaces was essentially dependent on the form of adsorbents. The composition of bacteria is also highly dependent on the pore size: on spheroidal (2 μm) adsorbents lived only *Alteromonas* cells, and on other forms *Vibrio* bacteria were detected – in the later stages of the biofilm development it was replaced by *Alteromonas* cells.

2.1.5 Acrylic

Poly(methyl methacrylate) (PMMA), also known as acrylic glass or plexiglass, is a transparent thermoplastic material, quite often used in sheet form as break-resistant alternative to glass. Lee et al. [98] isolated from the surface acrylic coupons (plates) submerged in the sea near the Goje Island (Korea), a total of 115 bacteria, 70 of which were identified according to their 16S rDNA sequences. These biofilm structures were dominated by Alphaproteobacteria, followed by gamma-proteobacteria, gram-positive bacteria with low GC-vapor content, gram-positive bacteria with high % GC, and bacteria belonging to Cytophaga/Flexibacter/Bacteroides groups.

The authors of the experiment proposed bacterial isolates as standard strains for testing new antifouling agents. They can also be used to attract the larvae of invertebrate animals when they settle on substrates – this is important for successful mariculture. After 6 days of experiment, a microbial film formed on the plates, in later stages covered with young individuals of serpulids and balanids. Six months from the beginning of the experiment, the surface of the plates was thickly covered by adult Bryozoans, mussels, and macroscopic algae.

The experiments of Dobretsov et al. [99] examined how the color of the substrate influenced the settlement of micro- and macrofouling communities – on acrylic glasses and tiles. The time of exposition of samples in seawater was 5, 10, and 20 days. On black and white substrates, the density of bacteria was almost the same, excluding the 10th day, on which the number on black substrates was higher. 454 pyrosequencing of 16S rRNA genes of bacteria from the surfaces of white and black substrates showed that alpha-Proteobacteria and Firmicutes formed the body of the communities.

Analysis of characterizing and differentiating groups (SIMPER) showed that bacterial phylotypes (uncultivated gamma-proteobacteria, *Actibacter, Gaetbulicola, Thalassobius* and *Silicibacter*) and diatoms (*Navicula directa, Navicula* sp. and *Nitzschia* sp.) characterized the differences between fouling cenoses formed on white and black substrates.

Mejdandžić et al. [100] used plexiglas plates in the Northwest Adriatic Sea: they observed the succession of bottom microphytes in the formation of biofilms. The qualitative composition and quantitative structure of the cenoses of diatoms and bacteria were studied on plexiglass plates immersed in the sea for 30 days. For this purpose epifluorescence was combined with electron microscopy. Among the pioneer species that colonized the substrate were planktonic diatoms *Dactyliosolen fragilissimus, Proboscia alata, Thalassionema nitzschioides,* and *Leptocylindrus danicus*. A week later from the beginning of the experiment, benthic diatoms *Licmophora, Cocconeis,* and *Achnanthes* became widespread among colonizers, which replaced plankton forms. After a month of exposure, *Pseudo-nitzschia pseudodelicatissima* was the dominant species – the density of colonial species positively correlated with the temperature. These experiments also produced results to understand the mutual influence of diatoms and bacteria in the formation and growth of biofilms.

Microplastic should be considered as a persistent contaminant deposited in the natural environment for a long time. Studies of aquatic ecosystems around the world revealed how severe and widespread the microplastic pollution is. Microphytes, as some of the most important producers of primary organic substance in aquatic ecosystems, can suffer from contamination by microplastics. In turn, this causes deformation of water trophic networks. However, there is not much known about the toxic effects of plastics on microalgae. The purpose of the review, performed by Prata et al. [101], was to identify the effects of plastics on microphytes (based on available literature) as well as to establish gaps in this area of expertise. It is obvious that microplastic particles have some influence on cell growth, chlorophyll content, photosynthesis activity, and release of active oxygen forms (ROS).

Several factors influence the toxicity of microplastic particles, including their concentration in the environment, type of polymer, size, availability of additives (staining, flavoring, etc.), chemical composition surface, and electric characteristics. The type and size of polymer particles also refer to factors capable of having a toxic effect on microphytes. Lagarde et al. [102] reported inhibition of *Chlamydomonas reinhardtii* cell growth in the presence of polypropylene (PP) particles, but not high-density polyethylene (HDPE). The type of polymer and particle size also have a direct impact on their distribution across the water column and may lead to their sedimentation [103] or buoyancy [104], modulating exposure of these particles in algae environments. Typically, smaller microplastic particles are more toxic to microalgae. For example, 0.05 μm polystyrene (PS) particles caused more pronounced decreasing in cell density in the population, as compared with particles 0.5 and 6 μm [105], and polyvinyl chloride (PVC) adversely affected growth, chlorophyll rate, and photosynthesis where a particle size was 1 μm, but not 1 mm [94].

The polymer itself and the size of its particles affect the chemistry of the surface, changing its toxicity [107]. The positively charged microplastic caused, in general, a more toxic effect on microphytes than the negatively charged [106–108]. Such is the likely effect of anionic cellulose produced at the cell wall surface, which contains carboxyl and sulfate group. They repel microplastics with a negative surface charge. But at the same time, positively charged particles are adsorbed. That is achieved by electrostatic interaction, hydrogen bonding, and hydrophobic interactions – depending on the morphology of algae [106].

In the same way, the adsorption of substances on its surface is associated with the properties of microplastic. For example, polyethylene does not affect the toxicity of copper [103], but increases toxicity of such medicines as procainamide and doxycycline [109]. Adsorption of chemicals to microplastics may increase their exposure to the environment [110]. At the same time, most microplastic particles used for the purpose of toxicity analysis are marked with fluorescent tags for better identification and quantitative accounting. For example, this method is used in fluorescent microscopy and fluorometry. Consequently, observed toxicity test results may be affected, so long as the toxicity of the tags themselves is not taken into account. Anyway, there is a need to investigate how the properties of microplastics affect its toxicity to microalgae, and how they interact with other substances in the environment.

The authors of the review also note that most concentrations tested in the available literature issues or publications substantially exceed those found in the environment. Most of these studies show little or weakly fixed level of microplastics influence on microalgae. The present concentrations of microplastics in the environment should not cause toxic contamination. But even in the current situation, microplastics are able to destroy the population regulatory mechanisms, reducing the availability and absorption of nutrients, or reducing the population of species that consume microphytes. Thus, there is a need to expand the range of studies that encourage understanding of the microplastics impacts on microalgae: on their adaptation strategies, population dynamics, and the properties of microparticles of polymers affecting their toxicity.

On the surface of plastic debris that was transported from lands into sea and oceans, organisms of different groups are able to live. Potentially harmful organisms could be transferred by them as well. Such organisms have the potential to travel through the water flows on plastic items. Allochthonous species may also appear among them. In addition, plastic transports toxic compounds adsorbed to it. Both those and others are spread by plastic over the water areas and cause harmful effects on the ecological environment and human health.

Casabianca et al. [111] examined plastic samples floating on the sea surface using qPCR molecular analysis to quantify attached microphyte taxa, including harmful. At the same time, 42 samples of multidimensional plastic obtained using manta net of 330 μm mesh size, as well as by manual methods in different areas of the Mediterranean Sea were taken into account.

Such examinations of plastic samples were done for the first time. Obtained informational results are important for improving water plastic monitoring practices and show how the community of colonizers of plastic surface in aquatic environments can cause adverse effect of plastic litter through the transfer and possible settlement of alien and toxic species capable of entering trophic networks in new locations. The diatoms proved to be the most numerous group of plastic colonizers (at a maximum of 8.2×10^4 cells/cm^2). At the same time, the dinoflagellates numbered maximum 1.1×10^3 cells/cm^2. Especially widespread harmful species were diatoms *Pseudo-nitzschia* spp. (at least 12 toxic species) and dinoflagellate *Ostreopsis* cf. *ovata* – with a maximum abundance of 6,606 and 259 cells/cm^2, respectively. Other toxic microphytes, including the allochthonous *Alexandrium pacificum*, were observed among 1–73 cells/cm^2. An important result of the study was to establish a direct correlation between the abundance of harmful microphytes colonizing the surface of plastic and their production of toxins. Concentrations of potentially toxic substances (for different families of toxins) produced by microphytes-colonizers were 101–102 ng/cm^2 on plastic fragments. Levels of adhesion to the plastic substrate in several species (22 strains) of microphytes (0.3–1.8 days $^{-1}$) were also studied, showing the ability of these species to rapidly colonize available plastic surfaces.

Among the plastic debris polluting the seas and oceans of the World, microplastic particles (size <5 mm) are of particular interest. They are the ones that are often swallowed accidentally by marine animals who mistake them for food. This way such particles enter the food chains. They can have toxic effects on living organisms, or might simply accumulate in their digestive system, leading to starvation [112, 113]. Microplastics particles are washed out into the sea with domestic wastewater, or are otherwise produced as a result of plastic items degradation caused by solar UV radiation and waves activity. Microplastics have been found already throughout the water column of seas and oceans, from the surface to the top layer of bottom sediments. However, the ways of spreading these microparticles through marine areas have not yet been studied enough.

The studies by Long et al. [114] have shown that microplastic particles easily constitute aggregates with microalgae. These structures change the rate at which microplastic passes through seawater. In laboratory tests, polystyrene (PS) balls were lowered to the bottom at a rate of 4 m/day. In turn, as part of clusters of microphytes, these particles dropped down at a rate of several hundred meters per day. Funded by the EU MICRO1 project, these studies focused on the role played by microphytes in transporting plastic from the sea surface to the bottom. Microphytes produce adhesive substances in the form of lumps by colliding with one another. These units can stick to other particles, including microplastic. They eventually sink to the bottom, captivating microplastic with them. Researchers from the group of M. Long developed a flow tank cistern with roller systems to simulate what happens to microplastic in the sea. Roller-containers rotated on the roller table, simulating varying turbulent processes and allowing microphytes to collide and form aggregates. In turn, these containers were located on a stream of water that contained microgranules that attached themselves to the aggregates.

Three microphyte cultures were used in the experiments: *Rhodomonas salina, Chaetoceros neogracile,* and a mixture of both species. At first, each culture was placed to the individual roller tanks, which rotated to form aggregates. Seawater, which contained PS balls with 2 μm in diameter, was then conveyed in each container. Then the process was stopped, and 50–100 units were extracted from each container to measure their size and sinking rate in a glass column with saltwater. These aggregates were also broken up to study the content of microgranules. In parallel, the sinking velocity of granules as such was measured. The *C. neogracile* sinking velocity decreased from 473 m/day (without granules) to 165 m/day with granules. Thus, the PS microspheres decreased the sinking rate of the aggregates, increasing their buoyancy. In the case with *R. salina* culture, the sinking velocity of aggregates with plastic increased from 76 to 125 m/day. The mixed culture of both species formed aggregates sinking with microballs somewhat slower than without them (122 vs 144 m/day). According to the researchers, these changes in the rate of sinking of aggregates can be explained as well due to different physical, chemical, and biological characteristics of microphyte species. In particular, the lightweight aggregates of *R. salina* had large pores in their structure, which included 18 times more PS microballs (per 1 mL) compared to aggregates *C. neogracile*. Microgranules are heavier than *R. salina* aggregates themselves, so their inclusion increased the sinking velocity.

The results of the study suggest that microalgae aggregates may be responsible for transporting microplastics into the deep ocean. They also explain the reason that other studies have found surprisingly little microplastic in surface water and its high concentrations on the seabed. Against this background, it is alarming that sea snow (conglomerates of slowly decomposable organic substance involving microalgae aggregates and bacteria colonies that fall from the ocean's surface down to the sediment), which is a major food source for many marine organisms, can be at high risk of microplastic contamination.

Another, later study by Long et al. [115] also refers to the interaction of microphytes with microplastic particles. The authors strove to understand what is the microplastics' further destiny in the sea, and how the marine ecosystem is affected by microplastic. Thus they continued with the examination of interaction between microplastics and phytoplankton. Such processes may affect the availability of microplastics as food to marine animals and also generally affect the destiny of microplastic particles in the water column. In this paper, the authors evaluated the ability of planktonic microphytes to form heteroaggregates (compound aggregates) with polystyrene (PS) microparticles depending on the type of microphytes and their physiological states. PS microspheres (2 μm in diameter, 3.96 μg/L) were added to the cultures Prymnesiophyceae, *Tisochrysis lutea*, dinoflagellates *Heterocapsa triquetra* and *Chaetoceros neogracile* – at culture growth stage.

By using a flow cytometric approach, they identified and quantified PS microspheres, which allowed to control their distribution in cultures, as well as to distinguish between free microspheres and those included in the heteroaggregates. For the *C. neogracile* culture, the formation of such heteroaggregates was observed during the stationary growth phase. In *T. lutea* and *H. triquetra* cultures, particularly high "losses" of microplastics attached to the bulb walls were observed. These "losses" were directly related to the age of cultures for both species. However, no influence of PS microspheres on the physiology of microphytes (in terms of their growth and chlorophyll fluorescence) was observed. These results once again demonstrate the capabilities of single-cell phytoplankton and the organic substance produced by it to interact with microplastics. These interactions can affect the distribution of microplastics in the water column and its bioaccessibility.

Michels et al. [116] also examined the aggregation of microplastics with biogenic particles and the effect of that process on its sinking rate – and, in general, on its destiny in the sea. By this time, laboratory experiments had already shown that fragments of microplastics, incorporated into aggregates from planktonic microphytes, could a) increase or decrease the sinking velocity of these aggregates, depending on their microphytic composition and the average density relative to the density of microplastics [114], or b) increase the sinking velocity of microplastics. The latter is more common when MPs fragments aggregate with biogenic particles with negative buoyancy. For example, if MPs aggregates with relatively high-density particles – diatoms and their dead shells – aggregation can lead to an increase in MP sinking rates. Typically, MP fragments enter the ocean through its surface. The ballasting described is likely capable of causing rapid sinking of these fragments from the surface layer and facilitates the distribution of MPs to deeper layers of the water column. In turn, the sinking of MPs by such ways may explain why concentrations of these fragments near the sea surface were less than expected [33]. Experiments by J. Michels and colleagues demonstrated that aggregates collected from MPs and biogenic particles can maintain structure stability for several days. This suggests that such aggregates can "survive" the long-lasting sinking to great depths without breaking apart before they reach the bottom of the sea. Thus, apart from the potentially possible transport of MPs through food networks, the sinking in aggregates – quite possibly – should be considered as the main transport route on which MP gets into deep sea sediments where they were observed [117–119]. As a result – and this is important – MP becomes fairly easy available to benthic organisms and may accumulate for a long time in marine sediments.

Diatoms produce polysaccharides that increase the efficiency of particle coagulation [120, 121]. It can be assumed that aggregation of diatoms and sticky MP particles were probably very efficient. In those areas of the ocean where diatoms predominate among phytoplankton, mass aggregation may lead to accelerated deluge of MPs and quite pronounced removal of both MPs and associated organic components of aggregates from the surface layer.

The results of this investigation showed that the contribution of MPs to the system stimulates self-assembly process of aggregates and clearly increases aggregation rates of organic material, at least sporadically. This clearly indicates that MP particles are able to significantly alter the vertical export of biogenic particles to the ocean water column and thus affect globally important biogeochemical processes.

In turn, the formation of a biofilm on the surface of MP particles can reduce hydrophobicity of plastic. Due to this, the plastic density which is less than the density of seawater shows more neutral buoyancy (the ability to float) [27]. Thus, plastics such as polyethylene usually float on the surface after they enter the ocean. However, with biofilms, they gain properties of more neutral buoyancy, especially when biofilms contain many relatively dense and small diatoms. This effect is probably enhanced by the aggregation of plastic fragments with more dense biogenic particles, such as dead diatom frustules or foraminifer shells.

Microbial communities that colonize plastic are different from those in the surrounding seawater and are also different for different types of plastic and geographical origin [7, 42, 49, 56]. Accordingly, the aggregation of MPs with biogenic particles can modify the microbial communities of emerging aggregates. The aggregation and further conjoint sinking result in a distribution along the vertical column of these microbial communities living on MPs and as such are entering the food chain of deeper layers of the ocean.

The microbial communities on plastics may include pathogens, for example, bacteria belonging to genus *Vibrio* [7, 122]. Therefore, it is possible that such aggregation and deluge processes may increase the risk to organisms that inhabit deeper areas of the ocean: they are exposed not only to MPs, but also to bacterial pathogens.

The potential for aggregation of microplastic is substantially increased by the formation of biofilms on its surfaces, which is probably a typical situation in the oceans where bacterial communities colonize surfaces of plastics. It is highly likely that MP fragments are actively involved in the natural aggregation processes and thus affect particle distribution according to their size and organic substance export rates.

2.2 Microorganisms and Plastics: Biofilm Communities and Biodegradation of Synthetic Polymers

An extensive study by Masó et al. [123] examined fouling of plastic debris from pelagic and benthic habitats sampled across coastal waters of Spain, Italy, and Greece (Mediterranean Sea). Scanning electron microscopy (SEM) techniques were used to characterize organisms and their cenoses. In total, 42 samples of plastic were analyzed: 26 from the sea surface and 16 from the bottom. According to the results of observations, the most widely distributed and ubiquitous colonizers of plastic were diatoms – both at the bottom and in the pelagic zone.

An analysis of samples showed that frequencies of 8 large taxonomic groups of colonizers enable qualitative differentiation of the fouling on pelagic and bottom fragments of marine plastic debris (MPD). Diatoms have been observed on almost all pelagic and benthic MPD fragments (100% and 94%). Dinoflagellates were present on more than half of pelagic MPD (58%), but only on 13% of benthic fragments. Coccolithophores have been observed on both pelagic and benthic MPD – with a relatively high frequency (35% and 50%). Fungi are the second most frequently observed group on pelagic fragments (85%), but relatively rare on the bottom (13%).

Pelagic and benthic MPD had a very distinctive appearance. Most fragments from the pelagic area show typically well-developed biofilms containing lots of bacteria, fungi, and diatoms. It should be noted that bacteria were present in both benthic and pelagic biofilms. However, benthic plastic fragments were often covered by sediment particles (mostly by loam), which made the identification of colonizers more difficult. Benthic MPD was distinguished by a large number of sessile or pedunculated protozoans, as well as colonies of Bryozoans. Foraminifera have been observed in several communities from among benthic MPD.

Differences between the fouling of benthic and pelagic plastic fragments were indicated by diatoms from several genera: *Ceratoneis* (Syn.: *Cylindrotheca*) (73% and 6% of caught specimens from different biotopes), *Cocconeis* (54% vs. 38%), *Navicula* (50% vs. 13%), *Achnanthes* (23% and 6%), Amphoroids (including *Amphora* and *Halamphora*) (23% vs. 19%), *Fragilariopsis* (15% and 6%), and *Diploneis* (0% and 6%). Diatoms belonging to different genera have only been observed in pelagic samples: *Thalassionema* (42% fragments), *Licmophora* (19%), *Thalassiosira,* and *Mastogloia* (8% each) as well as *Cyclotella, Striatella,* and *Thalassiothrix* (4% each). The most striking difference of pelagic plastic biofilms was the high frequency of the attached diatom *Ceratoneis closterium* (in 73% of fragments). The authors, however, point out that a substantial number of diatoms could not be identified due to the poor preservation of their frustules or low visibility of their frustules' ornament. The resting spores have been observed on several pelagic fragments (in 8% of the cases). In some cases, attached diatoms have been observed on hydrozoans that colonized benthic fragments.

The dinoflagellates on pelagic MPD have been observed such as *Prorocentrum minimum, Prorocentrum micans, Ceratium* sp., *Pentapharsodinium tirrenicum, Dinophysis* sp., *Coolia* sp. and *Prorocentrum lima*. Cells of *Coolia* sp. have been observed only on fragments obtained from the area of Cape Creus – on pelagic fragments and not only there. *Heterocapsa* sp. and other atecate forms were rare.

Coccolithophores were observed on pelagic and benthic fragments, but whole coccospheres were found only on pelagic fragments. Individual coccoliths were quite abundant and detected on benthic fragments. *Emiliania huxleyi* was often found – and was quite abundant – on both benthic and floating MPD. Also, on pelagic fragments *Syracosphaera pulchra, S. halldalii,* and *S. molischii* have been frequently observed. In addition, *Syracolithus confusus, Calcidiscus leptoporus,* *Coronosphaera mediterranea* HOL (formerly *Calyptrolithina wettsteinii*), *Zygosphaera hellenica, Calyptrosphaera dentata*, *Umbilicosphaera sibogae* coccoliths, and *Scyphae apsteinii* were found on floating plastic. In turn, *Rhabdosphaera clavigera, Helicosphaera carteri* HOL (formerly *Syracolithus catilliferus*) have been observed in both habitats, whereas *Umbellosphaera* sp. and *Calcidiscus* sp. were found only on the bottom.

Diatom Ceratoneis closterium, frequently observed on the floating plastic, is a harmful microphyte associated with the mucilage in the Mediterranean Sea. In theory, the distribution of MPD in coastal and open waters could provide new habitats to be easily colonized by these species.

The results of this work highlight the relevance of increased knowledge about the effects of colonization of plastics found in the sea and the potential impact of debris accumulation on marine biodiversity ecosystems.

The behavior of microplastics in the aquatic environment is remarkably similar to that of an artificial microbial reef with diverse communities of eukaryotes and bacteria colonizing its surface. It is not yet clear whether these communities are specific to certain type of plastic on which they develop.

In their large-scale work Dudek et al. [124] conducted a 6-week incubation experiment using 6 most common plastic polymers. Plastic fouling took place in the waters of the Caribbean Sea, within the Bokas del Torro archipelago (Panama). The prokaryote community composition, reconstructed on the basis of 16S rRNA, showed that neither the type of plastic nor the time of environmental exposure had a significant impact on the formation of bacterial biofilm. However, reconstruction of eukaryotic communities based on 18S rRNA gene sequences has shown that they can depend significantly on the polymer type and incubation time. These data were confirmed by scanning electron microscopy techniques, which made it possible to establish differences between diatom communities on plastic by the time the incubation period was completed.

Communities developing on floating plastic may eventually determine its degradation, sedimentation at the bottom of water bodies, as well as its entry into food networks. But the integrity and significance of the factors that determine their development, such as the environment, the type of plastic, and the time of formation under these conditions are not yet clear enough.

Experiments with controlled incubation in a tropical sea bay, in Panama, showed that only eukaryote cenoses were formed differently depending on both – the type of plastic and interaction time. At the same time, it was emphasized that diatom cenoses were exactly those colonizers which showed a preference for certain types of plastic.

By the end of the experiment, diatoms on the surface of plastic had reached a number and density at which species ratios in communities could already be assessed. It turned out that it is possible to distinguish individual diatom cenoses on microplastics fragments. Such diatoms as *Cocconeis placentula*, *Fragilara* sp., and *Navicula* sp. have grown apart on all types of plastic, but their abundance on different plastic was not the same. At the same time several species of diatoms algae showed a preference for individual types of polymers. *Mastogloia* sp. I, *Mastogloia* sp. II, *Mastogloia fimbriata,* and *Cocconeis* sp. only appeared on 2–4 types of plastic. *Nitzschia sicula* and *Striatella* sp. were exclusively present on PET whereas *Amphora* sp. was only observed on LDPE, *Mastogloia corsicana* – on PP, and *Pseudonitzschia* sp. – on PS. The authors applied hierarchical cluster analysis to see if the composition of diatoms was influenced by the type of plastic polymer. HDPE and LDPE carried diatoms of similar composition, so as PP and PVC, and both groups contrasted with resident communities on PET and PS surfaces. The community on PS was most clearly different from the rest, whereas Pseudo-nitzschia sp. was present exclusively on this polymer.

Biodegradation of synthetic polymers is now seen as a most promising increasing pollution prevention approach preserving the natural environment. The focus is on eco-friendly recycling strategies. Moog et al. [125] offer an extremely interesting mechanism for the application of *Phaeodactylum tricornutum* plankton diatom culture as a bioengineered "chassis" for the production of the enzyme PETase, by which the decomposition of PET and PETG (polyethylene terephthalate glycol) is possible.

PET is a thermoplastic polymer manufactured on an industrial scale from fossil natural raw materials for more than 60 years. This material is now very widely used for the production of synthetic clothing and plastic bottles. The well-developed mechanisms of industrial PET processing are not yet able to cope with the flow of products from this material that turn into garbage and pollute the natural environment, including the water bodies of the planet.

The bacterium *Ideonella sakaiensis*, capable of degrading PET and using products of its decomposition as the sole source of carbon, was isolated as culture for these purposes in 2016. This bacteria extracts PETase – a key enzyme responsible for splitting PET into monomers. Application of this enzyme belonging to hydrolase group of enzymes provides many application opportunities for developing biodegradation processes and PET recycling, as well as for remediation of environmental plastic waste.

The authors took advantage of the photosynthetic diatom *Ph. tricornutum* as a bioengineered "chassis" – a basis for introducing a plasmid with a gene encoding an engineered version of PETase *I. sakaiensis* (IsPETaseR280A) and created a microbial cell factory capable of synthesizing and releasing to the environment this version of PETase. Already in the early stages of these experiments, it was shown that the PETase produced by them at 30°C is active against PET and the PETG copolymer – with an approximately 80-fold increase in the turnover of low-crystalline PETG compared to the bottle PET. The authors also showed that PETase produced by diatoms remains active in relation to industrial grinding of PET in a culture environment based on salt water at a temperature of 21°C. The resulting PET decomposition products were mainly terephthalic acid (TPA) and mono (2-hydroxyethyl) terephthalic acid (MHET). Under the selected reaction conditions, these substances were formed in a micromolar volume.

Therefore, Moog et al. [125] offer a promising eco-friendly solution for the mechanism of PET latter biodegradation – a saltwater-based environment, using a diatom as a model system instead of bacteria. Their work in this direction has shown that P. tricornutum can be turned into a productive basis for PET biodegradation. In general, their results show the potential of diatom biosystems for applications in the field of biodegradation of PET – especially in areas heavily polluted by PET.

Industrial methods of polyethylene decomposition that have become widespread thus far – chemical treatment, incineration, and landfill technologies remain extremely dangerous for most living beings. However, no better solution for complete PE degradation has yet been formulated. However, the biological PE decomposition can also be evaluated in terms of perspective – and developed in the near future.

A study by Kumar et al. [126] examined the ability of microalgae to degrade PE. In particular, they studied diatoms, green microalgae and cyanobacteria, which grew on plastic bags, found in the water bodies in the peri-urban areas. These plastic bags were found in three different locations (Maduravoyal, Vanagaram, and Punamalli), Chennai, Tamil Nadu, India.

The microphytic biofilms living on the PE surface were obtained from the surface of bags. In turn, three most widely expanded types of microphytes have been identified for biological processing of low density PE (LD) and high density (HD) sheets. Among green algae, it was *Scenedesmus dimorphus*, among cyanobacteria – *Anabaena spiroides*, and diatoms were represented by *Navicula pupula*. A cross-cut of the PE sheet showed that microphytes colonized both sides of it. The destruction of its surfaces due to erosion and reproduction of cells was observed. Cultures of these three mentioned species of microphytes on PE sheets have been treated with cultural media to enhance their growth. As a result, on LDPE, microphytes proliferated more strongly than on HDPE. In this case, the culture of *Anabaena spiroides* was most efficient in destroying the polymer. The degradation of LDPE sheets using *Anabaena spiroides* was obvious – this was confirmed by observations using SEM. Also, this species showed higher growth rates than other microphytes.

Production of biodegradable plastic has been growing in recent years. Amid an increasing pollution of the land environment by these polymers, the flux of plastics to the sea is expected to increase. A number of laboratory experiments have shown that such polymers start to degrade in the early days or weeks after entering the natural environment. However, not too much is known yet about the early composition and activity of biofilms formed on fragments of biodegradable and conventional plastic found in the sea, and how these films are related to their degradation.

A study by Eich et al. [45] focused on the early stages of biofilms formation on consumer plastic bags and their possible impact on the degradation of polymers. Samples of PE and biodegradable plastic were exhibited in the Mediterranean Sea for 15 and 33 days. The samples were evenly distributed on the bottom in sandy shallow water, at a depth of 6 m, as well as in the water column, at a depth of 3 m. This was done to compare how different environments in which plastic was placed affect its pollution and degradation.

Biofilms were observed on both types of plastic and in both habitats. However, the abundance and diversity of diatoms differed significantly depending on both the exposure environment and the type of polymer. In general, diatoms were more common on samples colonized in pelagic areas. The authors suggest that different specific properties of polymers contributed to the growth of different biofilm communities on different types of plastic. In addition, the difference in habitat conditions between the bottom and the water column (lighting power and force of water movement) could affect unequal colonization in these environments.

The oxygen release rate was negative in all cases. Thus, biofilm in the early stages of development actively consumes oxygen, regardless of the type of plastic and habitat. In turn, mechanical tests of polymers did not reveal significant degradation after a month's exposure to the sea. However, observations with SEM showed potential signs of surface degradation, varying for different types of plastic.

The study showed that the early development of biofilm and its composition depends on the type of plastic and the environment in which it is being fouled. It has also been shown that after two weeks of exposure in the marine environment (both on the bottom and in the water column) the biodegradable plastic shows signs of degradation.

In Indonesia more than 4.6 million tons of different types of plastic are produced annually [127]. A substantial part of it then becomes garbage and fluxes into water bodies as well. PET and PP are the most widely used materials in the production of various packaging and fibers. Destruction of these types of plastic into microparticles is a threat to the natural environment, especially when microplastic interacts with freshwater microphytes.

The goal of the study by Khoironi et al. [127] was to assess the effects of microplastic on the growth of cyanobacteria Spirulina sp., and estimation of the contribution of these microorganisms to plastic degradation.

The interaction of cyanobacteria and microplastics was observed in 1 L glass bioreactors in which *Spirulina* sp. culture and microplastics PET and PP 1 mm particles in different concentrations (150 mg/500 mL; 200 mg/500 mL and 275 mg/500 mL) were deposited. Experiments took place over 112 days. As a result, it was shown that particle fracture strength had been decreasing by 0.9939 MPA/day for PET, and 0.1977 MPA/day for PP. EDX-analysis of microplastic showed that the reduction of carbon concentration in PET (48.61%) was higher compared to PP (36.7%). In turn, FTIR analysis of Spirulina sp. showed that CO_2 release by cells in the presence of PET microparticles was higher than in the presence of PP microparticles.

Spirulina sp. growth rate in the presence of microplastics was lower than in the absence of its particles in the experimental environment, and increased concentration of these particles significantly reduced growth rate of this cyanobacterium (by 75%). The results of the experiments led the authors to conclude that the process of plastic decomposition is due in no small part to biodegradation.

3 The Results of Studies of Plastic Colonization by Microorganisms, Obtained by Our Group

In this section we discuss the results of our research on plastic colonization. Since 2016, our team has provided a series of studies, having systematically joined the efforts of algologists, materials scientists, and nanotechnologists. The primary directions in our investigation are in the areas of interaction between microphytes and different types of plastic: in the global seas, continental waters, and under experimental conditions. Here we present an overview of key achievements along two lines of investigation:

1. which species of microphytes and how exactly they colonize the surface of plastic under the "wild" conditions of the seas and continental waters, and what really happens during this process;
2. how microphytes from natural populations colonize plastic under experiments in nature environment.

3.1 Microplaston on Plastic Waste in Natural Reservoirs

Similarly to such terms as microplankton, micropsammon, microepilithon, and microepiphyton, we therefore consider very appropriate the introduction of a new term for micro-fouling (a set of microorganisms that settle on a substrate and form a community on it) of plastic materials – "microplaston" [128–131].

In our work to observe the interaction between microphytes and different types of plastic in natural waters, we have investigated the surface of polymer samples covered with a layer of foulings in the "wild" environment of Lake Baikal (PE and PP) and Lake Issyk-Kul (PE), in the streams of the Leningrad region (PP), in various areas of the Black Sea (PE, PP, PET, UDHPE), as well as in the Eastern part of the Mediterranean Sea (PE, PP, PET, nylon). All the plastic samples discussed in this section belonged to the category of household waste, accidentally found in the water areas.

Analysis of the composition, structure, and spatial organization of microphytic fouling (biofilms) was carried out using the methods of light and electron microscopy. In particular, the light microscopes Leica DMLS, Leica DT 2500, and Carl Zeiss Primo Star were used, as well as scanning electron microscopes Hitachi TM 1000, Tescan LYRA, and JSM-6380LA. The identification of polymers was carried out by the marking applied to their surface during the manufacture of products.

Below we briefly state the primary results of these observations.

In 2017, samples of synthetic polymers were obtained floating on the surface of Lake Baikal (Russia, Eastern Siberia): these were relatively large fragments of PE packages and broken off disposable PP cups. A developed fouling, represented by a complex mosaic of various diatom species (on PP), as well as a mosaic of diatoms

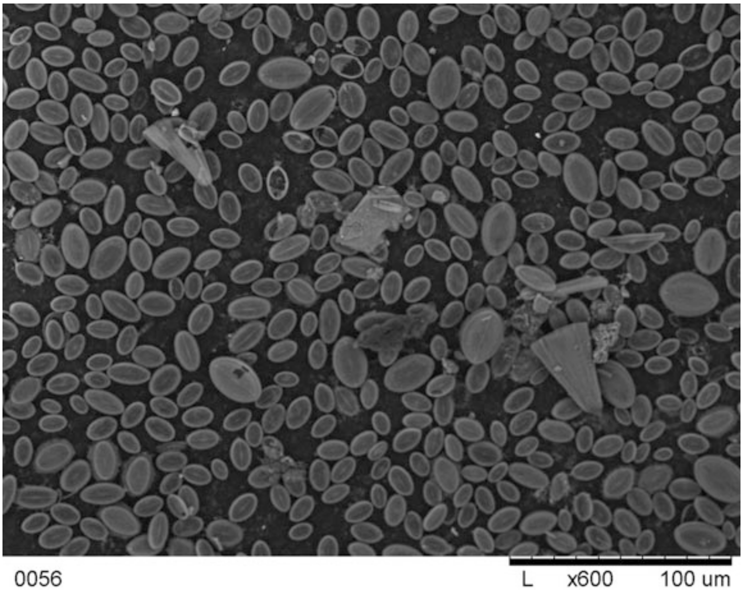


Fig. 1 Diatom fouling on the PP surface in Lake Baikal. The lower tier is formed by colonial settlements of the *Cocconeis* species

and cyanobacteria (on PE), was discovered on the surface of both types of samples. Microplaston populations on large PP fragments looked as if they were the simplest in their composition and, at the same time, the most hierarchically organized (Fig. 1).

These cenoses were organized almost entirely by species of the genus *Cocconeis* – but by six different species and subspecies. Furthermore, there were small ascending tree-like colonies of *Gomphoneis* and, separately, short branches of colonies of two *Didymosphenia* species. Of particular interest in this case were the extended colonies of *Cocconeis* species that developed directly on the surface of plastic. We shall note two striking features of them. First, the different *Cocconeis* species and varieties formed a mosaic carpet, in which the cells of the largest species *Cocconeis placentula* var. *placentula* were located sparsely, at a distance of 2–3 (and significantly more) lengths of their frustules from each other. They were surrounded by curve-patterns, formed by medium-sized cells of *C. lineata, C. placentula, C. fluviatilis,* and *C. euglypta*, as well as small-sized *C. lineata*, *C. euglypta,* and *C. neothumensis* (Fig. 2). Two other species – *C. baikalensis* and *C. neodiminuta* – were much rarer.

At the same time, compact groups of cells within curve-patterns of mixed colonies were formed not by single-species, as might be expected, but by cells of similar sizes – that is, by mixed species of the same size group. Within such groups, cells of different types have often coexisted. Most often, such compact groups have built medium-sized cells, and small ones filled the space around them, as well as "gaps" within the lines of the ornament – rather on a residual basis, when there was

Fig. 2 *Cocconeis* cells on the surface of the PP. Colonies of iron bacteria are visible, bordering their frustules

space for them. In turn, these curving lines, or not too strictly repeated “patterns” of colonies of *Cocconeis* spp. were surrounded by a solid border of slimy colonies of iron bacteria (Fig. 2).

In many areas along the edges of the torn PP cups, where the fouling seemed to be the oldest, and where there has been influence of hydrodynamics, we observed a large number of shallow, but also curving cracks inside the PP surface. With high probability, they were formed under the influence of photooxidation of the polymer by solar rays under the parallel influence of hydrodynamics. However, formation of such cracks along the contours of the matrix of iron bacteria surrounding the lines of the ornament of diatom colonies indirectly suggests that the matrix, as a tightly binding substance, could also participate in formation of cracks, forming their outlines. However, the images clearly show that the PP did not crack over the entire thickness, but in layers. Then *Cocconeis* cells – mostly medium and small-sized – rushed into these cracks, crawling under the upper layer of PP, which was slightly deformed due to the cracks and loss of elasticity.

Continuing to actively divide there, as if in greenhouses with a transparent polymer coating, these cells formed compact groups, gradually – and inevitably – pushing the pieces of plastic that covered them from above, from the bulk of the polymer (Fig. 3). Then the pieces would fall off. Having microscopic dimensions, they immediately passed into the category of microplastics (MP), already partially colonized by *Cocconeis* spp. In the most highly degraded marginal areas, where the cracks became more extensive and deeper, they accumulated fine bio-mineral

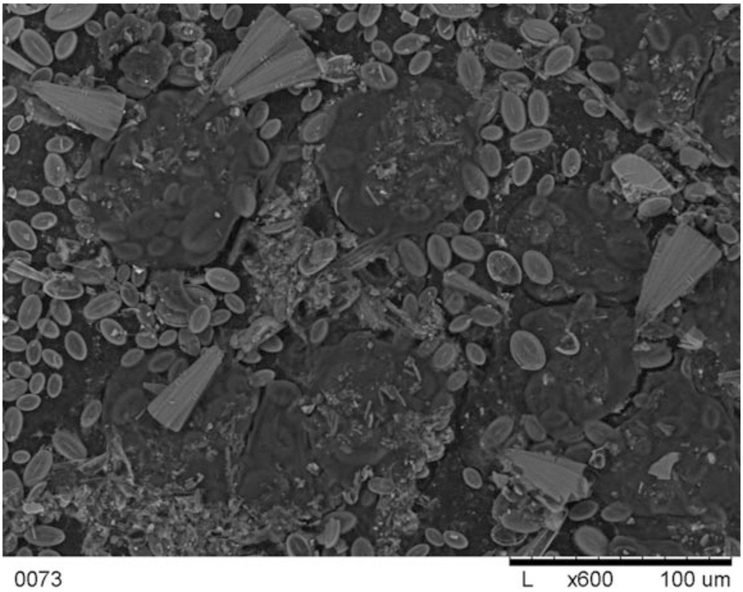


Fig. 3 Diatom fouling along the degrading edge of PP: groups of *Cocconeis* cells can be seen crawling along cracks under the scales of the upper polymer layer

detritus, among which colonies of other diatoms were formed: small- and medium-sized *Encyonema* spp., *Nitzschia frustulum* and large-celled *Navicula serotina*. These compact groups of diatom frustules also extended the cracks.

Thus, PP collapsed layer by layer along the edges of the cup that traveled on the surface of Lake Baikal.

Of particular note is an interesting feature of *N. serotina* cells, which were found sporadically and in small compact groups on the surface and in the cracks of PP. This mobile diatom, developing on PP, lost its motility: its cells as part of compact clusters (possibly formed in safe places due to the division of one cell that came there first) were immovable. For a comparison, all types of motile diatoms, developing as part of the periphyton (or microepiphyton) of natural substrates, such as filamentous algae, organs of aquatic plants or hydrobionts, stones, wood and charcoals, and even on such artificial materials as steel and glass, do not lose their ability to relocate and do not form compact clusters of nonmotile cells. We cannot yet explain why the cells have lost their mobility. However, our other studies have shown that this is not an isolated example – the same happens with many species of mobile diatoms on other types of plastic.

Biofilms developed on PE pieces floating on the surface of Lake Baikal are much more diverse than on PP. These polymer fragments being affected by microplaston are destructed otherwise. The fouling community is represented here by several tiers: from the lowest to the highest. The lower tier is formed by diatoms from the genus *Cocconeis* (Fig. 4): large *C. placentula* and *C. baikalensis* reside separately

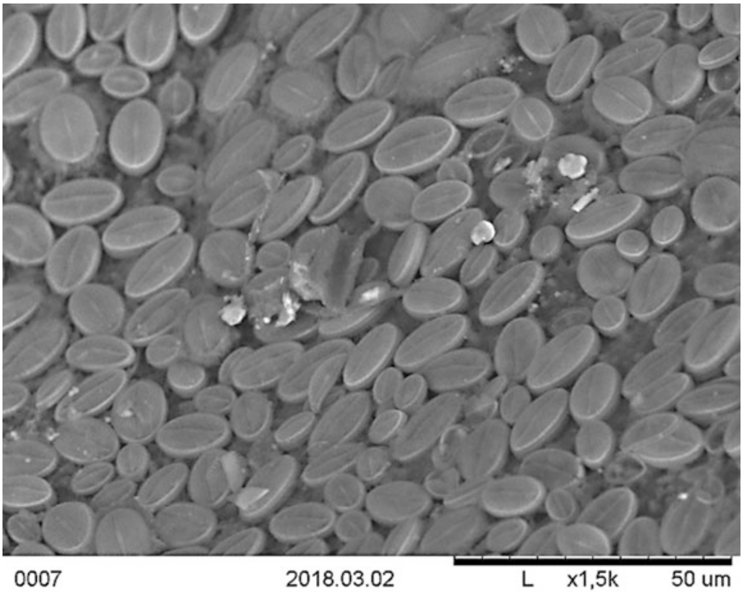


Fig. 4 The lower layer of fouling on the PE surface in Lake Baikal, formed almost exclusively by species from the genus *Cocconeis*

(scattered about) or in small compact groups. They are surrounded by curving, quasi-periodically repeating "patterns" of medium-sized cells: *C. lineata* and *C. euglypta*, among which *C. fluviatilis* and *C. placentula* are occasionally found. Small-celled *C. lineata* and *C. euglypta* are less common here, only among medium-sized cells, including as part of the lines formed by them, and also they build their own lines – between neighboring lines of medium-sized ones. *C. neothumensis* and *C.* cf. *pseudothumensis* cells are very rare. The *Cocconeis* "ornament" itself seems to be significantly denser. In this case, the competition of diatoms in this tier with cyanobacteria may have its impact. The entire space between *Cocconeis* cells, as well as surface areas free of them, is filled with small cells of cyanobacteria from the Chamaesiphonaceae family (*Xenococcus minimus*), tightly sitting on the polymer. In some places there are flat rosettes of green multicellular microphytes.

The closest analogue can perhaps be the forest, where we identified the tier of "mosses and low-growing grasses." A tier of "low-shrubs and bushes" rises above it. At first, this tier is formed by the colonies of diatoms from the genus *Gomphonema* (*G. rimetii* and *G. russicum*), the matrix of which is branched to varying degrees (Fig. 5). They rise above open (plain) areas of the surface at (40) 50–200(250) microns, forming extended to varying degrees "crowns" of cells crowning the branches of the matrix in form of fan-shaped structures. Secondly, colonies of cyanobacteria *Gloeocapsopsis magma* develop in this tier sometimes abundantly and form dense settlements. They form powerful folded structures up to 100–200(250) microns high (Fig. 6).

Fig. 5 Fragment of fouling on the surface of PE, formed with the participation of *Gomphonema* species

Fig. 6 Folded colonies of cyanobacteria *Gloeocapsopsis magma* on the PE surface

Larger microplaston structures are created in cracks. We've said before that polymers in natural waters become brittle. This occurs under the influence of solar ultraviolet light that penetrates the water column by several centimeters, as well as under the influence of water movement and temperature changes [3, 4]. Pieces of PE floating near the surface of Lake Baikal are not an exception: over time, they lose

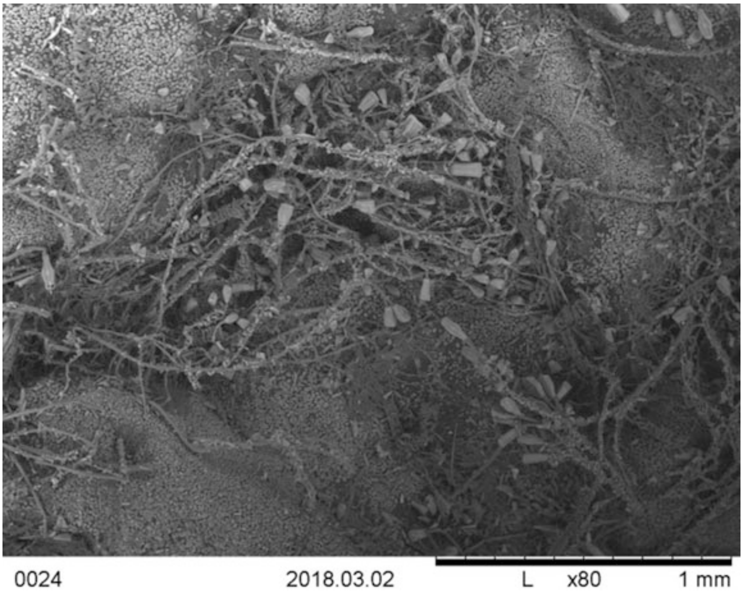


Fig. 7 Branched colonies of diatoms *Didymosphenia* spp. growing along cracks on the PE surface

their elasticity and become covered with micro-cracks. In these areas affected by erosion, within cracks, a "tree" layer of fouling is being developed. "Pillow-shaped" (hemispherical) colonies of cyanobacteria and filamentous green algae (Chlorophyceae) grow actively along small cracks and deepenings within the polymer surface. In turn, the bottom and walls of deeper cracks become a safe house for very large (up to 1,000–2,000(2,500) microns high), strongly branching colonies of diatoms from the genus *Dydimosphenia* (Fig. 7). Colonies of six species develop on PE: *Didymosphenia clavamagna, D. curvata, D. dorogostaiskyi, D. geminata, D. laticollis* and, occasionally, *D. grunowii*. In general, these are almost all *Didymosphenia* species identified for Lake Baikal.

Among natural substrates, they prefer rocks of coastal shallow waters, forming a layer of branched colonies (0.6–0.8 cm thick) to a depth of 1.5–2 m, as well as on floating fragements of "tree"– creating a thinner coating. On pieces of a PE film, they successfully develop within cracks.

The matrix of these colonies serves as a substrate for many species of attached diatoms (Fig. 8). They create a powerful microepiphyton on its surface, the structure of which is dominated by *Fragilaria pectinalis* – its small cells settled down on the matrix in a relatively dense layer. Among them, there are many small *Encyonema minutum* and *E. brevicapitatum, Amphora* cf. *pediculus, Amphora* sp. 1 (a small diatom not yet described by taxonomists), *A. inariensis*, and medium-sized *A. metzeltinii* and *A. indistincta*, which also grow directly on the matrix. Here, cells of different *Cymbella* species (*C. stuxbergioides, C. stuxbergii, C. pseudostuxbergii, C. paraintermedia, C. microlacusbaicalensis,* and *C. amplificata*) and *Rhoicosphenia* (3 species) settle down in the polysaccharide mucilage stalks.

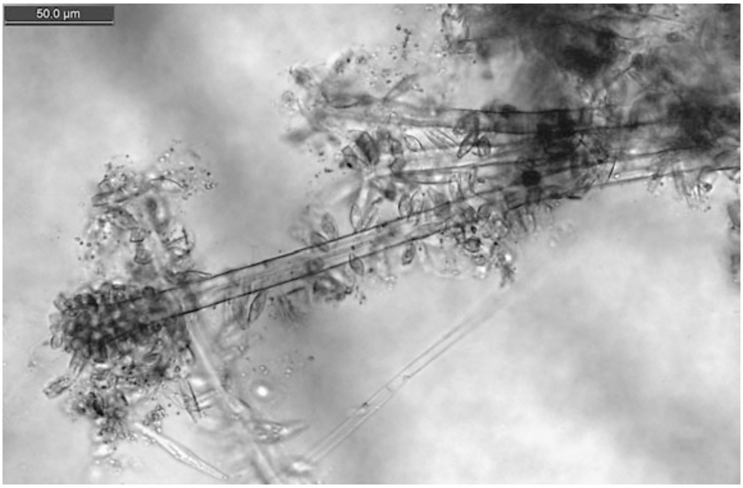


Fig. 8 Microepiphyton on the surface of matrix stems of *Didymosphenia* spp

Where the *Didymosphenia* matrix is branching, small tree-like colonies of *Gomphonema*, already described for open areas of the polymer, as well as *Gomphonella olivacea*, often develop. Among microepiphytic-spinney, some motile diatoms from the genera *Navicula* (*N. baicaloradiosa, N. serotina, N. permakarevichae*, etc.) and *Nitzschia* (from small *N. citrus* var. *appendiculata* to very large ones, such as *N. nevrovae* and *N. werumiana*) move in different directions. Cyanobacteria are represented in the fouling by thin species of *Heteroleibleinia* and compact mucous colonies of *Aphanocapsa* sp., developing directly on the matrix.

There are also "lianas" – branching tubular colonies of *Nitzschia pavida* – among the "arboreal" tier.

Note that the cells of the *Navicula* and *Nitzschia* species, living within cracks directly on the polymer, lost their motility– the same as on PP. This was especially true for *Navicula*, which formed compact groups of nonmotile cells on the walls of cracks.

The open end holes within PE are colonized especially densely by cyanobacteria and diatom colonies (Fig. 9). Trichomes of cyanobacteria *Leptolyngbya* spp. are predominant here, filling the spaces within inside cracks and other perforations with solid springy thickets.

We describe the communities in such detail in order to formulate an extremely important feature of this heterogeneous and multi-tiered fouling. Growing on PE, the biofilm does not leave any free places on its surface. The significant diversity of ecological niches of microphytes also determines the micro-topological diversity of their colonies. Each species – and here we see 75 species and subspecies only among diatoms – finds a set of micro-habitats that are most convenient for its existence. Different types of microphytes build tiers, mastering the space of the polymer

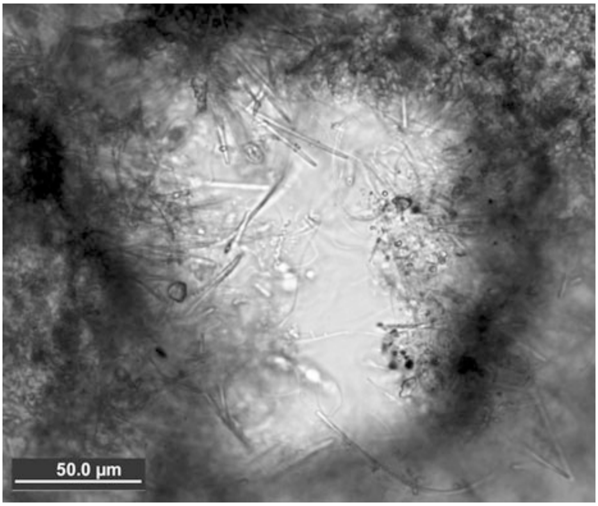


Fig. 9 Dense fouling of microphytes with a predominance of filamentous cyanobacteria (*Leptolyngbya* spp.), covering the walls of the through hole in PE

surface itself (relatively flat areas, depressions, cracks and perforations), and above it, on *Didymosphenia* colonies. Pieces of PE are clothed with biofilms so tightly that they penetrate all the cracks, and actively grow there, extending them. As plastic becomes more brittle, new cracks are formed through extending existing ones.

Thus, the microphytes participate actively in the fragmentation of PE-film pieces into small fragments (ultimately, into MP). At the same time, MP particles are already colonized, and often carry macrocolonies of *Didymosphenia* spp. with a well-developed and very extensive microepiphyton. Because of this the MP particles, in combination with their fouling, acquire an additional weight, due to which they sink. Of course, getting into the deep layers of water, cells of many microphytes die, because hundreds of meters deep in the ocean is not their habitat. However, their frustules themselves appear to have considerable weight, whereby macrocolonies and *Cocconeis* frustules are quite firmly attached to the particles. So, even having lost living cells, such aggregates (MP + fouling) can no longer achieve positive buoyancy and cannot return to the surface.

The populations of microphytes on the surface of coastal rocks mentioned above include much larger number of species. A significant number of those are not able to shift to floating plastic. Yet, quite a few Baikal species – mostly endemic – settle comfortably on PE and PP. They build cenoses on these substrates with their own spatial organization (“ornaments,” tiers, hothouse thickets – taking into account the features of the surface micro-topology). These cenoses collected from benthic microphytes no longer appear to be benthos. Now they live not only at the bottom. Now they inhabit the surface of the plastic that migrates near the surface of the water – and form a near-surface microplaston.

Our observations show that many species that reside on floating polymers acquire other opportunities for space development here as compared to natural substrates.

Fig. 10 "Spots" with a relatively stable structure, formed by four species of diatoms (*Cocconeis lineata, C. euglypta, Planothidium* sp., and *Achnanthidium* sp.) and repeated on the surface of the PP in the stream, like a pattern on the fabric

In 2018, in the streams of the Izhora plateau (Russia, Leningrad region), we studied microphyte populations on PP yogurt cups. This fouling can also be considered as an example of gradual colonization of the polymer. We immediately noticed that the population of epipsammon (inhabitants on the surface of sand grains) at the bottom of the streams included many more species than the cenoses on the PP. At the same time, observing PP increase in these biotopes, we noticed the formation of structures that are organized more regularly, quasi-periodically repeated, and, as a result, more extensive than on sand grains.

The lower layer, directly adjacent to the PP, was organized here by four species. They built spotty formations with a fairly stable structure, almost regularly – like a pattern on a fabric – repeated on the flat surface of the plastic. The spotty formations were based on medium-sized and small cells of *Cocconeis lineata* and *C. euglypta*, between them resided medium-sized cells of *Planothidium* sp., and already among them small cells of *Achnanthidium* sp. (Fig. 10).

At the same time, *Cocconeis* cells appeared first at the point of spotty formations and began to divide, settling in a small group. Then they were joined by *Planothidium* cells, and also began to divide, filling the space between them. After that, representatives of *Achnanthidium* visited the company. Their small oblong cells, dividing and spreading, filled the remaining empty places. Thus, in order for the newly formed *Cocconeis* cells to find free space, they had to crawl to the edge of the mixed colonies and attach themselves there. They were followed by the entire "retinue." So the spotty formation grew, gradually closing in on each other (Fig. 11).

On the same substrate cyanobacteria from the family Chamaesiphonaceae were residing (Fig. 12) (a different species than in Baikal).

They developed their own, rather dense and compact spotty formations that spread across the substrate independently of the diatoms. In those areas that

Fig. 11 As grows "spots" in different directions, they gradually merge, forming an almost continuous ornamental layer on the PP surface

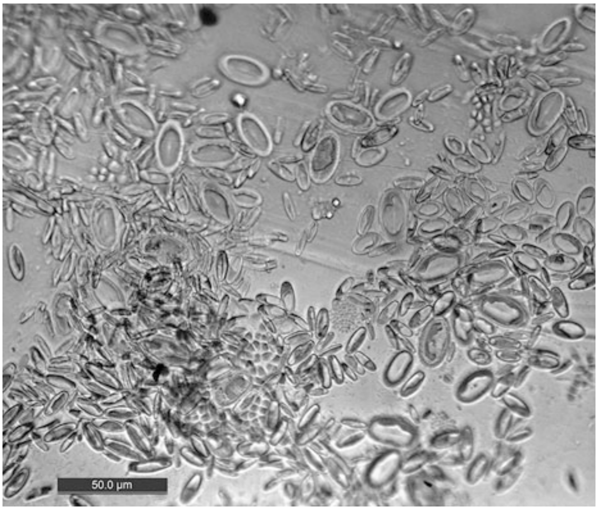
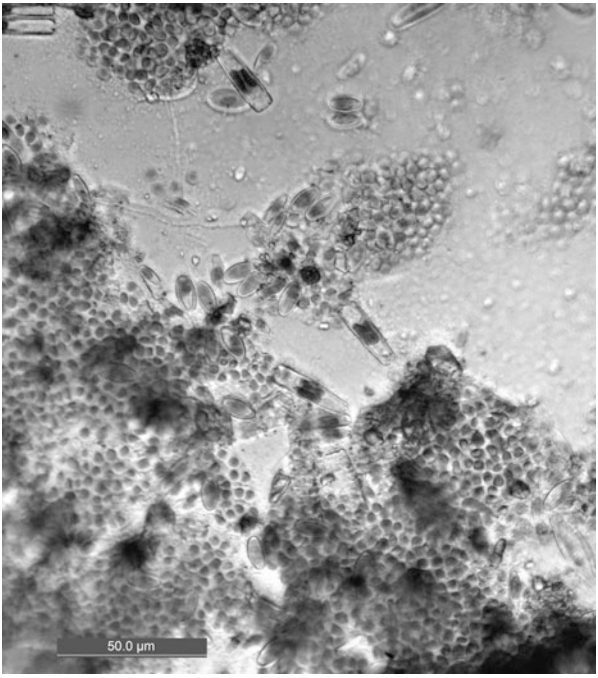


Fig. 12 "Spots" of microphytic fouling on the PP surface, formed mainly by colonial settlements of cyanobacteria from the Chamaesiphonaceae family, and also – in small quantities – by diatoms *Cocconeis* spp., *Gomphonema parvulum,* and *Planotnidium* sp



cyanobacteria managed to develop first, the remaining space was inhabited by small groups of *Planothidium* sp. or *Gomphonema parvulum*, becoming initiators of the growth of small spotty formations and surrounded by small *Achnanthidium* sp. *Cocconeis* cells settled here rarely and individually. It is entirely possible that

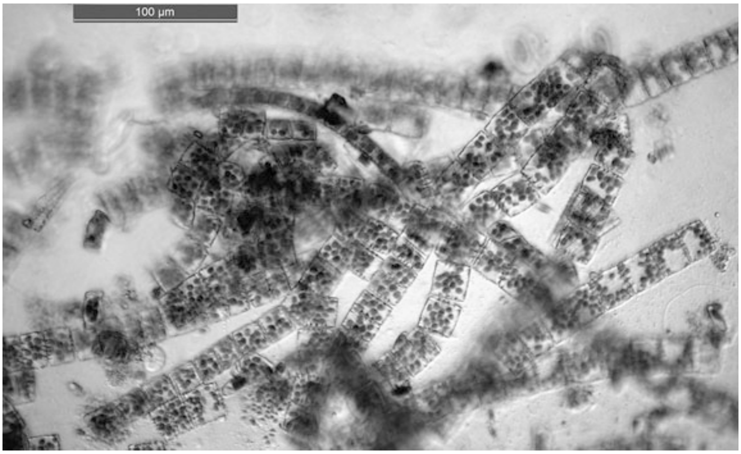


Fig. 13 Chains of colonial diatoms *Diatoma mezodon* forming the upper layer of fouling on PP in a fresh stream

competition for space in this case is mediated by allelo-chemical interactions, and *Cocconeis* species lose in this confrontation.

In other places, where the growth of the carpet of cyanobacteria was not so fast, and there was still a lot of space between the growing spotty formations of diatoms of the lower layer, diatoms of the second, "high-grass" layer were actively growing. These were fan-shaped colonies of *Meridion circulare* and bundles of *Ctenophora pulchella* cells that rose vertically from the surface of the PP. In the same places development of diatom colonies of the third, "shrub" layer occurred. These are branching chains of *Diatoma mesodon* cells (Fig. 13), the height of which could reach hundreds of microns.

There were no signs of destruction of the PP surface that had occurred with the participation of microphyte fouling. In general, judging by the dates on the preserved labels, the yogurt cups were left in the streams for no more than 2 months.

Turning to the fouling of polymers in the brackish reservoir space, we can discuss our research in Lake Issyk-Kul (Kyrgyzstan). In autumn 2019, samples of floating PE covered with developed brown biofilms were collected on the lake shore. The analysis showed that the polymer surface fouling was formed by two types of diatom populations. Cenoses of the first type were organized on the basis of large cells of *Cocconeis placentula* and *C. lineata* (all three size groups), *C. euglypta* (medium-sized), and *Gomphonema angustum* and *Achnanthidium* spp. (2 species). Colonies of these species had been growing on the surface of relatively "flat" areas, almost unspeckled with small folds and dents, comparable in size to the large cells of the diatoms themselves (Fig. 14). At the same time, *G. angustum* cells resided individually or in pairs, without forming colonies. *C. lineata* cells developed compact groups, often interspersed with two other species, but the formation of specific "patterns" was not observed here. In turn, small sparse colonies of *Amphora indistincta*, *A. pediculus*, and *A. pediculus* ssp. 1 developed on "flat" areas with a

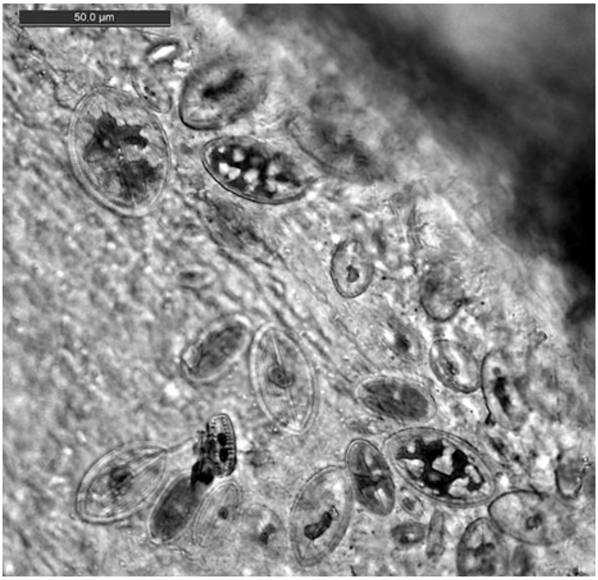


Fig. 14 Diatom fouling of a relatively flat ("champaign") area on the surface of PE from Lake Issyk-Kul

higher degree of speckling, with the presence of individual cells of *Diploneis subovalis*, *D. separanda*, and *Amphora copulata*.

A completely different picture developed in areas of high folding, where the surface was densely covered with large cellules and folds (Fig. 15). Here, using low-lying areas of cells and folds, *Denticula kuetzingii* developed fairly dense colonies. These were close groups of dozens or hundreds of cells attached to the substrate by a short string at one end. A few *G. angustum*, *Mastogloia* cf. *smithii*, and *Achnanthidium* spp. were sometimes observed in these colonies. On the contrary, the *Amphora* species (*A. ovalis*, *A. copulata*, *A. inariensis*, *A. indistincta*, *A. pediculus*) and *Fallacia* sp. (Fig. 16) actively settled along the edges of small flat "plateaus" along the tops of the folds.

For *D. subovalis*, a large motile species, it can be said that they were growing on this polymer substrate, moving without much reference to the micro-topology of the surface. The same would be true for the small "stray" *Nitzschia elegantula*. We also note that individual living *Denticula* cells were also observed in "flat" areas, among colonies dominated by *Cocconeis* spp., and small groups of them – along the edges of folded areas, with the transition to "plains."

In general, using the example of this PE fouling, we can talk about different micro-topological orientation of individual species of diatoms that can reside on the surface of plastic. There were no signs of biodegradation involving microphytes in these samples. However, it had a rather low durability and tore almost like a sheet of writing paper.

Our group started observations of microbial growth on garbage plastic in the sea in 2016. In September 2016, a large fragment of PE film was lifted from the water off the coast of Gurzuf. By its appearance and thickness, it most closely resembled a rain

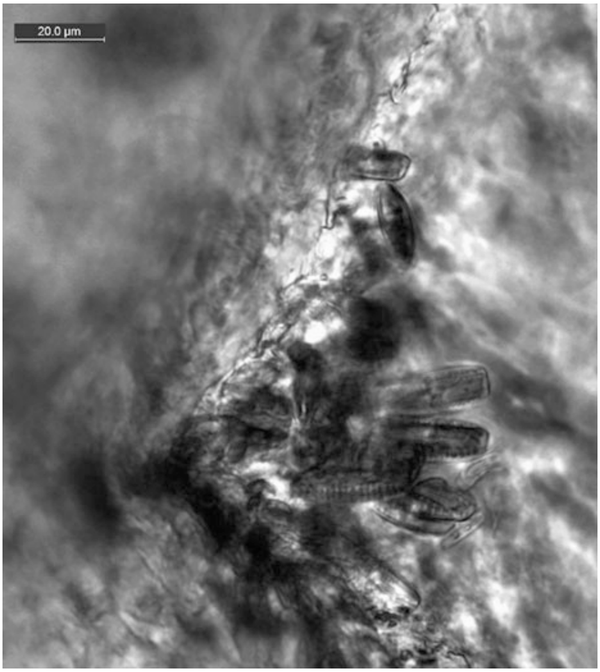


Fig. 15 Colonial settlement of *Denticula kuetzingii* in an area with dense foliation of the PE surface. A needle outgrowth of attached diatoms grows densely along the edge of the fold

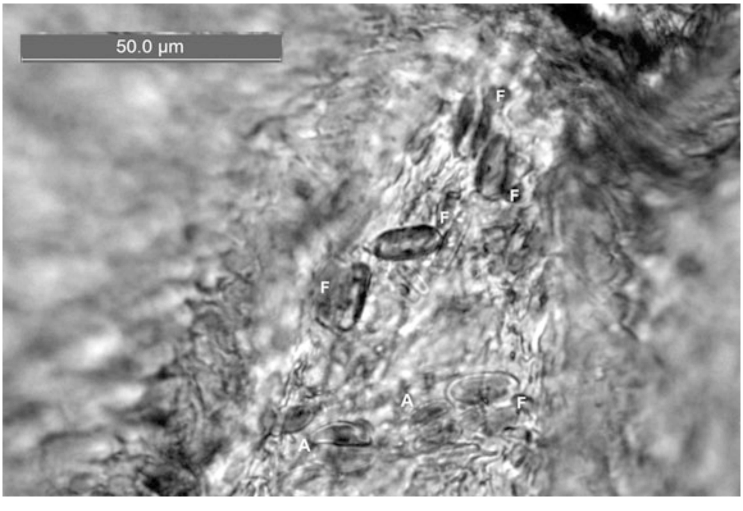


Fig. 16 Sparse groups of diatom cells from the genera *Amphora* (resemble coffee beans, marked with the letter "A") and *Fallacia* (ellipses, letter "F") along the edge of a small "plateau" formed by a wide fold on the surface of the PE

Fig. 17 Cyanoprokaryotes from the genus *Calothrix* forming dense microscopic clumps on the surface of PE in the Black Sea

cloak. It may have been blown out to the sea by the wind. It was not possible to determine how long this piece of PE had been floating on the surface of the sea.

On both sides of the surface of the transparent film, a golden-brown fouling of different densities was clearly visible. The fragment was badly torn along the edges, and also tore well when pulled by hands. At the same time, the stretching effect characteristic of the new PE raincoat was not observed.

When studying the fouling under a microscope, the thickness and transparency of the film allowed us to consider the structure of the periphyton on both its surfaces. At the same time, it was not necessary to turn over the wet specimen, which was covered with integumental glass. This indicates that the microbial fouling, developing in the sea on this substrate, was well lit from all sides, without experiencing the effect of significant shading due to the folds of the film.

The algocenosis was represented by only 14 taxa [132] (6 taxa of cyanoprokaryotes from the genera *Calothrix* and *Dichothrix* (Fig. 17) and 8 species of diatoms from the genera *Mastogloia*, *Halamphora*, *Cocconeis*, *Navicula*, and *Nitzschia*).

Here we observed the scarce diversity of the communities. However, it has evolved in significantly more aggressive conditions than, for example, the cenoses of the rocky supralittoral. For comparison, in the same period of 2016 and on the same part of the coast, in cenoses on rocks irrigated by the surf, we revealed a purple-cyanoprokaryotic-diatom cenoses, which included at least 30 species of micro- and macrophytes. Among them were *Ceramium* sp. and *Lophosiphonia* sp., 8 species of cyanobacteria morphologically similar to *Calothrix* (*Scytonematopsis crustacean*, *Calothrix fusca*, *C. fusca* f. *parva*, *C. contarenii*, *C.* aff. *Vivipara*, *C. parietina*, *C. scopulorum*, and *Calothrix* sp. 1), 2 species of *Leptolyngbya*, *Schizothrix* *cresswellii*, *S. telephoroides*, and 5 species of crustal forms of cyanoprokaryotes (*Entophysalis granulosa*, *E. major*, *Pleurocapsa minuta*, and *Placoma vesiculosa*) that developed in the lowest layer of the communities.

Among the diatoms here were the species *Halamphora*, *Mastogloia*, *Cocconeis*, *Navicula*, *Nitzschia*, *Licmophora*, *Achnanthes*, and *Rhopalodia*.

Despite the relatively small number of species, the algocenosis on the PE-film had a clearly pronounced tiering and strictly ordered architectural elements. All five species of *Calothrix* and *Dichothrix gypsophila* were trichomal forms clad externally with multi-layered transparent sheaths that easily transmit most of the light flux (polysaccharide covers hiding the lower part of their trichomes). This morphological feature – an advantageous adaptation – allowed them to develop here in relatively "hothouse" conditions, reducing the aggressive effects of light, temperature changes, and mechanical impacts of waves – in the hydro-dynamically active and light-saturated environment of the subsurface layer of the open sea.

The upper stage of the cenosis was formed by the largest species with rising trichomes: *C. scopulorum* and *C. polarietina*. Their dense turf covered up to 60–70% of the film surface, often with a layer up to 0.7–1.2 mm thick, and stratified multilayer covers (sheaths) protected the cenosis, preserving the microphyte thickets from aggressive environmental influences. The second, lower layer of trichomes developed by spreading turf of *D. gypsophila*, up to 0.3–0.4 mm high. Finally, rising above the surface of the film by 70–100 μm, also on both sides, three more species grew in small turfs: *C. fusca, C. fusca f. parva*. *Calothrix brevissima* grew in very small groups, only a few trichomes, in the most thin-walled cases.

Cyanobacteria trichomes did not grow in a continuous layer on the surface, but were grouped by shallow deformations and cracks, and especially densely – in the places where the cracks branched out.

Three species of diatoms from the genus *Mastogloia*, forming polysaccharide capsules, were localized in different layers of the cenosis. The most abundant among them was *M. lanceolata*, represented by three dimensional modifications. Of these, the smallest and broadest, 27.9–34.3 μm long (31.2 μm on average) and 13.2–16.1 μm wide (14.8 μm on average), was observed in thick-walled capsules only on the surface of the covers of *C. scopulorum* and *C. parietina*. In turn, the medium-sized shape is 34.3–41.8 μm long (average 39.3 μm) and 14.1–17.3 μm wide (average 15.9 μm) in thinner-walled capsules, the surface of *D. gypsophila* sheaths, as well as the surface of the film under them and in narrow open spaces, was inhabited. Finally, the largest form [45.5–47.6 μm long (average 46.4 μm) and 16.9–18.5 μm wide (average 17.3 μm)], clad in thick-walled capsules, inhabited mainly wide open areas of the surface of the PE film, not overgrown with cyanoprokaryotes.

Encapsulated *Mastogloia pusilla* and *M.* aff. *Urveae* were significantly less common: the abundance ratio in percentage looked like *M. lanceolata*: *M. pusilla*: *M.* aff. *Urveae* ≈ 80.39:15.69:3.92. The last two species had cell sizes, respectively, 21.3–25.5 × 6.5–7.4 μm (average 24 × 7 μm) and 14.2–23.9 × 6.6–11.2 μm (average 19 × 8.9 μm). Both species inhabited the surface of the covers of *C. scopulorum*, *C. parietina*, and *D. gypsophila* in the lower part of the turf; they were not observed directly on the film. In addition to encapsulated forms of diatoms protected from the aggressive environmental impacts by their secreted polymers (polysaccharides), the community structure was marked abundantly by four species of sessile (attached) diatoms, which used that as a substrate (and cover) sheath in the turf bases of large species *Calothrix* and *D. gypsophila*. These were *Halamphora* aff. *coffeaeformis*, *Halamphora* aff. *tenerrima*, *Halamphora* aff. *luciae*, and *Cocconeis* aff. *neothumensis*.

The sample-averaged parameters of the length of the frustules of non-encapsulated sessile forms did not exceed 15 μm (maximum – no more than 21 μm). These were relatively small species that were compactly placed under the cover of polymer sheaths of cyanobacteria. Especially dense and widely extended on the substrate, colonies – almost closed "carapace" coverings – developed small *Cocconeis* aff. *neothumensis* cells. Cells of other species were rarely present in its colonies. The colonies of small *H.* aff. *tenerrima* and *H.* aff. *luciae* had the character of chains rising along the sheaths of *Calothrix* spp. Medium-sized *H.* aff. *coffeaeformis* almost did not develop colonies. They were found mainly in the peripheral area of other species colonies. The approximate ratio of species by abundance (in %) can be expressed as follows: *C.* aff. *Neothumensis*: *H.* aff. *Tenerrima*: *H.* aff. *Coffeaeformis*: *H.* aff. *Luciae* ≈ 32.71: 44.86: 17.29: 5.14. Note that these species were not marked directly on the PE film. Single cells of highly mobile species of *Navicula pontica* and *Nitzschia dissipata* were also found in the microplaston.

Thus, the cenosis on the surface of the PE film studied during the formation of the maximum diversity of microphyte fouling in the Black Sea supralittoral (in early October) was much inferior in terms of the richness of flora to the population on the surface of rocks irrigated by surf. However, it should be noted that this extremotolerant population was characterized by a significant ordering of components: the presence of layerings, microbiotopic localization of certain species, and dimensional modifications of others. This architecture of the population indicates the formation of specific adaptations of microphytes to inhabit a poorly studied PE substrate in the Black Sea, which is very widespread in its water area.

We have reason to believe that compact groups of trichomes growing on deformed areas of the PE film surface, including cracks, could have a mechanical impact on the substrate, leading to its destruction. Their tight attachment and expansion in width could contribute to the mechanical expansion of deformations, and taking into account the loss of elasticity of the film, this should lead to deepening and expansion of cracks. An additional abrasive impact on the substrate could be caused by diatomaceous fouling, which developed in bulk on the covers in the bases of the images.

The periphyton of different types of marine plastic in the Eastern Mediterranean Sea was studied by us using materials from the coastal zone of Crete [129], and from the Eastern coastal zone of the Aegean Sea [133].

In August 2018, the fragmented remains of PET bottles were found on the rocky coast of Crete. They were torn into large flaps with lobed, finely torn edges. All samples were collected on the rock bottom of water-filled pits bordering the rocky shore, at a depth of about 3 m. The surface of these plastic fragments was densely covered with whitish-brown-pink-crimson spotty formations of crustal growth [121]. Here, we first encountered polymer growth with the participation of crustal macro-algae.

The macroscopic appearance of the community was formed by two types of crustal macrophytes: *Lithophyllum byssoides* and *Hydrolithon boreale.* On PET, these two crustal red algae cells formed almost continuous "frustules" here, filling up to 80% of the substrate surface. Initially, in natural biotopes, the first type grows within the lower splash zone on coastal rocks. Here the waves beat against the rocks even with minimal rolling, and *L. byssoides* grows in a continuous strip of densely folded limestone layers of pink-scarlet color. The habitual biotope for the second red algae – the surface of the leaves of the sea grass *Cymodocea nodosa*, which grows at depths of 5–8 m. In addition, a few micro-curtains of crustal green Alga *Ulvella scutata*, as well as red *Acrochaetium parvulum* and *Acrochaetium hlulekaense*, which tend to creep and rise above the surface of the substrate, were observed on PET.

Occasionally small shoots of filamentous algae *Polysiphonia* sp. and *Rhizoclunium* sp. were found, up to 0.7 cm high. Large coenocytic globular cells of the green alga *Blastophysa rhizopus* – or cells with irregular, curved lobes, 30–90 μm in diameter – were scattered singly or in small clusters both directly on the PET and along the cracks of the layers of calcareous purple. Clusters of small, up to 200–300 μm, palm-shaped colonies of cyanobacteria (*Chroococcus varius* and *Aphanocapsa litoralis*) were also localized within these cracks (breaks).

The outgrowth also included numerous thin-strichomous (thin-stranded) cyanobacteria *Limnothrix* spp., as well as rare trichomes *Calothrix* aff. *fusca, Spirulina meneghiniana, Leptolyngbya lagerheimii, Oscillatoria crassa*, *Hyella tenuior*, and *Pseudanabaena* sp.

Free-living and attached diatoms in the mass covered areas of the PET surface that was not affected by macrophyte crusts. These were *Licmophora remulus, L. debilis, Amphora tenuissima, A. wisei, A. ostrearia* var. *vitrea, A. hyalina, A. securicula, Halamphora abuensis, H. subholsatica, Nitzschia nienhuisii*, *N. angularis* var. *affinis, N. spathulata, Brachysira estonarium, Cocconeis molesta, C. scutellum, C. distans* and – occasionally – three species of *Navicula* spp., one of which built tubular colonies.

Mastogloia species, which grew both on crustal red algae and on PET, resided here most frequently and in the widest range of microtopes: *Mastogloia erythraea*, *M. crucicula*, *M. ovata*, *M. ovum-paschale*, *M. acutiuscula var. elliptica*, *M. horvathiana*, *M. cribrosa,* and *M. ovulum.* Among diatoms of other genera, *N. nienhuisii* was especially numerous, marked as short ribbon-like colonies, *C. molesta*, which in some places created monoid "cloak-like coverings" (fields, large extended spotty formations) with almost regular cell arrangement, and *L. debilis*, which grew sparsely on most accessible surfaces.

Large colonies of heterotrophic bacteria that can participate in the formation of the appearance of the fouling were not observed.

The PET surface under the red algae and in areas overgrown with attached diatoms and cyanobacteria bore numerous traces of micro-deforming impacts and micro-abrasions. At the same time, polymer capsules of various *Mastogloia* species were surrounded by microscopic PET folds, among which micro-cracks were noticeable.

Microphyte growth of various types of plastic from bays in the Eastern Aegean Sea was studied on samples obtained in 2019. These were both whole plastic products and fragments of them. All samples were lifted from the bottom in different places, where they lay at depths of 0.4–2 m. Of course, taking into account coastal hydrodynamics and seasonal storms, we cannot say that the growth of these objects occurred from the beginning to the end in the places where they were found. Besides, it was almost impossible to determine the time period during which an item in question was exposed to seawater. This study took into account the characteristics of the substrate in the composition of the plastic (type of polymer), thickness, type of product, and the degree of wear. A total of 22 samples of colorless (transparent) plastic were studied: 4 disposable colorless PP cups (PP cups), 3 thin-walled PET bottles (thin-w (thin-walled) PET bottles), 3 thick-walled PET bottles (thick-w PET bottles), 3 files made out of low-pressure corrugated polyethylene (LDPE files), 3 HDPE bags (LDPE bags), 3 fragments of plastic film (LDPE sheet), and 3 lumps of 0.7 mm thick nylon (nylon) tangled fishing line.

To compare the fouling composition and structure of polymers with natural substrates, we selected five large shells left from Bivalvia molluscs found lying on the shell bottom at a depth of 2–3 m. From each sample of the artificial polymers with a relatively flat surface (hereinafter referred to as “flat sample”), six fragments with an area of 2 × 4 cm were cut out, and 25 pieces of 4 cm long were cut from the fishing line.

Totally, 2 species of cortical macrophytes (*Hydrolithon boreale* and *Myrionema* cf. *latipilosum*), 8 species of cyanoprokaryotes (*Calothrix* cf. *parietina, Calothrix* sp. 1, *Scytonema* sp. 1, *Gloeocapsa* sp. 1, *Merismopedia* cf. *elegans*, *Heteroleibleinia* sp. 1, *Heteroleibleinia* sp. 2, and *Chroococcus* sp. 1), as well as one haptophyte species that forms cortical palmelloid colonies of various densities on solid substrates (*Ruttnera* cf. *lamellosa*) were marked for the entire set of the studied substrates. Against this background, 155 species and subspecies (ssp.) of diatoms were identified, representing 39 genera. Among them, the genus *Mastogloia* was the most diverse (42 species and ssp., or 27.1% of the total diatom flora found – more than a quarter), followed by *Nitzschia* (22 species and ssp., or 14.2%), *Amphora* (12 species and ssp., or 7.7%), *Halamphora* and *Cocconeis* (10 species each, or 6.5%). The rank distribution of the number of genera represented by one or another number of species could be represented in the form of a power function $y = 12.5\times^{-0.836}$ ($R^2 = 0.66$). Genera represented by 1–2 species in the entire sample were predominant.

32 species and subspecies were recorded in diatomaceous taxocenes that inhabited the surface of Bivalvia shells, and on different types of plastic samples – from 5 (PP-cup) to 95 (LDPE-bag) (on average – 39 per each type of samples). In all epiplaston taxocenes, the basis of flora diversity was formed by *Mastogloia* species

(up to 3 to 26, on average – 14.7). Among 6 diatom species recorded in different combinations on 6–7 types of substrates out of 8, 5 belonged to this genus: *Mastogloia decipiens, M. adriatica, M. angulata, M. acutiuscula* var. *elliptica* and *M. similis.* The sixth species was *Berkeleya micans*, not marked only on clam shells and on thick-w PET bottles.

Algo-bacterial cenoses on the surface of the studied samples differed in tiering. The lower layer, which was directly spread out on the substrate, was formed everywhere by extended red algae thallomas *of H. boreale* and the soles of pheophyte *layers of M.* cf. *latipilosum,* as well as flat colonies of cyanobacteria *M.* cf. *elegans* on PP cups. On all substrates in the lower tier, there were numerous encapsulated cells of different *Mastogloia* species, while the volumes of their transparent capsules were not always proportional to the volumes of the cells themselves: for example, in *Mastogloia adriatica*, multilayer polymer covers formed a capsule up to 5–8 times larger than the cell itself. *M. vasta, M. robusta*, and especially the large species *M. recta* were also distinguished by their massive capsules. *Cocconeis*, *Amphora*, and *Halamphora* species were also included in the structure of the lower tier: either directly on the main substrate, or on the surface of *H. boreale*, where the encapsulated *Mastogloia ovalis*, *M. pusilla*, and *M. urveae* could also migrate. Cells of *Rhopalodia, Protokeelia, Fallacia, Auricula, Seminavis,* and *Thalassiophysa* species that lead a semi-attached lifestyle were found mainly on the main polymer substrate, rather than on cortical macrophytes. At the same time, epiphytic *Pteroncola, Pseudogomphonema, Planothidium, Plagiodiscus*, and *Astartiella* resided mainly on extended macrophyte layers.

The second tier of cenoses was formed by large palm-like colonies of *Gloeocapsa* sp. 1 and *Ruttnera* cf. *lamellosa*, which rose above the polymer substrate. The third tier was formed by rising trichomes of *Scytonema* sp. 1 (everywhere except shells and nylon), *C.* cf. *parietina* (on polymers other than nylon and PP cups), and also abundantly, on LDPE files – large trichome (thick-filamentous) bundles of *Calothrix* sp. 1. Thin *Heteroleibleinia* trichomes were included in this layer on the surface of *H. boreale,* sometimes forming a dense fouling. The species *Licmophora, Grammatophora, Guinardia, Striatella, Hyalosira, Achnanhes*, and *Dimergramma* developed as epiphyte species on the ascending *Myrionema* filaments – on different samples of "flat" polymers.

The most powerful and high tier, which evidently indicates the late stages of fouling development, was formed on the surface of "flat samples" of polymers by branched colonies of diatoms *Berkeleya micans*. The same tier included dendritic colonies of *Mastogloia cocconeiformis* (occasionally on thick-w PET bottles) and zigzag chains of *Cyclophora tenuis* (occasionally on nylon and abundantly on LDPE packages). Other species of diatoms traversed freely among the tiers.

To distinguish groups (floristic and coenotic) among diatom taxocenes, 72 species and subspecies found in more than one type of samples were used. The average similarity of these populations in terms of species composition (according to the Sørensen index) was 35.45% – it was relatively low, and populations on different types of substrates differed significantly in composition. However, the sample material did not have a significant overall effect on the composition of the fouling diatoms (Global R: −0.021, $p = 49.8\%$). In turn, the average similarity of populations in the quantitative structure (according to the Bray-Curtis index) was even lower – 26.57%. At the same time, only two groups of taxocenes were identified with a significant level of differences (Global R: 1, $p = 2.9\%$), i.e., populations on all polymer samples were divided into mature, dominated by *Berkeleya micans* (PP cups, LDPE sheets, LDPE files, and thin-w PET bottles) and young, on which fouling from its colonies had not yet developed (thick-w PET bottles and LDPE packages). On nylon, *B. micans* did not develop at all. The material of the samples did not significantly affect the quantitative structure of the populations.

Excluding large upper-tier colonial species from statistical analysis, we obtained an even lower average similarity of taxocenes in the quantitative structure (20.95%). At the same time, the material of the samples again did not have a general reliable effect on the ratio of diatom abundance (Global R: 0.354, $p = 16.6\%$). However, we managed to identify three groups of populations at a level close to highly reliable (Global R: 0.524, $p = 0.4\%$). The first group, with an average intra-group similarity of 33.99%, was characterized by abundant species *Mastogloia adriatica, M. acutiuscula* var. *elliptica, M. similis, M. ovalis, M. decipiens, Rhopalodia guettingeri,* and *Navicula parapontica* (on LDPE bags, nylon and Bivalvia shells); the second (with an average similarity of 37.18%) *Mastogloia decipiens, M. acutiuscula* var. *elliptica, M. pumila, M. ovalis,* and *M. ignorata* (on LDPE files, LDPE sheets, and thin-w PET bottles). The taxocenes on PP cups and thick-w PET bottles were not similar in the structure to each other or to other populations.

We can make a general conclusion that in the coastal zone of the Aegean Sea, different types of artificial polymers developed mature, often multi-tiered and complexly integrated, multi-species algo-bacterial cenoses. In a number of cases, they were similar in the composition and structure to cenoses on Bivalvia shells from the same area. At the same time, the substrate material did not have a significant impact on the organization of populations. We observed clear signs of destruction only on LDPE files, LDPE bags, and LDPE films: in all cases, the surface was covered with wide-scale cracks. Given the scale and shape of micro-cracks, as well as their localization, it can be assumed that their formation was related to the mechanical impact exerted by colonies of microphytes and crustal calcareous macrophytes.

3.2 Microplaston in Experiments: Colonization of Synthetic Polymers in the Natural Environment

In August–September 2018, our group conducted a large-scale experiment with fouling samples on different types of plastic. For this purpose, an experimental set-up was installed in Quarantine Bay of Sevastopol (Black Sea), within the limits of a mussel farm [134].

Primarily, microbial fouling was observed on the surface of a PET bottle floating on the surface of the sea in Quarantine Bay. The bottle's exposure time was 1.5 months, and it was tied with a thin rope to the underwater structure of the mussel farm. Growth had occurred under conditions of periodic eutrophication: every night there was a discharge of household water into the bay from the city's sewage system.

The thickness of the fouling was – in the densest area – about 1.3–1.5 mm. There was also a pronounced layering of the population. However, it was not possible to establish clearly interpreted features of the general spatial structure. The cenosis was formed by mosaically scattered spotty formations with a different spatial organization. It is quite possible that the development of the fouling in conditions of intensive water circulation or turbulence and excessive lighting (as aggressive factors) was fairly flavored with a constant influx of biogenic substances (nitrogen and phosphorus compounds). This regular injection of "fertilizers" helped to reduce competition for nutrient resources when microphytes populated the substrate. For example, in the upper tier there were extensive, almost monoid turf filamentous (trichomous) cyanobacteria *Leptolyngbya foveolarum*, as well as very large colonies of diatoms *Neosynedra provincialis*, formed by branching chains of long cells, in some places entwined, like lianas, with trichomes *Symploca elegans*. Among the colonies of *N. provincialis*, tubular colonies of diatoms such as *Berkeleya* aff. *Sparsa* and *Parlibellus delognei* were often present. Small loose colonies of coccoid cyanobacteria *Asterocapsa salina* and *Chroococcus* cf. *montanus* also developed here, as well as compact aggregates of diatoms *Halamphora eunotia* and *H. obscura.* There were single cells of *Halamphora tenerrima.* We note that these *Halamphora* species also developed in the lowest layer of the population, where they resided in extended colonies, in company with *Amphora helenensis* and *Seminavis strigosa*.

In addition, fully independent spotty formations on the PET surface developed palm-like (slimy) colonies of the diatom *Proschkinia bulnheimii* – their branches in some places penetrated into the "brushwood" of colonies of other species. Among "brushwood" of colonial forms, individual trichomes *Phormidium* sp., *Geitlerinema* sp., *L. foveolarum, Limnothrix* aff. *Pseudovacuolata* and *Spirulina subsalsa*, motile diatoms *Navicula pontica* and *Entomoneis punctulata*, as well as dinoflagellates *Prorocentrum lima* were often found.

Among the microphyte colonies, small colonies of heterotrophic bacteria developed in abundancy.

We see that under the described conditions, a multi-species and structurally diverse periphyton developed on PET – and quite quickly. There were no damages on the PET surface that corresponded to the destructive activity of microphytes.

This fouling was radically different from what we observed on the PE film. In addition, it was much more diverse than the biofilm formed on the concrete scaffolds of the breakwater located in the bay. Here, in the zone of intense impact of the rolling wave, along the conditional line of the water edge, a cenosis of cyanobacteria grew in the composition of *Asterocapsa salina, Placoma vesiculosa, Gloeocapsopsis* sp. 1, *Gloeocapsopsis* sp. 2, and *Calothrix* sp. 1. For the most part, they built compact colonies of very tightly spaced cells immersed in a common polymer matrix.

Of course, the variety of cenoses in this biotope was much more strongly impacted by rolling waves – their kinetic energy when hitting the breakwater blocks could be much higher than that of those that passed under the PET bottle dangling on the surface 200 m from the shore. This factor can be regarded as significantly more extreme than swaying on the waves.

Our other observations of the periphyton of plastic in the Black sea were only experimental. We will discuss them below, in terms of results of experimental research.

Specimens of various types of opaque polymers, such as thin white polylactide (plate thickness 1 mm), thick white polylactide (plate thickness 2.5 mm), porous ultra-high-molecular-weight high-pressure polyethylene (UHMWPE), yellow polypropylene (PP) disposable cups with a thickness of about 0.7 mm (corrugated and smooth), as well as white opaque polyethylene (PE) intended for food storage, were exposed in the form of small, 2–4 cm^2 fragments of rectangular shapes placed co-located (simultaneously, all together) inside cylindrical containers made of transparent PET. The containers themselves were open to the flow of water from both ends, had a diameter of 5.5 cm and a wall thickness of 1 mm. To prevent the samples, residing inside the containers, from falling out of them, the outside of the containers was covered with a thin, yellow fine PP mesh (with a mesh size of 4 mm), which did not interfere with the free flow of water through the container. Containers packed in a chain inside a mesh PP tube were stored at different depths. The first of these was located at a distance of 138–146 ($\pm$15) cm from the surface (based on the length of the container, stored vertically, and small oscillations of the equipment in a storm), while the eighth – in a depth of 1,199–1,209 ($\pm$15) cm. Other six containers were evenly distributed between 1 and 8 in the water column. The sea depth at the point of the experiment was 18 m, so all the samples imitated pelagic plastic debris. The samples were exposed for one and a half months, from the beginning of August till the end of the second decade of September.

Analysis of the composition and structure of microphyte cenoses formed on the surface of samples of opaque polymers exposed inside the installation containers, as well as the walls of the containers themselves (PET) and the mesh (PP) covering the containers outside, revealed a number of common features and characteristic differences in the characteristics of these populations.

1. All polymer samples during the period of exposure in the open water of the Black Sea coastal zone, on a regularly eutrophied section of the water area, were largely overgrown with bacterial films encrusted with limestone. At the same time, bacterial films had already developed on top of the forming diatom and, in some cases, cyanoprokaryotic fouling. Further growth of diatomaceous taxocenes had occurred already under these films, inside their thickness, or, sometimes, on their surface. However, based on the results of the SEM survey, as well as on the analysis of permanent preparations, made by taking into account the dissolution of the bacterial film with chemical reagents, the main part of the diatom fouling was formed on the surface of the polymer samples themselves – that is, it could depend on the chemical composition and nature of the sample surface during its

formation. The walls of the PET containers themselves acquired a matte surface character during the exposure, possibly due to photooxidation and biological fouling together, which is why their fragments, cut out for analysis, were also classified as opaque polymer samples.

2. In total, 74 species and subspecies diatoms were observed in the cenoses formed on all the studied polymers, one of which (*Halamphora abuensis*) was represented by two clearly distinct size morphs. Two species of cyanobacteria from the genus *Aphanocapsa* were also observed on the surface of white polylactide plates.
3. Among the entire population of diatoms, 15 species and subspecies (20.27% of the found diatomaceous flora) were observed on all types of samples. In turn, 21 species and subspecies (28.38% of the flora) were characterized by the occurrence on samples of only one type.
4. Species composition on the surfaces of samples of different types had significant differences. The average similarity of diatomaceous taxocenes by species composition (according to the Sørensen index) was 51.79% – this is the average level of similarity. This means, among the entire set of taxocenes on different specimens of plastic (among all their totality), there were communities both significantly similar to each other in the species composition, as well as very different ones. Multidimensional scaling (MDS-analysis: based on the data of the Sørensen index; Kruskal stress formula: 1; minimum stress: 0.01) and clustering (Cluster-analysis: based on the data of the Sørensen index; cluster mode: group average) were performed on the basis of a sample of 40 species remaining after exclusion from the general list of 15 species marked on all samples, and another 21 species, each of which was marked only on one type of the sample. The results of these procedures revealed two large floral groupings. The first of them developed on the surface of white opaque PE, as well as on the yellow PP disposable cups – both on corrugated and smooth cups. Let's denote this grouping as Af. The second floristic grouping was formed on the surfaces of porous UHMWPE, polylactide (thick and thin plates), as well as on the walls of PET containers and on the PP mesh that was entwined on the containers from outside. We will denote this grouping as Bf. ANOSIM-1 similarity analysis (PRIMER-6 software package) showed the general nature of differences between groups at a significant level ($p = 1.8\%$) with a slight predominance of inter-group differences over intra-group ones (Global $R = 0.523$). Using the SIMPER procedure (the PRIMER-6 software package), sets of species that characterize floral groups were identified. These species were most widely distributed on samples of polymers of the corresponding group.

For the Af grouping, these were: *Tabularia fasciculata*, Amphora helenensis, Brachysira sp. 1, Halamphora subturgida, *Navicula arenaria* var. rostellata, *Nitzschia distans*, Hyalosira sp. 1, Halamphora tenerrima, *Navicula salinicola*, *Nitzschia fonticola*, Berckeleya scopulorum, Licmophora ehrenbergii, and *Licmophora dalmatica*.

For the Bf grouping: *Mastogloia pusilla* var. subcapitata, Halamphora cf. turgida, *Licmophora paradoxa*, *Tabularia fasciculata*, Halamphora holsatica, Halamphora tenerrima, Mastogloia cf. cuneata, *Nitzschia amphibia*, *Navicula salinicola*, Halamphora obscura, Halamphora kolbei, Amphora helenensis, *Nitzschia fonticola*, Halamphora salinicola, Halamphora subturgida, Navicula pontica, Pteroncola inane, Navicula subagnita, Berckeleya scopulorum, Halamphora subsalina, and Halamphora thumensis. The sequence of the species mentioned is determined by their spread in the group's taxocenes.

5. The quantitative structure of taxocenes was studied on the basis of the data we obtained from the data on the relative abundance of species (on the frequency of their occurrence in populations). Against the background of the general thinning of diatom fouling, which had occurred against the background of an increase in the depth of the container location, the change in the ratio of species abundance on samples of a certain type was insignificant, ranging from 12.5% to 27.1%.

To analyze the similarity and differences in the quantitative structure of communities, 55 species distributed on samples of more than one type (from 2 to 8 types of samples) were taken into account. The ubiquitous species in this case did not introduce background statistical noise into the analysis. This did not happen for the reason that, despite the widespread distribution on the samples, their participation in building populations on different samples was not the same: somewhere they developed as dominants, while somewhere as an abundant or isolated species. At the same time, the species recorded on only one specimen were excluded from consideration. It makes sense to talk about them separately, and they are not related to the allocation of taxocene groups in this case.

The analysis of similarities and differences between taxocenes was based on the Bray-Curtis similarity index. The average similarity of diatom populations in the structure (ratio of species by abundance) was 53.11% – this is also the average level. This means that in the sample we examined, there were both very similar taxocenes in the structure and significantly different ones. In turn, the results of MDS-analysis (based on the data of Bray-Curtis similarity index; Kruskal stress formula: 1, minimum stress: 0.01) and Cluster-analysis (based on the data of Bray-Curtis similarity index; cluster mode: group average) revealed 4 groups of taxocenes (or 4 coenotic groups). They differed from each other in the participation of certain species in the formation of their structure. ANOSIM-1 similarity analysis revealed the general nature of such differences at a level close to highly reliable ($p = 0.2\%$). At the same time, inter-group differences clearly prevailed over intra-group differences (Global $R = 0.983$).

In particular, the first coenotic grouping – Az – combined taxocenes formed on the surface of both polylactide samples and both the samples of yellow PP (plastic cups). The SIMPER procedure enabled us to identify sets of species that characterize a particular group. For the Az grouping, these were the following species: *Nitzschia* cf. *palea*, *Berkeleya sparsa*, *Nitzschia* cf. *paleacea*, *Nitzschia dissipata*, *Nitzschia pusilla*, *Halamphora abuensis* morph. 2, *Halamphora abuensis* morph. 1, *Nitzschia inconspicua*, *Berckeleya micans*, *Navicula duerrenbergiana*, *Mastogloia cuneata*, and *Navicula perminuta*. In the sets, species are ranked by their average contribution to the similarity of populations within the group – by their participation in the organization of the taxocene structure.

The second grouping – Bz – includes populations formed on the walls of the containers (PET) and on the outer grid (PP). They were characterized by the following species: *Halamphora abuensis* morph. 1, *Berkeleya sparsa*, *Nitzschia* cf. *paleacea*, *Halamphora abuensis* morph. 2, *Nitzschia* cf. *palea*, *Nitzschia pusilla*, *Halamphora subturgida*, *Berckeleya micans*, *Nitzschia dissipata*, *Mastogloia cuneata*, *Mastogloia pusilla* var. *subcapitata* and *Halamphora gasseae*.

The third group – Cz – consisted of taxocenes on samples of porous UHMWPE. It was characterized by the following species: *Berkeleya sparsa, Nitzschia* cf. *palea, Nitzschia pusilla, Mastogloia pusilla* var. *subcapitata, Halamphora abuensis* morph. 2*, Halamphora abuensis* morph. 1*, Nitzschia dissipata, Berckeleya micans, Mastogloia cuneata, Halamphora subsalina, Halamphora thumensis, Nitzschia* cf. *paleacea, Navicula duerrenbergiana,* and *Halamphora gasseae.*

Finally, the fourth group of populations – Dz – was formed by fouling on samples of white opaque PE. Species in the following sequence were essential for their structure: *Nitzschia dissipata, Nitzschia pusilla, Mastogloia cuneata, Halamphora abuensis* morph. 1*, Nitzschia* cf. *palea, Mastogloia pusilla* var. *subcapitata, Nitzschia fonticola, Hyalosira* sp. 1*, Halamphora abuensis* morph. 2*, Halamphora salinicola, Nitzschia inconspicua, Halamphora gasseae, Halamphora* cf. *sardinensis, Berkeleya sparsa, Psammodictyon panduriforme, Tabularia fasciculata, Nitzschia* cf. *paleacea, Navicula duerrenbergiana, Brachysira* sp. 1*, Halamphora obscura, Pteroncola inane, and Staurosira punctiformis.*

We can draw two primary conclusions based on these results.

First, 74 species and subspecies of diatoms were found on the exposed polymer samples – this is quite a lot, the overall species richness of the microplaston flora was high. In the general list of species, there were 15, for which it did not matter on which polymer to grow. 21 species and subspecies were also marked (more than a quarter of the total list), which, on the contrary, were significantly selective in their preferences for the substrate. Based on the distribution of species found on opaque samples of more than one type, but not all (from 2 to 7 types of samples), we can distinguish two population groups of diatom-foulings on the composition of the flora: Af – on disposable polypropylene dishes and white polyethylene, and Bf – on PET, UHMWPE, polylactide and PP2 – non-food materials.

Second, the chemical composition of the polymer samples, as well as the nature of their surface, influenced the quantitative structure of diatom fouling. At the same time, despite the similar composition of the sets of abundant and dominant species that developed populations, they differ in the sequence of species significance in the formation of the appearance of taxocenes. The fouling in each container included species that preferred only a certain type of polymer. There were also widespread species that successfully overgrew certain samples to varying degrees, regardless of their physically close location within the container.

4 Conclusions

The results of our colleagues' research on micro-fouling of synthetic polymers in different water bodies of the World, presented in Sect. 2 of this chapter, allow us to make a series of generalizations:

1. Microorganisms have not bypassed the plastic side, assimilating substrates in the water areas that may be suitable for life.
2. Colonization of plastic by microorganisms in water areas occurs. It wasn't obvious, though. Of course, theoretically it can be assumed that any material, being in water, can serve as a substrate for living things. However, in practice this is not always or not immediately turns out to be the case.
3. The colonization of various types of plastic by microorganisms in the aquatic environment has been studied to different degrees.
4. Travel of microorganisms on plastic particles through seas and oceans can lead to their dispersal across the World. It is not yet entirely clear whether such movements could be the reason of biological invasions. However, microplaston can aggravate the harmful effects of plastic waste through the transfer and possible dispersal of alien and toxic species that can enter food webs in new places.
5. There are indications of the toxic effects of plastic on the growth and functioning of microorganisms. Generally, smaller microplastics are more toxic to microalgae. Most studies show an insignificant or weakly recorded level of influence of microplastics on microalgae, at least at the current concentration of its particles in the environment.
6. There is reason to believe that plastic can cause toxicity of some potentially toxic microphytes. A direct correlation has been shown between the abundance of such microphytes colonizing the surface of plastic and their production of toxins.
7. In pure cultures, species of bacteria have been isolated that can use some types of plastic as carbon sources. However, it is still unknown to what extent they can exhibit these properties in natural conditions.
8. The most numerous group of microscopic eukaryotes that can inhabit plastic are diatoms.
9. The current level of knowledge about the biodegradation of plastic with the participation of microorganisms is still replete with gaps.

The results of our observations allow us to describe the interactions between microphytic fouling (microplaston) and synthetic polymers that enter natural water bodies for various reasons:

1. Microphytes from benthic communities are able to colonize and mechanically destroy a synthetic polymer substrate. These are microalgae species adapted to life on the surface of solid substrates for a long series of generations. In the coastal zone, they enter the pelagic zone and spread to the very surface during storms, when waves stir the soil at the bottom surface. In the process of settling, they come to the plastic. Some of them are able to live and reproduce on its surface, others are not.

2. Not all species of benthic or periphytic microphytes inhabiting the area where the growth of marine plastic debris is formed are able to colonize polymers.
3. Different polymers in the same habitat can accommodate different combinations of benthic or periphytic species.
4. Colonial settlements of microphytes in polymer areas of different microlandscapes have different morphology – and, quite often, are formed with the participation of different species of microalgae.
5. Microphytes, which are mobile on natural substrates, often lose the ability to move on polymers – and form colonial settlements similar to those of initially immobile forms leading an attached lifestyle.
6. The spatial organization of colonial microphyte settlements is capable of governing the mechanisms and forms of polymer destruction.

In conclusion, it is important to note that plastic has been entering the ecosystems of the planet's water bodies for many decades and interacts with the inhabitants of the seas and fresh waters. However, by now we are only in the beginning of a journey in most areas of research highlighting these interactions. But experience shows that these areas are rapidly developing against the backdrop of environmental challenges and risks posed by plastic to aquatic communities and – through them – to humans.

Acknowledgments This work was supported by RFBR Grant 18-44-920012/18 and RFBR Grant 19-55-80004. The work was performed at User Facilities Center of M.V. Lomonosow Moscow State University under financial support of Ministry of Education and Science of Russian Federation. The authors express their deep gratitude for the translation of the text to the agency "Lingvohaus" and personally to Anna Zheleznyak – an extra-class translator.

References

1. Andrady AL, Song Y (1991) Fouling of floating plastic debris. Under Biscayne bay exposure conditions. Mar Poll Bull 22:117–122
2. Stevens LM (1992) Marine plastic debris: fouling and degradation. A thesis submitted in partial fulfillment of the requirements for the degree of master of science in environmental science and zoology. University of Auckland, Auckland, p 110
3. Barnes DKA, Galgani F, Thompson RC, Barlaz M (2009) Accumulation and fragmentation of plastic debris in global environments. Philos Trans R Soc Lond Ser B Biol Sci 364:1985–1998
4. Andrady AL (2015) Persistence of plastic litter in the oceans. In: Bergmann M, Gutow L, Klages M (eds) Marine anthropogenic litter, pp 57–72
5. Gracia CA, Rangel-Buitrago N, Flórez P (2018) Beach litter and woody-debris colonizers on the Atlantico department Caribbean coastline, Colombia. Mar Pollut Bull 128:185–196
6. Harrison HP, Sapp M, Schratzberger M, Osborn AM (2011) Interactions between microorganisms and marine microplastics: a call for research. Mar Technol Soc J 45:12–20
7. Zettler ER, Mincer TJ, Amaral-Zettler LA (2013) Life in the "Plastisphere": microbial communities on plastic marine debris. Environ Sci Technol 47:7137–7146
8. Auta HS, Emenike CU, Fauziah SH (2017) Distribution and importance of microplastics in the marine environment: a review of the sources, fate, effects, and potential solutions. Environ Int 102:165–176

9. Andrady AL (2011) Microplastics in the marine environment. Mar Pollut Bull 62:1596–1605
10. Thompson RC, Olsen Y, Mitchell RP, Davis A, Rowland SJ, John AW, McGonigle D, Russell AE (2004) Lost at sea: where is all the plastic? Science 304:838
11. Jacquin J, Cheng J, Odobel C, Conan P, Pujo-pay M, Meistertzheim A, Jean-francois G (2018) Microbial ecotoxicology of marine plastic debris: a review on colonization and biodegradation by the ' plastisphere'. Front. Microbiol 10:865
12. Amaral-Zettler LA, Zettler ER, Mincer TJ (2020) Ecology of the plastisphere. Nat Rev Microbiol 18:139–151
13. Jambeck JR, Geyer R, Wilcox C, Siegler TR, Perryman M, Andrady A et al (2015) Plastic waste inputs from land into the ocean. Science 347:768–771
14. Charette M, Smith W (2010) The volume of Earth's Ocean. Oceanogr. 23
15. Hidalgo-Ruz V, Gutow L, Thompson RC, Thiel M (2012) Microplastics in the marine environment: a review of the methods used for identification and quantification. Environ Sci Technol 46(6):3060–3075
16. Eriksen M, Lebreton LCM, Carson HS, Thiel M, Moore CJ, Borerro JC et al (2014) Plastic pollution in the world's oceans: more than 5 trillion plastic pieces weighting over 250,000 tons Afloat Sea. PLoS One 9:1–15
17. Duis K, Coors A (2016) Microplastics in the aquatic and terrestrial environment: sources (with a special focus on personal care products), fate and effects. Environ Sci Eur 28:2
18. Zalasiewicz J, Waters CN, Sul JAI, Corcoran PL, Barnosky AD, Cearreta A et al (2016) The geological cycle of plastics and their use as a stratigraphic indicator of the Anthropocene. Anthropocene 13:4–17
19. Willis K, Denise Hardesty B, Kriwoken L, Wilcox C (2017) Differentiating 874 littering, urban runoff and marine transport as sources of marine debris in coastal and estuarine environments. Sci Rep 7:44479
20. Worm B, Lotze HK, Jubinville I, Wilcox C, Jambeck J (2017) Plastic as a persistent marine pollutant. Annu Rev Environ Resour 42:1–26
21. Bakir A, Rowland SJ, Thompson RC (2014) Enhanced design of persistent organic pollutants from microplastics under simulated physiological conditions. Environ Pollut 185:16–23
22. Gewert B, Plassmann MM, Macleod M (2015) Pathways for degradation of plastic polymers floating in the marine environment. Environ Sci Process Impacts 17:1513–1521
23. Ghiglione JF, Martin-Laurent F, Stachowski-Haberkorn S, Pesce S, Vuilleumier S (2014) The coming of age of microbial ecotoxicology: report on the first two meetings in France. Environ Sci Pollut Res Int 21:14241–14245
24. Ghiglione JF, Martin-Laurent F, Pesce S (2016) Microbial ecotoxicology: an emerging discipline facing contemporary environmental threats. Environ Sci Pollut Res 23:3981–3983
25. Debroas D, Mone A, Ter Halle A (2017) Plastics in the North Atlantic garbage patch: a boat microbe for hitchhikers and plastic degraders. Sci Total Environ 599:1222–1232
26. Dussud C, Meistertzheim AL, Conan P, Pujo-Pay M, George M, Fabre P, Coudane J, Higgs P, Elineau A, Pedrotti ML et al (2018) Evidence of niche partitioning among bacteria living on plastics, organic particles and surrounding seawaters. Environ Pollut 236:807–816
27. Lobelle D, Cunliffe M (2011) Early microbiofilm formation on marine plastic debris. Mar Pollut Bull 62:197–200
28. Oberbeckmann S, Loeder MGJ, Labrenz M (2015) Marine microplastic- associated biofilms – a review. Environ Chem 12:551–562
29. Dussud C, Hudec C, George M, Fabre P, Higgs P, Bruzaud S et al (2018) Colonization of non-biodegradable and biodegradable plastics by marine microorganisms. Front Microbiol 9:1571
30. Bryant JA, Clemente TM, Viviani DA, Fong AA, Thomas KA, Kemp P et al (2016) Diversity and activity of aommunities inhabiting plastic debris in the North Pacific gyre. mSystems 1
31. Oberbeckmann S, Kreikemeyer B, Labrenz M (2018) Environmental factors support the formation of special bacterial assemblages on microplastics. Front Microbiol 8:2709

32. Kettner MT, Rojas-Jimenez K, Oberbeckmann S, Labrenz M, Grossart H-P (2017) Microplastics alter composition of fungal communities in aquatic ecosystems. Environ Microbiol 19:4447–4459
33. Cózar A, Echevarria F, Gonzalez-Gordillo JI, Irigoien X, Ubeda B, Hernandez-Leon S et al (2014) Plastic debris in the open ocean. Proc Natl Acad Sci 111:10239–10244
34. Lebreton L, Slat B, Ferrari F, Sainte-Rose B, Aitken J, Marthouse R et al (2018) Evidence that the great Pacific garbage patch is rapidly accumulating plastic. Sci Rep 8:4666
35. Fazey FMC, Ryan PG (2016) Biofouling on buoyant marine plastics: an experimental study into the effect of size on surface longevity. Environ Pollut 210:354–360
36. Kalogerakis N, Karkanorachaki K, Kalogerakis GC, Triantafyllidi EI, Gotsis AD, Partsinevelos P et al (2017) Microplastics generation: onset of fragmentation of polyethylene films in marine environment Mesocosms. Front Mar Sci 4:84
37. De Tender CA, Devriese LI, Haegeman A, Maes S, Ruttink T, Dawyndt P (2015) Bacterial community profiling of plastic litter in the Belgian part of the North Sea. Environ Sci Technol 49:9629–9638
38. De Tender CA, Devriese LI, Haegeman A, Maes S, Vangeyte J, Cattrijsse A et al (2017) Temporal dynamics of bacterial and fungal colonization on plastic debris in the North Sea. Environ Sci Technol 51:7350–7360
39. Lee J-W, Nam J-H, Kim Y-H, Lee K-H, Lee D-H (2008) Bacterial communities in the initial stage of marine biofilm formation on artificial surfaces. J Microbiol 46:174–182
40. Pompilio A, Piccolomini R, Picciani C, D'Antonio D, Savini V, Di Bonaventura G (2008) Factors associated with adherence to and biofilm formation on polystyrene by Stenotrophomonas maltophilia: the role of cell surface hydrophobicity and motility. FEMS Microbiol Lett 287:41–47
41. Dang H, Lovell CR (2016) Microbial surface colonization and biofilm development in marine environment. Microbiol Mol Biol Rev 80:91–138
42. Oberbeckmann S, Loeder MG, Gerdts G, Osborn AM (2014) Spatial and season variation in diversity and structure of microbiofilms on marine plastics in northern European waters. FEMS Microbiol Ecol 90:478–492
43. Briand J-F, Djeridi I, Jamet D, Coupé S, Bressy C, Molmeret M, Le Berre B, Rimet F, Bouchez A, Blache Y (2012) Pioneer marine biofilms on artificial surfaces included antifouling coatings immersed in two contrasting French Mediterranean coast sites. Biofouling 28:453–463
44. Harrison JP, Schratzberger M, Sapp M, Mark Osborn A (2014) Rapid bacterial colonization of low-density polyethylene microplastics in coastal sediment microcosms. BMC Microbiol 14:232
45. Eich A, Mildenberger T, Laforsch C, Weber M (2015) Biofilm and diatom succession on polyethylene (PE) and biodegradable plastic bags in two marine habitats: early signs of degradation in the pelagic and benthic zone? PLoS One 10(9):e0137201
46. Dussud C, Meistertzheim A-L, Conan P, Pujo-Pay M, George M, Fabre P et al (2018) Evidence of niche partitioning among bacteria living on plastics, organic particles and surrounding seawaters. Environ Pollut 236:807–816
47. Webb HK, Crawford RJ, Sawabe T, Ivanova EP (2009) Poly(ethylene terephthalate) polymer surfaces as a substrate for bacterial attachment and biofilm formation. Microbes Environ 24:39–42
48. Pramila R, Ramesh KV (2011) Biodegradation of low density polyethylene (LDPE) by fungi isolated from municipal landfill area. J Microbiol Biotechnol 1:131–136
49. Oberbeckmann S, Osborn AM, Duhaime MB (2016) Microbes on a bottle: substrate, season and geography influence community composition of microbes colonizing marine plastic debris. PLoS One e0159289:11
50. Pomeroy LR, Le B, Williams PJ, Azam F, Hobbie JE (2007) The microbial loop. Oceanography 20:28–33
51. Fenchel T (2008) The microbial loop – 25 years later. J Exp Mar Biol Ecol 366:99–103

52. Hutchins DA, Fu F (2017) Microorganisms and ocean global change. Nat Microbiol 2:17058
53. Goldstein MC, Carson HS, Eriksen M (2014) Relationship of diversity and habitat area in North Pacific plastic-associated rafting communities. Mar Biol 161:1441–1453
54. Kirstein IV, Kirmizi S, Wichels A, Garin-Fernandez A, Erler R, Löder M et al (2016) Dangerous hitchhikers? Evidence for potentially pathogenic Vibrio 690 spp. on microplastic particles. Mar Environ Res 120:1–8
55. Keswani A, Oliver DM, Gutierrez T, Quilliam RS (2016) Microbial hitchhikers on marine plastic debris: human exposure risks at bathing waters and beach environments. Mar Environ Res 118:10–19
56. Amaral-Zettler LA, Zettler ER, Slikas B, Boyd GD, Melvin DW, Morrall CE, Proskrowski G, Mincer TJ (2015) The biogeography of the plastisphere: implications for policy. Front Ecol Environ 13:541–546
57. Hoeksema BW (2012) Evolutionary trends in onshore-offshore distribution patterns of mushroom coral species (Scleractinia: Fungiidae). Contrib Zool 81(4):199–221
58. Barnes DKA, Fraser KPP (2003) Rafting by five phyla on man-made flotsam in the Southern Ocean. Mar Ecol Prog Ser 262:289–291
59. Harrison JP, Boardman C, O'Callaghan K, Delort A-M, Song J (2018) Biogradability standards for carrier bags and plastic films in aquatic environments: a critical review. R Soc Open Sci 5:171792
60. Krueger MC, Harms H, Schlosser D (2015) Prospects for microbiological solutions to environmental pollution with plastics. Appl Microbiol Biotechnnol 99:8857–8874
61. Cosgrove L, Mc Geechan PL, Robson GD, Handley PS (2007) Fungal communities associated with degradation of polyester polyurethane in soil. Appl Environ Microbiol 73:5817–5824
62. Koitabashi M, Noguchi Masako T, Sameshima-Yamashita Y, Kitamoto HK et al (2012) Degradation of biodegradable plastic mulch films in soil environment by phylloplane fungi isolated from gramineous plants. AMB Express 2:40
63. Gajendiran A, Krishnamoorthy S, Abraham J (2016) Microbial degradation of low-density polyethylene (LDPE) by Aspergillus clavatus strain JASK1 isolated from landfill oil. 3 Biotech 6:52
64. Magnin A, Hoornaert L, Pollet E, Laurichesse S, Phalip V, Avérous L (2018) Isolation and characterization of different promising fungi for biological waste management of polyurethanes. Microbial Biotechnol 12(3):544–555
65. Gonda KE, Jendrossek D, Molitoris HP (2000) Liebezeit G, Dittmann S, Kröncke I (eds) Fungal degradation of the thermoplastic polymer poly-ß-hydroxybutyric acid (PHB) under simulated deep sea pressure BT – life at interfaces and under extreme conditions. Springer, Dordrecht, pp 173–183
66. Shah AA, Hasan F, Hameed A, Ahmed S (2008) Biological degradation of plastics: a composite review. Biotechnnol Adv 26:246–265
67. Mohee R, Unmar GD, Mudhoo A, Khadoo P (2008) Biogradability of biodegradable/degradable plastic materials under aerobic and anaerobic conditions. Waste Manag 28:1624–1629
68. Kale SK, Deshmukh AG, Dudhare MS, Patil VB (2015) Microbial degradation of plastic: a review. J Biochem Technol 6:952–961
69. Bhardwaj H, Gupta R, Tiwari A (2013) Communities of microbial enzymes associated with biodegradation of plastics. J Polym Environ 21:575–579
70. Pathak VM (2017) Review on the current status of polymer degradation: a microbial approach. Bioresour Bioprocess 4:15
71. İpekoglu B, Böke H, Çizer Ö (2007) Assessment of material use in relation to climate in historical buildings. Build Environ 42:970–978
72. Dussud C, Ghiglione J-F (2014) Bacterial degradation of synthetic plastics. In: Briand F (ed) Marine litter in the Mediterranean and black seas. CIESM Publisher, p 180
73. Caruso G (2020) Microbial-colonization in marine environments: overview of current knowledge and emerging research topics. J Mar Sci Eng 8(2):78

74. Moore CJ, Moore SL, Weisberg SB, Lattin GL, Zellers AF (2002) A comparison of neustonic plastic and zooplankton abundance in southern California's coastal waters. Mar Pollut Bull 44:1035–1038
75. Lattin GL, Moore CJ, Zellers AF, Moore SL, Weisberg SBA (2004) Comparison of neustonic plastic and zooplankton at different depths near the southern California shore. Mar Pollut Bull 49:291–294
76. Cole M, Lindeque P, Halsband C, Galloway TS (2011) Microplastics as contaminants in the marine environment: a review. Mar Pollut Bull 62:2588–2597
77. Ballent A, Pando S, Purser A, Juliano MF, Thomsen L (2013) Modelled transport of benthic marine microplastic pollution in the Nazaré canyon. Biogeosciences 10:7957–7970
78. Browne MA, Galloway T, Thompson R (2007) Microplastic – an emerging contaminant of potential concern? Integrr Environ Assess Manag 3:559–566
79. Corcoran PL (2015) Benthic plastic debris in marine and fresh water environments. Environ Sci Process Impacts 8:1363–1369
80. Corcoran PL, Biesinger MC, Grifi M (2009) Plastics and beaches: a degrading relationship. Mar Pollut Bull 58:80–84
81. Cai L, Wu D, Xia J, Shi H, Kim H (2019) Influence of physicochemical surface properties on the adhesion of bacteria onto four types of plastics. Sci Total Environ 671:1101–1107
82. Carson HS, Nerheim MS, Carroll KA, Eriksen M (2013) The plastic-associated microorganisms of the North Pacific gyre. Mar Pollut Bull 75:126–132
83. Reisser J, Shaw J, Hallegraeff G, Proietti M, Barnes DK, Thums M, Wilcox C, Hardesty BD, Pattiaratchi C (2014) Millimeter-sized marine plastics: a new pelagic habitat for microorganisms and invertebrates. PLoS One 9:e100289
84. Salta M, Wharton JA, Blache Y, Stokes KR, Briand JF (2013) Marine biofilms on artificial surfaces: structure and dynamics. Environ Microbiol 15:2879–2893
85. Khandeparker L, D'Costa PM, Anil AC, Sawant SS (2014) Interactions of bacteria with diatoms: influence on natural marine biofilms. Mar Ecol 35:233–248
86. Amin SA, Parker MS, Armbrust EV (2012) Interactions between diatoms and bacteria. Microbiol Mol Biol Rev 76:667–684
87. Abell GC, Bowman JP (2005) Colonization and community dynamics of class Flavobacteria on diatom detritus in experimental mesocosms based on southern ocean seawater. FEMS Microbiol Ecol 53:379–391
88. Delacuvellerie A, Cyriaque V, Gobert S, Benali S, Wattiez R (2019) The plastisphere in marine ecosystem hosts potential special microbial degraders inclusion Alcanivorax borkumensis as a key player for the low-density polyethylene degradation. J Hazard Mater:380
89. Hung OS, Thiyagarajan V, Zhang R, Wu RSS, Qian PY (2007) Attachment of Balanus amphitrite larvae to biofilms originating from contrasting environments. Mar Ecol Prog Ser 333:229–242
90. Balasubramanian V, Palanichamy S, Subramanian G, Rajaram R (2012) Development of polyvinyl chloride biofilms for succession of selected marine bacterial populations. J Environ Biol 33:57–60
91. Briand J-F, Barani A, Garnieri C, Réhel K, Urvois F, LePoupon C, Bouchez A, Debroas D, Bressy C (2017) Spatio-temporal variations of marine biofilm communities colonizing artificial substrata including antifouling coatings in contrasted French coastal environments. Microb Ecol 74:585–598
92. Misic C, Harriague AC (2019) Development of marine biofilm on plastic: ecological features in different seasons, temperatures, and light regimes. Hydrobiologia 835:129–145
93. O'Connor NJ, Richardson DL (1998) Attachment of barnacle (Balanus amphitrite Darwin) larvae: responses to bacterial films and extracellular materials. J Exp Mar Biol Ecol 226:115–129
94. Leroy C, Delbarre-Ladrat C, Ghillebaert F, Rochet MJ, Compère C, Combes D (2007) A marine bacterial adhesion microplate test using the DAPI fluorescent dye: a new method to screen antifouling agents. Lett Appl Microbiol 44:372–378

95. Chiu JMY, Thiyagarajan V, Tsoi MMY, Qian PY (2005) Qualitative and quantitative changes in marine biofilms as a function of temperature and salinity in summer and winter. Biofilms 2:183–195
96. Xu H, Min G-S, Choi J-K, Jung J-H, Park M-H (2009) An application to analysis of periphytic ciliate colonization for monitoring water quality using a modified artificial substrate in Korean coast waters. Mar Pollut Bull 58:1278–1285
97. Jeong S, Kim J-A, Kim H, Chung K-S, Yoon H-O (2018) Identification of preponderant marine bacteria and their biofouling charteristics on adsorbents of different sizes and shapes in seawater. J Mar Sci Technol 26:458–464
98. Lee YK, Kwon KK, Cho KH, Park JH, Lee HK (2005) Isolation and identification of Bacteria from marine biofilms. Key Eng Mater 277:612–617
99. Dobretsov S, Abed RM, Teplitski M (2013) Mini-review: inhibit of biofouling by marine microorganisms. Biofouling 29:423–441
100. Mejdandžić M, Ivanković T, Pfannkuchen M, Godrijan JM, Pfannkuchen DM, Hrenović J, Ljubešić Z (2015) Colonization of diatoms and bacteria on artificial substrates in the northeastern coastal Adriatic Sea. Acta Bot Croat 74:407–422
101. Prata JC, da Costa JP, Lopes I, Duarte AC, Rocha-Santos T (2019) Effects of microplastics on microalgae populations: a critical review. Sci Total Environ 665:400–405
102. Lagarde F, Oliver O, Zanella M, Daniel P, Hiard S, Caruso A (2016) Microplastic interactions with freshwater microalgae: hetero-aggregation and changes in plastic density approach strongly independent on polymer type. Environ Pollut 215:331–339
103. Davarpanah E, Guilhermino L (2015) Single and combined effects of microplastics and copper on the population growth of the marine microalgae *Tetraselmis chuii*. Estuar Coast Shelf Sci 167:269–275
104. Zhang C, Chen X, Wang J, Tan L (2017) Toxic effects of microplastics on marine microalgae *Skeletonema costatum*: interactions between microplastics and algae. Environ Pollut 220:1282–1288
105. Sjollema SB, Redondo-Hasselerharm P, Leslie HA, Kraak MHS, Vethaak AD (2016) Do plastic particles affect microalgal photosynthesis and growth? Aquat Toxicol 170:259–261
106. Bhattacharya P, Lin S, Turner JP, Ke PC (2010) Physical adsorption of charges plastic nanoparticles affect algal photosynthesis. J Phys Chem C 114:16556–16561
107. Casado MP, Macken A, Byrne HJ (2013) Ecotoxicological assessment of silica and polystyrene nanoparticles assessed by a multitrophic test battery. Environ Int 51:97–105
108. Nolte TM, Hartmann NB, Kleijn JM, Garnæs J, van de Meent D, Hendriks AJ, Baun A (2017) The toxicity of plastic nanoparticles to green algae as inflected by surface modification, medium hardness and cellular adsorption. Aquat Toxicol 183:11–20
109. Prata JC, Lavorante BRBO, Montenegro MDBSM, Guilhermino L (2018) Influence of microplastics on the toxicity of the pharmaceuticals procainamide and doxycyline on the marine microalgae Tetraselmis chuii. Aquat Toxicol 197:143–152
110. Besseling E, Wang B, Lürling M, Koelmans AA (2014) Nanoplastic affects growth of S. obliquus and reproduction of D. magna. Environ Sci Technol 48:12336–12343
111. Casabianca S, Capellacci S, Giacobbe MG, Dell'Aversano C, Tartaglione L, Varriale F, Narizzano R, Risso F, Moretto P, Dagnino A, Bertolotto R, Bargbone EN, Penna A (2019) Plastic-associated harmful microalgal assemblages in marine environment. Environ Pollut 244:617–626
112. Ryan P (1988) Effects of ingested plastic on seabird feeding: evidence from chickens. Mar Pollut Bull 19(3):125–128
113. Spear LB, Ainley DG, Ribic CA (1995) Incidence of plastic in seabirds from the tropical pacific, 1984–1991: relation with distribution of species, sex, age, season, year and body weight. Mar Environ Res 40(2):123–146
114. Long M, Moriceau B, Gallinari M, Lambert C, Huvet A, Raffray J, Soudant P (2015) Interactions between microplastics and phytoplankton gregates: impact on their respective fates. Mar Chem 175:39–46

115. Long M, Paul-Pont I, Hégaret H, Moriceau B, Lambert C, Huvet A, Soudant P (2017) Interactions between polystyrene microplastics and marine phytoplankton lead to species-species-specific hetero-aggregation. Environ Pollut 228:454–463
116. Michels J, Stippkugel A, Lenz M, Wirtz K, Engel A (2018) Rapid aggregation of biofilm-covered microplastics with marine biogenic particles. Proc R Soc B Biol Sci 285 (1885):20181203
117. Van Cauwenberghe L, Vanreusel A, Mees J, Janssen CR (2013) Microplastic pollution in deep-sea sediments. Environ Poll 182:495–499
118. Woodall LC, Sanchez-Vidal A, Canals M, Paterson GLJ et al (2014) The deep sea is a major sink for microplastic debris. R Soc Open Sci 1:140317
119. Bergmann M, Wirzberger V, Krumpen T, Lorenz C, Primpke S, Tekman MB, Gerdts G (2017) High quantities of microplastic in Arctic deep-sea sediments from the HAUSGARTEN observer. Environ Sci Technol 51:11000–11010
120. Kiørboe T, Andersen KP, Dam HG (1990) Coagulation efficiency and gregate formation in marine phytoplankton. Mar Biol 107:235–245
121. Engel A (2000) The role of transparent exopolymer parties (TEP) in the increase in apparent particle stickiness (a) during the decline of a diatom bloom. J Plankton Res 22:485–497
122. Foulon V, Le Roux F, Lambert C, Huvet A, Soudant P, Paul-Pont I (2016) Colonization of polystyrene microparticles by *Vibrio crassostreae*: light and electron microscopic investigation. Environ Sci Technol 50:10988–10996
123. Masó M, Fortuño JM, de Juan S, Demestre M (2016) Microfouling communities from pelagic and benthic marine plastic debris sampled across Mediterranean coast waters. Sci Mar 80S1:117–127
124. Dudek KL, Cruz BN, Polidoro B, Neuer S (2020) Microbial colonization of microplastics in the Caribbean Sea. Limnol Oceanogr Lett 5(1):5–17
125. Moog D, Schmitt J, Senger J et al (2019) Using a marine microalga as a chassis for polyethylene terephthalate (PET) degradation. Microb Cell Factories 18:171
126. Kumar RV, Kanna GR, Elumalai S (2017) Biogradation of polyethylene by green photosynthetic microalgae. J Bioemed Biodegrad 8:381
127. Khoironi A, Anggoro S, Sudarno S (2019) Evaluation of the interaction among microalgae Spirulina sp, plastics polyethylene terephthalate and polypropylene in freshwater environment. J Ecol Eng 20(6):161–173
128. Kalinina OY, Sapozhnikov PV (2019) The study results of plastic waste overgrowth with micro-algae. J Environ Earth Energy Study (JEEES) 4:46–53
129. Sapozhnikov PV, Kalinina OY, Snigirova AA (2019) Phytoperyphyton of the marine plastic (pet) near the Crete coast. In: Advances in modern phycology: book of abstracts of the VI international conference 93
130. Snigirova AA, Kurakin AP (2019) Microalgae on the plastic substrates in the coastal area of the Gulf of Odessa (the Black Sea). In: Advances in modern phycology: book of abstracts of the VI international conference 103
131. Sapozhnikov PV, Snigirova AA, Kalinina OY (2020) Microphytes assemblages on the neustoplastics from the north Black Sea. In: Marine litter in the Black Sea marine litter in the Black Sea. Publisher: Turkish Marine Research Foundation (TUDAV) Publication No: 56, Istanbul, Turkey
132. Sapozhnikov PV, Snigirova AA, Kalinina OY (2018) Arkhitektura fitoperifitona polietilenovoy plenki s poverkhnosti Chernogo morya. Vodorosli: problemy taksonomii, ekologii i ispolzovanie v monitoringe. In: Algae: problems of taxonomy, ecologyand use in monitoring: proceedings of the IV all-Russian scientific conference with international participation, pp 378–383 (In Russ.)
133. Sapozhnikov PV, Kalinina OY, Snigirova AA (2019) Algobakterialnye cenozy na poverkhnosti razlichnykh vidov plastic (microplaston) v pribrezhnoj zone Egejskogo morya (Distrikht Bodrum, Turtsia). MARESEDU-2019, pp 351–359 (In Russ.)
134. Ryabushko LI, Sapozhnikov PV, Bondarenko AV, Kalinina OY (2019) Diatom foulings of synthetic polymer materials in Karantinnay Bay (Crimea, the Black Sea). Issues Modern Algol 2(20):87–91

Freshwater Microplastic Pollution: The State of Knowledge and Research

Sarantuyaa Zandaryaa

Contents

1 Global Water Quality Challenges 256
2 Water Quality in the UN 2030 Agenda and Sustainable Development Goals: Relevance to Microplastics in Freshwater 258
3 What Are Microplastics? 258
4 Source and Pathways of Microplastics in Freshwater Environments 261
5 Occurrences of Microplastics in the World's Freshwater Resources 262
  5.1 Microplastics in Lakes 263
  5.2 Microplastics in Rivers 264
  5.3 Microplastics in Groundwater 264
  5.4 Microplastics in Drinking Water 265
6 Health and Ecological Risks of Microplastics in Freshwater: How Harmful Are Microplastics? 265
  6.1 Ecotoxicological Effects 265
  6.2 Potential Human Health Risks 266
  6.3 Microplastics as Carriers and Sources of Other Pollutants in Freshwater 266
7 Reducing Microplastic Pollution in Freshwater Environments 267
8 Knowledge Gaps and Research Needs on Microplastics in Freshwater Environments . . . 268
References 270

Abstract The presence of plastics and microplastics in freshwater ecosystems and biota has been reported in different parts of the world – even in most remote areas. Yet, scientific information on the extent of freshwater microplastic pollution is limited. Comprehensive assessments on plastics and microplastics in freshwater environments at global, regional, and basin scales are lacking. Human health and ecological effects of freshwater microplastic pollution remain unknown. Freshwater microplastic pollution is a new research area recently attracting attention from the academic community. Further research is needed to improve scientific knowledge on

---

S. Zandaryaa (✉)
UNESCO, Division of Water Sciences – Intergovernmental Hydrological Programme, Paris, France
e-mail: s.zandaryaa@unesco.org

Friederike Stock, Georg Reifferscheid, Nicole Brennholt, and Evgeniia Kostianaia (eds.),
Plastics in the Aquatic Environment - Part I: Current Status and Challenges,
Hdb Env Chem (2022) 111: 255–272, DOI 10.1007/698_2021_748,
© Springer Nature Switzerland AG 2021, Published online: 30 March 2021

microplastic pollution in the world's freshwater resources and to develop evidence-based appropriate policies and solutions to reduce microplastics in freshwater environments. The role of international scientific programmes such as the International Initiative on Water Quality of UNESCO's Intergovernmental Hydrological Programme (IHP) will be crucial for research promotion and knowledge generation to fill the knowledge gaps on freshwater microplastic pollution and its human health and ecological impacts.

The chapter focuses on the presence on microplastics in freshwater systems, their sources and pathways and associated potential human health and environmental risks. It presents a summary of the available scientific knowledge and information related to microplastics in freshwater environments, which were reviewed for a preliminary assessment of microplastics in freshwater environments conducted by UNESCO-IHP's International Initiative on Water Quality between 2015 and 2017. Some studies published more recently have been included. It highlights knowledge gaps and research needs on freshwater microplastic pollution and recommends appropriate policies and solutions to prevent and reduce the discharge of microplastics to freshwater environments.

Keywords Freshwater, Microplastics, Pollution, Sustainable development goals, UNESCO, Water quality

1 Global Water Quality Challenges

Water is vital for life on the Earth. Clean and safe water provides a wide range of ecological and societal benefits to humans and economic activities such as clean water supplies, healthy ecosystems and services, safe food produces, good human health and well-being, reduced water-borne diseases and associated economic burden, enhanced livelihoods for farmers and local communities, and opportunities for tourism development and recreational industries.

The quality of the world's freshwater resources is deteriorating due to nutrient, microbial, chemical, and heavy metal pollution. Surface and groundwater resources are heavily polluted by nutrients, resulting from agricultural runoff and discharges of untreated, or insufficiently, treated wastewater. The need for more food production for the growing world population is leading to extensive use of fertilizers and pesticides, which, in turn, is causing increasing nutrient and chemical pollution in freshwater resources. Rapid industrial and mining developments throughout the world represent the main source of heavy metal and chemical pollution in water resources. In addition to these conventional "*primary*" water pollutants, a new global water quality concern is arising over "*emerging pollutants*" found in the world's freshwater resources. Emerging pollutants broadly comprise pharmaceutical residues, substances used in personal care products, hormones, endocrine disruptors, chemicals, and microplastics. Emerging pollutants are not regulated, or routinely monitored. Conventional secondary-level wastewater treatment plants are not effective in fully removing emerging pollutants such as pharmaceuticals [1].

Water pollution is one of the main causes of water-borne diseases with serious human health effects. According to the World Health Organization, 829,000 people die each year from diarrhoea because of unsafe drinking water, sanitation, and hand hygiene. Improving safe drinking water and sanitation services is crucial for reducing deaths resulting from water-borne diseases [2]. In 2017, only 71% of the global population used safely-managed drinking water and just 45% used safely-managed sanitation services, leaving 2.2 billion persons without safe drinking water, including 785 million without even basic drinking water, and 4.2 billion without safely-managed sanitation [3].

Water pollution is also a threat to ecosystems and their functions, goods and services, which are essential to humans, as well as to water supplies, food production, and livelihoods. According to the *IPBES Global Assessment Report on Biodiversity and Ecosystem Services*, water pollution is one of the five key drivers of freshwater ecosystem degradation [4]. A good water quality state in water resources is, therefore, indispensable for the restoration and maintaining of both aquatic and terrestrial ecosystems and the ecosystem integrity.

Plastic pollution in both freshwater resources and oceans calls for an urgent attention due to its magnitude and global extent. In particular, the *IPBES Global Assessment Report on Biodiversity and Ecosystem Services* stresses that, globally, marine plastic pollution has increased tenfold since 1980, affecting at least 267 species, which can also affect humans through food chains [5]. Information on the extent and magnitude of freshwater plastic and microplastic pollution is lacking. Most research on plastic and microplastic pollution conducted during the past decades has focused on marine environments, whereas freshwater microplastic pollution is an emerging research area with very few studies published only recently. Even fewer studies focus on microplastics in freshwater environments. The presence of both plastics and microplastics in freshwater environments may have potentially significant effects on human health and aquatic life.

Freshwater microplastic pollution is more challenging because of its complexity and multidimensional characteristics in terms of sources, pathways, fate, and behaviour of microplastics in aquatic environments. Uncertainties and lack of knowledge about their effects on human health and aquatic organisms represent a major global concern. Therefore, further research in this area is needed to improve the scientific understanding and knowledge of microplastics in freshwater and their health and ecological risks. Monitoring data on microplastic occurrences in freshwater systems and aquatic organisms is also essential for setting evidence-based policies and management priorities to protect water quality, human health, and aquatic life from microplastic pollution.

2 Water Quality in the UN 2030 Agenda and Sustainable Development Goals: Relevance to Microplastics in Freshwater

Protecting and improving water quality is essential for the achievement of the Sustainable Development Goals (SDGs) of the UN 2030 Agenda for Sustainable Development [6]. Reducing microplastic pollution in freshwater ecosystems is essential for the achievement not only of the SDG 6 on clean water and sanitation, but also of a number of other goals.

The SDG 6 "*to ensure the availability and sustainable management of water and sanitation for all*" includes Target 6.3, which is specifically aimed to "*improve water quality by reducing pollution, eliminating dumping and minimizing release of hazardous chemicals and materials, halving the proportion of untreated wastewater and substantially increasing recycling and safe reuse globally*" [3]. The SDG 6 cannot be achieved without concerted action, globally and locally, to reduce water pollution by all types of pollutants, including microplastics.

It is indispensable to improve water quality and reduce water pollution globally for the achievement of several other SDGs. In particular, good water quality is the pre-requisite for the achievement of: SDG 2 on food security; SDG 3 on human health and well-being; SDG 5 on gender equality; SDG 14 on healthy oceans; SDG 15 on ecosystems and biodiversity.

The SDG 12 on responsible production and consumption explicitly calls for "*. . . the environmentally sound management of chemicals and all wastes throughout their life cycle . . .*" in order to "*significantly reduce their release to air, water and soil in order to minimize their adverse impacts on human health and the environment*" [6]. Microplastic pollution in freshwater is of direct relevance for the achievement of this SDG target because microplastics are made of chemicals and their release to the environment and freshwater systems primarily results from the lack of proper management and disposal of plastic wastes.

Table 1 describes the SDGs that are of direct relevance to microplastics in freshwater and oceans.

3 What Are Microplastics?

Plastics are indispensable to society and bring benefits in everyday life – from packaging to various uses in construction, automotive industry, medical devices, and electronics.

Plastics are insoluble in water and designed to be very persistent. These unique properties and low production costs have made plastics the most widely used material. The global production of plastics increased by about 200-fold times since 1950 [8]. However, these beneficial properties – the durability and water-resistance – make it also difficult, or impossible, for nature to assimilate the plastics. Plastics do

Table 1 The Agenda 2030 sustainable development goals related to microplastic pollution in freshwater and oceans

Sustainable Development Goals (SDGs)	SDG targets	Relevance to reducing microplastics in freshwater
SDG 6: *Clean water and sanitation*	*Target 6.3*: Improve water quality by reducing pollution, eliminating dumping and minimizing release of hazardous chemicals and materials, halving the proportion of untreated wastewater and substantially increasing recycling and safe reuse globally	Improving wastewater treatment will lead to significant reduction in microplastic pollution in freshwater ecosystems
	Target 6.6: Protect and restore water-related ecosystems, including mountains, forests, wetlands, rivers, aquifers, and lakes	Protection and restoration of water-related ecosystems requires the prevention and reduction of the release of microplastics to freshwater ecosystems
Other SDGs		
SDG 3: *Good health and well-being*	*Target 3.9*: Substantially reduce the number of deaths and illnesses from hazardous chemicals and air, water and soil pollution and contamination	Some chemicals used in plastics are a human health concern for their toxic, cancerogenic, or endocrine disrupting effects Microplastics in wastewater and freshwater systems may act as carriers of other hazardous pollutants in water such as heavy metals and pharmaceuticals [7]
SDG 11: *Sustainable cities*	*Target 11.6*: Reduce the adverse per capita environmental impact of cities, including by paying special attention to air quality and municipal and other waste management	Lack of municipal waste collection and management is a major source of microplastics in freshwater and coastal waters
SDG 12: *Responsible consumption and production*	*Target 12.4*: Achieve the environmentally sound management of chemicals and all wastes throughout their life cycle, in accordance with agreed international frameworks, and significantly reduce their release to air, water, and soil in order to minimize their adverse impacts on human health and the environment	All types of plastics and microplastics are made of chemicals. Some chemicals used in plastics and microplastics are known to be toxic, or cancerogenic, or hormone (endocrine) disruptors such as phthalates, Bisphenol A (BPA), polyvinyl chloride (PVC), etc.
SDG 14: *Healthy oceans*	*Target 14.1*: Prevent and significantly reduce marine pollution of all kinds, in particular from land-based activities, including marine debris and nutrient pollution	A significant portion of microplastics in freshwater systems are transported to coasts, seas, and oceans via rivers
SDG 15: *Ecosystems and biodiversity*	*Target 15.1*: Ensure the conservation, restoration, and sustainable use of terrestrial and inland freshwater ecosystems and their services, in	Water pollution, including environmental plastics and microplastics, is one of the main drivers of ecosystem degradation and biodiversity

(continued)

Table 1 (continued)

Sustainable Development Goals (SDGs)	SDG targets	Relevance to reducing microplastics in freshwater
	particular forests, wetlands, mountains, and drylands, in line with obligations under international agreements	loss Water quality improvements and reducing microplastics in freshwater, hence, will lead to ecosystem restoration and protecting biodiversity

Source: Original by the Author

not decay or decompose readily in the environment and thus may last in nature for hundreds of years [9]. As a result, overall lifecycles of plastics extend beyond product lifetimes; i.e. plastics remain in the environment for hundreds of years after the effective duration of the intended use of plastic products. This means that all plastics that have ever ended up in the environment will still exist with time in one form or another. Consequently, the very long lifetime and non-biodegradability of plastic materials represent a major environmental concern for adequate management of plastic waste. Plastics that enter the environment either may remain in their original form during a long time or break into small fragments, depending on their types and environmental conditions.

Generally, plastic particles found in the environment that are smaller than 5 mm are referred to as "microplastics" [10]. Yet, there is no internationally accepted definition of microplastics in terms of the size range. A definition of "nanoplastics" has been proposed for plastic particles within the size range from 1 to 1,000 nm that are unintentionally produced (i.e. from the degradation and manufacturing of plastic objects) and presenting a colloidal behaviour [11]. Hence, the definition of "microplastics" encompasses generally plastic particles within the size range between 1 μm (i.e. 1,000 nm) and 5 mm.

Depending on their sources, microplastics are classified as "*primary*" and "*secondary*". Plastics that are manufactured intentionally with sizes smaller than 5 mm are referred to as primary microplastics. Primary microplastics include pellets used in plastic manufacturing and microbeads used in cosmetics and personal care products. Secondary microplastics result from degradation and disintegration of larger plastic items into smaller fragments during their use (such as fibres that shed off during synthetic textile washing) or once after they enter the environment (for example, fragmentation of plastic bags and films). Physical, chemical, and biological forces such as wind, solar radiation, and water currents cause the breakdown of plastic items into smaller fragments, thus resulting in secondary microplastics.

Consequently, microplastics in the environment and freshwater are found in various polymer types and compositions, as well as in different shapes, forms, and sizes. The most common shapes and forms of microplastic particles found in the environment are:

- Fragments: pieces of high-density plastic such as bottles and plastic objects.

- Fibres: fibres of synthetic textile such as clothing and fishing nets.
- Films: small pieces of plastic bags, wrappers.
- Granules and pellets: resin microbeads and pellets.
- Foam: styrofoam pieces such as cushioning, cups, food packaging.

4 Source and Pathways of Microplastics in Freshwater Environments

Microplastics enter the environment and freshwater systems from various sources. Main sources of primary microplastics are the use of personal care and cosmetic products containing plastic microbeads such as toothpaste, scrubs, and gels. Researchers estimated that up to 94,500 microbeads could be released from an exfoliant in a single use [12]. Microplastic pellets – also called nurdles – are the second largest source of primary microplastic pollution. Pellets typically have a diameter of 2–5 mm and are used as raw material for the production and recycling of plastics. Huge amounts of spills of plastic pellets enter the environment and freshwater during their transport, loading, storage, production, and use in industrial facilities. A study estimated the total annual release of pre-production pellets from a polyethylene production facility in Sweden to the surrounding environment and surface water to be between 3 and 36 million (or, between 73 and 730 kg) pellets and these numbers could be multiplied with a factor of hundred and the mass by a factor of three, if smaller fractions of plastic particles down to 300 μm were included [13]. Pellets are not only a concern for their presence in freshwater environments, but also represent a significant vehicle for the transport of metals in aquatic environments [14].

Sources of secondary microplastics include: fragmentation of plastic wastes in the environment; discharge of synthetic fibres from textile during washing; and plastic particles from abrasion of tyres from roads. Fragmentation of plastic debris is the largest source of secondary microplastics in the environment. With the rapid increase of global plastic production during the past decades, the volume of plastic wastes and their inadequate discharge into the environment have increased exponentially too. Only 9% of all plastic ever discarded since 1950 has been recycled, while another 12% has been incinerated [9]. Lack of proper waste collection, management and disposal systems leads to plastic litter entering into surface waters directly or with storm runoff, which over time results in their fragmentation into microplastics. Although data are scarce, another major source of secondary microplastics appears to be fibres from washing clothes with more than 1,900 fibres produced per wash of a single synthetic garment, based on experiments sampling wastewater from domestic washing machines [15]. Rubber particles that are generated from tyre abrasion on roads may represent a significant source of secondary microplastics. It is estimated that between 60,000 and 111,000 t of microplastics enter the environment due to tyre abrasion in Germany each year, which corresponds to 0.75–1.38 kg per capita per year [16].

Microplastics enter freshwater environments via multiple pathways. Plastic waste littering is a principal pathway of microplastics into the freshwater resources. Plastic litter is the most acute environmental problem in many cities in developing countries and has reached an appalling level. Wastewater treatment plants represent another main pathway through which microplastics – mainly microbeads and synthetic fibres – are released into freshwater environments. Studies suggest that microplastics can be removed with efficiency over 85% in wastewater treatment plants with secondary-level treatment [17]. A study on microplastics in wastewater effluents downstream Paris urban area reported a very high efficiency of microplastic removal in the wastewater treatment plant from 469 fibres/L in raw wastewater to 31 fibres/L in treated wastewater [18].

Yet, low levels of wastewater collection and treatment in developing countries can lead to the continuous discharge of huge amounts of microplastics into freshwater with untreated, or insufficiently treated, wastewater. According to UN data on progress in the SDG 6 achievement, only less than half of all household wastewater flows were treated safely in 2019, based on preliminary estimates from 79 high- and higher-middle income countries [3]. No information on wastewater treatment is available in other countries. Hence, wastewater treatment plants represent a major environmental source of microplastics, considering large volumes of wastewater discharged continuously into freshwater environments.

Urban stormwater runoff is another pathway of microplastics entering rivers, lakes, streams, and oceans along with many other pollutants. In addition to rubber particles from tyre abrasion on roads, fragments of plastic paints derived from road marking appear to be washed off with stormwater runoff into surface waters [16, 19]. Extreme weather events such as floods and storms can exacerbate the discharge of microplastics from land into water bodies [20].

5 Occurrences of Microplastics in the World's Freshwater Resources

Freshwater microplastic pollution is a new research area, attracting attention from the academic community in recent years. Available scientific information, albeit scarce, indicates that microplastics are present ubiquitously in freshwater environments. Scientific understanding and data on the presence of microplastics in freshwater environments are scarce. Only recently have few studies started to investigate microplastics in freshwater systems, whereas most studies on microplastics focused on marine systems.

Microplastics in freshwater environments are studied predominantly in lakes and large rivers, based on the review of available research data on microplastics in freshwater between 1995 and 2016 [21]. Concentrations of microplastics in the water and sediments of freshwater lakes and rivers vary, showing a large variability among different studies. The available data are not comparable due to the use of different sampling methods and data metrics in different studies. Microplastic concentrations in lake and river sediments ranged from few hundred particles per m^3 to several hundred thousand particles per m^3. It has also been observed that microplastic concentrations in lake and river waters appear to be about three orders of magnitude lower compared to the levels of microplastics in sediments. In terms of pollution levels, microplastic concentrations in freshwater ecosystems can be orders of magnitude higher than in marine environments [22].

5.1 Microplastics in Lakes

Microplastics have been detected in large lakes in different continents of the world:

Asia

- China: Lakes in Tibetan Plateau, Taihu Lake.
- Mongolia: Lake Khovsgol.

Europe

- Italy: Lake Garda, Lake Bolsena, Lake Chiusi.
- Switzerland: six lakes.

North America

- Canada: Lake Ontario.
- USA: Lake Michigan.
- Canada, USA: Laurentian Great Lakes.

The presence of microplastics in lakes in areas with high and low levels of anthropogenic disturbance demonstrates the ubiquitousness of the problem. Not only microplastics are found in lakes near urban, industrial, and agricultural areas (such as in North American and European lakes), but also high levels of microplastic pollution have been detected in lakes in remote areas with low population densities (i.e. in lakes in the Tibetan Plateau, China and in a mountain lake in Mongolia). The presence of pelagic microplastic pollution in Lake Hovsgol – a large, oligotrophic mountain lake in a remote touristic location in Mongolia – has been reported, with an average microplastic density of 20,264 particles km^2 and the most abundant microplastic types being fragments and films [23]. This study indicated that microplastic density decreased with distance from the lake's most populated shores and was distributed by the prevailing winds, which demonstrate that without proper waste management, low-density populations can heavily pollute freshwater systems with consumer plastics.

5.2 Microplastics in Rivers

Microplastics are found in all rivers and streams in different parts of the world that were investigated. Similarly, the majority of the available studies on microplastics in rivers have been carried out in Europe and North America.

Asia

- China: Yangtze river and Three Gorges Dam.

Europe

- Germany: Rhine and Main rivers.
- Austria: Danube River.
- France: Seine River.
- Switzerland: Rhône, Aubonne, Venoge, Vuachère rivers.

North America

- Canada: Tributaries of Lake Ontario, Saint Lawrence River.
- USA: Rivers and streams in Indiana and Illinois, Raritan River, Tributaries of Great Lakes, North Shore Channel.

Some ecotoxicological studies on effects of microplastics on freshwater aquatic organisms indicated both higher detection frequencies and higher concentration levels of microplastics in fish in river segments near and downstream urban areas (*see* below Section on Health and ecological risks of microplastics in freshwater). This demonstrates that higher levels of microplastic pollution can be expected in freshwater systems near and downstream urban areas. No studies on the presence of microplastics in remote, presumably pristine rivers and streams, have been published yet.

5.3 Microplastics in Groundwater

Microplastics appear to be present also in groundwater, although in relatively low concentrations. Only a handful of studies are available on microplastics in groundwater [24]. A study indicated the presence of microplastics with sizes bigger than 20 μm in groundwater samples and at different positions within the drinking water supply chain, with concentrations ranging from 0 to 7 microplastics/m^3 in raw groundwater and an overall mean of 0.7 microplastics/m^3 in drinking water [25]. Water from springs and wells from two karst aquifers in Illinois, USA, contained microplastics with a maximum concentration of 15.2 particles/L, all of which were fibres [26]. Karst aquifers are open systems, susceptible to contamination by all types of pollutants, and groundwater in karst aquifers provides about one fourth of drinking water sources globally.

5.4 *Microplastics in Drinking Water*

Microplastics are present in drinking water – both in tap water and bottled waters. Several studies have reported the presence of microplastics in purified tap water and bottled waters in different countries of the world [27–29]. Analyses of 11 globally-sourced brands of bottled water, purchased in 19 locations in nine different countries, indicated that 93% of 259 total bottles contained microplastic contamination [28]. A study on small-sized microplastics and pigmented particles in bottled mineral water found pigment and additive particles in bottled mineral water samples, with a smallest analysed particle size of 1 μm [29]. This study suggested the bottle cleaning process and leaching out from the bottle material as possible contamination route of microplastics in bottled waters. It is estimated that annually a person ingests on average over 5,800 particles of synthetic debris that are contained in tap water, beer, and sea salt, with the largest contribution coming from tap water (88%) [27]. However, more research and data are needed on the occurrence of microplastics in drinking water in order to better understand potential exposure and to inform human health risk assessments [30, 31].

6 Health and Ecological Risks of Microplastics in Freshwater: How Harmful Are Microplastics?

The main concern associated with microplastics in freshwater environments is about their potential harmful effects on human health and aquatic life. However, a major issue in defining health and ecological risks of microplastics in freshwater ecosystems is a lack of scientific understanding and data.

6.1 *Ecotoxicological Effects*

The scientific understanding of microplastics effects on freshwater aquatic organisms is lacking. Whereas numerous researches have resulted in substantial understanding and knowledge of microplastics effects on marine aquatic organisms, ecotoxicological risks of microplastics to freshwater organisms have been studied only recently since the 2010s [32, 33]. Most studies have focused on microplastics in fish [34–37]. Higher numbers of microplastics were detected in fish at the urbanized sections of rivers [34]. A study indicated the ingestion of microplastics also in freshwater invertebrates such as *Lumbriculus variegatus*, *Daphnia magna*, *Notodromas monacha*, *Potamopyrgus antipodarum*, and *Gammarus pulex* [38]. Few studies have investigated ecotoxicological effects of environmentally-relevant concentrations of microplastics on freshwater aquatic organisms. Most ecotoxicological studies have been conducted in laboratories with microplastic concentrations relatively higher than those found in the natural environment [35].

The ability of microplastics to enter food webs at different trophic levels is a particular concern [33]. Some researchers have argued that some aquatic organisms have a limited ability to distinguish between food (for example, algae) and microplastics [20, 39], suggesting that aquatic organisms may ingest microplastics as food. A 28-day experiment on freshwater amphipods feeding with fibres and beads demonstrated that the exposure to microplastics significantly reduced the assimilation efficiency of these invertebrates, impairing the health and ecological functions of freshwater amphipods under continuous exposure to microplastics [40]. On the contrary, a laboratory experimental study on the uptake, retention, and impact of 2 μm polystyrene microplastics in the freshwater crustacean *Daphnia magna* suggested that microplastics may have little effect on food assimilation if ample food is present [41]. This demonstrates that more studies are needed to fully understand ecotoxicological effects of microplastics in freshwater organisms.

6.2 Potential Human Health Risks

Some studies indicate that microplastic particles may be ingested by humans with tap or bottled waters [27]. However, there is no evidence of risks to human health when microplastics are ingested. More research needs to be conducted to study the presence and abundance of microplastics in drinking water and food for human consumption and their potential human health risks [31]. Although no data are yet available, women, because of their physiological differences, may be more exposed and vulnerable to potential effects of microplastics.

6.3 Microplastics as Carriers and Sources of Other Pollutants in Freshwater

Microplastics may also act as carriers of other pollutants such as chemicals, metals, and pharmaceuticals in freshwater environments. Plastics are made typically from mixtures of resins and other chemicals and materials, called "*additives*", and therefore chemicals in plastics can potentially leach into the water and/or plastics may adsorb hydrophobic pollutants, so continuing their persistence and spread in the water [42]. Concerns have been raised over potential adverse effects of specific additives such as phthalate and bisphenol A plasticizers on human health and wildlife [43, 44]. Experimental investigations in animals indicated a wide variety of effects associated with exposure to chemical compounds in plastics, causing concern regarding potential risk to human health such as endocrine disruption and effects on neurological development and reproductive function [45]. Bisphenol A leaching from drinking water bottles has been indicated as a significant source of human exposure to this chemical [46]. The potential of microplastics as carriers of metals (such as lead, chromium, and zinc) has also been demonstrated, with an enhancement of metal adsorption in waters with high chemical and biological oxygen demands such as urban wastewater and irrigation runoff [7].

7 Reducing Microplastic Pollution in Freshwater Environments

It is practically impossible to remove microplastics once after they enter into freshwater environments. Hence, measures to reduce microplastic pollution in freshwater environments should be taken before they are discharged to freshwater from various sources.

Reducing microplastics at source is the most effective solution. Since microplastics enter freshwater systems predominantly via plastic litter and wastewater discharge, sustainable consumption at the level of individual consumers is the first and foremost means of reducing microplastics' input into the environment. Public awareness raising on environmentally-conscious choices of habits and consumer goods such as plastics and synthetic clothing – in particular avoiding the use of single-use plastics – will significantly contribute to the reduction of both primary and secondary microplastics such as microbeads, synthetic fibres and fragmentation of plastic wastes in freshwater systems. The gender mainstreaming and women's involvement in all aspects of sustainable consumption and consumer awareness should be promoted, as women can play a greater role in reducing microplastics' discharge to freshwater environments because of their roles in families and societies.

Transitioning to more responsible and sustainable production and manufacturing is necessary for the prevention of discharge of microplastics into the environment that pose significant health and ecological concerns. It can be achieved by promoting recycling and reuse of plastics. Substituting plastics with bio-degradable alternatives and materials is another effective way of reducing microplastics at source, where such options are available on an economically-viable basis. For example, replacing plastic microbeads in cosmetic and personal care products with natural and bio-degradable beads such as plant seeds has proven to be feasible and consumer-friendly. In response to growing concerns from the scientific community about environmental microplastic pollution, single-use plastics and microplastics (such as microbeads in personal care products) have been banned in several countries across the world [47]. Eliminating the use of non-essential, single-use plastics (such as plastic bags, cups, straws, cotton buds) is the next step in reducing plastic and microplastic pollution in freshwater environments. Many countries across the world have taken measures to phase out or ban single-use consumer plastics.

End-of-the-pipe solutions are still necessary, although costly to reduce freshwater microplastic pollution. Wastewater treatment plants appear to be highly effective in removing fibres from wastewater. Therefore, improving wastewater collection services and upgrading wastewater treatment levels are needed to reduce the amounts of synthetic fibres discharged with untreated and treated effluents. Because more than 80% of municipal wastewater in developing countries are not collected, or discharged with no, or little, treatment, there is an urgency to expand wastewater services to all populations in developing cities.

Improving solid waste collection, management and disposal, especially in developing countries, will lead to significant reductions of secondary microplastics in freshwater systems from plastic litter. Plastic waste reduction and recycling should be encouraged throughout the entire chain of waste management. Waste management systems based on the principles of a circular economy are of particular interest for creating and enhancing plastic recycling value chains, resource efficiency, and environmental sustainability, as well as for job creation opportunities.

Solutions to reduce microplastic pollution in freshwater environments should involve both policy approaches and technological innovations throughout different lifecycle stages of plastics – from production, manufacturing to consumption and use.

Freshwater microplastic pollution is a major source of marine microplastic pollution because rivers transport not only plastic debris from land, but also microplastics discharged with wastewater into river systems. Rivers are believed to be the main pathway of transport plastic waste from land-based activities into the ocean [45]. Consequently, reducing microplastic pollution in freshwater environments will result in a significant reduction of microplastics along the source-to-sea continuum and in coastal and marine waters, contributing to healthy oceans.

8 Knowledge Gaps and Research Needs on Microplastics in Freshwater Environments

Freshwater microplastics are an under-researched area with limited scientific information, in contrast to the wealth of knowledge and the vast number of studies on marine microplastics. There is an evident knowledge gap on freshwater microplastic pollution.

The small number of available studies and scientific information demonstrates huge gaps in knowledge and data on microplastics in freshwater environments. Although plastic pollution is increasingly emerging as a serious global environmental problem, the issue of freshwater microplastics has not been a priority for research until only recently.

More research and monitoring data on microplastics in different freshwater bodies are needed for a systematic estimation of the extent and levels of occurrences of microplastics in freshwater aquatic environments at global, national or basin levels. Microplastics need to be studied in all types of freshwater environments, including lakes and rivers as well as springs, streams, ponds, and wetlands.

There is a need for harmonisation of definitions, sampling methods and data metrics on microplastics in freshwater environments, which is essential for comparability and quality of research data. The application of different size ranges in defining microplastics and the use of different sampling methods limit the use of research data for the benefit of future research, policy development, and the public.

Research on potential ecotoxicological risks, exposure mechanisms and accumulation of microplastics in freshwater aquatic life is lacking. Although first scientific evidences show that microplastics are present in freshwater biota, there is limited scientific information available on microplastics exposure, accumulation, and ecotoxicological effects on freshwater aquatic organisms. Moreover, ecotoxicological studies should be evaluated for all major groups of freshwater aquatic organisms (fish, invertebrates, etc.) and for different levels of the aquatic food web.

There is a lack of scientific information on human exposure to microplastics through drinking water. Research is needed to study the presence and levels of microplastics in drinking water (tap water and bottled waters) and to evaluate their potential human health risks, if any.

Building research capacity and sharing knowledge with developing regions is essential to facilitate studies on microplastics in freshwater environments in these regions. The geographical distribution of research and available scientific information on microplastics in freshwater environments are primarily in developed countries, with over 80% studies between 1995 and 2015 conducted in Europe and North America [21]. Only few studies have been carried out in other regions such as Africa, Asia, and Latin America, where microplastic pollution in freshwater environments may be very serious due to the lack of appropriate solid waste and wastewater management. Researchers found microplastics in the Antarctic ice core [48]. Microplastics may be present ubiquitously in the world's freshwater, even in the most pristine environments.

Acknowledgements The initial UNESCO case study on microplastics in freshwater was conducted in the framework of the UNESCO Project on Emerging Pollutants in Water, with the funding from the Government of Sweden through its Programme Cooperation Agreement with UNESCO (2014-2018). The UNESCO's work on microplastics in freshwater has further benefitted from scientific expertise of researchers from around the world. In particular, UNESCO-IHP's International Initiative on Water Quality wishes to thank (in alphabetical order): Michiel Roscam Abbing, Plastic Soup Foundation (the Netherlands); Nicole Brennholt, Federal Institute of Hydrology (Germany); Michiel De Smet, European Commission; Nanna B. Hartmann, Technical University of Denmark (Denmark); Alice A. Horton, Centre for Ecology and Hydrology (United Kingdom); Liv Jaeckel, Technical University of Freiberg and HHL Leipzig Graduate School of Management (Germany); Johanna Kramm, Institute for Social-Ecological Research (Germany); Helen Lynn, University of Stirling (United Kingdom); Margriet Samwel, Women Engage for a Common Future (the Netherlands); Friederike Stock, Federal Institute of Hydrology (Germany); Maria Antonia N. Tanchuling, University of the Philippines (the Philippines); and Martin Wagner, Norwegian University of Science and Technology (Norway). The results of the UNESCO's work on microplastics in freshwater environments will be published in Microplastics in Freshwater Environments (upcoming) as volume 2 of the UNESCO Series on Emerging Pollutants in Water.

References

1. UNESCO and HELCOM (2018) Pharmaceuticals in the aquatic environment of the Baltic Sea region - a status report. UNESCO emerging pollutants in water series, vol 1. UNESCO Publishing https://unesdoc.unesco.org/ark:/48223/pf0000247889?
2. WHO (2020) Water, sanitation and hygiene (WASH). World Health Organization (WHO). https://www.who.int/health-topics/water-sanitation-and-hygiene-wash
3. UN (2020) Sustainable development goal 6: ensure availability and sustainable management of water and sanitation for all – Progress and Info. United Nations, sustainable development. https://sdgs.un.org/goals/goal6
4. IPBES (2019) In: Brondizio ES et al (eds) Global assessment report on biodiversity and ecosystem services of the Intergovernmental Science-Policy Platform on Biodiversity and Ecosystem Services (IPBES), Chapter 2.1: status and trends – drivers of change. IPBES Secretariat. https://ipbes.net/global-assessment
5. IPBES (2019) In: Díaz S et al (eds) Summary for policymakers of the global assessment report on biodiversity and ecosystem services of the Intergovernmental Science-Policy Platform on Biodiversity and Ecosystem Services (IPBES). IPBES Secretariat. https://ipbes.net/sites/default/files/2020-02/ipbes_global_assessment_report_summary_for_policymakers_en.pdf
6. UN (2020) Sustainable development goals. United Nations. https://sdgs.un.org/goals
7. Godoy V et al (2019) The potential of microplastics as carriers of metals. Environ Pollut 255 (3):113363. https://doi.org/10.1016/j.envpol.2019.113363
8. Plastics Europe (2016) Plastics – the facts 2016: an analysis of European plastics production, Demand and waste data. https://www.plasticseurope.org/en/resources/publications/3-plastics-facts-2016
9. Geyer R, Jambeck JR, Law KL (2017) Production, use, and fate of all plastics ever made. Sci Adv 3(7):e1700782. https://doi.org/10.1126/sciadv.1700782
10. Arthur C, Baker J, Bamford H (2009) Proceedings of the international research workshop on the occurrence, effects and fate of microplastic marine debris (Sept. 9–11, 2008). NOAA Technical Memorandum NOS-OR&R30. https://www.semanticscholar.org/paper/Proceedings-of-the-International-Research-Workshop-Arthur-Baker/7f8c9dd6437a34f96812836c66ca83d69f7ea9eb?p2df
11. Gigault J et al (2018) What is a nanoplastic? Environ Pollut 235:1030–1034. https://doi.org/10.1016/j.envpol.2018.01.024
12. Napper IE et al (2015) Characterisation, quantity and sorptive properties of microplastics extracted from cosmetics. Mar Pollut Bull 99:178–185. https://doi.org/10.1016/j.marpolbul.2015.07.029
13. Karlsson TM et al (2018) The unaccountability case of plastic pellet pollution. Mar Pollut Bull 129(1):52–60. https://doi.org/10.1016/j.marpolbul.2018.01.041
14. Turner A, Holmes LA (2015) Adsorption of trace metals by microplastic pellets in fresh water. Environ Chem 12:600–610. https://doi.org/10.1071/EN14143
15. Browne MA et al (2011) Accumulation of microplastic on shorelines worldwide: sources and sinks. Environ Sci Technol 45(21):9175–9179. https://doi.org/10.1021/es201811s
16. Essel R et al (2015) Sources of microplastics relevant to marine protection in Germany. Federal Environment Agency of Germany. TEXTE 64/2015, Project no. 31969, Report no. (UBA-FB) 002147/E. http://www.umweltbundesamt.de/publikationen/sources-of-microplastics-relevant-to-marine
17. Conley K et al (2019) Wastewater treatment plants as a source of microplastics to an urban estuary: removal efficiencies and loading per capita over one year. Water Res X3:100030. https://doi.org/10.1016/j.wroa.2019.100030
18. Dris R, Gasperi J, Tassin B (2014) Assessing the microplastics in urban effluents and in the Seine River (Paris). https://hal-enpc.archives-ouvertes.fr/hal-00968276

19. Horton AA et al (2017) Large microplastic particles in sediments of tributaries of the river Thames, UK – abundance, sources and methods for effective quantification. Mar Pollut Bull 114(1):218–226. https://doi.org/10.1016/j.marpolbul.2016.09.004
20. Cole M et al (2011) Microplastics as contaminants in the marine environment: a review. Mar Pollut Bull 62(12):2588–2597. https://doi.org/10.1016/j.marpolbul.2011.09.025
21. UNESCO (2021) Microplastics in freshwater environments, UNESCO emerging pollutants in water series, vol 2. UNESCO Publishing (inpress)
22. Mani TA et al (2015) Microplastics profile along the Rhine River. Sci Rep 5:17988. https://doi.org/10.1038/srep17988
23. Free CM et al (2014) High-levels of microplastic pollution in a large, remote, mountain lake. Mar Pollut Bull 85:156–163. https://doi.org/10.1016/j.marpolbul.2014.06.001
24. Geological Society of America (2020) Microplastics in groundwater (and our drinking water) present unknown risk: presentation at the 2020 annual meeting of the Geological Society of America. Science Daily. http://www.sciencedaily.com/releases/2020/10/201026153939.htm
25. Mintenig SM et al (2019) Low numbers of microplastics detected in drinking water from ground water sources. Sci Total Environ 648:631–635. https://doi.org/10.1016/j.scitotenv.2018.08.178
26. Panno SV et al (2019) Microplastic contamination in karst groundwater systems. Groundwater 57(2):189–196. https://doi.org/10.1111/gwat.12862
27. Kosuth M, Mason SA, Wattenberg EV (2018) Anthropogenic contamination of tap water, beer, and sea salt. PLoS One 13(4):e0194970. https://doi.org/10.1371/journal.pone.0194970
28. Mason SA, Welch VG, Neratko J (2018) Synthetic polymer contamination in bottled water. Front Chem 6:407. https://doi.org/10.3389/fchem.2018.00407
29. Ossmann B et al (2018) Small-sized microplastics and pigmented particles in bottled mineral water. Water Res 141:307–316. https://doi.org/10.1016/j.watres.2018.05.027
30. Koelmans AA et al (2019) Microplastics in freshwaters and drinking water: critical review and assessment of data quality. Water Res 155:410–422. https://doi.org/10.1016/j.watres.2019.02.054
31. WHO (2019) Microplastics in drinking-water. World Health Organization (WHO). https://www.who.int/water_sanitation_health/publications/microplastics-in-drinking-water/en/
32. Wagner M et al (2014) Microplastics in freshwater ecosystems: what we know and what we need to know. Environ Sci Eur 26:12. https://doi.org/10.1186/s12302-014-0012-7
33. Anderson JC, Park BJ, Palace VP (2016) Microplastics in aquatic environments: implications for Canadian ecosystems. Environ Pollut 218:269–280. https://doi.org/10.1016/j.envpol.2016.06.074
34. Silva-Cavalcanti JS et al (2017) Microplastics ingestion by a common tropic freshwater fishing resources. Environ Pollut 221:218–226. https://doi.org/10.1016/j.envpol.2016.11.068
35. de Sá LC et al (2018) Studies of the effects of microplastics on aquatic organisms: what do we know and where should we focus our efforts in the future? Sci Total Environ 645:1029–1039. https://doi.org/10.1016/j.scitotenv.2018.07.207
36. Slootmaekers B et al (2019) Microplastic contamination in gudgeons (Gobio gobio) from Flemish rivers (Belgium). Environ Pollut 244:675–684. https://doi.org/10.1016/j.envpol.2018.09.136
37. Hurt R, O’Reilly CM, Perry WL (2020) Microplastic prevalence in two fish species in two US reservoirs. Limnology and oceanography letters 5: special issue: microplastics in marine and freshwater organisms: presence and potential effects. https://doi.org/10.1002/lol2.10140
38. Imhof HK et al (2013) Contamination of beach sediments of a subalpine lake with microplastic particles. Curr Biol 23:R867–R868. https://doi.org/10.1016/j.cub.2013.09.001
39. Von Moos N, Burkhardt-Holm P, Köhler A (2012) Uptake and effects of microplastics on cells and tissue of the blue mussel Mytilus edulis L. after an experimental expo-sure. Environ Sci Technol 46:11327–11335. https://doi.org/10.1021/es302332w
40. Blarer P, Burkhardt-Holm P (2016) Microplastics affect assimilation efficiency in the freshwater amphipod Gammarus fossarum. Environ Sci Pollut Res 23:23522–23532. https://doi.org/10.1007/s11356-016-7584-2

41. Aljaibachi R, Callaghan A (2018) Impact of polystyrene microplastics on Daphnia magna mortality and reproduction in relation to food availability. Peer J 6:e4601. https://doi.org/10.7717/peerj.4601
42. Andrady AL, Neal MA (2009) Applications and societal benefits of plastics. Philos Trans R Soc B Biol Sci 364:1977–1984. https://doi.org/10.1098/rstb.2008.0304
43. Meeker JD, Sathyanarayana S, Swan SH (2009) Phthalates and other additives in plastics: human exposure and associated health outcomes. Philos Trans R Soc B 364:2097–2113. https://doi.org/10.1098/rstb.2008.0268
44. Oehlmann J et al (2009) A critical analysis of the biological impacts of plasticizers on wildlife. Philos Trans R Soc Lond Ser B Biol Sci 364:2047–2062. https://doi.org/10.1098/rstb.2008.0242
45. Talsness CE et al (2009) Components of plastic: experimental studies in animals and relevance for human health. Philos Trans R Soc Lond Ser B Biol Sci 364(1526):2079–2096. https://doi.org/10.1098/rstb.2008.0281
46. Le HH et al (2008) Bisphenol A is released from polycarbonate drinking bottles and mimics the neurotoxic actions of estrogen in developing cerebellar neurons. Toxicol Lett 176(2):149–156. https://doi.org/10.1016/j.toxlet.2007.11.001
47. UN Environment (2018) Legal limits on single-use plastics and microplastics: a global review of national laws and regulations. https://www.unenvironment.org
48. Kelly A et al (2020) Microplastic contamination in east Antarctic sea ice. Mar Pollut Bull 154:111130. https://doi.org/10.1016/j.marpolbul.2020.111130

From Land to Sea: Model for the Documentation of Land-Sourced Plastic Litter

Stephanie Cieplik

Contents

1 Global Challenge 274
2 Background/Motivation 274
3 Plastic Waste in Germany 275
4 Improperly Disposed-of Plastics and Littering in Germany 276
5 Subject of the Investigation of the Model 279
6 Methodology of the Model 280
7 Assumptions Made in the Model 280
8 Overview of the Discharge Pathways 281
9 Special Report: Compost and Digestate 282
10 Special Report: Littering 283
11 Data Basis 284
12 Limitations of the Model 284
13 Main Results 286
14 Conclusions 288
References 288

Abstract The model "From Land to Sea – Model for the documentation of land-sourced plastic litter" was developed on behalf of BKV GmbH, Frankfurt, Germany. This model systematically records for the first time discharges of improperly disposed-of plastic litter from Germany that gets into the North Sea, the Baltic Sea, and the Black Sea. All discharge pathways and sources are taken into account. A distinction is made between discharges of microplastic and macroplastic.

Keywords Discharge pathways/discharge sources, Germany's contribution, Microplastic/macroplastic, New methodology, Volume estimate

---

S. Cieplik (✉)
BKV GmbH, Frankfurt am Main, Germany
e-mail: stephanie.cieplik@bkv-gmbh.de

Friederike Stock, Georg Reifferscheid, Nicole Brennholt, and Evgeniia Kostianaia (eds.),
Plastics in the Aquatic Environment - Part I: Current Status and Challenges,
Hdb Env Chem (2022) 111: 273–288, DOI 10.1007/698_2021_781,
© BKV GmbH 2021, Published online: 3 July 2021

1 Global Challenge

Since the 1950s, plastics have achieved unparalleled growth. Because of their enormous variety of potential uses and outstanding technical properties, they have become established in numerous fields of application. We encounter plastics in all areas of our life. They have successfully taken over from conventional materials, for example, in car production, in household appliances, in the building sector, in sports and leisure articles, in medicine, and in packaging.

Because of their wide range of application, plastics have become indispensable in our day-to-day lives. They are valuable resources that bring people benefits in many areas, and they also make a valuable contribution to sustainable solutions. Having said this, the obvious question is of course what happens to the plastics at the end of their useful life. Established and well-functioning waste management systems can provide for the recycling and, if necessary, disposal of the plastics. Regional circumstances, as well as the responsible action and behaviour of the public and institutions also play a decisive role.

Often, however, plastics turn up in the environment, and marine litter in particular is a much-discussed topic at present. Plastics naturally do not belong in the environment. Plastics are useful materials and thus much too valuable to end their life floating as litter unused in the sea. However, it is currently estimated that between 4.8 and 12.7 mt of plastic waste end up in the world's seas every year [1].

Whether and how the litter already present in the seas can be collected for orderly disposal is one of the challenges of our age. Another is avoiding the creation of litter in the first place. Only cooperation between all the parties involved – science, research, politics, administration, NGOs, trade and industry – at all possible levels can lead to a solution to the marine litter problem.

2 Background/Motivation

The Marine Strategy Framework Directive came into force in Europe on 15 July, 2008. Its aim is to encourage the sustainable use of the seas and to conserve marine ecosystems. Based on this directive, the member states of the European Union are obliged to take the necessary measures to achieve or retain a healthy state of the marine environment at the latest by the year 2020. An important criterion for evaluating a good environmental status of the seas is marine litter. Whenever there is any talk of marine litter, plastics are always at the focus of discussion because of their long life, the additives they contain, and their decomposition into microplastics.

Against this background, the BKV GmbH wants to contribute to the necessary fact-oriented clarification of the true situation – by providing the required information and carrying out corresponding projects. Here, the BKV focuses its attention on the so-called land-sourced littering, in other words, the plastics that get from the land into the water. The main emphasis of this project is on the collection, compilation, and processing of facts and data, especially with regard to mass flows and the routes by which the plastics are transported into the sea.

On behalf of BKV GmbH the Conversio Market & Strategy GmbH has presented for the first time a model approach for the documentation of land-sourced plastic litter with regard to its discharge pathways and discharge sources into the seas. The model was supported by the Association of the Austrian Chemical Industry (FCIO), the Germany's Plastics Packaging Industry Association (IK), PlasticsEurope Deutschland, and the German Engineering Federation (VDMA), here the Association of Plastics and Rubber Machinery.

In the first step, the methodology was applied to the discharges of improperly disposed-of plastic waste from Germany into the North Sea, and subsequently supplemented by the discharges into the Baltic Sea and the Black Sea. This provides an overall picture of discharges into the sea of improperly disposed-of plastic litter that can be attributed to Germany.

The aim of the project is, based on the methodical approach, to systematically record, structure, and quantify the main discharge pathways and sources for plastics. Only if the discharge pathways and sources as well as the corresponding mass flows of the plastics into the sea are identified and analysed, it will be possible to make a useful contribution to the prevention of further input of litter into the seas.

3 Plastic Waste in Germany

Worldwide, approximately 348 million metric tonnes of plastic (The term "plastics" here covers thermoplastics, polyurethanes, thermosets, elastomers, adhesives, coatings, sealants and PP fibres. It does not include PET-, PA- or polyacrylic fibres) were produced in 2017 (Fig. 1). Of this, around 7% or over 20 million tonnes came from Germany and 18% from Europe, equivalent to over 64 million tonnes. According to current forecasts, a period of consolidation will be expected in the following years [2].

In Germany, around 14.4 mt of plastic were used in 2017 for the production of plastic goods. Of this, 12.6 mt were virgin material and 1.8 mt were recyclate (Fig. 2). Taking the export surplus into account, 11.8 mt remained in Germany for private and industrial consumption [3].

In 2017, 11.8 mt of plastic products were used in Germany as new products. In the same year, around 6.2 mt of plastic products became waste that was correctly disposed of. This volume was made up of 5.2 mt of post-consumer waste and 0.95 mt of production and processing scrap. This means that the volume of waste has risen by approximately 1.9% a year since 2015 [4].

Depending on the particular application, plastic products are used for a different length of time before they are sent for disposal. More than 95% of packaging material – generally very short-lived products – returns as waste in the same year as it was produced. On the other hand, only 19% of building products become waste in the same year because of their much longer useful life. For example, plastic pipes

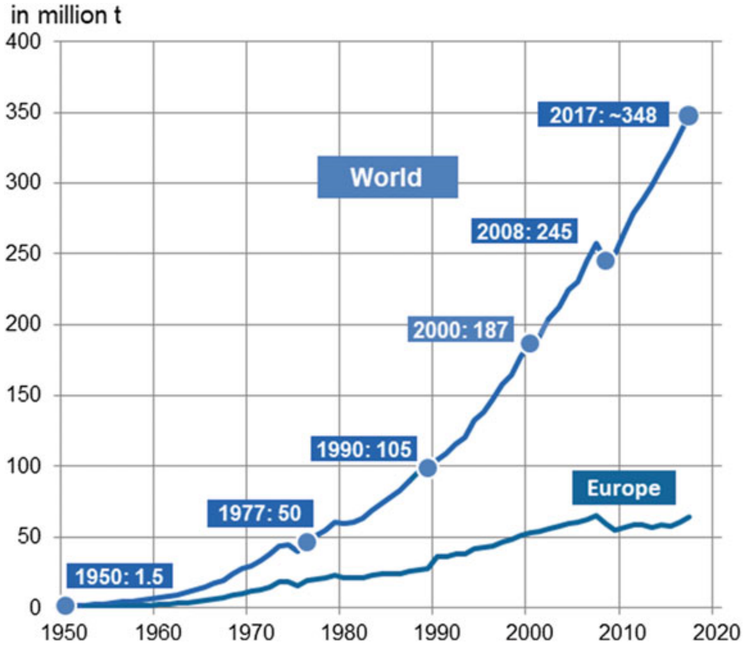


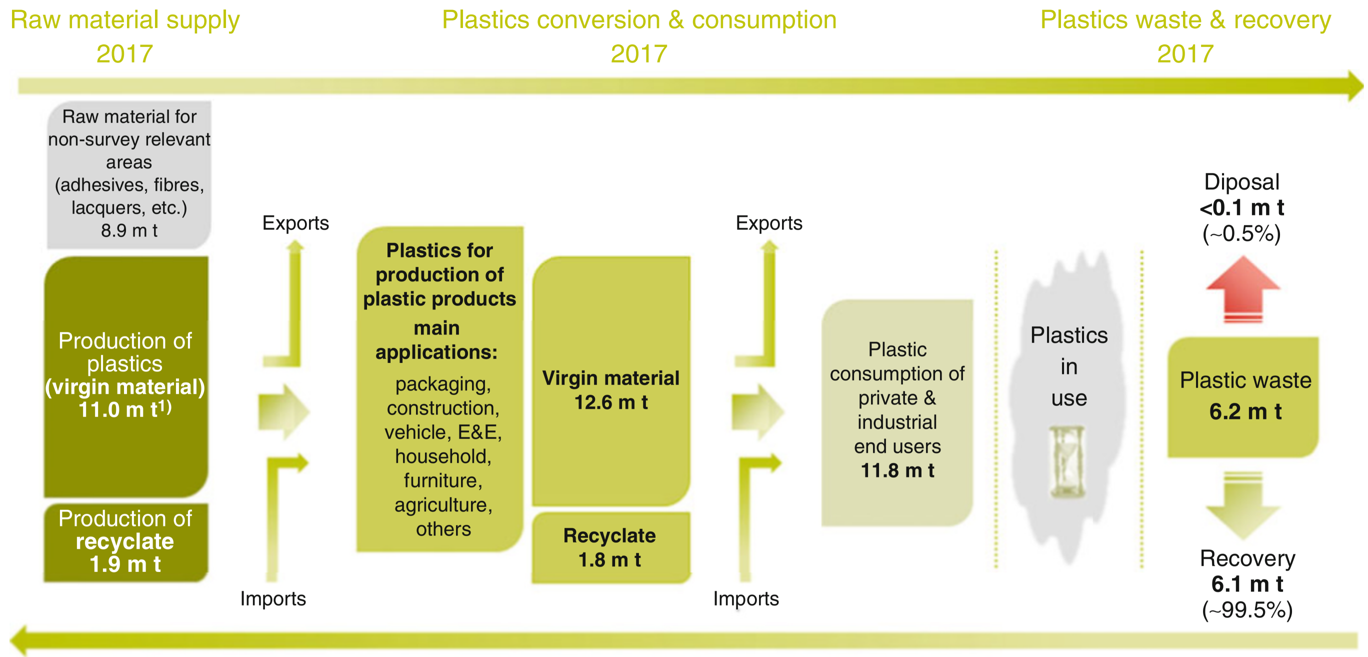
Fig. 1 World plastics production 1950–2017. © PlasticsEurope Market Research Group (PEMRG)/Consultic Marketing & Industrieberatung GmbH, 2016

remain in use for an average of 80 years and plastic window frames for 50–60 years before they are disposed of. The same applies to plastic products in vehicle construction. The average service life of a vehicle in Germany is 10–12 years, followed by an continued use of older vehicles abroad. As a consequence of this plastics from the automotive segment account for around 21% of the waste.

4 Improperly Disposed-of Plastics and Littering in Germany

Plastics enter the environment in various ways. During the manufacture of products, for example, plastic granules can be released in the form of microplastics. Microplastic in the environment also arises through the actual use of the products for their intended purpose. This includes, for example, abrasion of synthetic fibres during the wearing or washing of garments, and the use of detergents and cleaning products by private and commercial consumers [5].

In addition to the properly disposed-of waste, Germany also registers a certain amount of improperly disposed-of waste: the so-called littering. Littering is understood here to mean the contamination of the public sphere by people carelessly or irresponsibly discarding or leaving behind litter on roads, rivers in public places or in



[1] LD/LLDPE, HD/MDPE, PP, PVC, PS, EPS, PA, PET, ABS, ASA, SAN, PMMA, other thermosets, other plastics, including PU

Fig. 2 Material flow analysis plastics in Germany 2017: Overview of the main results. © Conversio, Material flow analysis plastics in Germany, 2017, 2018

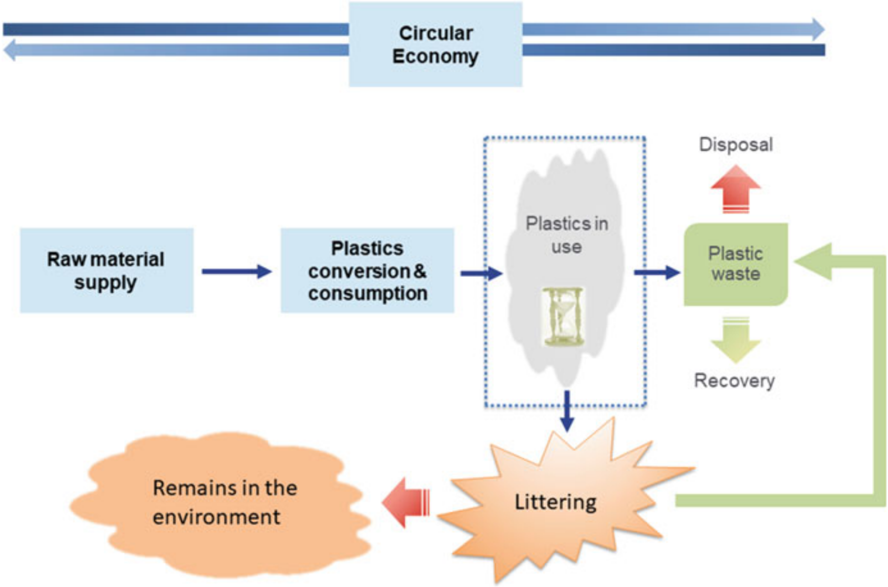


Fig. 3 Plastic waste in Germany and littering

the countryside. Littering involves all kinds of materials, whether metals, glass, paper, or plastics.

Although littering is a problem affecting many kinds of material, plastics receive an above-average amount of criticism. One reason for this is the comparatively long-life expectancy of plastics in nature. Furthermore, plastics – especially plastic bags and packaging – tend to attract more attention because of their size and appearance. Also, the fact that plastics are very light in weight has consequences with regard to their importance for littering: plastic articles – especially plastic bags – are very easily picked up by the wind and distributed over a wide area. They float on rivers, lakes, and seas and thus become distributed worldwide. Furthermore, microplastics are produced through the weathering and fragmentation of plastic articles. Removing such microplastics from the environment or from the sea is virtually impossible.

In Germany, a relevant cause of littering is the careless behaviour of people who simply throw such products away – out of laziness, convenience, indifference, lack of responsibility, provocation, or simply ignorance. In many places, a lack of waste bins contributes to the increase of littering. Only a change in behaviour of everyone involved will, in the long run, be able to solve this problem [6].

A large proportion of the improperly disposed-of waste in Germany is returned by one way or another to the intended collection systems (Fig. 3). Examples of this include the uncontrolled disposal of waste that is collected by the relevant authorities at big public events or at tourist destinations. Such refuse is also often collected by tram or railway maintenance depots, and also, for example, at motorway service areas. According to estimates, around 75% of the improperly disposed-of waste is returned at some time to the waste management system [7].

Despite this, not all uncontrolled disposed-of waste can be subsequently collected. Reasons for this include gaps in the documentation of the disposal infrastructure and accumulations of waste at remote areas in the wild. Accordingly, the risk exists with such waste that it will end up in the sea by one route or another.

5 Subject of the Investigation of the Model

The focus of the model is on improperly disposed-of plastic litter. The term plastic is taken to mean polymer materials. Tyre and brake abrasion are at present not taken into account, nor are polymers in paints and other surface coatings. This is attributable to conventional definitional classifications (Tyres are made from rubber. According to the official statistics, tyres thus count as "rubber goods". Tyre abrasion can therefore not be counted as plastic waste. In the case of brake abrasion, the focus here is primarily on the fine dust pollution. Fine dust does not count as plastic waste either. In the study, by definition, primarily plastic products are taken into account as moulding compounds, i.e. polymers in surface coatings are not included. Resins are also taken into account only as moulding compounds).

In the model, a distinction is made between microplastic and macroplastic. There is no uniform definition for the term "microplastic". Within the framework of the model, plastics that are <5 mm when discharged into one of the discharge pathways or discharge sources count as microplastics (primary microplastics). Plastics that are >5 mm on discharge are recorded as macroplastics.

There is one special aspect in the model when distinguishing between primary and secondary microplastic. For designing the model, the plastic discharge via the various discharge pathways must be separated analytically from the discharge of plastic into the seas. The key factor for the classification as microplastics or macroplastics is thus their size at the moment of discharge into the discharge pathway. They are regarded as primary microplastics if, at the time of discharge into one of the discharge pathways, the plastics are <5 mm. Secondary microplastics are created by decomposition processes. Plastics that are >5 mm when entering a discharge pathway and do not decompose until they are in the discharge pathway (secondary microplastics) are recorded within the model calculation as macroplastics. Consequently, secondary microplastics are also taken into account in the model, but, in line with the assumptions made in the model, are shown as macroplastics. The "development" from macroplastic to microplastic is not a subject of the model.

A differentiation of the microplastics covered by the model according to the particle size has not so far been carried out because of the available data. Within the framework of the model, the particle size on entering one of the discharge pathways is decisive. According to this, the classification as microplastics or macroplastics is made. In contrast, previous studies have so far relied on results based on measurements from one of the discharge pathways. Although these measurements provide a differentiation of the microplastics according to the particle size, it is not possible – or only to a limited extent – to draw conclusions about the particle size on entering the discharge pathway, about the place of origin (e.g. household, compost/digestate and industry/trade) of the plastic waste, or about its discharge pathway.

6 Methodology of the Model

The model serves primarily to estimate more reliably the origin, quantity, and nature of the plastic litter ending up in the sea. It presents individually the possible discharge pathways of plastic waste (micro- and macroplastics) that finds its way into the sea. It examines the land-sourced litter that gets into the North Sea, Baltic Sea, and Black Sea that can be attributed to Germany. Litter from marine shipping, cruise ships, and the fishing industry (sea-sourced litter) and discharges from other regions into the seas are not yet taken into account in the model.

In the first step, the main discharge pathways and discharge sources were identified and a data model established. Based on this data model, a database was drawn up. In the second step, an analysis was made of the discharge volumes based on secondary and primary data. Alongside scientific studies and investigations, statistical data, among other things from EUROSTAT and the German Statistical Office, were used and primary data were generated in the form of expert discussions. The process of evaluating other literature and sources and their potential subsequent utilisation in the model consistently continues.

7 Assumptions Made in the Model

The model subdivides land-sourced litter according to particular discharge pathways or discharge sources, namely “rivers”, “river shipping”, “coastal regions”, “ports”, and “landfills” (Fig. 4).

The model refers consistently to discharge pathways even if, for example, a landfill or a port is more a source than a pathway. Other point sources, such as wastewater treatment plants, or diffuse sources, such as discharges from agriculture, are assigned to the discharge pathways, and are included in the calculation of the discharge volume of the respective pathway into the sea. Depending on the relevance, certain sources are shown in the model separately and in more detail.

Only one part of the litter that gets into the environment also ends up in the sea, after a certain delay. Some of it remains in the countryside, becomes deposited in riverbeds and wetlands, or is collected again and disposed of (e.g. with the help of screens or sieves on weirs and wastewater treatment plants). In addition to discharges of litter into the environment, different “loss factors” therefore have to be taken into account when calculating the transport into the sea.

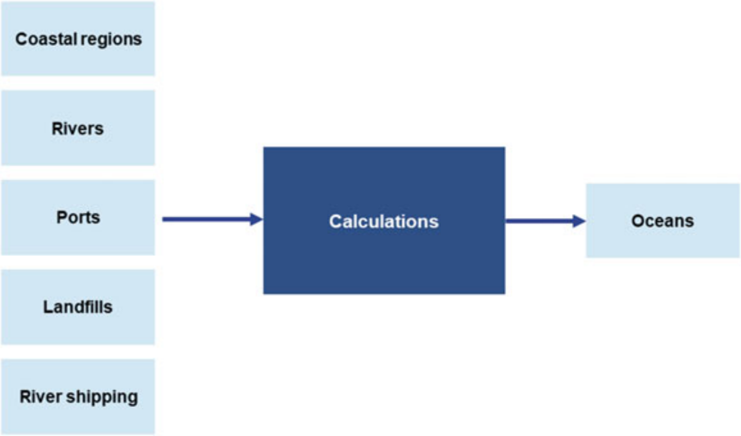


Fig. 4 Discharge sources and pathways

8 Overview of the Discharge Pathways

In the model, five main discharge pathways or discharge sources are identified.

Microplastics and macroplastics can get into the sea via rivers. What is unclear, however, is at what point the plastic enters the rivers. To ensure that the model takes into account not only the main rivers that flow directly into the sea but also other smaller rivers, it also covers the catchment areas of the rivers as defined by “river basin districts”. River basin districts comprise one or several neighbouring river catchment areas. They encompass rivers from their source to their mouth (at the point where they flow into the sea) as well as all their tributaries and streams plus the groundwater in this district. The study focuses on river basin districts in Europe in which the main river flows into the North Sea, Baltic Sea, or Black Sea, or borders directly the North Sea, Baltic Sea, or Black Sea. Consequently, it covers not only the respective countries bordering these seas, but also countries that are part of an international river basin district and are thus of importance for discharges into the North Sea, Baltic Sea, or Black Sea.

In the study, the discharge pathway labelled as “rivers” covers waste that, in some way or other, gets from the land into a river. For example, microplastics in effluent can either find their way directly into the receiving water via the waste water from a treatment plant or can be blown or leached out from the sewage sludge that has been spread over fields. Improperly disposed-of macroplastics can get into streams and rivers through the wind, rainwater, or illegal dumping.

In the system used here, the discharge pathway “rivers” includes only plastic litter that enters the discharge pathway inland. The discharge of plastic litter into streams and rivers near the coast falls under the discharge pathway “coastal regions”. The reason for this is that the transport losses are smaller with waste that is discharged near the coast.

Because of their proximity to the sea, coastal regions must be regarded separately. Here, plastic litter can either get directly into the sea or can be carried into the sea via surface water. In contrast to the discharge pathway “rivers”, the loss factors here for plastic litter are much smaller because of the proximity to the sea. The model takes into account both the coast and the coastal regions of the North Sea and Baltic Sea. The EUROSTAT definition of a coast is used here: “a coastal region in the European Union is a region of the NUTS 3 level, which has a coastline or in which more than half the population lives less than 50 km from the sea”. When considering the discharge pathway covered by “coastal regions”, tourism was also taken into account.

In river shipping, litter (macroplastics) can first find its way into a river and finally into the sea through the wind, drifting, careless/irresponsible disposal, or illegal dumping. The model includes inland shipping in all countries whose river basin districts are relevant for the discharge of plastic waste into the North Sea, Baltic Sea, or Black Sea.

Waste disposed in landfills near to the coast can get into the seas by the wind or by drifting. The discharge of plastic litter from landfills into the sea can be selectively documented. This model covers active landfills near the coast with a maximum distance of 5 km to the North Sea or Baltic Sea. For discharges into the Black Sea attributable to Germany, the discharge pathway of “landfills” does not play a role because of the lack of a coastline.

Litter occurring in ports can end up in the sea through the wind, drifting, careless disposal, or illegal dumping. The model covers ports in countries with direct access to the North Sea and Baltic Sea that are on the coast or near to the coast. This discharge pathway is not relevant for discharges into the Black Sea.

9 Special Report: Compost and Digestate

Composts and digestates can contain microplastics to a differing extent. As a rule, plastics get into the recycling of biowaste through the collection of biowaste from households and trade and commerce. Especially in the collection of biowaste from private households, incorrect consumer behaviour plays a decisive role. Through carelessness, plastics and other foreign substances find their way into the organic waste bin. In the commercial sector, it is, above all, incorrectly disposed-of plastic packaging that plays the largest part in the amount of plastics found in compost and digestate. When compost and digestate are spread on the land in agriculture, forestry, landscaping, and private households, it is quite possible that microplastic particles will get into rivers or directly into the sea through the wind, through drifting, or by being leached out.

In the model, compost and digestate from household and commercial sources are shown as a potential discharge source for plastics into the seas. Every year, composts and digestates, with approximately three metric tonnes, account for less than 1% of the total discharges into the North Sea, Baltic Sea, and Black Sea attributable to

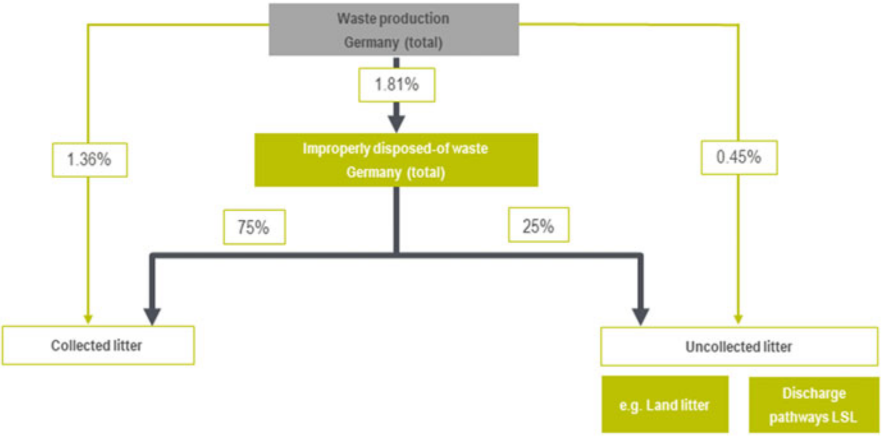


Fig. 5 Littering in Germany – Calculation approach. © Conversio, From Land to Sea – Model for the documentation of land-sourced plastic litter, 2018

Germany (The calculations are based on data from the RAL quality assurance of the Federal Compost Association (BGK) and on data from the Federal Statistical Office).

For discharges into the North Sea, the Baltic Sea, and the Black Sea, a special report on compost/digestate has been compiled as a supplement to the model. It describes the individual calculation steps in detail [8].

10 Special Report: Littering

To determine the percentage of macroplastic in marine litter, a calculation based on improperly disposed-of plastic waste – litter – is being carried out because there is a lack of adequate data and information (Fig. 5). Although a direct correlation between the quantity of correctly recorded waste and incorrectly recorded waste is only possible to a certain extent, it can, in view of the lack of available data, serve as an initial aid.

There have been few studies regarding the proportion of litter in the total amount of waste in Germany. To date, no countrywide study of improperly disposed-of waste has been carried out. The basis for the calculation in the model is therefore a study by the Bavarian Office for Environmental Protection (Bayerisches Landesamt für Umwelt – LfU) In 2001, a report entitled “Special assessment of the waste figures for 2001 – Illegal waste disposal” was published.

The assumptions made in the model about littering were examined in more detail and verified in a separate study in 2018. Especially the assumptions made on the basis of the LfU study from 2001 were checked with the aid of additional sources and methods. To this end, literature research and expert discussions were held. Furthermore, a sensitivity analysis was carried out to coordinate the obtained results.

The results of the special report on littering show that only minor changes need to be made to the assumptions made in the model with regard to littering. These will be undertaken in the course of the next update of the report and handbook in 2019.

11 Data Basis

The model is based on the data and information obtained and forwarded by third parties (Fig. 6). The absolute discharge quantities determined here should be seen as an estimate based on the existing study situation and discussions with relevant market experts. Against this background it is understandable that the quality and quantity of the available data considerably vary between the studied areas of microplastics and macroplastics.

In the microplastics segment, the model is based on a broad set of data, as many studies and investigations have already been carried out on this subject. Nevertheless, because a large number of variables are used for the calculation of individual factors, there are also some individual data gaps in the field of microplastics. For this reason, the decision was taken to work with estimates from experts.

For some factors that extend over several discharge pathways and also several applications (e.g. transport losses), only rudimentary data is so far available. Consequently, when looking at the volumes involved in the discharge of plastics into the sea, an assessment of the data accuracy according to the application or discharge pathway is only possible to a limited extent.

In the macroplastics segment, hardly any studies so far exist that can be integrated into the model. For this reason, the figures have so far been derived from the amount of improperly disposed-of waste in Germany.

12 Limitations of the Model

A model is always a simplified representation of the reality. For this reason, not all factors can be taken into account in the model “From Land to Sea – Model for the documentation of land-sourced plastic litter”.

As already described, the limitations to the model result, on the one hand, from the frame of reference and the definitional classifications, and, on the other hand, from the frequent lack of the necessary data and information on relevant issues, which prevents them from being included in the model. This leads, for example, to a situation in which a differentiated classification according to the type and size of plastic in the model is currently not possible. Also, the importance of tourism, the discharges of plastic particles through the air, the effects of natural phenomena such as floods, and special events such as music festivals cannot be taken into account

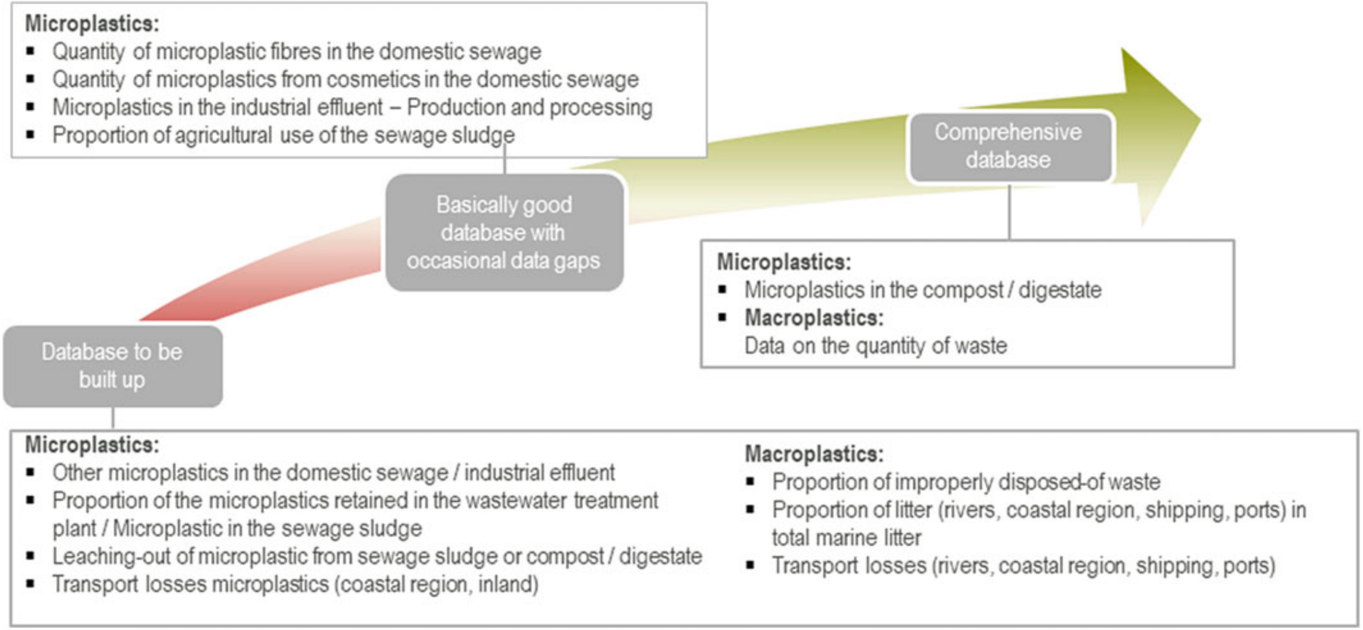


Fig. 6 Evaluation of the data. © Conversio, From Land to Sea – Model for the documentation of land-sourced plastic litter, 2018

separately in the model at the present time due to a lack of usable information. However, all this information also goes wherever possible into the model via the factors that were established for calculating the discharge of plastics into the sea attributable to Germany.

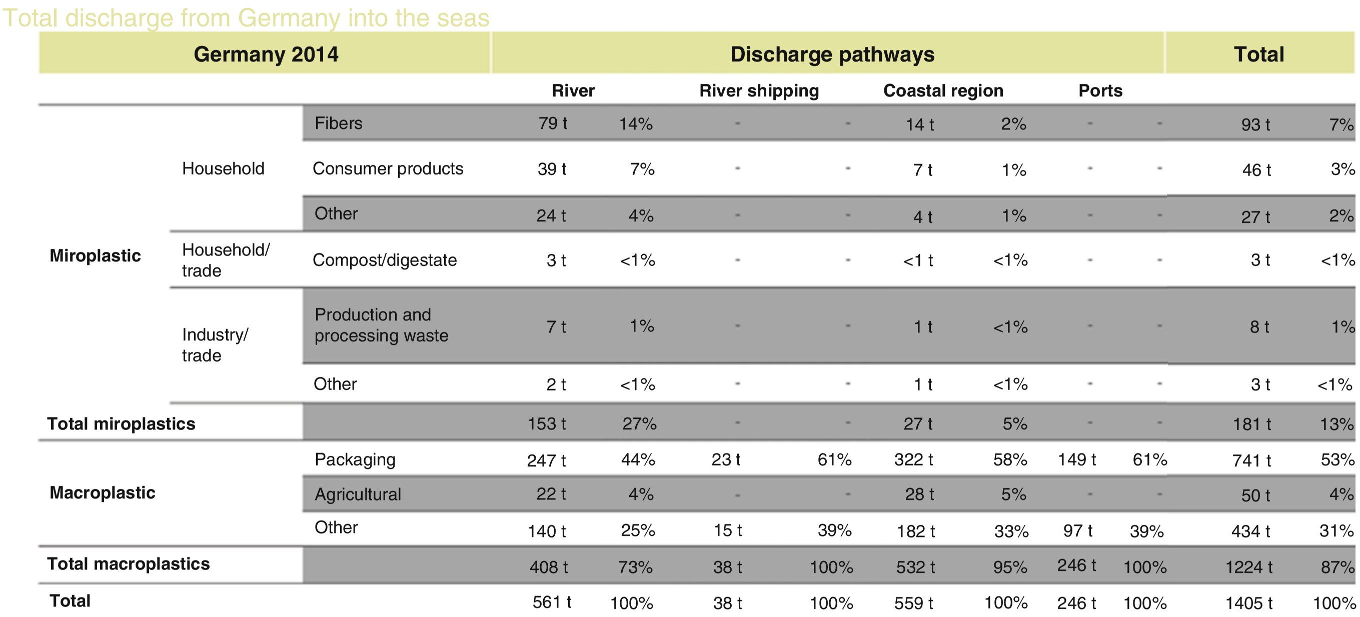
13 Main Results

Most of the plastics discharged into the sea from Germany are macroplastics (Fig. 7). This result must always be considered taking into account the distinction used in the model between microplastics and macroplastics, as well as the given definition of "plastics". Furthermore, the proportion of macroplastics in the total discharges differs from one sea to another. By far the highest proportion of macroplastics is discharged into the Baltic Sea. This is explained by the high percentage of the coastline of the river basin districts that are connected with the Baltic Sea. The majority of the population which has to be considered within the calculations for the Baltic Sea live on the coast. Because of the proximity to the sea and the partial direct discharge into the sea, the transport losses are lower. Consequently, more macroplastics get into the sea and are not, en route, permanently eliminated again from the discharge pathway.

As regards the discharge pathways, the two pathways "rivers" and "coastal regions" dominate. The majority (approx. 80%) of the total discharge gets into the seas, i.e. North Sea, Baltic Sea, and Black Sea via these two pathways.

Virtually all the litter entering the Black Sea that is attributable to Germany is transported via the discharge pathway "rivers". Because Germany does not directly border the Black Sea, only the "rivers" and "river shipping" pathways are of relevance here, but not the "coastal regions" pathway.

Discharges via landfills are of no relevance in Germany, because in Germany it is forbidden to dump carbon-containing waste – and thus plastic-containing waste – on landfills of classes I, II, and III (Definition and descriptions according to German law: § 2 Number 7 and 8 of the German "Verordnung über Deponien und Langzeitlager (Deponieverordnung – DepV)" and Annex 3 of the German "Verordnung über Deponien und Langzeitlager (Deponieverordnung – DepV)". Waste can only be deposited on landfills of classes I, II or III if it fulfils the criteria according to law). Only a neglectable amount of plastics end up on landfills of classes IV – an underground landfill for hazardous waste. For this reason, an account of the discharge pathway "landfills" has been dispensed with in the report on the model.



Total discharge from Germany into the seas

Germany 2014			Discharge pathways								Total	
			River		River shipping		Coastal region		Ports			
Miroplastic	Household	Fibers	79 t	14%	-	-	14 t	2%	-	-	93 t	7%
		Consumer products	39 t	7%	-	-	7 t	1%	-	-	46 t	3%
		Other	24 t	4%	-	-	4 t	1%	-	-	27 t	2%
	Household/ trade	Compost/digestate	3 t	<1%	-	-	<1 t	<1%	-	-	3 t	<1%
	Industry/ trade	Production and processing waste	7 t	1%	-	-	1 t	<1%	-	-	8 t	1%
		Other	2 t	<1%	-	-	1 t	<1%	-	-	3 t	<1%
Total miroplastics			153 t	27%	-	-	27 t	5%	-	-	181 t	13%
Macroplastic		Packaging	247 t	44%	23 t	61%	322 t	58%	149 t	61%	741 t	53%
		Agricultural	22 t	4%	-	-	28 t	5%	-	-	50 t	4%
		Other	140 t	25%	15 t	39%	182 t	33%	97 t	39%	434 t	31%
Total macroplastics			408 t	73%	38 t	100%	532 t	95%	246 t	100%	1224 t	87%
Total			561 t	100%	38 t	100%	559 t	100%	246 t	100%	1405 t	100%

Fig. 7 Results of the calculations of the discharge. © Conversio, From Land to Sea – Model for the documentation of land-sourced plastic litter, 2018

14 Conclusions

The report and handbook belonging to the model are continuously updated and further revised. In addition to continuously adapting the above-mentioned framework conditions, the model parameters are also checked and, wherever necessary, modified. The evaluation of the model is accompanied by consultations with external experts.

The open structure of the methodology makes it possible to also apply it to other regions or countries and to incorporate further discharge pathways. The advantage of the model lies, in particular, in the easy and flexible adaptation of variables and calculations. The aim is, therefore, to transfer the model to other regional conditions. The report and handbook for the model are available in German and English and can be obtained free of charge via the BKV website. Like the model, the "Special study on littering" and the "Special study on compost/digestate" can be ordered free of charge on the BKV website (https://www.bkv-gmbh.de/studies.html).

References

1. Jambeck et al (2015) Plastic waste inputs from land into the ocean
2. PlasticsEurope Market Research Group (PEMRG) and Conversio Market & Strategy GmbH (2018) Plastics – the facts 2018
3. Conversio Market & Strategy GmbH (2018) Material flow analysis plastics in Germany 2017
4. Conversio Market & Strategy GmbH (2017) Post-consumer plastic waste management in European countries 2016 – EU 28 + 2 countries
5. Fraunhofer UMSICHT (2018) Kunststoffe in der Umwelt: Mikro- und Makroplastik, Kurzfassung der Konsortialstudie
6. Öko-Institut e.V. and ZEUS-GmbH (2020) Status Quo, Handlungspotentiale, Instrumente und Maßnahmen zur Reduzierung des Litterings
7. Conversio Market & Strategy GmbH (2018) Verification of the factor 0.3% within the model "From Land to Sea – Model for recording land-sourced plastic waste"
8. Conversio Market Strategy GmbH (2020) Sonderbetrachtung Kompost und Gärrückstände

Plastic Waste Management: Current Status and Weaknesses

Oksana Horodytska, Andrea Cabanes, and Andrés Fullana

Contents

1 Introduction 290
2 Waste Management in Developed Countries 293
  2.1 Collection 293
  2.2 Sorting 296
  2.3 Treatment 297
3 Waste Management in Developing Countries 299
  3.1 Latin America 299
  3.2 Asia 300
  3.3 Africa 301
4 Circular Economy and Weaknesses 302
5 Conclusion 304
References 305

Abstract The weaknesses of existing plastic waste management strategies lead to the pollution of the natural environment. Although around 75% of plastic litter come from developing countries, an important 25% is originated in western countries mainly due to the limited efficiency of the collection systems and low recycling rates. Global plastic production has almost doubled over the last decade, and it is predicted that it will continue to grow. This chapter provides an extensive review of current waste management routes and existing recycling and recovery options. Two types of plastic products have been considered: rigid and flexible materials. These materials show different behaviour and usually are treated separately. Plastic waste sources can also be diverse, but they are commonly grouped into post-industrial and post-consumer. In this chapter, the focus has been placed on post-consumer plastics since a higher amount of this type of waste is being generated and its treatment is more challenging.

Keywords Circular economy, Plastics, Recycling, Upcycling, Waste management

---

O. Horodytska (✉), A. Cabanes, and A. Fullana
Chemical Engineering Department, University of Alicante, Alicante, Spain
e-mail: oksana.hka@ua.es; andrea.cabanes@ua.es; andres.fullana@ua.es

Friederike Stock, Georg Reifferscheid, Nicole Brennholt, and Evgeniia Kostianaia (eds.),
Plastics in the Aquatic Environment - Part I: Current Status and Challenges,
Hdb Env Chem (2022) 111: 289–306, DOI 10.1007/698_2019_408,

© Springer Nature Switzerland AG 2019, Published online: 18 September 2019

1 Introduction

Plastics can now be found in every aspect of our lives. There are several good reasons for such success. Excellent mechanical and thermal properties, low manufacturing costs, versatility, and lightness are just some of the advantages that are worth mentioning. Usually plastic materials are divided into thermosets and thermoplastics. Thermosetting polymers present highly crosslinked structures which provide the materials with high mechanical and physical strength and heat stability. Well known thermosets are epoxy and phenolic resins, polyurethanes (PUR), and acrylonitrile butadiene styrene (ABS). These materials are mainly used when heat and chemical resistance is required (e.g. automotive manufacture, construction equipment, electrical components). Thermoplastic materials consist of linear or branched chains linked by intermolecular interactions. This is a flexible structure which allows thermoplastics to flow when the temperature is high and to solidify when the temperature decreases. Some of the polymers belonging to this group are polyethylene (PE), polypropylene (PP), polyvinylchloride (PVC), and polyethylene terephthalate (PET).

More than 8.3 billion tons of plastic have been produced worldwide since the 1950s [1]. The global plastic production rose sharply from 0.35 million tons in 1950 to 348 million tons in 2017. China is the world's largest plastics producer (Fig. 1), accounting for 29.4% of global production in 2017, followed by Europe (18.5%) and North America (17.7%). European plastic demand reached 51.2 million tons in

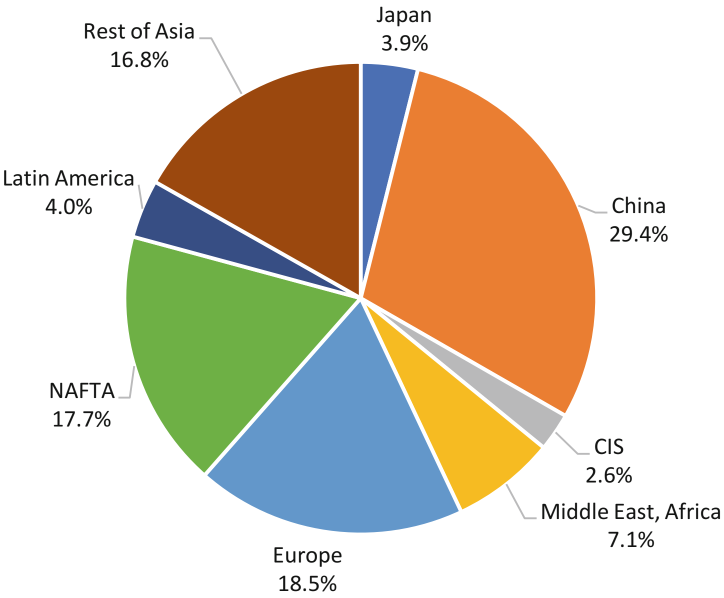


Fig. 1 Distribution of global plastic production in 2017 [2]

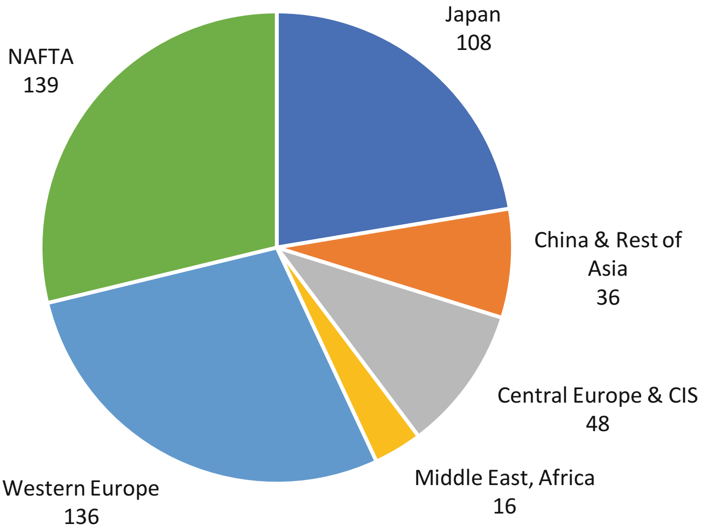


Fig. 2 Plastic consumption rates per region in 2015 (kg/person) [3]

2017, with Germany in the lead, followed by Italy, France, Spain, and the UK [2]. Globally, North America, Western Europe, and Japan show the highest consumption rates (Fig. 2). The regions which produce the most plastic are not necessarily the largest consumers. For instance, Japan is a region with one of the lowest plastic production rates; however, it is one of the biggest consumers. On the other hand, China is the largest producer, but the consumption rate is lower compared with other regions (almost four times smaller than North America or Europe) [3].

Plastic products applications vary from one sector to another. There exist a wide range of polymers, each one with specific properties that make them ideal for each application. Polymers such as PE, PP, PVC, and PET are the materials most in demand, followed by polystyrene (PS) and PUR. The packaging sector has become the most important application representing almost 40% of the total [2]. Polyethylene is one of the most versatile polymers owing to a changeable degree of chain branching. The polymerization conditions can be altered to produce the desired structure. Low-density polyethylene (LDPE) has a branched structure that makes the material clear, flexible, easy of processing, and with good heat sealability. It is widely used in secondary and tertiary packaging (shrink and stretch wrap films), reusable bags, and agricultural films. Fewer branches increase the density of the material. This is the case for medium-density polyethylene (MDPE) which is slightly stronger, stiffer, and less permeable than LDPE. On the other side, there is the high-density polyethylene (HDPE), which is a linear and crystalline PE. It has a good water vapour barrier (better than LDPE), resistance to different chemical compounds, and good tensile and impact strength. Also, it is a non-transparent material with poor gas barrier characteristics. HDPE is widely used in hard packaging (e.g. milk, juice, water bottles; cosmetic containers; pharmaceutical bottles) and flexible food packaging (e.g. cereals, snacks, crackers). PE materials can be also produced by copolymerization to obtain, for example, ethylene vinyl acetate (EVA) or linear low-density polyethylene (LLDPE). The second most commonly used thermoplastic after PE is PP. It has a lower density, a higher melting point and a higher rigidity. It is also a good barrier to moisture, so it is used for dry food and bakery packaging. Hard PP can be found in automotive parts, microwave containers, and pipes. The optical, mechanical, and barrier properties can be improved by orientation methods [4]. There is a wealth of information on plastic's properties and applications, but it is beyond the scope of this chapter. Therefore, only a few of them have been described.

High production rates and a lack of consumer awareness have led to uncontrolled plastic waste generation. Globally, around 150 million tons are released every year [5]. In the USA, 34.56 million tons of post-consumer plastic waste (including residential, commercial, and institutional sources) was generated in 2015. Recycling rates remain low (slightly higher than 9%), while almost 76% of plastic solid waste (PSW) is disposed in landfills [6]. The remaining fraction is used as an energy source. In 2016, the European Union countries plus Norway and Switzerland generated 27.1 million tons of post-consumer plastic waste. Of this, 31.1% was recycled, 41.6% was recovered as energy, and 27.3% ended up in landfills. It was the first time that recycling overtook landfilling. The rates, however, vary among countries. For instance, Switzerland, Austria, or Germany has committed to recycling and energy recovery by implementing landfill restrictions. Meanwhile, Bulgaria, Greece, or Malta still buries more than 70% of their waste in landfills [2]. In developing countries, the percentage of plastics in municipal solid waste (MSW) streams is on the increase, mainly due to changes in people's lifestyle. The summary of annual PSW generation in some Asian cities (from Indonesia, India, Thailand, Malaysia, Iran, and Bangladesh) can be estimated around 1 million tons [7]. However, accurate data are difficult to obtain. Seven major cities in India produce nearly 500,000 tons of plastic waste per year, representing 4% of the total MSW [8]. In a recent study, researchers estimated that in 2015, the Asian continent was the largest contributor to global plastic waste, generating 82 million tons. The following regions are Europe (31 million tons), North America (29 million tons), Latin America and Africa (19 million tons each), and Oceania (0.9 million tons) [9].

Waste treatment methods can be divided into mechanical recycling, chemical recycling, and energy recovery. Here a distinction is drawn between post-industrial and post-consumer waste. The former is generated during the converting operations (rejects and offcuts), and it is usually clean and homogeneous. The latter consists of a mixture of products at the end of their service lives. There are two types of post-consumer waste. On one hand, there is the so-called commercial waste which is mainly secondary and tertiary packaging from retail industry area. The composition is usually known and homogeneous, and the amount of physical impurities and chemical contaminants is low. On the other hand, there is domestic post-consumer waste coming from kerbside collection. This waste is dirty, highly contaminated, and heterogeneous.

2 Waste Management in Developed Countries

A number of actions are necessary to properly manage the waste produced by human activity, such as collection, transportation, sorting, and disposal or treatment. The European Union Waste Framework Directive [10] states that operations intended to manage waste must not cause any damage to the environment or human health. The waste hierarchy has been established to determine priority among waste treatment processes (prevention or minimization, preparing for reuse, recycling, recovery as energy, and lastly disposal). However, the waste hierarchy cannot be strictly applied because it depends strongly on waste composition and characteristics.

2.1 Collection

Plastic recycling always begins with waste collection whether at households or in the industry, and the ratio of collected plastics will severely impact the recycling efficiency of a locality and will undoubtedly keep from diffuse pollution of plastic litter leaking into the environment. In Europe (EU28 + NO/CH), the average plastic waste collection rate increased by 12% between 2006 and 2016. Germany contributed with a 41% improvement and Poland with 40%. Other European countries did not match these figures (the UK 13%, France 9%, Italy 2%). Actually, in Spain, waste collection decreased by 7% [11].

Regarding commercial and post-industrial plastic waste collection, companies are responsible of segregating the different waste fractions, and these are more likely to do so if incentives are given in return by the local authorities. Even so, companies usually recycle their plastic waste as it is virtually clean and, therefore, highly valuable for recyclers. Moreover, specialized recycling companies generally offer a collection and recycling service for purchasing the post-industrial and commercial plastic scrap [12].

Concerning municipal solid waste, there are multitudes of schemes when it comes to collection, but most of them can be summarized in four main groups: kerbside collection, drop-off collection, buy-pack centres, and deposit collection methods [13].

- *Kerbside collection* of plastic solid waste is referred to waste disposal within communities, meaning buildings and individual houses, that separate plastic and metal container packages from the rest, organics, paper, etc., and dispose them in either plastic bags or containers to be collected on a specified day of the week or the month, according to the municipality regulations [14] (Fig. 3a).
- *Drop-off or bring point collection methods* consist of different containers, the use of which is clearly indicated and where organics, paper, rest of food, and plastic are disposed separately (Fig. 3b). Generally, these containers are placed near residential areas in order to be accessible for citizens to dispose their household plastic waste among other recyclable materials. However, nowadays, the use of

(a) (b)

(c)

Fig. 3 Different waste collection options. (**a**) Kerbside collection in Erlangen (Germany). (**b**) Bring point collection in Alicante (Spain). (**c**) Multi-material collection system in Lausanne (Switzerland). Pictures by Andrea Cabanes and Oksana Horodytska

drop-off collection is increasing not only in neighbourhoods but also in public areas such as parks, schools, universities, etc., so that all the waste disposed can be separated and recycled, not only at homes. In populated areas, bring points are more frequently seen than kerbside collection, since the amount of disposal points and transportation resources are significantly reduced [14].

- *Buy-pack containers*. The operability of buy-pack containers is similar to the drop-off collection systems, with the exception that this scheme aims to encourage more citizens to separate their plastic fraction by means of an incentive that is received for the disposal of each plastic container, e.g. PET bottles. At the same time, the separation avoids an extra contamination in the plastic package and subsequently facilitates the recycling process. This kind of collection system is normally placed in supermarkets [14].
- *Deposit programmes* aim to reduce plastic waste pollution and impel citizens to return their recyclable containers. The way it works is quite similar to buy-pack containers, but in this case the end-user has to pay an additional cost for the plastic container that will be refunded after taking it back to the so-called reverse vending machine. Consequently, most of packed food stuff in the supermarket will have an added cost, driving citizens whether to buy non-packed food or to return the package in order to get the deposit back [15, 16].

This collection scheme seeks to increase the recyclability not only of the plastic waste produced at home but also of the waste produced outdoors. Meaning that, for example, there is a greater chance that people will contribute to return the plastic bottles consumed outside home to get the deposit back instead of discarding them in the general containers placed on the street. In the last years, this collection scheme has been growing across European countries, North America, and Australia [17].

Nevertheless, in both *kerbside and drop-off* collection methods, plastic is not always collected separately, and, in some cases, all municipal solid waste is collected in the same fraction to be sorted later in the corresponding facilities. However, the single-fraction collection system is decidedly more inefficient in terms of materials recovery and, therefore, yields in a lower recycling ratio of plastic packaging due to the increase of contaminants attached to the material, which are originated as a consequence of being in direct contact with rests of food, care products, cleaning products, and so forth. Regardless the inconveniences for plastic recovering, there exist several solid waste collection systems ranging from zero separation fraction to specific materials separation, as would be the case of PET bottles [18].

- *Single fraction – mixed solid waste*: all solid waste is disposed in one single bin, and the mixed waste-processing facilities are responsible for collection, transporting, and separation of plastic, metal, paper, and organic fractions.
- *Two fractions – dry waste and organic*: in this case citizens are responsible for separating the organic biodegradable fraction from the rest of waste. This scheme would decrease the level of contamination in the packages, but still a complex sorting process is required to recover the different types of materials.
- *Three fractions – organic, paper and cardboard, and rest of dry waste*: in this case paper and cardboard will result in a lower level of contamination which may facilitate the recycling process, and the rest of dry waste contains mainly plastic and metal packaging. This is the most usual system found around Europe.
- *Multi-material collection systems* are applied, for example, in Switzerland separating PET bottles, aluminium, and paper from the rest of waste (Fig. 3c). The aim of multi-material collection is to recover valuable materials while minimizing the content of contaminants.

Once the waste is fractionated, there are mainly two types of trucks that can be utilized for waste transportation: multisection or single section trucks [18]:

- Multisection trucks offer the possibility of collecting all waste at once, which seems an efficient option for multifraction systems. However, there is a clear limitation to consider, as the truck must discharge whenever one of the chambers is completely full, even though the rest of compartments remain empty. Therefore, if the amount of waste is not balanced among the different fractions to be collected, this would not be an appropriate solution for waste transportation.
- The other option is to use single section trucks. In this case, if each waste fraction has to be transported in a different truck to avoid contamination in recyclable materials (such as plastics and cardboard), then the inconvenience would be the

necessity of a larger number of vehicles, which also involves a higher initial investment in the collection system.

The frequency of collection in both cases varies depending on the local authorities, but in general, it mostly depends on the weather conditions. For instance, Southern Mediterranean countries in Europe collect the household waste almost daily, since the elevated temperatures favour the decomposition of organics and lead to unpleasant odours emitted to the environment [18].

2.2 Sorting

Sorting of waste is carried out in so-called material recovery facilities (MRFs) and depends on the existing collection schemes. In general, there are two types of MRFs. On the one hand, facilities that receive mixed waste from single stream collection are called mechanical-biological treatment (MBT) plants (in EU) or dirty MRFs (in the USA). On the other hand, there are dual-stream or clean MRFs where only potentially recyclable materials are treated.

Technologies and equipment used vary among facilities and depend on the input waste characteristics. Some commonly used technologies are bag splitters, ballistic separation, air separators, and optical detection systems.

Single-stream MRFs require a previous sorting operation to separate the fibres (paper and cardboard) and containers or packaging (plastics, glass, and metals). A trommel screen and disc screens are typically used for this purpose. After that, different materials follow their own sorting process. Plastics recovered for recycling are mainly rigid containers. Plastic films, wraps, and shopping bags from commingled waste are usually considered contaminants and sent to landfills or energy recovery. Additionally, magnetic separation is used to remove ferrous metals, and eddy current systems are used to remove non-ferrous metals.

Plastic sorting processes consist of several mechanical operations. First, bag opening takes place, and materials are released onto a downstream conveyor. Lightweight (2D) and heavier (3D) products are separated using ballistic or air separators. Ballistic technology consists of an inclined screen where the heavy rigid products roll downwards and lighter films and papers are pushed upwards. Air separation is based on the difference in material density since lighter materials can be removed from the conveyed stream using air at relatively low velocity. Optical sorting is used to distinguish between different materials, for instance, plastic and paper. Near-infrared optical detection (NIR) is a burgeoning technology based on the wavelength reflected after light is incident upon the material. Sorting of a range of polymers (e.g. PE, PP, PET) is a big advantage. However, there are still several limitations associated with NIR. Multilayer, coated, or black plastics pose specific challenges and lead to missorting. Rigid plastics from different sources (WEEE, packaging, bottle caps, etc.) can be sorted into pure polymer fractions using electrostatic plastic separators. These systems operate on the principle of the different tribo-electric behaviour of involved plastic materials.

Effective sorting is the key to increasing plastic recycling rates. However, it is still underdeveloped especially for flexible plastics recovery. Rejected plastic from sorting facilities are usually sent to landfill and can end up in nature.

2.3 Treatment

Plastic recycling processes can be divided into different groups based on polymer classification (thermosets or thermoplastics), waste source (post-industrial or post-consumer), and plastic type (rigid or flexible).

Thermosetting plastics (ABS, epoxy and polyester resins, etc.) cannot be recycled using temperature owing to their ability to withstand heat. Therefore, the most common waste treatment is energy recovery through combustion or pyrolysis. Alternatively, mechanical recycling is carried out via grinding or pulverization. In this case, the plastic flakes produced can be utilized as fillers in new products. Finally, several chemical recycling technologies have been developed to depolymerize plastic materials using solvents and supercritical fluids. For instance, Connora Technologies has developed a low-energy recycling technology for thermosetting resins recovery including the conversion of thermosets into thermoplastics [19].

Thermoplastics have excellent mechanical properties, lightness, versatility, and relatively low processing costs. Accordingly, they are used in many applications such as packaging, agriculture, and building and construction. Thermoplastics have become single-use or short-life products due to the ease of production and low costs. This has led to an increase in the volume of thermoplastic waste, and it is more likely to escape from municipal collection schemes. Waste treatment methods for these materials are described hereafter.

Post-industrial waste can be successfully recycled through mechanical recycling processes. Direct re-extrusion is used when the input material contains a minimal level of contamination. The quality of the recycled products is similar to the original material, and, thus, it can be used for the same application. This is known as closed-loop recycling or in-house recycling since converting companies can recover their own scrap without leaving their facilities. However, when some contaminants are present, such as ink or adhesives, more complex technologies are required. Usually, plastic washing lines are used to remove the contaminants and extrusion machines with ultrafine filtration, homogenization, and degassing might be needed. As a result, the materials suffer degradation and some loss of properties. Hence, it is suitable only for less demanding applications. This is known as open-loop recycling.

Both closed-loop and open-loop systems contribute to reduction of virgin plastic consumption. However, closed-loop recycling is a preferred option from an environmental point of view because of higher quality of the recycled pellets. This means that these products can be used during several life cycles. On the contrary, products made of more degraded recycled pellets from open-loop system are more likely to end up in landfills or incineration plants after the use phase.

Post-consumer waste is more difficult to recycle through mechanical operations mainly due to high levels of contamination. Usually, only commercial and separately collected or efficiently sorted plastic waste (e.g. PET bottles, shopping bags, etc.) can be recycled using open-loop systems. Generally, the process consists of several mechanical operations such as grinding, washing, drying, and pelletization. The recycled pellets can be used only for nondemanding applications (trash bags, pipelines, flowerpots, etc.). Chemical recycling can be used for highly contaminated mixed plastic waste. However, these processes are not fully developed and can be energy intensive. Therefore, post-consumer domestic waste usually goes to landfills or, at best, to energy recovery plants.

Rigid and flexible plastics undergo different recycling processes. Properly collected and sorted rigid plastics (e.g. water bottles, pots, and trays) can be easily recycled through mechanical recycling methods. PET bottles closed-loop recycling is one of the most successful material recovery systems. This can be attributed to high-grade resins used in original products manufacturing, inert character of PET, and efficient collection schemes that ensure that the contamination level is minimized. Mechanical recycling consists of grinding the material into small flakes, washing (or decontaminating) with detergents, and re-extrusion. Recycled PET is suitable for new bottles manufacturing, but sometimes it is used for production of new products different from the original one. Depolymerisation of PET residues (chemical recycling) is also applied to reuse the monomers in new polymerization processes [20].

Mechanical recycling of flexible plastics is somewhat more challenging particularly due to their low bulk density and low thickness. Washing and drying efficiencies are lower than with hard plastics because the equipment has not been adequately designed to handle flexible plastics. Only post-industrial and selectively collected monolayer plastic films are currently mechanically recycled. The quality of the recovered materials depends on the input waste characteristics. For instance, the presence of ink on the plastics surface leads to coloured pellets suitable solely for nondemanding applications (downcycling). In recent years, several technologies have been developed to remove the ink before extrusion [21]. The University of Alicante has patented an innovative process based on an aqueous solution of nonhazardous chemicals. This is an environmentally friendly technology that produces clear pellets of high quality which can be used for the same application as the original products (upcycling) [22].

Currently, monolayer films recycling shows good results, whereas, multilayer films cannot be recovered yet at the industrial scale. The problem lies in the combination of noncompatible materials (polymers, aluminium, paper, etc.) that produces severe defects during extrusion. There are three main branches of research focused on multilayer films recycling. The first one is the use of compatibilizers to improve the interactions between different polymers inside the mixture. The second one is delamination (separation of the layers) and recovery of the different materials individually. Lastly, there is the technique of dissolution-precipitation, which involves the use of solvent or non-solvent systems to selectively dissolve one of the materials and separate them afterwards [23].

3 Waste Management in Developing Countries

All successful waste management scenarios begin with collection and sorting operations. The problem of environmental pollution and waste accumulation in developing countries can be precisely attributed to the lack or inefficiency of municipal collection strategies. Unfortunately, inexistent waste collection services and limited capacity of landfills force the inhabitants and authorities to throw the waste directly to rivers and canals.

One clear similarity among different developing countries is the presence of an informal recycling sector. This is an unregulated waste management activity performed by local individuals or groups to improve their economic situation. The so-called waste pickers collect potentially recyclable materials directly from the streets and dumps and sell them to local recycling companies [24]. Sorting technologies are almost non-existent, and classification and separation are carried out by hand. Recyclable waste from the informal waste recycling sector can be sorted in situ. Although in some cities this is the only way to recover recyclable materials because there is no formal separate collection system, from the social perspective, informal recycling is usually associated with atrocious working conditions. Fortunately, in some countries of Latin America (Brazil, Colombia, and Argentina), municipal authorities are starting to recognize the labour of these waste pickers and to integrate them in formal waste management strategies [25]. Mechanical recycling is the preferred option and PET, and polyolefins are the most recycled materials. Recycling processes, as in developed countries, consist of grinding, washing, drying, and re-granulation. To increase the profitability, some companies invest in sophisticated equipment from reputable European machinery manufacturers.

3.1 Latin America

In countries with emerging economies such as Brazil, waste generation has increased over the last decades mainly due to the rise of the average income. Consequently, national authorities were forced to implement MSW regulations (Lei 12305, 2010) [26]. Several big metropolitan areas (e.g. Rio de Janeiro, João Pessoa) count with separate collection systems and material recovery facilities [27]. In general, the plastic recycling sector in Latin America is experiencing considerable growth in recent years. Several recycling companies exist in different countries (e.g. Mexico, Brazil, Peru, and Honduras) aimed mainly at food-grade PET production. Recyclers usually receive sorted materials directly from industries or from intermediates that work with waste pickers. Kerbside collection of domestic waste remains a weak point for most cities and regions. Furthermore, even if a municipal collection strategy exists, mixed domestic waste is usually sent to landfills (controlled or uncontrolled). Only a small fraction ends up in sorting facilities where the predominant method is still manual separation.

3.2 Asia

Within the Asian region, China is the country with the highest ratio of plastic debris leaking into the marine environment, followed by Indonesia and the Philippines. These countries have experienced an economic growth in the last years, which has been directly reflected on an increase of solid waste pollution, and the plastic leak is basically the result of a scarce waste collection system or a lack of it [28].

For many years China has been the destiny of not only their own plastic waste but Europe's and America's plastic waste, resulting in the generation of roughly 440 million tons yearly. In China the overall collection rate is under 40%, leading to 260 million tons of uncollected plastic waste leaking into the environment, which contributes to 84% of the global plastic pollution. Nevertheless, there is a significant difference between urban and rural areas. In urban areas, the collection rate of plastic waste is reaching 65%, while in rural areas, more than 95% of plastic waste is totally uncollected, and residents resort to traditional waste management habits, such as uncontrolled waste disposal, burning, or dumping in the river, which leads to a high load of contamination in both land and oceans. In urban areas, the most common recovery system for plastics is at incinerators or sanitary landfilling. All in all, only 11% of all plastic waste generated is collected by waste pickers to be recovered in recycling facilities [29].

On the other side, the collection system in the Philippines is, at least initially, considerably more efficient than in China; specifically, the mean rate of plastic waste collection is nearly 85%. In this case, the collection system efficiency differs between rural and urban areas too, although the collection rate drop-off in rural areas is not that high when compared to China, above 40%. Cities with high population density, e.g. Metro Manila, reach 90% of plastic waste collection. The waste management success in the Philippines is due to the regulation "Ecological Solid Waste Management Act of 2000" (Republic Act No. 9003), which targets zero waste and recycling of all valuable products by promoting the segregation of materials at source, dividing them into four main groups: compostable, recyclable, nonrecyclable, and special waste. The objective is to collect them separately and make the most of plastic materials. However, in 2009, there was no evidence of a realistic change in the country, and almost every city continued to perform uncontrolled landfilling and open incineration. Fortunately, owing to the Global Alliance for Incinerator Alternatives (GAIA), the regulation Waste Management Act of 2000 was implemented in 25 localities, and after 2011, open burning and dumping was totally banned. Domestic waste is nowadays segregated for composting and recycling [30]. In spite of the progress made, the Philippines greatly contributes to plastic litter in the ocean. Surprisingly, the issue of plastic waste management in the Philippines is not at the collection point but after collection. It has been reported that 74% of the plastic leakage in the Philippines is originated from the collected fraction. Moreover, the main treatment to plastic and household waste is open burning and uncontrolled landfilling. Only 25% of the plastic waste stream is recovered, although the recovery ratio changes significantly from PET bottles (90%) to flexible packaging (<5%). The plastic recovery is mainly done by waste pickers at different points: at household, during collection while the trucks are moving, and at material recovery facilities. The waste officially collected is discarded in open dump sites (600 available) and sanitary landfills (70 available) [29].

By 2014, it was estimated that Indonesia recycled and composted only 7% of its household waste, 5% was treated with uncontrolled incineration, 10% was illegally landfilled, and 9% of MSW was leaked to the environment, which includes land regions as well as rivers and oceans. The 69% left was landfilled, which is the most common treatment for household waste in this region [31]. More specifically, in Jakarta, waste pickers are the major force in segregating plastics from the rest of waste, since the official collection system only considers dumping plastics into the landfill without any treatment. However, there is an alternative to municipal waste management called "waste bank", where citizens can deliver their presorted waste separated in paper, metals, plastic, and organic fractions in return for cash refund. Companies responsible for the waste banks gain profit by selling the sorted waste to recyclers [32]. This waste is then recycled in the case of plastics, metals, and paper or composted in the case of organic fractions. However, the municipality does not take part in these waste banks, and there are only individuals who are interested in sorting their household waste to increase their income. A recent study estimated that Jakarta recovers 34% of all plastic waste through waste pickers and the waste bank but only 24% of the total plastic waste generated is recycled. The rest remains in landfills or leaks into the environment [33].

The situation in India is similar to the rest of the Asian countries mentioned above. Waste pickers are also a relevant figure in the waste management system, contributing to 20% of the total waste collection. In this region, their practice is not only picking and sorting MSW but also managing waste pickers associations, where valuable materials such as plastics are recycled. On the other side, the official waste collection system in India consists mainly of collection and transportation to dump sites, where organic and valuable materials are burnt together [8].

3.3 Africa

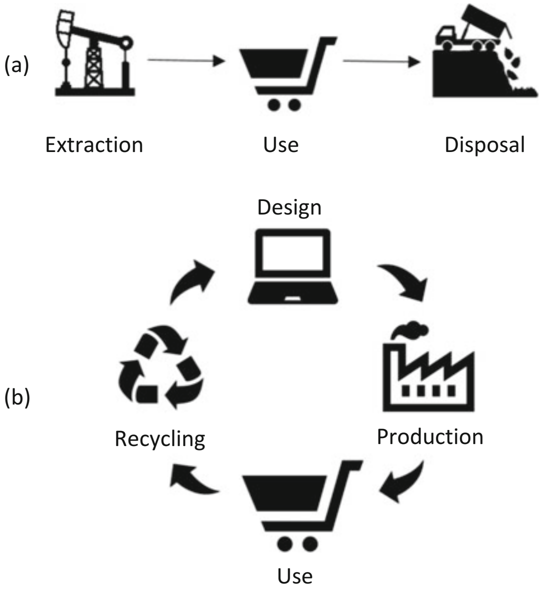
In Africa's developing countries, the same scenario is repeated: waste management basically means transportation from household collection points to open dump, where the waste is either burnt or just landfilled and plastics recycling is only a future project. On top of it, most of these countries are running out of landfilling space; besides, their waste management plan is not able to cope with all the waste generated by their growing population [34].

4 Circular Economy and Weaknesses

The current economy model based on extraction, use, and disposal is not sustainable from the environmental point of view, especially for fossil-based non-biodegradable plastics. Several associations and academic institutions have joined forces to develop the idea of a new economy model, the circular economy (Fig. 4). The fundaments lie in trying to mimic the natural ecosystems. Zero waste, diversity, use of renewable energy, and interaction between systems are the main principals of this economy approach. Regarding plastics and other non-biodegradable materials, the following actions should be taken after the product's service life (in order of preference): maintenance, reuse, refurbishment, and, finally, recycling. Closed-loop mechanical recycling and upcycling provide the highest economic and environmental benefits. Therefore, recycled pellets should be suitable for high-demanding applications. On the contrary, energy recovery and landfilling must be minimized.

The selection of the most appropriate recycling method depends on the quality of plastic waste and the degree of degradation. Currently, upcycling is feasible only with clean, non-contaminated waste. Contamination and inappropriate use of plastic goods significantly diminish material's quality. Furthermore, degradation also occurs during recycling operations. There are different types of plastic contaminants. For example, coating, inks, adhesives, and additives, intentionally added (IAS)

Fig. 4 Schematic representation of the linear economy model (**a**) and the circular economy model (**b**) [35]



during manufacturing, produce defects (bubbles, voids, gels, etc.) when the material goes through re-extrusion at a high temperature. Other contaminants (labels, missorted noncompatible polymers, etc.) and dirtiness adhere to the plastics during the use phase and collection. These are denominated non-intentionally added substances (NIAS). To ensure effective recycling into value-added secondary products, plastic waste requires decontamination.

Both IAS and NIAS pose serious problems for the implantation of closed-loop recycling processes. The packaging sector generates an enormous volume of plastic waste, which cannot be recycled into new packages (especially food packages) due to high consumer's safety requirements. The NIAS, which can be degradation or reaction products, impurities, etc., can migrate from the packaging layer into the food and may constitute a hazard to human health. These substances are hard to detect, and, if detected, it is difficult to establish their origin. High sensitivity advanced analytical techniques are required. Headspace solid phase microextraction (HS-SPME) and gas chromatography-mass spectrometry (GC-MS) are common techniques used to identify volatile and semi-volatile organic compounds. Liquid extraction and GC-MS are used to identify semi-volatile and non-volatile compounds. And, finally, liquid chromatography (HPLC) and high-resolution mass spectrometry (HRMS) techniques such as time of flight (TOF) identify non-volatile compounds when mass spectra libraries are not available [36]. Some plastic manufacturers try to minimize the generation of NIAS during their processes by optimization of manufacturing parameters. In the recycling sector, there is little progress, but the elimination of undesirable substances might be the key for recycled products upgrading.

The fraction of volatile NIAS that confer an aroma to the plastic represent a barrier for the circular economy. Even if post-consumer recycled plastic is intended to be only used for non-food packaging applications, the unpleasant odour produced as a consequence of food and cosmetics contamination among others permeates into the polymer matrix and is not removed through the current recycling process. For this reason, this kind of materials is nowadays downcycled rather than recycled, since consumers are likely to refuse any package made of post-consumer recycled plastic. To put it simply, none would buy a shampoo bottle releasing a cheesy aroma, which presents a roadblock for the plastics recycling industry [37]. As evidence of this, a recent study revealed that the volatiles released from post-consumer high-density polyethylene (HDPE) confer a soapy and citrus-like odour to the recycled plastic, while virgin HDPE do not emanate this kind of odours at all. The study also compared the input plastic waste from the kerbside collection with the resulting pellet after a conventional recycling process, showing that odours are still present at levels of perception in the output material, and therefore, indicating that odours are not mitigated with the current recycling technologies [38].

The techniques used for identification of odorants include gas chromatography coupled to an olfactometer (GC-O) for both identification and sensory characterization of the volatile organic substances causing malodours. Additionally, there exist sensory analyses that do not involve any equipment and in which the only requisite is to have a trained panel of human evaluators comprised of a minimum of eight individuals. In this case, the analysis is based on odour description and hedonic and intensity rating. For the odour description, the panellist has to agree with a list of odour attributes by sniffing the headspace of the sample to be analysed, and then each attribute has to be scored. In the case of hedonic rating, each panellist has to decide whether they like it (maximum score) or dislike it (minimum score); and, last but not least, the odour intensity of the sample is rated from 1 (low intensity) to 10 (high intensity) [39].

Deodorization of recycled plastic would undoubtedly open new market opportunities in the packaging, building, and construction sectors, contributing, thus, to the introduction of plastics in the circular economy. The technologies developed until now are at a research or pilot scale rather than at an industrial scale, which are mainly based on supercritical CO_2 extraction and oxidation of volatile organic compounds (VOCs) attached onto the polymer surface [37].

5 Conclusion

Plastic production will continue growing worldwide because of the excellent properties, ease of production, and low costs. It also contributes to diminishing some negative environmental impacts. For instance, plastic packaging helps to reduce the amount of food waste produced globally. The negative side is that millions of tons of non-degradable waste are generated every year. However, plastic waste should be perceived as a resource rather than a problem.

In general, developed countries are more committed to sustainable waste management. A number of strategies, which include collection, sorting, and waste treatment operations, have been developed. Collection systems vary from one country to another, and there is no consensus on which one offers the biggest environmental benefits. Separate collection of potentially recyclable materials increases the quality of the waste steam. Unfortunately, operating costs also increase, decreasing the profitability of the entire system. Sorting processes are necessary to avoid contamination and improve the recycling rates. Waste treatment methods can be divided into mechanical recycling, chemical recycling, and energy recovery. Landfilling is considered the least preferred option. In fact, several European countries have banned plastic waste going to landfills.

Regarding developing countries, the informal recycling sector significantly contributes to the recovery of valuable materials. Waste pickers collect recyclable materials from the streets, houses, and landfills and sell them to recyclers. Generally, they work in atrocious conditions and do not get the credit that they deserve. Latin America is making a lot of progress in waste management. Plastic recycling has become a business opportunity and existing recycling companies compete with their European and American counterparts. In Asia and Africa, waste management strategies only exist in big cities and metropolitan areas. The costs of sorting and recycling processes are usually too high; therefore, collected waste is merely landfilled.

Although much progress has been made, recycling rates are far from achieving the goal of a circular economy. Materials should flow in a closed-loop cycle. That means that the waste generated by one system should be used as feedstock in another system. Currently, the value of recycled materials decreases due to contamination and degradation. As a result, recovered products are used in less demanding applications (downcycling). The European Union is trying to force the plastic manufacturers to introduce a minimum recycled material content in their products. But, the quality of recyclates must be significantly improved to achieve that. The NIAS and odours pose a serious problem, since current technologies are not prepared to meet the required degree of decontamination.

References

1. Pilz H, Brandt B, Fehringer R (2010) The impact of plastics on life cycle energy consumption and greenhouse gas emissions in Europe: executive summary. Denkstatt, Vienna
2. PlasticsEurope (2018) Plastics – the Facts 2017. An analysis of European plastics production, demand and waste data. Association of Plastics Manufacturers, European Association of Plastics Recycling, Brussels
3. PlasticsInsight (2016) Global consumption of plastic materials by region (1980–2015). Company annual report
4. Selke SEM, Culter JD (2016) 4 – Major plastics in packaging. In: SEM S, Culter JD (eds) Plastics packaging3rd edn. Hanser, Munich, pp 101–157
5. Singh N, Hui D, Singh R, Ahuja IPS, Feo L, Fraternali F (2017) Recycling of plastic solid waste: a state of art review and future applications. Compos Part B Eng 115:409–422. https://doi.org/10.1016/j.compositesb.2016.09.013
6. EPA (2018) Advancing sustainable materials management: 2015 fact sheet. United States Environmental Protection Agency, Washington, DC
7. Dhokhikah Y, Trihadiningrum Y (2012) Solid waste management in Asian developing countries: challenges and opportunities. J Appl Environ Biol Sci 2:329–335
8. Kumar S, Smith SR, Fowler G, Velis C, Kumar SJ, Arya S, Rena N, Kumar R, Cheeseman C (2017) Challenges and opportunities associated with waste management in India. R Soc Open Sci 4(3):160764
9. Lebreton L, Andrady A (2019) Future scenarios of global plastic waste generation and disposal. Palgrave Commun 5:6
10. Council Directive (2008) Council Directive 2008/98/EC of 19 November 2008 on waste and repealing certain directives. OJ L 312, 22/11/2008 P 0003–0030; 2008
11. Plastics Europe (2018) Plastics – the Facts 2018. An analysis of European plastics production, demand and waste data. Association of Plastics Manufacturers, Brussels
12. Dreher P, Faulstich M, Weber-Blaschke G, Berninger B, Keilhammer U (2004) VI.5 – Recycling of plastic waste, rubber waste and end-of-life cars in Germany. Waste Manag 4:815–863. https://doi.org/10.1016/S0713-2743(04)80035-7
13. Zafar S (2018) Plastic waste collection methods. https://www.cleantechloops.com/plastic-waste-collection-methods/
14. Seyring N, Dollhofer M, Weißenbacher J, Herczeg M, McKinnon D, Bakas I (2015) Final Report. Assessment of separate collection schemes in the 28 capitals of the EU
15. Zafar S (2016) Methods for plastic wastes collection. https://www.ecomena.org/plastic-waste-collection/
16. Reality Check Team (2018) Plastic recycling: how do bottle deposit schemes work? https://www.bbc.com/news/science-environment-43571269

17. Morawski C (2018) In my opinion: a business case for deposit return. https://resource-recycling.com/plastics/2018/05/23/in-my-opinion-a-business-case-for-deposit-return/
18. Ibrahim-Shehu S (2017) Separation of plastic waste from mixed waste: existing and emerging sorting technologies performance and possibilities of increased recycling rate with Finland as case study. Dissertation, Lappeenranta University of Technology
19. La Rosa A, Blanco I, Banatao D, Pastine S, Björklund A, Cicala G (2018) Innovative chemical process for recycling thermosets cured with Recyclamines® by converting bio-epoxy composites in reusable thermoplastic – an LCA study. Materials 11:353
20. Welle F (2011) Twenty years of PET bottle to bottle recycling – an overview. Resour Conserv Recycl 55:865–875. https://doi.org/10.1016/j.resconrec.2011.04.009
21. Horodytska O, Valdés FJ, Fullana A (2018) Plastic flexible films waste management – a state of art review. Waste Manag 77:413–425. https://doi.org/10.1016/j.wasman.2018.04.023
22. Fullana A, Lozano A (2015) Method for removing ink printed on plastic films, EP20130770017
23. Kaiser K, Schmid M, Schlummer M (2018) Recycling of polymer-based multilayer packaging: a review. Recycling 3:1
24. Wilson DC, Rodic L, Modak P, Soos R, Carpintero A, Velis K, Iyer M, Simonett O (2015) Global waste management outlook. UNEP, Nairobi
25. Fernández L (2013) Integrating the informal waste recycling sector in Latin America. The 2013 Global Dialogue on Waste
26. Goulart Coelho LM, Lange LC (2018) Applying life cycle assessment to support environmentally sustainable waste management strategies in Brazil. Resour Conserv Recycl 128:438–450. https://doi.org/10.1016/j.resconrec.2016.09.026
27. Ibáñez-Forés V, Coutinho-Nóbrega C, Bovea MD, de Mello-Silva C, Lessa-Feitosa-Virgolino J (2018) Influence of implementing selective collection on municipal waste management systems in developing countries: a Brazilian case study. Resour Conserv Recycl 134:100–111. https://doi.org/10.1016/j.resconrec.2017.12.027
28. Jambeck JR, Geyer R, Wilcox C, Siegler TR, Perryman M, Andrady A, Narayan R, Law KL (2015) Plastic waste inputs from land into the ocean. Science 347(6223):768–771
29. Ocean Conservancy (2015) Stemming the tide: land-based strategies for a plastic-free ocean. Ocean Conservancy and McKinsey Center for Business and Environment, London
30. Larracas A. GAIA zero waste from dream to reality in the Philippines. http://www.no-burn.org/zero-waste-from-dream-to-reality-in-the-philippines/
31. Damanhuri E (2017) Country chapter. State of the 3Rs in Asia and the Pacific. United Nations center for Regional Development, Nagoya
32. Nurhasana R (2014) Study on economic and environmental benefits of waste bank initiatives in DKI Jakarta province. Jurnal Dampak 11:127–137
33. Ratna Putri A, Fujimori T, Takaoka M (2018) Plastic waste management in Jakarta, Indonesia: evaluation of material flow and recycling scheme. J Mater Cycles Waste Manag 20:2140–2149
34. Alemma-Ozioruva A (2017) Challenges of managing waste disposal in Nigeria. https://guardian.ng/saturday-magazine/challenges-of-managing-waste-disposal-in-nigeria/
35. Ellen MacArthur Foundation (2016) The new plastics economy. Rethinking the future of plastics. World Economic Forum
36. Nerin C, Alfaro P, Aznar M, Domeño C (2013) The challenge of identifying non-intentionally added substances from food packaging materials: a review. Anal Chim Acta 775:14–24. https://doi.org/10.1016/j.aca.2013.02.028
37. SUSCHEM (2018) Plastics strategic research and innovation agenda in a circular economy. SUSCHEM, Brussels
38. Strangl M, Schlummer M, Maeurer A, Buettner A (2018) Comparison of the odorant composition of post-consumer high-density polyethylene waste with corresponding recycled and virgin pellets by combined instrumental and sensory analysis. J Clean Prod 181:599–607. https://doi.org/10.1016/j.jclepro.2018.01.137
39. Strangl M, Fell T, Schlummer M, Maeurer A, Buettner A (2017) Characterization of odorous contaminants in post-consumer plastic packaging waste using multidimensional gas chromatographic separation coupled with olfactometric resolution. J Sep Sci 40(7):1500–1507

Plastic Pollution in Slovenia: From Plastic Waste Management to Research on Microplastics

Gabriela Kalčíková and Andreja Žgajnar Gotvajn

Contents

1 Introduction 308
  1.1 A Brief History of (Plastic) Waste 308
  1.2 From Macroplastics to Microplastics 309
2 Plastics and Slovenia 310
  2.1 Waste and Plastic Waste Generation 311
  2.2 Plastic Waste Management 313
3 Plastics and Microplastics Research in Slovenia 315
4 Conclusions 318
References 319

Abstract Despite the increasing environmental awareness, there is still an enormous amount of plastic waste generated which often ends up in the environment. Slovenia is not an exception, and in the past years, there was a lot of effort to minimize plastic pollution in terrestrial and aquatic environments. Since waste management is closely connected to plastic pollution, the first part of the chapter summarizes waste management practice and plastic waste handling in Slovenia. According to European statistical data, Slovenia belongs among countries, where a good waste management practice is well established; waste collection, recycling rate, and waste management are comparable to other developed countries. The book chapter also focuses on the plastic waste in Slovenia and how plastic pollution is treated from a governmental perspective as well as from the perspective of nonprofit organizations. The last part of the chapter is aimed to present research on plastic pollution and microplastics in Slovenia; to introduce current research efforts and trends in Slovenia; to discuss monitoring results and the impact of microplastics on the local environment; and to link these results to the global microplastic research.

G. Kalčíková (✉) and A. Žgajnar Gotvajn
Faculty of Chemistry and Chemical Technology, University of Ljubljana, Ljubljana, Slovenia
e-mail: gabriela.kalcikova@fkkt.uni-lj.si; andreja.zgajnar@fkkt.uni-lj.si

Friederike Stock, Georg Reifferscheid, Nicole Brennholt, and Evgeniia Kostianaia (eds.),
Plastics in the Aquatic Environment - Part I: Current Status and Challenges,
Hdb Env Chem (2022) 111: 307–322, DOI 10.1007/698_2019_402,
© Springer Nature Switzerland AG 2019, Published online: 13 September 2019

Keywords Microplastics, Plastics, Pollution, Waste management

1 Introduction

1.1 A Brief History of (Plastic) Waste

In early history, when humans roamed the Earth, there was no such thing as waste, because nature was able to recycle everything that was left on the ground or in water. Food was immediately consumed, and waste quickly disappeared by the action of natural processes of scavenging and microbial decomposition [1]. Containers for food packaging were not needed; later, some natural materials as clay or reed fibers were used to make pottery that were reused and repaired until they were broken. Already in 1500 BC, Egyptians produced glass from silica and sodium carbonate to prepare glasses, bowls, and containers for storage. In the first and second century BC in China, mulberry bark was used for the packaging of food [2]. However, due to the low human population, there was no need for a special waste management practice and problems associated with waste, i.e., diseases, air pollution, and groundwater contamination were negligible for a long time [3].

With the development of civilization and increased population density, problems connected to waste became more important, and, as a result, some rules and practices emerged to encourage some early programs of waste management. The most common way how to manage waste during the ancient time was to place it away from populated areas into large pits or rivers, while some civilizations (as, e.g., Ancient Rome) had organized waste collection workforces already in the first century AD. It would be expected that the development of waste management will go further, but with the beginning of the Middle Ages, the common practice was to discard waste directly out of the window. Increased population in medieval cities, waste crisis, and finally the plague pandemic in the fourteenth century AD helped to reinvent rudimentary rules of waste management [1, 3].

Another important milestone regarding waste management progress was the industrial revolution. It was characterized by the development of new processes, materials, and products, but it also led to the generation of an enormous amount of waste. New types of industrial wastes could not be handled by traditional ways, and thus the real breakthrough in waste management just started [3]. Engineers were challenged to develop technologies that would alleviate and solve these problems. Among other measures, this led to the construction of the first waste incinerators in 1876 [4] and construction of a sanitary landfill in 1912 [5]. During this time, there were also major changes for many ordinary consumers. In the nineteenth century, metal containers became the primary material for food storage, and their usage expanded in 1959 when they started to be used for beer and soda. At the end of the nineteenth century, the typical shopping bags were made of jute, because paper as a packaging material was still scarce and paper bags were not very strong. Several further inventions helped to make paper bags stronger and inexpensive, and soon they became an essential shopping item in every grocery store [2].

However, the real revolution in packaging, and consequently in waste generation, started at the beginning of the twentieth century with the invention of plastics. The first plastic material made was a synthetic resin called Bakelite, and the commercial success of the invention encouraged the chemical industry to start research, development, and production of many new plastic materials. Further, and mainly due to the worries about a shortage of natural resources during World War II, there was a great interest in the development of artificial materials. Although the majority of plastic materials were originally used for military purposes after the war, new commercial products burst onto the market [2, 6]. Polystyrene, a by-product of military research that was supposed to bring a new rubber material from styrene monomers, became the key material of hot cups, plates, fast food containers, and other common packaging products. Similarly, polyethylene was discovered during military research. After the war, it replaced many metal cans and paper products and became the most important material for packaging ever made [6]. Polyethylene plastic bags were patented in 1965 [7], and together with PET bottles that were introduced in 1977, they quickly became superior to other consumer packaging products [8].

In overall, since the 1950s, plastics became an absolutely indispensable part of the consumer society; they permeated to everyday life, and thereafter almost every product could be replaced by a plastic substitute [6]. The extraordinary global expansion of plastic can be seen from the dramatic increase of plastic production from less than 2 million tons manufactured in 1950 to more than 335 million tons made annually today, with Europe among the leading plastic producers (19%) [9]. As of 2015, there were 5 billion tons of plastics produced, which is enough to wrap the Earth in a layer of plastic foil [10].

1.2 From Macroplastics to Microplastics

Today's society became a plastic consumption society – low production costs, as well as the favorable properties of plastics, make them suitable for an enormously broad range of applications and easily available for consumers [11]. More products are available for purchase, and they are wrapped in packaging which is of little value to consumers. It means that the majority of packaging materials are used once and then thrown away [2]. Since plastic packaging is the major sector of the plastic industry – e.g., in 2016, 39.9% of produced plastics in the EU were used for packaging – thus, the generation of massive amounts of plastic waste is inevitable. As a consequence, plastics contaminate aquatic habitats worldwide – it occurs in coastal areas and open oceans from the poles to the equator including even the most remote habitats; they are accumulated in sediments, dispersed in the water body, or floating on the water surface [11] – making plastic the most significant part of marine litter [12]. Although plastics are considered as environmentally nondegradable materials, they are subject to environmental aging – a combination of photo- and thermal-oxidative degradation by ultraviolet (UV) radiation, mechanical weathering, and biodegradation, which results in plastic fragmentation [13]. Such fragments are called microplastics.

Although the first scientific articles that identified microplastics in the environment as an issue were published in 1972 [14], efforts of the scientific community on this subject only started in 2004, with the paper "Lost at Sea: Where Is All the Plastic?" in the *Science* journal [15]. Since then, microplastics have become a truly global issue, because they can be found practically everywhere around the world and have thus become contaminants of emerging concern [16].

In the last years, microplastics have received great attention not only in the research community but also in society. High-profile media attention has expanded the issue of microplastics to the public and, therefore, driven by concerns with respect to both ecological harm and human health, triggered calls for policy action. However, human decisions and behaviors are the reason why microplastics occur in the environment and the economy drives the production of (micro)plastics [17]. If microplastic pollution is to be reduced, the society needs to understand the overall microplastic issue and to link the production of microplastics to their behavior and decisions. Efforts should emphasize the controlling sources and inputs of plastics into the environment by better educating and by improved waste management [18].

2 Plastics and Slovenia

In line with the introduction, the aim of this book chapter is to introduce an overview of plastic waste and management in Slovenia, the role of the government and nongovernmental organizations in the plastic issue, and, in the end, research efforts regarding plastics and microplastics that have been going on in Slovenia in recent years.

Slovenia is a country located in the south of Central Europe; it lies in the eastern Alps and borders the Adriatic Sea at its northern end. Although Slovenia was part of socialistic Yugoslavia for most of the twentieth century, it managed to sidestep most of the problems associated with the breakup of Yugoslavia. Since gaining independence, Slovenia has striven quickly to attain a level of development that enabled it to become a member of the European Union. Despite the common history of Slovenia and Western Balkans, Slovenia integrated economically and politically with Western Europe, and thus it may be considered a country bridging these two "worlds" – not only geographically but also socioeconomically. Therefore, Slovenia is an exceptional example of a post-socialistic country that, in the last decade, has developed into a country with strong priorities in nature conservation and environmental protection.

2.1 Waste and Plastic Waste Generation

In the past decades, changes in lifestyle, economic and commercial growth, technological development, and other factors led to increases in waste generation in many countries [19]. Waste is regarded as an inevitable, valueless by-product of human activities [20]. Waste has many negative environmental consequences, and thus the prevention of waste generation got into forefronts of environmental strategies around the world [21].

Plastics are an important part of many waste types: they can be found in industrial, municipal, and household wastes as well as in packaging waste. Therefore, it is difficult to evaluate a total amount of generated plastic waste by a country or to compare such data among countries due to differences in data collection and management (landfilling, incineration without energy recovery, incineration with energy recovery and recovery other than energy recovery), statistical analysis, missing data, and also the definition of various types of waste. For example, statistical data in the EU define the amount of generated plastic waste by the population as the amount of plastic waste collected by recycling centers of each municipality. It means that it is the plastic waste that inhabitants brought to the recycling center and not all the plastics collected by municipalities. In Slovenia, it is 2 kg per capita, while in the EU (28) (data for 28 member states) it was 5 kg per capita in 2014 [22]. But the majority of plastic waste is found in municipal and packaging waste.

Most waste generated by the population is collected as municipal waste. The main compositional categories of municipal solid waste are paper and cardboard, organic waste, plastics, metals, glass, textiles, and other minor fractions of waste [23]. In Slovenia, the most abundant fraction of mixed municipal waste is plastics (24%), followed by organic waste (18%) and paper (14%) [24]. However, the municipal waste composition varies a lot among countries and also depends on local conditions and on the waste collection and management system. It is strongly affected by socioeconomic factors, level of industrialization, geographic location, climate, level of consumption, collection system, population density, the extent of recycling, legislative controls, and public attitudes as well as by seasons, for example, in the amount of organic yard waste [1, 25].

The amount of generated municipal waste is also very variable. Although the move toward the reduction of waste in Europe was introduced by the Waste Framework Directive already in 1975 emphasizing the importance of waste prevention and minimization [26], there is still no common trend of municipal waste reduction. For example, over the last 22 years, in Switzerland and Slovakia, the amount of municipal waste per capita increased by 18% and 28%, respectively (Fig. 1). In the EU (27) the overall generation of municipal waste is relatively stable, similar to many member states (e.g., Poland and Germany). In Slovenia, the amount of municipal waste per capita was reduced by 21% [22] (Fig. 1).

Figure 1 also shows a significant difference in the amount of waste generated by each country, and it seems that some countries within the EU generate a significantly lower amount of municipal waste than others (e.g., Germany and Poland, Fig. 1).

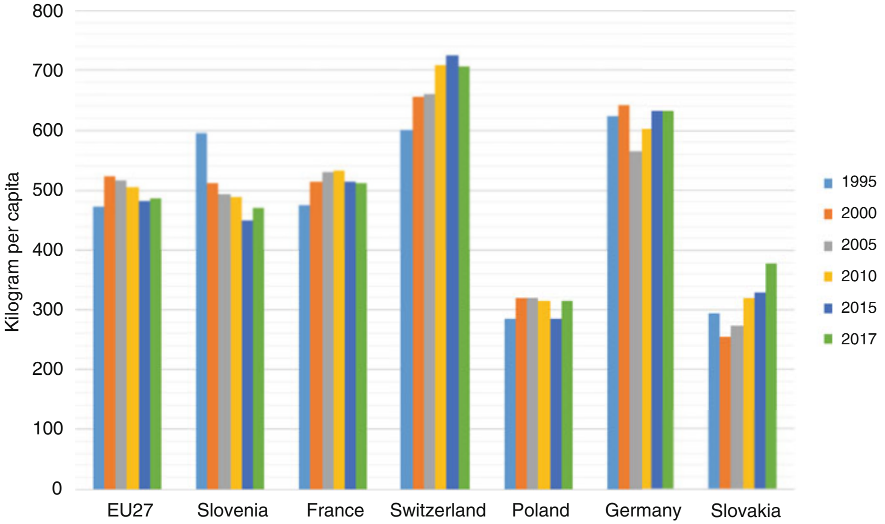


Fig. 1 The development of municipal waste generation (kg per capita) in different countries of the European Union from 1995 until 2017 [22]

However, countries define municipal waste differently; some countries also include bulky or garden waste. Then, it appears that these countries generate more municipal waste per capita than a country that excludes these waste fractions. Some countries also include only waste from households, whereas other countries also include waste from commercial activities [27].

Plastic packaging comprises about two-thirds of all the plastics put on the market [9], and most of the packaging is disposable. Thus, it can be assumed that plastic packaging waste is a relevant indicator of plastic waste generation by the population. In 2016, 170 kg of packaging waste was generated per capita in the EU, varying from 55 kg per capita in Croatia and 221 kg per capita in Germany and Slovenia with about 108 kg per capita [22]. Generation of packaging waste can also be linked to an economic situation of a country. The more the population grows, the more goods are consumed, and packaging becomes an important part of the waste. For example, when the gross domestic product (GDP) – a quantitative indication of the mean living standard of a nation – is compared to packaging waste generation in Slovenia from 2007 to 2016 (Fig. 2), there is a significant correlation. In this case, Slovenia was dragged into a deep recession by the European financial crisis from 2008, and it was quickly reflected in the package waste generation. After 2014, the financial situation stabilized, and GDP and amount of packaging waste again steeply increased (Fig. 2).

In Slovenia, the amount of plastic packaging waste generated per capita is stable with 22.66 kg per capita in 2007 and 22.45 kg per capita in 2016. Packaging plastics represent about 21% of the total packaging waste [22].

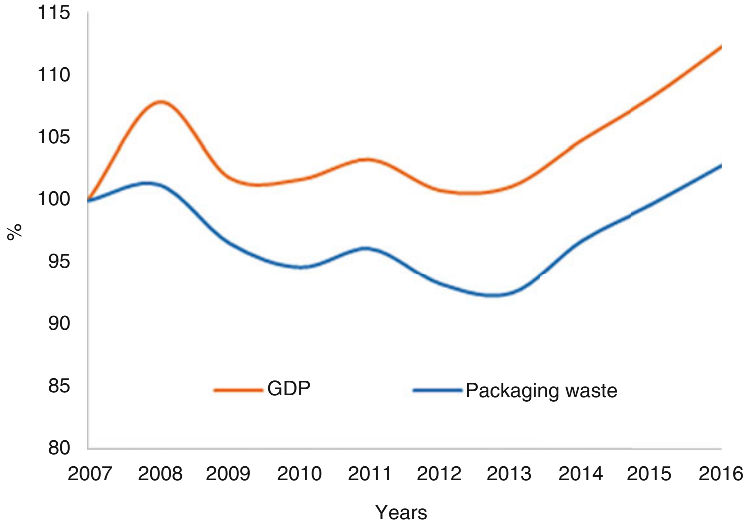


Fig. 2 Comparison of packaging waste generation in Slovenia and gross domestic product (GDP) from 2007 to 2016 (calculated from 100% in 2007 selected as a starting year) [28]

2.2 Plastic Waste Management

Solid waste management is an important tool to reduce environmental pollution caused due to the generation of plastic waste. It includes many actions such as planning, organization, administration, and financial and legal implications of various activities regarding generation, on-site storage, collection, transfer, transportation, processing, and recovery, as well as the ultimate disposal of solid waste [3, 29]. The first effort to define the priority of the EU waste management began in 1975 with the first Waste Framework Directive (1975/442/EEC, [26]), but the modern concept of waste hierarchy was introduced in 2008. The EU parliament implemented a new five-step waste hierarchy to the legislation (Directive 2008/98/EC, [30]): the priority of waste management is the waste prevention, reuse, recycling, and another recovery (e.g., energy recovery), and the last option is disposal.

Over the last decade, Slovenia has successfully implemented the EU waste legislation that helped in diverting waste from landfills and has established a recycling system throughout the country, achieving a high recycling rate. Slovenia has a high number of waste management facilities: 386 for recycling, 10 for energy recovery, and 14 landfills for municipal waste disposal; the number of waste treatment facilities increased by 3% from 2014 to 2016 [31]. In 2010, only 22.4% of municipal waste was recycled in Slovenia, while in 2017 it was 57.7%. On the other hand, landfilling of municipal waste significantly decreased over the last decade, from 72.0% in 2010 to 16.4% in 2017 [22, 31]. 69.4% of all packaging waste and 62% of plastic packaging waste were recycled in Slovenia in 2016; it is one of the highest recycling rates in the EU (Fig. 3). The transition from landfilling to recycling was strongly supported by the modernization of waste management facilities in Slovenia. In 2015, the most modern and one of the largest waste management centers in Europe was opened in Ljubljana. This Regional Waste Management Center (RCERO) collects waste from 50 municipalities, which represents one-third of the Slovenian population. It is aimed to treat mixed municipal waste that does not contain paper and cardboard, plastics, and organic waste, because they are collected and treated separately. The treatment results in 95% of mixed municipal waste being utilized and only 5% of municipal waste being disposed in a landfill [32].

However, the positive trend in the implementation of the hierarchy of waste management in Slovenia can be linked also to other aspects. For example, social aspects play an important role in successful waste management [33]. These social aspects include communication and acceptance of waste treatment methods (e.g., a large campaign about the new waste management facility – the example of RCERO), public participation in planning and implementation (e.g., sorting of waste, separation of plastics), and consumer behavior (e.g., their own decision not using single-use plastics). In Slovenia, sorting of waste and separation of plastics are strongly supported by legislation. For example, in the capital city of Ljubljana, individuals who do not separate waste properly face a fine of 200–800 euros [34].

Another very important factor is early education. The Slovenian government has been supporting many projects regarding waste management education since the modern history of Slovenia. For example, in 1996, a program was launched, which was called "Eco-Schools," designed to implement sustainable development education in schools by encouraging children and youth to take an active role in how their school can be run for the benefit of the environment [35]. The Ministry of the

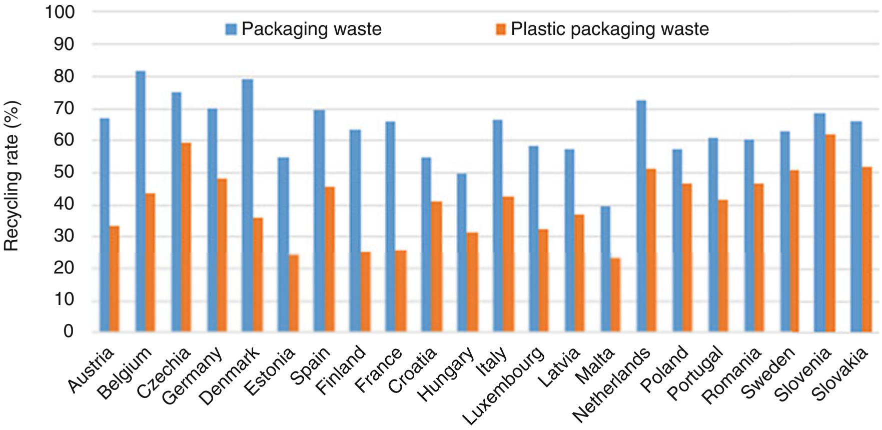


Fig. 3 Comparison of the recycling rate of packaging waste and plastic packaging waste in selected European countries in 2016 [22]

Environment and Spatial Planning ran a large campaign on the impact of lightweight plastic carrier bags on the environment in 2018. The aim of this campaign was to increase knowledge about single-use plastics and generation of microplastics in the environment as well as to reduce consumption of lightweight plastic carrier bags [36].

There are also many nonprofit and nongovernmental organizations spreading the awareness of waste and waste plastic generation. The largest action against waste in Slovenia was organized by the environmental organization “Ecologists Without Borders.” They have organized several events; one of them is “Let’s Clean Slovenia in One Day!” targeting illegal dumps and collecting waste around the country. During the last event in 2018, about 28,000 volunteers removed 77 tons of waste from the environment [37]. Before the event, various promotional and organizational activities were offered and aimed to educate about waste, plastics, and appropriate waste handling [38]. The same organization supports Slovenian municipalities to join the initiative “Zero Waste Slovenija.” There are nine municipalities in the network (17% of the population) that have adopted the Zero Waste strategy and limited the generation of mixed municipal waste. Together, they prevented of at least 15,750 tons of mixed municipal waste, thus saving about three million euros avoiding waste management costs [39].

The successful story of plastic waste management and handling in Slovenia is related to a combination of many factors including advanced technological development, adaptation of new legislations, and participation of people as well as constantly increasing awareness regarding environmental issues. All these aspects are crucial to ensure the success of proper waste management systems. However, there is still a lot of work to do to prevent plastics from entering the environment. Most of these efforts are conducted by scientists, and therefore the last part of the book chapter is aimed to introduce their work on the plastic and microplastic issues.

3 Plastics and Microplastics Research in Slovenia

In the last years, there has been a strong interest in the development of strategies to prevent plastics from entering the environment and to understand the ecological consequences of their presence in various ecosystems. The interest in plastics research basically follows the line of the waste hierarchy: (1) to search for new materials to prevent generation of plastic waste (*prevention*), (2) to develop new methods for plastic waste reuse and recycling (*recycling*, *reuse*), and (3) to describe impacts of plastics that already entered the environment (*disposal*).

Most important is the prevention of plastic waste generation. Thus researchers have been looking for other materials that could become an appropriate replacement of petroleum-based plastics. They have designed many plastic materials that could have a potentially lower impact on the environment. One of such examples is the so-called oxo-plastics or oxo-degradable plastics, which are conventional plastics that contain additives which promote oxidation of the material under certain conditions. It has been designed and implemented to increase the rate of natural removal of plastic waste disposed in the environment. They were claimed to degrade rapidly into harmless products under common environmental conditions. However, they will break down into smaller parts, potentially contributing to environmental contamination by microplastics [40]. Therefore, many research efforts rather focused on biodegradable plastics produced from natural materials [41]. Biodegradable plastics are degraded under natural conditions, and they represent a source of carbon and energy for microorganisms [42]. They can be prepared from several sources, including animal and plant materials. Currently, starch-based bioplastics are the most commonly manufactured on an industrial scale [43]. Similarly, in Slovenia, research on biodegradable polymers was included in schemes of many research groups. Pepic et al. [44] worked for example on synthesis and characterization of biodegradable aliphatic copolyesters with poly(ethylene oxide) soft segments being one of the most promising biodegradable materials due to their susceptibility to biological attacks and their degradation products being soluble, biodegradable, and nontoxic. A further step toward the commercial application of biodegradable polymers proceeds in the framework of the EU project BioApp (Interreg Europe), where Slovenia is one of the project partners. The project focused on the utilization of waste biomass; shells and exoskeletons from shellfish production are used for the production of biopolymers that are a base for the development of a product for commercialization (e.g., biopolymer packaging materials) [45].

In order to tackle the problem of already generated plastic waste, researchers are looking for alternative innovative applications for reuse or recycling of waste plastics [46]. In Slovenia, this part of the research is covered by various projects, e.g., MOVECO – Mobilizing Institutional Learning for Better Exploitation of Research and Innovation for the Circular Economy in the framework of the Interreg Danube Transnational Program. The aim of the program is to minimize waste generation and to keep products and resources in the economy as long as possible so that both the economy and the environment can benefit from it [47]. Another project led by a Slovenian partner (TECOS, the Slovenian Tool and Die Development Centre) is a new circular economy through the valorization of postconsumer plastic waste and reclaimed pulp fiber (CEPLAFIB). The project is heading toward the development of new materials out of plastic packaging waste and waste newspaper that could be used for high-tech products in the automotive, construction, and packaging industries [48].

The last aspects of plastic research in Slovenia are monitoring and impact studies that are carried out in several Slovenian institutions. One of the first and largest projects focusing on monitoring of (micro)plastics was the DeFishGear Project (IPA Adriatic Cross-border Cooperation Programme) in 2013. The aim of the project was to improve knowledge on the occurrence, amounts, sources, and impacts of marine litter in the Adriatic Sea and to address the emerging threat of microplastics [49]. Results of visual observation of floating macroplastics (items > 2.5 cm) showed that the average number of floating macroplastics in Adriatic waters is 251 ± 601 per km^2. The majority of macroplastics were plastic bags (29%) followed by plastic pieces (22%) and sheets (15%) [50]. The results are one or two orders of magnitude higher than most of previously reported in the Mediterranean and the Adriatic Sea, e.g., 15 items/km^2 [51], but the difference can be related to variations in the methodology for macroplastic observation, e.g., oceanographic vessel speed or observation height. Microplastics (particles > 330 μm) were also very abundant at the sea surface of the Adriatic Sea. The average number was 315,009 ± 568,578 per km^2, and the majority of microplastics were made of polyethylene (67%) and polypropylene (18%). Such results are in accordance with many authors that use a similar sampling technology, i.e., manta net (330 or 333 μm) towing (Table 1). The abundance of microplastics related to the Slovenian part of the Adriatic Sea can be derived from the data of Gajšt et al. [59]. The authors sampled microplastics (manta net, with mesh size 300 μm) at the sea surface of the Gulf of Trieste close to the Slovenian cities Piran, Portorož, and Koper. The average number of microplastic particles was 472,000 ± 210,000 per km^2. Viršek et al. [60] also sampled microplastics (manta net, with mesh size 308 μm) around the Slovenian coast. The results of the first sampling (August 2014) showed 259,310 ± 57,096 microplastic particles per km^2, while during the second sampling (May 2015), 1,304,811 ± 609,426 microplastic particles per km^2 were found. Such results reveal one of the highest microplastic abundances among similar studies conducted around the world (Table 1). The authors determined also bacterial community drifting on the microplastic surface and among many bacterial species found invasive fish pathogen *Aeromonas salmonicida* [60].

Slovenian beaches are also affected by microplastics. Laglbauer et al. [61] found 178 microplastic particles per kg of sediment at Slovenia beaches (>250 μm). However, it is difficult to compare these results with other studies due to a significant variation in sampling methods (Table 1). The authors suggested that most of the microplastics originated from outflows and untreated wastewaters that flow through the river into the sea [61]. It is in agreement with Kalčikova et al. [62] who evaluated

Table 1 Occurrence of microplastics in the marine environment – water surface and beach sediments

Location	Sample	Sampling size (μm)	Number of microplastic particles	Reference
Northwestern Mediterranean Basin	Water surface	>333	116,000 per km^2	[52]
Ligurian Sea	Water surface	>333	2,100–578,000 per km^2	[53]
South Pacific Gyre	Water surface	>333	26,898 per km^2	[54]
North Pacific Gyre	Water surface	>333	334,271 per km^2	[55]
Baltic beaches of Kaliningrad region	Beach sediment	>500	1.3–36.3 per kg	[56]
Belgium coast	Beach sediment	>38	92.8 per kg	[57]
Islands Kachelotplate and Spiekeroog	Beach sediment	>1.2	210 per kg (granular) 461 per kg (fibers)	[58]

a high amount of polyethylene microbeads entering waterways with treated wastewater from the largest Slovenian municipal wastewater treatment plant. According to their calculations, about 1 kg of polyethylene microbeads (about 112,500,000 particles) is daily released into the Ljubljanica River with treated wastewaters. Similarly, many other authors have considered wastewater treatment plant effluents as an important route for microplastics into the environment (e.g., [63]). However, the presence and abundance of microplastics in Slovenian freshwaters have not been systematically investigated yet.

Besides monitoring studies, Slovenian researchers also focus on the impact of microplastics on various freshwater and terrestrial organisms. Jemec et al. [64] investigated the ingestion and effects of polyethylene terephthalate microfibers on the water flea *Daphnia magna*. The results showed that water fleas are able to ingest very long fibers up to 1,400 μm and confirmed that fibers have a significant impact on the mortality of daphnids if they are not pre-fed with algae. Further, Kokalj et al. [65] extended their work to various types of microplastics that are commonly found in the environment (microplastics from two facial cleansers, a plastic bag, and textile fleece) and tested their uptake by water flea *Daphnia magna* and brine shrimp *Artemia franciscana*. The results showed that both organisms are able to ingest microplastics. Daphnids preferably ingested smaller particles. The most abundant particles in a gut were up to 100 μm, while brine shrimps did not show any preference toward microplastic sizes. The results of the study also showed that these microplastics do not represent an acute hazard for tested organisms, as over the duration of the study, no significant mortality was observed. Similarly, no effect of microplastics from a facial cleanser and a plastic bag was observed on feeding and energy reserves of terrestrial isopods [66]. Kalčikova et al. [67] exposed freshwater floating plants duckweed *Lemna minor* to microplastics from two facial cleansers and observed impact of microplastics on plants' root and fronds. The growth of fronds was not affected, while microplastics significantly reduced the growth of plants' roots and caused mechanical abrasion of roots. This study also showed that microplastics are highly adsorbed onto plant surfaces.

4 Conclusions

Slovenia is one of the EU member states where an environmentally sustainable structure has been successfully implemented and adopted. The significance of plastic pollution is well recognized and understood by the population and the politicians, and thus waste generated by the population has decreased. At the same time, the recycling rate has significantly increased in the last years. Although good waste management practice can be directly linked to the reduction of plastic items and consequently microplastics in the environment, there are many more possible routes to the environment. Despite the good waste management strategy, research results showed that Slovenia's coast is significantly polluted by microplastics. Their occurrence can be linked to several sources, but the most important ones seem to be wastewater treatment plant effluents acting as a point source of microplastic pollution. A significant amount of fibers, microbeads, and disintegrated consumer products originate from these effluents and consequently pollute freshwater and marine ecosystems. Therefore, more efforts should be undertaken to reduce plastic pollution not only by implementation of appropriate solid waste management but also by improving wastewater treatment in order to reduce the plastic stream from the society into the environment.

Acknowledgments The authors are grateful to the Slovenian Research Agency (Research program Chemical Engineering – P2-0191) for supporting their research and to Mr. Predrag Korica for help with statistical data about plastic waste and plastic waste management.

References

1. Letcher TM, Vallero DA (2011) Waste: a handbook for managers. Academic Press, Burlington
2. Vaughn J (2009) Waste management: a reference handbook. Abc-Clio, Santa Barbara
3. Pichtel J (2005) Waste management practices: municipal, hazardous, and industrial. Taylor & Francis, Boca Raton
4. Bilitewski B, Hardtle G, Marek K (1997) Waste management. Springer, New York
5. Pipkin BW (2014) Geology and the environment. Brooks/Cole, Cengage Learning/National Geographic Learning, Pacific Grove/Boston
6. Miller C (2013) The atlas of U.S. and Canadian environmental history. Routledge, New York
7. United Nation Environment (2018) From birth to ban: a history of the plastic shopping bag. https://www.unenvironment.org/news-and-stories/story/birth-ban-history-plastic-shopping-bag. Accessed 16 Feb 2019
8. Smith AF (2013) Food and drink in American history: a "full course" encyclopedia. Santa Barbara, Abc-Clio
9. Plasticseurope (2018) Plastics – the facts 2017 – an analysis of European plastics production, demand and waste data. Plasticseurope, Brussels
10. Zalasiewicz J, Waters CN, Ivar Do Sul JA, Corcoran PL, Barnosky AD, Cearreta A, Edgeworth M, Gałuszka A, Jeandel C, Leinfelder R, Mcneill JR, Steffen W, Summerhayes C, Wagreich M, Williams M, Wolfe AP, Yonan Y (2016) The geological cycle of plastics and their use as a stratigraphic indicator of the anthropocene. Anthropocene 13:4–17
11. Derraik JGB (2002) The pollution of the marine environment by plastic debris: a review. Mar Pollut Bull 44:842–852
12. Andrady AL (2011) Microplastics in the marine environment. Mar Pollut Bull 62:1596–1605
13. Driedger AGJ, Dürr HH, Mitchell K, van Cappellen P (2015) Plastic debris in the Laurentian Great Lakes: a review. J Great Lakes Res 41:9–19
14. Carpenter EJ, Smith Jr KL (1972) Plastics on the sargasso sea surface. Science 175:1240–1241
15. Thompson RC, Olsen Y, Mitchell RP, Davis A, Rowland SJ, John AWG, Mcgonigle D, Russell AE (2004) Lost at sea: where is all the plastic? Science 304:838
16. Avio CG, Gorbi S, Regoli F (2017) Plastics and microplastics in the oceans: from emerging pollutants to emerged threat. Mar Environ Res 128:2–11
17. SAPEA (2019) A scientific perspective on microplastics in nature and society. SAPEA, Berlin
18. Peng J, Wang J, Cai L (2017) Current understanding of microplastics in the environment: occurrence, fate, risks, and what we should do. Integr Environ Assess Manag 13:476–482
19. Renou S, Givaudan JG, Poulain S, Dirassouyan F, Moulin P (2008) Landfill leachate treatment: review and opportunity. J Hazard Mater 150:468–493

20. Pariatamby A (2014) Msw management in Malaysia-changes for sustainability. In: Pariatamby A, Tanaka M (eds) Municipal solid waste management in Asia and the Pacific Islands: challenges and strategic solutions. Springer, Singapore
21. Minelgaitė A, Liobikienė G (2019) Waste problem in European Union and its influence on waste management behaviours. Sci Total Environ 667:86–93
22. Eurostat (2019) Waste statistics. https://ec.europa.eu/eurostat/web/waste/overview. Accessed 11 Feb 2019
23. Malinauskaite J, Jouhara H, Czajczyńska D, Stanchev P, Katsou E, Rostkowski P, Thorne RJ, Colón J, Ponsá S, Al-Mansour F, Anguilano L, Krzyżyńska R, López IC, Vlasopoulos A, Spencer N (2017) Municipal solid waste management and waste-to-energy in the context of a circular economy and energy recycling in Europe. Energy 141:2013–2044
24. Mol (2016) The environmental report on waste. http://www.mop.gov.si/fileadmin/mop.gov.si/pageuploads/podrocja/cpvo/okoljsko_porocilo_op_odpadki_priloga1.pdf. Accessed 16 Feb 2019
25. Williams PT (2005) Waste treatment and disposal. Wiley, Chichester
26. Waste Framework Directive (1975) Council Directive 75/442/EEC of 15 July 1975 on waste
27. European Environmental Agency (2013) Managing municipal solid waste – a review of achievements in 32 European countries. https://www.eea.europa.eu/publications/managing-municipal-solid-waste. Accessed 28 Feb 2019
28. Republic of Slovenia Statistical Office (2019) Waste and GDP statistics. https://www.stat.si/statweb/en. Accessed 17 Feb 2019
29. Subramanian MN (2016) Plastics waste management: processing and disposal. Smithers Rapra Technology, Akron
30. Waste Framework Directive (2008) Directive 2008/98/EC of the European Parliament and of the Council of 19 November 2008 on waste and repealing certain directives
31. Republic of Slovenia Statistical Office (2016) Seventeen operating landfill sites in Slovenia in 2016. https://www.stat.si/statweb/en/news/index/7501. Accessed 27 Feb 2019
32. Rcero (2019) Regional waste management center. http://www.rcero-ljubljana.eu/. Accessed 21 Feb 2019
33. Kaplan Mintz K, Henn L, Park J, Kurman J (2019) What predicts household waste management behaviors? Culture and type of behavior as moderators. Resour Conserv Recycl 145:11–18
34. Official Gazette of Republic of Slovenia (2008) 72/2008
35. Ekošola (2019). https://ekosola.si/. Accessed 28 Feb 2019
36. Mop (2018) I have a chice: so i bring my own bag. http://www.mop.gov.si/si/delovna_podrocja/odpadki/akcije_ozavescanja_javnosti/. Accessed 16 Feb 2019
37. STA (2018) Let's clean Slovenia in one day! 2018: 28,000 people removed 77 tonnes of waste. https://www.sta.si/2555227/ocistimo-slovenijo-2018-28-000-ljudi-odstranilo-77-ton-odpadkov?q=o%c4%8dist,sloven. Accessed 28 Feb 2019
38. Ecologists without Borders (2018) Let's clean Slovenia in one day! http://www.ocistimo.si/o-projektu/ocistimo-slovenijo-2018. Accessed 15 Feb 2019
39. Ecologists without Borders (2018) Zero waste municipalities. https://ebm.si/zw/obcine/. Accessed 16 Feb 2019
40. European Commission (2018) Report from the commission to the European Parliament and the Council on the impact of the use of oxo-degradable plastic, including oxo-degradable plastic carrier bags, on the environment
41. Luyt AS, Malik SS (2019) Can biodegradable plastics solve plastic solid waste accumulation? In: Al-Salem SM (ed) Plastics to energy. William Andrew, Norwich
42. Chinaglia S, Tosin M, Degli-Innocenti F (2018) Biodegradation rate of biodegradable plastics at molecular level. Polym Degrad Stab 147:237–244
43. Harding KG, Gounden T, Pretorius S (2017) "Biodegradable" plastics: a myth of marketing? Procedia Manuf 7:106–110
44. Pepic D, Zagar E, Zigon M, Krzan A, Kunaver M, Djonlagic J (2008) Synthesis and characterization of biodegradable aliphatic copolyesters with poly(ethylene oxide) soft segments. Eur Polym J 44:904–917

45. Bioapp (2019) A trans-regional platform for the transfer of technological biopolymers from the research sector to the market. https://www.ita-slo.eu/en/bioapp#objectives. Accessed 27 Feb 2019
46. Mohammadinia A, Wong YC, Arulrajah A, Horpibulsuk S (2019) Strength evaluation of utilizing recycled plastic waste and recycled crushed glass in concrete footpaths. Constr Build Mater 197:489–496
47. Moveco (2019) Mobilising institutional learning for better exploitation of research and innovation for the circular economy. http://www.interreg-danube.eu/approved-projects/moveco. Accessed 28 Feb 2019
48. Ceplafib (2019) Implementation of a new circular economy through the valorisation of postconsumer plastic waste and reclaimed pulp. https://www.tecos.si/index.php/en/applied-research/running-projects/item/631-life-ceplafib-predelava-odpadne-plasticne-embalaze-in-casopisnega-papirja-v-nove-kompozitne-materiale-za-proizvodnjo-embalaznih-avtomobilskih-in-gradbenih-komponent. Accessed 1 Mar 2019
49. Defishgear (2019) Towards litter-free adriatic and ionian coasts and seas. http://www.defishgear.net/project/objectives. Accessed 23 Feb 2019
50. Zeri C, Adamopoulou A, Bojanić Varezić D, Fortibuoni T, Kovač Viršek M, Kržan A, Mandic M, Mazziotti C, Palatinus A, Peterlin M, Prvan M, Ronchi F, Siljic J, Tutman P, Vlachogianni T (2018) Floating plastics in adriatic waters (Mediterranean Sea): from the macro- to the micro-scale. Mar Pollut Bull 136:341–350
51. Di-Méglio N, Campana I (2017) Floating macro-litter along the Mediterranean French coast: composition, density, distribution and overlap with cetacean range. Mar Pollut Bull 118:155–166
52. Collignon A, Hecq J-H, Glagani F, Voisin P, Collard F, Goffart A (2012) Neustonic microplastic and zooplankton in the North Western Mediterranean Sea. Mar Pollut Bull 64:861–864
53. Pedrotti ML, Petit S, Elineau A, Bruzaud S, Crebassa J-C, Dumontet B, Martí E, Gorsky G, Cózar A (2016) Changes in the floating plastic pollution of the Mediterranean sea in relation to the distance to land. PLoS One 11:1–14
54. Eriksen M, Maximenko N, Thiel M, Cummins A, Lattin G, Wilson S, Hafner J, Zellers A, Rifman S (2013) Plastic pollution in the south pacific subtropical gyre. Mar Pollut Bull 68:71–76
55. Moore CJ, Moore SL, Leecaster MK, Weisberg SB (2001) A comparison of plastic and plankton in the North Pacific central gyre. Mar Pollut Bull 42:1297–1300
56. Esiukova E (2017) Plastic pollution on the Baltic beaches of Kaliningrad region, Russia. Mar Pollut Bull 114:1072–1080
57. Claessens M, Meester SD, Landuyt LV, Clerck KD, Janssen CR (2011) Occurrence and distribution of microplastics in marine sediments along the Belgian coast. Mar Pollut Bull 62:2199–2204
58. Liebezeit G, Dubaish F (2012) Microplastics in beaches of the east frisian islands spiekeroog and Kachelotplate. Bull Environ Contam Toxicol 89:213–217
59. Gajšt T, Bizjak T, Palatinus A, Liubartseva S, Kržan A (2016) Sea surface microplastics in Slovenian part of the Northern Adriatic. Mar Pollut Bull 113:392–399
60. Viršek MK, Lovšin MN, Koren Š, Kržan A, Peterlin M (2017) Microplastics as a vector for the transport of the bacterial fish pathogen species *Aeromonas salmonicida*. Mar Pollut Bull 125:301–309
61. Laglbauer BJL, Franco-Santos RM, Andreu-Cazenave M, Brunelli L, Papadatou M, Palatinus A, Grego M, Deprez T (2014) Macrodebris and microplastics from beaches in Slovenia. Mar Pollut Bull 89:356–366
62. Kalčíková G, Alič B, Skalar T, Bundschuh M, Gotvajn AŽ (2017) Wastewater treatment plant effluents as source of cosmetic polyethylene microbeads to freshwater. Chemosphere 188:25–31

63. Murphy F, Ewins C, Carbonnier F, Quinn B (2016) Wastewater treatment works (Wwtw) as a source of microplastics in the aquatic environment. Environ Sci Technol 50:5800–5808
64. Jemec A, Horvat P, Kunej U, Bele M, Kržan A (2016) Uptake and effects of microplastic textile fibers on freshwater crustacean *Daphnia magna*. Environ Pollut 219:201–209
65. Jemec Kokalj A, Kunej U, Skalar T (2018) Screening study of four environmentally relevant microplastic pollutants: uptake and effects on *Daphnia magna* and *Artemia franciscana*. Chemosphere 208:522–529
66. Jemec Kokalj A, Horvat P, Skalar T, Kržan A (2018) Plastic bag and facial cleanser derived microplastic do not affect feeding behaviour and energy reserves of terrestrial isopods. Sci Total Environ 615:761–766
67. Kalčíková G, Žgajnar Gotvajn A, Kladnik A, Jemec A (2017) Impact of polyethylene microbeads on the floating freshwater plant duckweed *Lemna minor*. Environ Pollut 230:1108–1115

Marine Litter Assessment on Some Beaches Along the Southeastern Adriatic Coastline (Albania)

Jerina Kolitari and Laura Gjyli

Contents

1 Introduction ... 324
2 Materials and Methods ... 325
  2.1 Study Area ... 325
  2.2 Methods ... 327
  2.3 Data Analysis and Processing ... 327
3 Results and Discussion ... 329
  3.1 Litter Densities of Surveyed Beaches and Beach Cleanliness ... 329
  3.2 Marine Litter Composition of Surveyed Beaches ... 332
  3.3 Marine Litter Sources of Surveyed Beaches ... 341
  3.4 Options to Manage the Litter of Beaches ... 345
4 Conclusions ... 347
References ... 348

Abstract Marine litter is a truly global challenge, changing all oceans and seas of the world. Every year, millions of tons of litter end up in the coastal and marine environment worldwide, resulting in environmental, economic, health, and safety impacts. This study investigated the abundance, composition, and sources of marine litter stranded on four beaches located at Durrës Bay and in the Gulf of Drin, which also includes Rodoni Bay and Shëngjini Bay along the Albanian southern-eastern

---

Jerina Kolitari and Laura Gjyli contributed equally to this work.

J. Kolitari (✉)
Laboratory of Fisheries and Aquaculture, Durrës, Department of Aquaculture and Fishery, Agricultural University of Tirana, Tirana, Albania
e-mail: j.kolitari@gmail.com

L. Gjyli
Department of Applied and Natural Sciences, Aleksandër Moisiu University of Durrës, Durrës, Albania
e-mail: lauragjyli@yahoo.com

Friederike Stock, Georg Reifferscheid, Nicole Brennholt, and Evgeniia Kostianaia (eds.),
Plastics in the Aquatic Environment - Part I: Current Status and Challenges,
Hdb Env Chem (2022) 111: 323–352, DOI 10.1007/698_2020_627,

© Springer Nature Switzerland AG 2020, Published online: 12 September 2020

Adriatic coastline. During the winter 2015, 12 beach transects were surveyed, covering 12,000 m^2 and extending over 1,000 m of the coastline. The mean litter density of the total four beaches studied was 219 items/100 m and 0.219 items/m^2. The majority of litter items (58%) were plastic or artificial polymer materials. Other bottles and containers (drums) were the most frequently found items with a percentage of 6%, followed by cartons/tetra pack (others) with 5.7% and by cigarette butts and filters with 3.7%. The sites investigated differed in terms of human-induced pressures with two sites classified as semi-urban: one site as urban and one as rural. Litter from shoreline sources such as tourism and recreational activities, including poor waste management practices, accounted for 37.5% of litter collected, accounting for the vast majority of litter items. Sea-based sources of litter (fisheries and aquaculture, shipping) amounted to 8% of total litter items on all beach locations.

Keywords Adriatic beaches, Albanian coastline, Marine litter, Plastic pollution, Single-use plastic

1 Introduction

The Mediterranean basin is one of the world's biodiversity hotspots and one of the most polluted and affected seas worldwide by marine litter. In the Mediterranean Sea, marine litter is a major threat for living marine organisms [1–3]. Research indicates that plastic pollution may impact biodiversity, ecosystem services, food security, and human health. Briefly, plastic pollution is a global threat [4]. Plastic marine litter observed in the Mediterranean Sea is widely distributed over various environmental compartments such as water, coastlines, bottom sediments, and biota and poses a considerable risk to ecosystems and human health [5].

The Adriatic Sea is an elongated basin, with its major axis in the NW-SE direction, between Italy and the Balkans, located in the central Mediterranean. The Adriatic Sea is characterized by one of the greatest floating plastic particles pollution among Mediterranean regions [6, 7]. The mean particles' half-life (i.e., the time after which 50% of the particles still remain on the sea surface) is estimated to be approximately 43.7 days [8]. Thus, the Adriatic Sea is a highly dissipative system with respect to floating plastics (in contrast to the global ocean, where the half-lifetime of particles equals 19 years). This result suggests that the main sink of floating plastics is partitioned between the shoreline and the seafloor, posing an additional risk to such ecosystems [7].

About 60% of the Albanian population lives in coastal areas. In 1991, closure of the industries helped diminish environmental pollution including in the coastal water. However, there is also an increase in urban pollution in the coastal area caused by the tourism development mainly in the Adriatic coast and by the increasing number of inhabitants in the main Albanian cities like Tirana, Durrës, and Vlora

[9]. Durrës County has a coastline of some 62 km along the Adriatic Sea, extending from Lalzi Bay till the area of Golem in Durrës Bay. Recent publications also include the quality of the coastal waters at Durrës Bay [10–14]. Plastic was the dominant substance of marine litter in Albanian Adriatic coastal waters in our previous studies [15, 16]. The shoreline of the Gulf of Drin has a length of approximately 60 km and is spotted of cliffs and beaches fed by fluvial imputes. The region is drained by numerous rivers and has formed a characteristic ecosystem and biodiversity.

The first aim of this study is to focus on assessing the abundance, composition, and sources of marine macro-litter (>2.5 cm) on four beaches located along the Albanian coastline – at Durrës Bay, inside the Gulf of Drin, such as Rodoni and Shëngjini Bays, and also Velipoja, as part of the southeastern Adriatic Sea coastline. The second aim is to understand the situation about marine macro-litter on Albanian beaches and to compare them to the Clean Coast Index or other similar studies.

At the national level, this study directly feeds into the implementation of the Regional Plan for Marine Litter Management in the Mediterranean of the Barcelona Convention [17].

2 Materials and Methods

2.1 *Study Area*

The beach litter surveys were carried out on beaches located at Durrës Bay, Rodoni Bay, the Gulf of Drin, and Shëngjini Bay in Albania (Fig. 1a, b). Durrës County is located on a flat alluvial and coastal plain in the southeastern Adriatic Sea, at one of the narrowest points opposite the cities of Bari and Brindisi in Italy. Durrës is the second largest city in the country with a population of some 200,000 people, and its harbor ranks as the largest passenger port in Albania and one of the largest in the Adriatic Sea [18].

Along the coastline of Durrës Bay, almost all parts have been subjected to human-induced pressures. Undoubtedly, construction works and man-made structures are predominant, while tourism and recreational activities are very intensive. Ishmi village is located 52.6 km from Durrës city center. A river with the same name, the Ishmi River flows through Durrës County and is the biggest river in this area. It is formed by several rivers which have their sources northeast of Tirana in the Skanderbeg Mountains beyond the Kruja area. Durrës plain is divided from Tirana by a long range of hills known as the Kodra e Gjatë to the east of the port city. The Ishmi River discharges into the Adriatic Sea to the southwest of Laç in Rodon Bay, which is bounded on the western edge by the Cape of Rodon and forms part of the Gulf of Drin. According to Pano [19], the drainage basin of the Ishmi covers a total area of 673 km^2. The average discharge at the mouth of the river is 20.9 m^3/s. The highest annual discharge is over six times the annual minimum.

The Gulf of Drin is an ocean basin of the Adriatic Sea within the Mediterranean Sea at the northern coast of Albania. Roughly in a shape of a scythe, it extends

Fig. 1 (**a**, **b**) Locations of studied beaches along the Albanian southern-eastern Adriatic coastline. Maps with the surveyed transects on four studied beaches: (**a**) Plepa, (**b**) Ishmi, (**c**) Velipoja, (**d**) Shëngjini (https://geoportal.asig.gov.al/map/ modified by Erion Gjyli)

immediately from the Delta of Buna in the north, across the port city of Shëngjini, to the Cape of Rodon in the south. The shoreline of the gulf is a combination of sandy shallow beaches, sand dunes, capes, salty and freshwater wetlands, estuaries, pine and coastal forests, reed beds, and coastal meadows. The Velipoja region is located about 28 km away from Shkodër town. Shkodër is the fourth most populated city in Albania and the largest city in the Shkodër County with a population of some 136,000 people. The Velipoja complex consists of Viluni lagoon, Velipoja Managed Reserve, Franz Josef promontory/island, and Buna delta. The freshwater marshes of Domni and Murteme extend along the road to Velipoja. Velipoja as part of the Gulf of Drin is also a touristic center with a wide range of accommodation options and vast opportunities to see the natural wonders of the region and to relax, especially during summer. The sediments of the Buna River also play an important role in the morphology of the seashore and the coastline with the strong coastal erosion in the Velipoja plain. A pine belt has been planted to stabilize the dunes [20].

The towns of Lezhë and Shëngjini (combined about 30,000 inhabitants) are appropriate centers to start wetland excursions to the Drin delta. Lezhë is situated at the main road from Tirana to Shkodër and Shëngjini town as the city port is about 8 km distant from Lezhë toward the sea. Both cities, especially Shëngjini, offer a wide range of accommodation options and are attractive to tourists not only in terms of nature but also as a vacation site, especially in summer. The sites investigated differed in terms of human-induced pressures with two sites classified as semi-urban, one site as urban and one as rural (Table 1) in accordance with the approach proposed by Semeoshenkova et al. [21].

Marine litter was assessed on all four investigated free-access beaches: Plepa, Ishmi, Velipoja, and Shëngjini (Fig. 1a, b). The first beach is located in the Durrës Bay, the second in Rodon Bay, the third in the Gulf of Drin, and the fourth in Shëngjini Bay, near the port of Shëngjini. The three last beaches are part of the Gulf of Drin, which extends immediately from the Delta of Buna in the north across the port city of Shëngjini to the Cape of Rodon in the south.

2.2 Methods

The beach litter surveys were performed in line with the operational guidelines produced by the EU MSFD Technical Group on Marine Litter and described by Galgani et al. [22]. Three 100 m transects were randomly positioned along the strandline of four beaches, covering a width of 10 m toward the back of the beach and ensuring that they are separated at least by a 50 m stretch. Three sampling units (100 m × 10 m) were assessed on each beach. During winter, in January and February 2015, 12 beach transects were surveyed, covering 12,000 m^2 and extending over 1,000 m along the coastline. All litter items on the beach surface larger than 2.5 cm in the longest dimension were collected in the area defined by the strandline and all the way toward the back of the beach. In the end all beaches were cleaned.

2.3 Data Analysis and Processing

The density of litter items per m^2 was calculated: $C_M = n/(w \times l)$, where C_M is the density of litter items per m^2, n is the number of litter items recorded, and w and l are the width and length of the sampling unit in meter, respectively [23]. The number of items per 100 m stretch was also calculated. The cleanliness of the beach was assessed through the Clean Coast Index (CCI): $CCI = C_M \times K$, where C_M is the density of litter items per m^2 and K is a constant that equals to 20. According to the CCI scale: 0–2 are very clean beaches, 2–5 clean, 5–10 moderately clean, 10–20

Table 1 Key features of four surveyed beaches affecting the beach marine litter densities

Area-Bay	Coordinates of the areas	Beach	Beach type	Development behind the beach	Primary area usage	Secondary area usage	Prevailing winds	Prevailing currents	Distance from the closest harbor[a] (km)	Distance from the closest town[b] (km)	Presence of cleaning activities	Distance from the closest river mouth[c] (km)
Durrës Bay	41°17′05.9″N 19°30′35.7″E	Plepa	Semi-urban	Yes	Tourism and recreation	Residential area	NNW	S	5.5	9.1	Seasonal	17.35
Rodon Bay	41°34′58.3″N 19°27′36.2′E	Ishmi	Rural	No	Tourism and recreation	Coastal fishing	NW	SW	20.0	50.2	Seasonal	8.0
The Gulf of Drin	41°51′45.4″N 19°24′53′E	Velipoja	Semi-urban	Yes	Tourism and recreation	Coastal fishing	NW	SW	15.0	29.9	Seasonal	3.3
Shëngjini Bay	41°48′21′N 19°35′59′E	Shëngjini	Urban	Yes	Tourism and recreation	Residential area	NW	SW	0.8	0.9	Seasonal	6.67

[a] The distance from the closest harbour is calculated for two ports, respectively: Durrës port, Romano port and Shëngjini port

[b] The distance from the closest town is calculated for three towns, respectively: Durrës, Shkodër and Lezhë

[c] The distance from the closest river mouth is calculated for four rivers in the area: Erzeni River, Ishmi River, Buna River and Drin River

dirty, and >20 extremely dirty [24]. According to the Master List of Categories of Litter Items, the marine litter items recorded of our study were classified into eight major groups of material types: as artificial polymer materials, rubber, cloth/textile, paper/cardboard, processed/worked wood, metal, glass/ceramics, and unidentified items and/or chemicals [22]. The collected marine litter items were also classified into three major groups of items: single-use plastics, non-single-use plastics, and non-plastic marine litter items. According to UNEP [25], the following 14 items were considered as single-use plastics: shopping bags, including pieces (G3), drink bottles ≤0.5 l (G7), drink bottles >0.5 l (G8), food containers including fast food containers (G10), plastic caps/lids from drinks (G21), cigarette butts and filters (G27), crisps packets/sweets wrappers (G30), lolly sticks (G31), cups and cup lids (G33), cutlery and trays (G34), straws and stirrers (G35), cotton bud sticks (G95), sanitary towels/panty liners/backing strips (G96), and toilet fresheners (G97). The sources of marine litter were classified into eight major categories: (1) shoreline, including poor waste management practices, tourism, and recreational activities; (2) fisheries and aquaculture; (3) shipping; (4) fly-tipping; (5) sanitary and sewage-related; (6) medical-related; (7) agriculture; and (8) non-sourced as described by Vlachogianni et al. [26]. The sources of marine litter were also classified as three major categories: (1) sea-based sources where fisheries, aquaculture, and shipping-related items are included; (2) land-based sources where shoreline, including poor waste management practices, tourism and recreational activities, medical-related, and agricultural-related items are included; and (3) mixed sources where sanitary and sewage-related, fly-tipping, and non-sourced items are included as described by Vlachogianni et al. [26].

For the statistical processing and the visualization of results, Microsoft Excel 2013 and Minitab 17 were used.

3 Results and Discussion

3.1 *Litter Densities of Surveyed Beaches and Beach Cleanliness*

The mean litter density of the four beaches studied was 219 items/100 m (range: 152–313 items/100 m) and 0.219 items/m^2 (range: 0.152–0.3131 items/m^2) (Fig. 2). The largest abundant beach in terms of items per 100 m stretch was observed in Ishmi with 313.3 items/100 m (0.313 items/m^2). The second largest was observed in Plepa with 227.3 items/100 m (0.227 items/m^2), followed by Velipoja with 183.3 items/100 m (0.183 items/m^2) and Shëngjini with 152.3 items/100 m (0.152 items/m^2).

Comparing litter density in items/m^2 of our southeastern Adriatic beaches with beaches in the north-western Adriatic coast of Italy (five beaches), the highest value was at Volano, two times higher than Ishmi and three times higher than Plepa litter

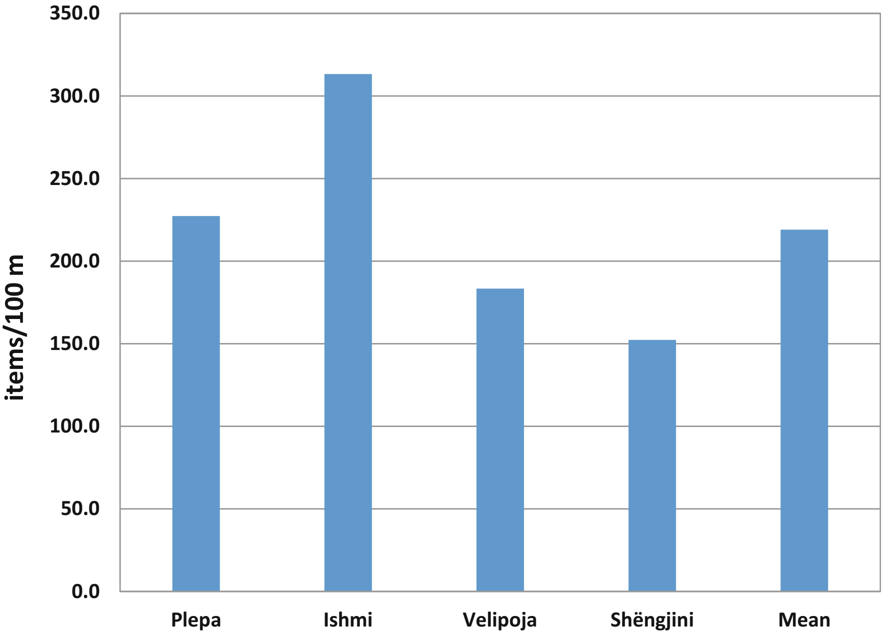


Fig. 2 Abundance of litter items for the beaches Plepa, Ishmi, Velipoja, and Shëngjini and the mean of total beaches (items per 100 m stretch)

density (0.16 items/m^2), whereas in Velipoja and Shëngjini, the situation is almost the same as in Bevano, Casalborsetti, Bellocchio, and Rosolina (Italy) [27].

Comparing the mean litter density of our study to marine litter in Mediterranean coastal and marine protected areas in northern Mediterranean countries [28], the mean litter density in items per m^2 on the beaches in Albania is three times lower than in Mediterranean coastal and marine protected areas of northern Mediterranean countries. If we compare the mean litter density of our study to beaches of the countries bordering the Adriatic and Ionian Seas [26], three times less litter (items/m^2) was detected on the Albanian beaches than on beaches along the Adriatic and Ionian Seas. According to Munari et al. [27], the litter density along the northwestern Adriatic coastline of Italy (five beaches) is almost similar to the southeastern Adriatic coast of our study (four beaches). In Pelagos Sanctuary of Italy and on Black Sea beaches of Turkey, the mean litter densities are, respectively, five and four times higher than in our study [29, 30].

On Slovenian beaches and on the coast of Ecuador, the mean litter densities are both six times higher than in our study [31, 32], whereas on Mumbai and Spain beaches, they are, respectively, 314 times higher and two times lower than in our study [33, 34].

If we compare the mean litter density in items/100 m of our study to the Lithuanian Baltic coast, the density of marine litter is about the same as on our beaches, while comparing the mean litter density of our study to the German Baltic coast, the density of marine litter is five times higher on the German Baltic coast than shown by our study [35].

Comparing the mean litter density of our study to marine litter in Japan and Israel, it is visible that the mean litter density in our study is two times lower than on Japanese beaches and 18 times higher than on Israel beaches [36, 37].

Comparing the results with other studies of Albania, the mean litter density of our study in items/m^2 is two times lower than along the Albanian coastline and almost the same in Zvernec, a protected area along the Albanian coastline [16, 28] (see Table 2).

The mean Clean Coast Index labelled our southeastern Adriatic beaches as a “Clean” beach area (CCI = 4.4). Comparing the mean Clean Coast Index of our southeastern Adriatic beaches with beaches along the north-western Adriatic coastline in Italy (five beaches, CCI = 4.5), both are labelled as “Clean” beaches [27].

Table 2 Beach marine litter densities in the Mediterranean and worldwide

Study area	No of surveyed beaches	Mean litter density (items/m^2 or items/100 m)	References
North-western Adriatic coast, Italy	5	0.2 items/m^2	[27]
Volano, north-western Adriatic coast, Italy	1	0.57 items/m^2	[27]
Bevano, north-western Adriatic coast, Italy	1	0.16 items/m^2	[27]
Casalborsetti, north-western Adriatic coast, Italy	1	0.14 items/m^2	[27]
Bellocchio, north-western Adriatic coast, Italy	1	0.13 items/m^2	[27]
Rosolina, north-western Adriatic coast, Italy	1	0.12 items/m^2	[27]
Mediterranean coastal and marine protected areas, northern Mediterranean countries	22	0.61 items/m^2	[28]
Zvernec, protected area Albania	1	0.13 items/m^2	[28]
Adriatic and Ionian Seas, all countries	31	0.67 items/m^2	[26]
Pelagos sanctuary, Italy	5	1.06 items/m^2	[29]
Turkey, Black Sea	10	0.884 items/m^2	[30]
Slovenian coast	6	1.25 items/m^2	[31]
Ecuador	26	1.31 items/m^2	[32]
Mumbai coast, India	4	68.83 items/m^2	[33]
Alicante, Spain	56	0.116 items/m^2	[34]
Lithuanian coast	4	222 items/100 m	[35]
German Baltic coast	31	47 items/100 m	[35]
Japanese coast	18	341 items/100 m	[36]
Israel	8	12.1 items per 100 m	[37]
Albania	5	0.14 items/m^2	[16]
Albania	4	0.219 items/m^2 or 219 items/100 m	Our study

The Clean Coast Index classified Ishmi as a “Moderate” beach (CCI = 6.3), whereas the other surveyed beaches were labelled as “Clean”: Shëngjini with CCI = 3; Velipoja with CCI = 3.7; Plepa with CCI = 4.5. Comparing the Clean Coast Index of the southeastern Adriatic beaches to the five surveyed beaches along the north-western Adriatic coast in Italy, the beaches of both studies are labelled as “Clean,” except for the beaches Volano (“Dirty” (CCI = 11.4), north-western Adriatic coast in Italy) and Ishmi (“Moderate” clean beach, southeastern Adriatic beaches) [27].

3.2 *Marine Litter Composition of Surveyed Beaches*

The marine litter items recorded were classified into eight major groups of material types: artificial polymer material, rubber, cloth/textile, paper/cardboard, processed/worked wood, metal, glass/ceramics, and unidentified items and/or chemicals (Fig. 3). The majority of litter items at the aggregated level (“aggregate level”

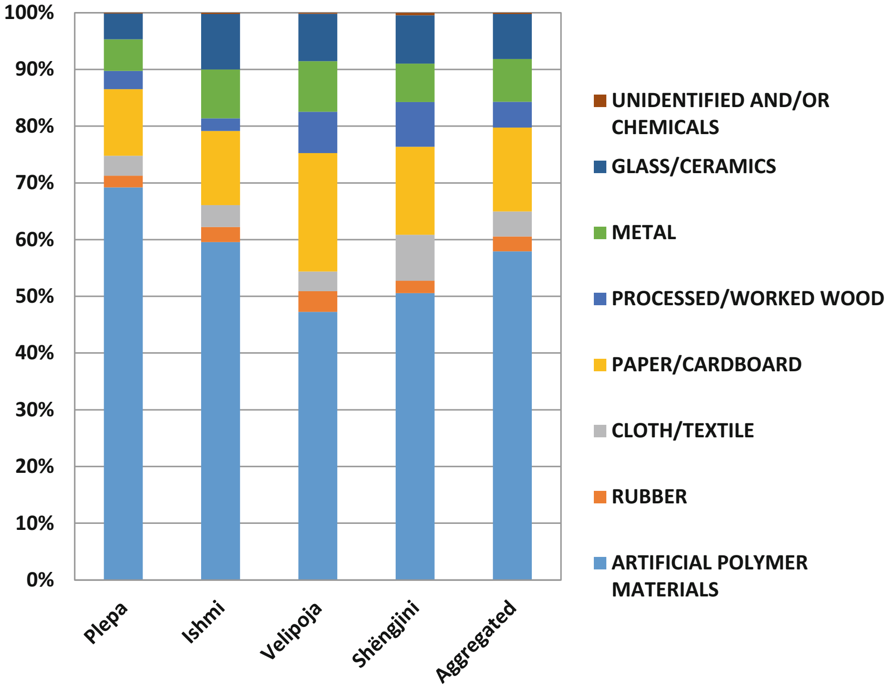


Fig. 3 Results of the percentage (%) of total litter items per material category type (artificial polymer materials, rubber, cloth/textile, paper/cardboard, processed/worked wood, metal, glass/ceramics, and unidentified items and/or chemicals) on beaches: Plepa, Ishmi, Velipoja, Shëngjini, and the aggregated level

means the sum or the assemblage of many individuals level units/sum total.) were made of artificial polymer materials (58%, or 1,523 items of 2,629 total items). Similar results about the majority of litter items carried out on Slovenian beaches reported an amount of 64% of artificial polymer materials [31]. Italian beaches along the north-western Adriatic coastline had a greater majority of marine litter made of plastic (artificial polymer materials; 81%) [27]. In Durrës Bay and Lalzi Bay, artificial polymer materials (plastics) reached 65% [16].

At the beach level of our study, the percentage of plastics ranged from 47 to 69%, whereas Schernewski et al. [35] show that plastic dominates all transects on Lithuanian beaches with 64–91%. According to the recent study of Vlachogianni et al. [26], in almost all countries of the Adriatic-Ionian region (with the exception of Albania where plastics accounted for 54.3%), plastic items were in the range of 74–92% of total items recorded, while at the regional level of all countries of the Adriatic-Ionian Seas, the amount of plastics reached 91%.

According to Liubartseva et al. [5], in the majority of Mediterranean countries, more than 50% of plastics come from their own terrestrial sources (esp. Turkey, Morocco, Israel, Spain, France, Syria, Egypt, Albania, and Tunisia).

The second most abundant material type of litter items at the aggregated level of our southeastern Adriatic was paper/cardboard (15%, or 389 items). It was also the second most abundant group on Italian beaches along the north-western Adriatic coastline [27], amounting to 7%. In contrast, in almost all countries of the Adriatic-Ionian region, the second most abundant group was glass/ceramics (3.2%) [26]. According to Schernewski et al. [35], the second most common litter was rubber on Lithuanian beaches.

The third most abundant group of items at the aggregated level of our southeastern Adriatic beaches comprises both glass/ceramics and metal and amounts to 8%. The third most abundant group of items at the aggregated level according to Munari et al. [27] also consists of glass and ceramics (3.9%) on the beaches along the north-western Adriatic coastline in Italy. According to Schernewski et al. [35], glass was also the third most abundant group on monitored Lithuanian beaches. The third most abundant group of items at the aggregated level was made of metal (1.5%) in almost all countries of the Adriatic-Ionian region [26].

This group was followed by items made of processed/worked wood (5%) and cloth/textile (4%), as well as rubber (3%) on our southeastern Adriatic beaches. No items classified as unidentified or chemicals related were found. According to Munari et al. [27], on the beaches along the north-western Adriatic coastline in Italy, foamed plastic (3.3%), rubber (1.4%), and wood (1.2%) were found. In almost all countries of the Adriatic-Ionian region, paper (1.4%), cloth/textile (1.1%), and rubber items 0.6% were counted [26].

On each studied beach, the highest percentage of plastics was recorded at Plepa (69%, 472 items), followed by Ishmi (60%, 560 items) and Shëngjini (51%, 231 items), whereas the lowest percentage of plastics was recorded for Velipoja (47%, 260 items). On Velipoja beach, paper/cardboard included 21%, on Plepa beach only 12%.

Cluster analysis was performed to identify the linkage and similarities of the four beaches. Figure 4 shows an example of a dendrogram that used the absolute abundance values of the eight groups of material types of each beach as input data

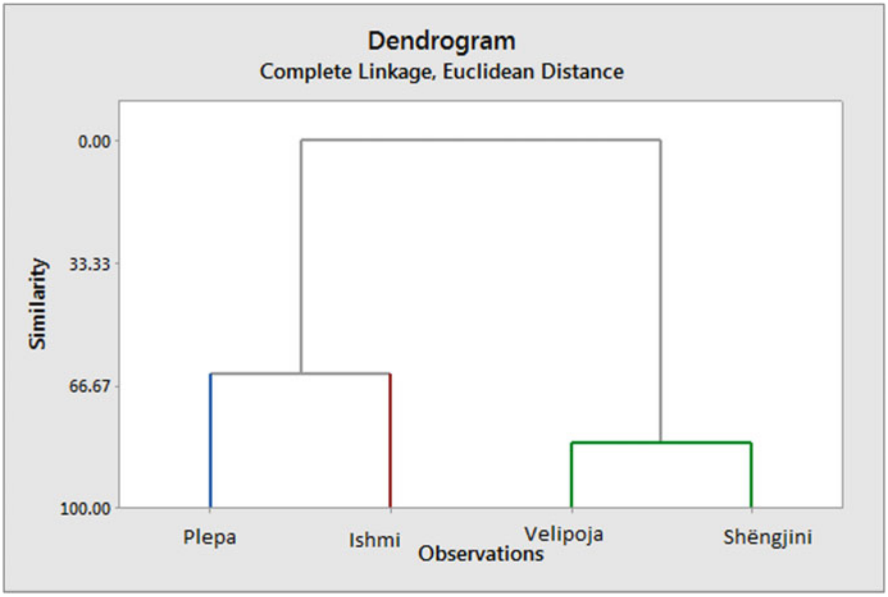


Fig. 4 Dendrogram resulting from a cluster analysis applying the complete linkage method using the Euclidean distance measure of proximity. Absolute abundance values of material types of each beach were used as input data (artificial polymer materials, rubber, cloth/textile, paper/cardboard, processed/worked wood, metal, glass/ceramics, and unidentified items and/or chemicals). Beaches of the same cluster are marked with the same color

(such as artificial polymer materials, rubber, cloth/textile, paper/cardboard, processed/worked wood, metal, glass/ceramics, and unidentified items and/or chemicals). Cluster analysis of the studied sites revealed three distinct groups. The third one refers to the beaches of Velipoja and Shëngjini, which are characterized by a strong similarity (82.5%) in terms of the total abundance of items (550 and 457, respectively) and of artificial polymer materials items (260 and 231, respectively). Ishmi beach is the second cluster due to the highest abundance of marine litter with 940 items, in particular artificial polymer materials with the most abundant item beach use-related cosmetic bottles and containers, e.g., sunblocks (G11), followed by “other bottles & containers (drums)” (G13), but also had the most quantity of paper/cardboard, with the most abundant items cartons/tetra pack (others) (G151); glass and ceramic in particular jars, including pieces (G201) and metal of all beaches, in particular cans (food) (G176). Plepa beach is in the first cluster due to the high abundance of marine litter with 682 items, in particular artificial polymer materials with the most abundant items “other bottles & containers (drums)” (G13), followed by cigarette butts and filters (G27), other plastic/polystyrene items (identifiable) (G124), shopping bags, including pieces (G3), and food containers including fast food containers (G10).

The top 30 items accounted for 73% of all 2,629 items found (Table 3). Among 120 litter item categories recorded, other bottles and containers (drums) (G13) were

Table 3 Top 30 items found on the four studied beaches: Plepa, Ishmi, Velipoja, and Shëngjini, labelled according Galgani et al. [22]: Annex 8.1 – Master List of Categories of Litter Items

	Material type	*Code TSG_ML (Technical Subgroup on Marine Litter), General Code	General name	Total	%
1	Artificial polymer materials	G13	Other bottles and containers (drums)	158	6.01
2	Paper/cardboard	G151	Cartons/tetra pack (others)	150	5.71
3	Artificial polymer materials	G27	Cigarette butts and filters	98	3.73
4	Artificial polymer materials	G3	Shopping bags, including pieces	92	3.50
5	Artificial polymer materials	G11	Beach use-related cosmetic bottles and containers, e.g., sunblocks	84	3.20
6	Artificial polymer materials	G21	Plastic caps/lids drinks	81	3.08
7	Glass/ceramics	G201	Jars, including pieces	80	3.04
8	Artificial polymer materials	G10	Food containers incl. Fast food containers	78	2.97
9	Artificial polymer materials	G7	Drink bottles ≤0.5 l	73	2.78
10	Paper/cardboard	G156	Paper fragments	71	2.70
11	Artificial polymer materials	G8	Drink bottles >0.5 l	70	2.66
12	Artificial polymer materials	G66	Strapping bands	67	2.55
13	Artificial polymer materials	G124	Other plastic/polystyrene items (identifiable)	67	2.55
14	Metal	G177	Foil wrappers, aluminum foil	66	2.51
15	Artificial polymer materials	G80	Plastic pieces >50 cm	63	2.40
16	Paper/cardboard	G154	Newspapers and magazines	54	2.05
17	Artificial polymer materials	G72	Traffic cones	53	2.02
18	Cloth/textile	G139	Backpacks and bags	53	2.02
19	Artificial polymer materials	G5	Plastic bag collective role; what remains from rip-off plastic bags	47	1.79
20	Glass/ceramics	G202	Light bulbs	47	1.79
21	Metal	G175	Cans (beverage)	41	1.56
22	Artificial polymer materials	G23	Plastic caps/lids unidentified	39	1.48
23	Paper/cardboard	G158	Other paper items	37	1.41

(continued)

Table 3 (continued)

	Material type	*Code TSG_ML (Technical Subgroup on Marine Litter), General Code	General name	Total	%
24	Cloth/textile	G138	Shoes and sandals (e.g., leather, cloth)	36	1.37
25	Processed/worked wood	G172	Other wood >50 cm	36	1.37
26	Metal	G176	Cans (food)	35	1.33
27	Glass/ceramics	G210	Other glass items	35	1.33
28	Artificial polymer materials	G26	Cigarette lighters	33	1.26
29	Artificial polymer materials	G58	Fish boxes – expanded polystyrene	32	1.22
30	Processed/worked wood	G171	Other wood <50 cm	32	1.22

the most frequently found items with a percentage of 6% (158 items). The second most abundant group of items was cartons/tetra pack (others) (G151) with 5.7%, followed by cigarette butts and filters (G27) with 3.7%, shopping bags, including pieces (G3), with 3.5%, and beach use-related cosmetic bottles and containers, e.g., sunblocks (G11), with 3.2%. Plastic caps/lids drinks (G21), jars, including pieces (G201), food containers including fast food containers (G10), drink bottles ≤0.5 l (G7), paper fragments (G156), drink bottles >0.5 l (G8), strapping bands (G66), other plastic/polystyrene items (identifiable) (G124), foil wrappers, aluminum foil (G177), and plastic pieces >50 cm (G80) were among the top 15 items found.

On monitored Lithuanian beaches (five transects on a remote part of the Curonian Spit in Lithuania [35]), eight out of ten most common single items were made of plastic. Most abundant are "Other plastic/polystyrene items" (119 items), "String and cord" (diameter less than 1 cm; 110 items), and "Small plastic bags," e.g., freezer bags (108 items). At the aggregated level, in countries of the Adriatic-Ionian region, according to Vlachogianni et al. [26], among 159 litter categories, plastic pieces larger than 2.5 cm and smaller than 50 cm in the longest dimension (G79), accounted for the highest percentage 19.89% (14,040 items) of the total 70,581 litter items recorded in all surveys, followed by polystyrene pieces 2.5 cm> <50 cm (G82) with 11.93%. The third most abundant group of items was cotton bud sticks (G95) accounting for 9.17% of total items recorded, followed by plastic caps/lids from drinks (G21) with 6.67% and cigarette butts and filters (G27) with 6.6%. According to Munari et al. [27], among 35 litter categories, cigarette butts accounted for the highest percentage (22.9%) on five beaches of the north-western Adriatic coast in Italy, followed by unrecognizable plastic pieces (13.5%), bottle caps (9.2%), mesh bags (7.2%), plastic bottles, and cutlery (6.5% and 6.4%, respectively).

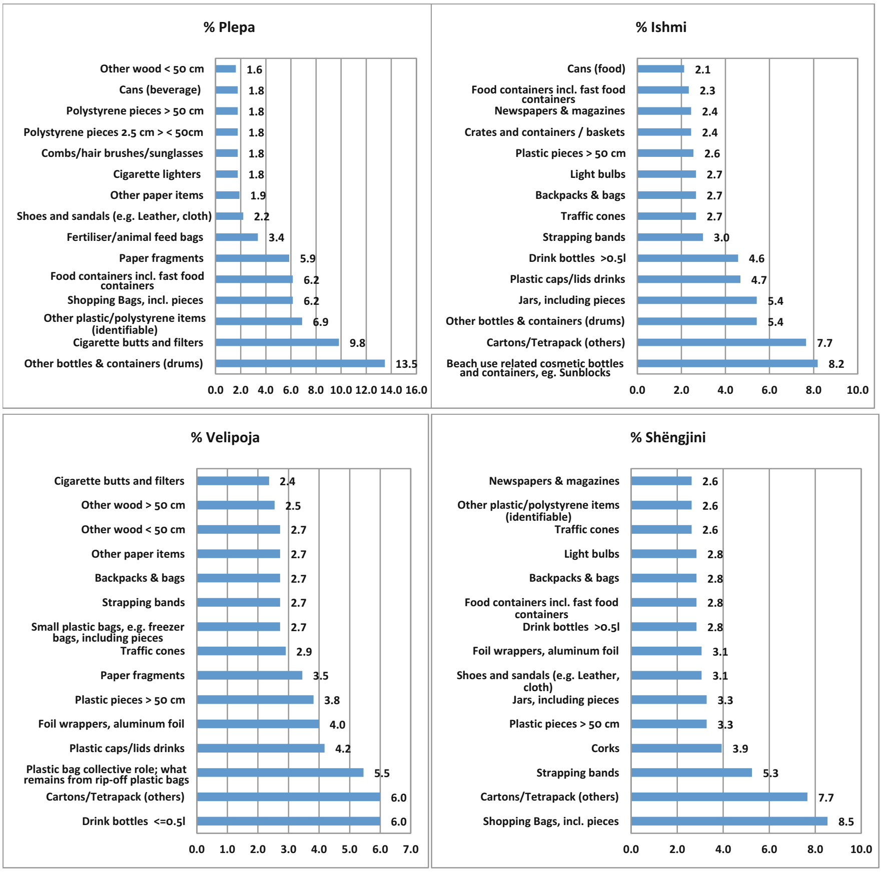


Fig. 5 Percentage of top 15 items found on each of the four studied beaches: Plepa, Ishmi, Velipoja, and Shëngjini

The percentage of top 15 marine litter items found in each of the four studied beaches: Plepa, Ishmi, Velipoja, and Shëngjini varied as shown in Fig. 5. At Plepa, the top 15 items accounted for 66.3% of all items recorded. The most abundant item was other bottles and containers (drums) (G13) with a percentage of 13.5% of all items recorded, followed by cigarette butts and filters (G27) with 9.8%, other plastic/polystyrene items (identifiable) (G124) with 6.9%, and shopping bags, including pieces (G3), and food containers including fast food containers (G10), both with 6.2%.

At Ishmi, the top 15 items made up 58.8% of all items found. The most abundant item were beach use-related cosmetic bottles and containers, e.g., sunblocks (G11) with 8.2%. The second most abundant items were cartons/tetra pack (others) (G151) with 7.7%, followed by other bottles and containers (drums) (G13) and jars, including pieces (G201), both with 5.4%, followed by plastic caps/lids (G21) with 4.7%.

At Velipoja, the top 15 items accounted for 54.4% of the total items. The most abundant items were drink bottles ≤0.5 l (G7) and cartons/tetra pack (others) (G151), both with 6%, followed by "plastic bag collective role; what remains from rip-off plastic bags" (G5) with 5.5%, plastic caps/lids drinks (G21) with 4.2%, and foil wrappers, aluminum foil (G177), with 4%. This is similar to Poeta et al. [38], who found plastic fragments, plastic bottles, and bottle caps to have the highest occurrence on the Tyrrhenian shores.

At Shëngjini, the top 15 items accounted for 57.3% of the total items found. The most abundant item was shopping bags, including pieces (G3) accounting for 8.5% of all items recorded, followed by cartons/tetra pack (others) (G151) with 7.7%, strapping bands (G66) with 5.3%, corks (G159) with 3.9%, and plastic pieces >50 cm (G80) with 3.3%.

On all our beaches, plastic (artificial polymer materials) was the first material type found, with other bottles and containers (drums) (G13) at Plepa; shopping bags, including pieces (G3) at Shëngjini; related cosmetic bottles and containers, e.g., sunblocks (G11) at Ishmi; and drink bottles ≤0.5 l (G7) at Velipoja. At Velipoja, however, plastic and paper/cardboard were both the first material types found, with drink bottles ≤0.5 l (G7) and cartons/tetra pack (others) (G151), respectively (both 6%).

Plastic and paper/cardboard were the second material types with cigarette butts and filters (G27) (plastic) at Plepa, cartons/tetra pack (others) (G151) (paper/cardboard) both at Ishmi and Shëngjini, followed by "plastic bag collective role; what remains from rip-off plastic bags" (G5) (plastic) at Velipoja.

The collected marine litter items were classified into three major groups: single-use plastics, non-single-use plastics, and non-plastic marine litter items. Results are presented at the aggregated level and the beach level (Fig. 6). According to UNEP [25], the following items were considered as single-use plastics: shopping bags, including pieces (G3), drink bottles ≤0.5 l (G7), drink bottles >0.5 l (G8), food containers including fast food containers (G10), plastic caps/lids from drinks (G21), cigarette butts and filters (G27), lolly sticks (G31), cups and cup lids (G33), cutlery and trays (G34), straws and stirrers (G35), sanitary towels/panty liners/backing strips (G96), and toilet fresheners (G97). Crisps packets/sweets wrappers (G30) and cotton bud sticks (G95) are not found on our studied beaches.

At the aggregated level, single-use plastics accounted for 29% of the items recorded. At the beach level, the abundance of single-use plastics varied from 16.4 to 58.7%.

The highest abundance of single-use plastics was recorded at Plepa with 58.7%, followed at Shëngjini with 20.6% and Ishmi with 18.8%, while the lowest abundance of single-use plastics was recorded at Velipoja with 16.4%. Same percentage with single-use plastics was found for non-single-use plastics (29%) at the aggregated level.

If we look at a recent study [28], single-use plastic marine litter items at the aggregated level in marine litter in Mediterranean coastal and marine protected areas and single-use plastics in northern Mediterranean countries accounted for one fifth (21%) of the items recorded. In our study this value is 1.5 times higher. Comparing

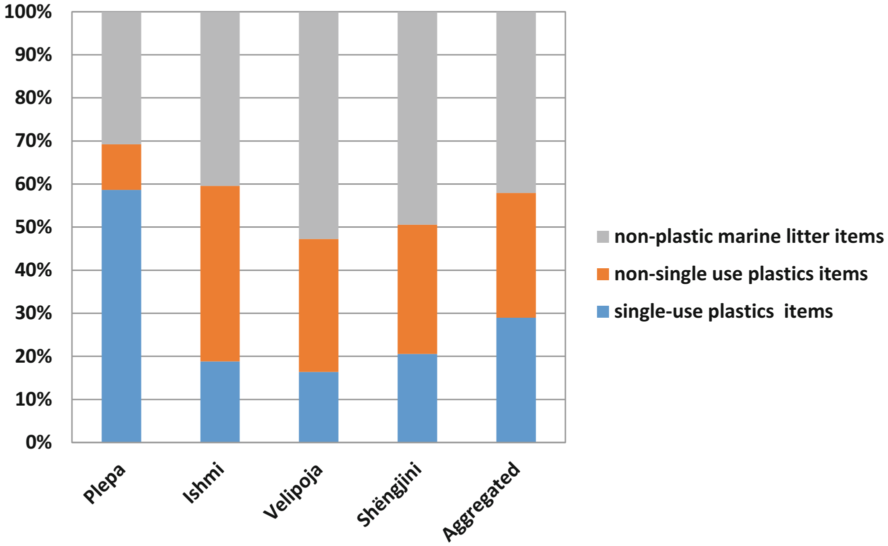


Fig. 6 Percentage of single-use plastic items recorded in beaches: Plepa, Ishmi, Velipoja, Shëngjini, and the aggregated level

our results to a study by Adamo et al. [39] who reported that single-use plastics represent 50% of the total marine litter items found on European beaches in 2016, it is evident that the level of single-use plastics of our study is 2.5 times lower than on other European beaches.

The European Commission has begun to implement regulations [40] on the reduction of plastic production, especially single-use plastic products. The ban will apply to plastic ear sticks, plastic cutlery, straws, mixing sticks for drinks, and balloon holders. All these products will have to be produced only from sustainable materials. EU member states will have to reduce the use of plastic food containers and drink cups and by 2025 will need to organize the collection of 90% of disposable plastic bottles, for example, through a return program [41].

Figure 7 shows an example of a dendrogram that used the absolute abundance values of the 12 single-use plastics at each beach as input data. The following items were considered as single-use plastics: shopping bags, including pieces (G3), drink bottles ≤0.5 l (G7), drink bottles >0.5 l (G8), food containers including fast food containers (G10), plastic caps/lids from drinks (G21), cigarette butts and filters (G27), lolly sticks (G31), cups and cup lids (G33), cutlery and trays (G34), straws and stirrers (G35), sanitary towels/panty liners/backing strips (G96), and toilet fresheners (G97). Crisps packets/sweets wrappers (G30) and cotton bud sticks (G95) are not found in the studied beaches.

Cluster analysis of the studied sites revealed four distinct groups. The fourth one refers to the beach of Shëngjini, which is dominated by shopping bags, including pieces (G3) (41% of the total abundance of single-use plastic items in this beach). The third cluster on Velipoja beach is dominated by drink bottles ≤0.5 l (G7) (37%

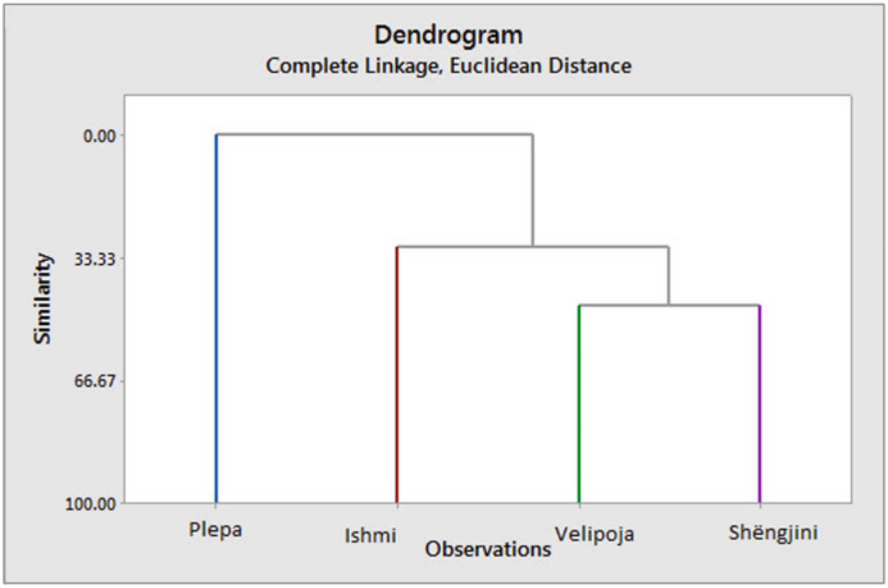


Fig. 7 Dendrogram resulting from a cluster analysis applying the complete linkage method using the Euclidean distance measure of proximity. Absolute abundance values of the 12 single-use plastics at each beach were used as input data. The following items were considered as single-use plastics: shopping bags, including pieces (G3), drink bottles ≤0.5 l (G7), drink bottles >0.5 l (G8), food containers including fast food containers (G10), plastic caps/lids from drinks (G21), cigarette butts and filters (G27), lolly sticks (G31), cups and cup lids (G33), cutlery and trays (G34), straws and stirrers (G35), sanitary towels/panty liners/backing strips (G96), and toilet fresheners (G97). Beaches of the same cluster are marked with the same color

of the total abundance of single-use plastic items on this beach). The second cluster on Ishmi beach is dominated by plastic caps/lids from drinks (G21) (24.9%) and drink bottles >0.5 l (G8) (24.3% of the total abundance of single-use plastic items on this beach). The first cluster of Plepa beach is dominated by cigarette butts and filters (G27) (36.6% of the total abundance of single-use plastic items in this beach).

With regard to smoking-related marine litter items, on the aggregated basis, 5% of the total litter items collected fell under one of the following category types of litter: tobacco pouches/plastic cigarette box packaging (G25), cigarette lighters (G26), cigarette butts and filters (G27), and paper cigarette packets (G152) (Fig. 8). Comparing our study to related studies in Mediterranean coastal and marine protected areas in northern Mediterranean countries, the level of smoking-related items is similar to the aggregated basis (3%) [28].

The highest percentage of smoking-related items was recorded on Plepa beach (12% of sampled items) (Fig. 8). The lowest percentage of smoking-related items was observed at Shëngjini beach (2%), followed by Ishmi and Velipoja (both 3%).

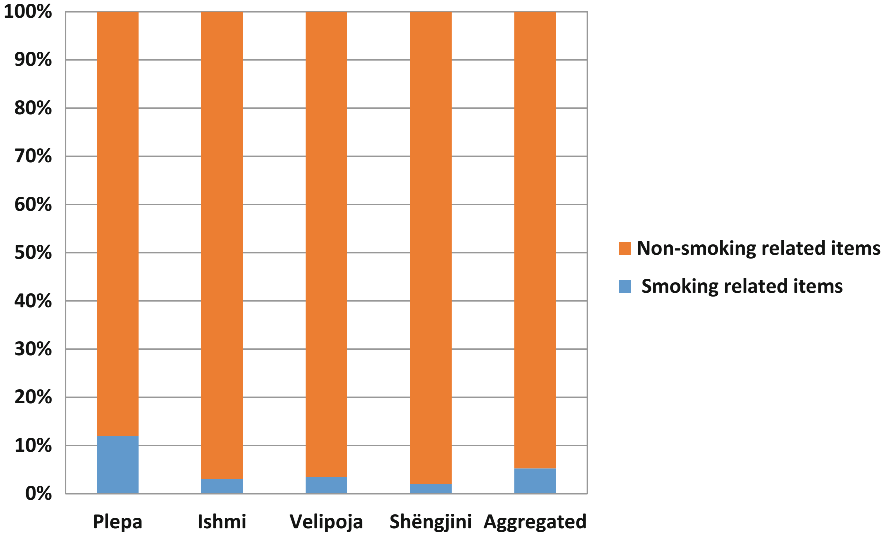


Fig. 8 Percentage of smoking-related items recorded in beaches: Plepa, Ishmi, Velipoja, Shëngjini, and the aggregated level

On Plepa beach, 9.8% of total items were recorded as cigarette butts and filters (G27) (Fig. 5), as the second most items found on the beach. Similar situations where cigarette butts are the first or second most frequent found type of litter are reported by other studies carried out at beaches located in Italy [27] Slovenia [31], Spain [34], and Albania [16].

3.3 *Marine Litter Sources of Surveyed Beaches*

At the aggregated level, 50.1% or 1,316 items of all litter items collected could be attributed to one of the following sources: tourism- and recreational-related items (including poor waste management), fishing-related items, shipping-related, fly-tipping, agricultural-related items, sanitary sewage-related, and medical-related (Fig. 9). Litter items from shoreline sources such as tourism and recreational activities, including poor waste management practices, accounted for 37.5% or 987 items of all litter items collected, while smoking-related items were 14% or 138 items of litter items from shoreline sources such as tourism and recreational activities. The related sources of the second most often found items were shipping-related items (4.8% or 125 items), while the third most were from fisheries and aquaculture (3.1% or 81 items). Fly-tipping-related items accounted for 2.4% or 64 items, while agricultural-related items, sanitary and sewage-related items, and medical-related items accounted for 0.9% or 23 items, 0.8% or 20 items, and 0.6% or 16 items, respectively.

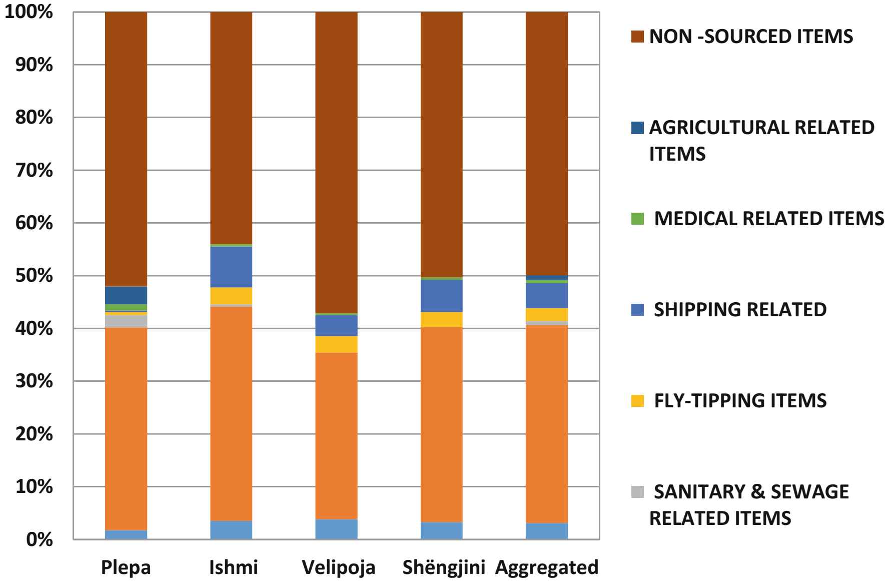


Fig. 9 Percentages of the sources of marine litter: fisheries and aquaculture, shoreline sources such as tourism and recreational activities, sanity and sewage-related items, fly-tipping items, shipping-related items, medical-related items, agricultural-related items, and non-sourced items recorded in beaches: Plepa, Ishmi, Velipoja, Shëngjini, and the aggregated level

Comparing the percentage of litter from shoreline sources such as tourism and recreational activities, including poor waste management practices in our study (37.5%) to other studies, litter from shoreline sources, such as tourism and recreational activities and poor waste management practices, was almost 1.5 times higher (27% [28]) in our study than in Mediterranean coastal and marine protected areas of northern Mediterranean countries. On German Baltic beaches, this percentage was 1.5 times higher (50% [35]) than in our study. On the beaches of the north-western Adriatic coast in Italy, marine litter sources were primarily the shoreline and recreational activities (37.9% [27]) such as in our study, with the same situation in countries of the Adriatic-Ionian region (33.4% [26]). Comparing our results to the global average in 2010 (68.2% [42]), the percentage of litter from shoreline and recreational activities was almost two times higher than the global average. Compared to the Mediterranean average (52% [43]), the Mediterranean average was 1.5 times higher than in our study. Comparing the percentage of litter from shoreline sources such as tourism and recreational activities, including poor waste management practices, our study reveals 1.5 times lower results than along the Albanian coastline-Durrës beaches (58.5% [16]).

At the individual beach level (Fig. 9), the inputs of litter from the different sectors and their comparative importance were quite similar, with shoreline sources such as tourism and recreational activities accounting for the vast majority of litter items collected (range: 31.6–40.6%). Ishmi (40.6%) accounted for the highest percentage of litter from shoreline sources such as tourism and recreational activities and poor waste management practices, followed by Plepa (38.4%), Shëngjini (37.0%), and Velipoja (31.6%).

The highest marine litter inputs from shipping were recorded at Ishmi with 7.8% and Shëngjini with 6.1%. Fishing-related items range between 3.3 and 3.8% in three beaches except Plepa (1.8%). The highest fishing-related items were found at Velipoja. The highest number of items from fly-tipping-related waste was recorded at Ishmi with 3.2%. Agricultural and sanitary and sewage-related items as well as medical-related items were found in very small percentages in all studied beaches or not at all. Agricultural-related items were not found at Ishmi, Velipoja, and Shëngjini and sanitary and sewage-related items not found at Velipoja and Shëngjini.

Figure 10 shows an example of a dendrogram that used as input data the absolute abundance values of the eight groups of sources in each beach as attributed to one of the following sources: fishing-related items and aquaculture, shoreline sources such as tourism and recreational activities (including poor waste management), sanity and sewage-related items, fly-tipped items, shipping-related items, medical-related items, agricultural-related items, and non-sourced items. Cluster analysis of the studied sites revealed three distinct groups. The third one refers to the beaches of Velipoja and Shëngjini, which are characterized by a strong similarity (70%) in

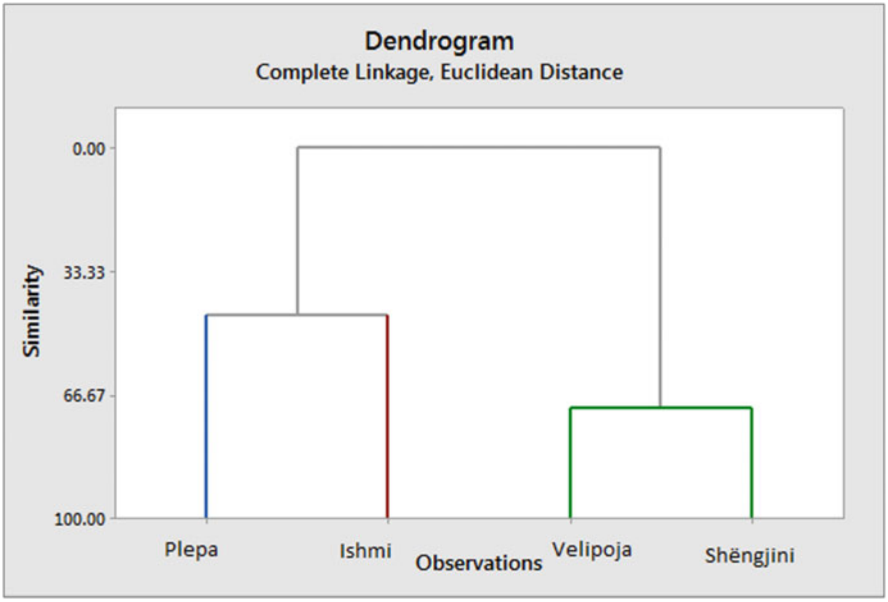


Fig. 10 Dendrogram resulting from a cluster analysis applying the complete linkage method using the Euclidean distance measure of proximity. Absolute abundance values of the sources of marine litter at the individual beach level: fisheries and aquaculture, shoreline sources such as tourism and recreational activities, sanity and sewage-related items, fly-tipping items, shipping-related items, medical-related items, agricultural-related items, and non-sourced items. Beaches of the same cluster are marked with the same color

terms of the total abundance of sources, especially tourism- and recreational-related items, respectively, with 174 items or 17.6% and 169 items or 17.1% from the aggregated tourism- and recreational-related items for all beaches (987 items). The second cluster is located at Ishmi that has the largest abundance of source items (940 items), especially tourism and recreational (382 or 38.7%) as well as shipping-related items (73 items or 58.4%) from the aggregated shipping-related items for all beaches. Plepa beach is in the first cluster due to the high abundance of source items (682 items), especially tourism- and recreational-related items with 262 or 26.5% of the aggregated tourism- and recreational-related items.

In our study, we assessed the contribution of the sea-based sources (fisheries and aquaculture, shipping), the land-based sources (shoreline, tourism and recreational activities, agriculture, medical-related), and the mixed sources (sanitary and sewage-related, fly-tipping, non-sourced items). At the aggregated level, the items coming from sea-based sources resulted in 7.8% versus 39% of items attributed to land-based sources (Fig. 11). Comparing the results to beaches in the Adriatic Sea and Mediterranean basin, the highest contribution of sea-based vs. land-based sources was observed for Italy (14.8% vs. 27%) and Greece (13.2% vs. 48.0%), while the lowest contribution was recorded for Montenegro (1.5% vs. 74.1%) and Bosnia and

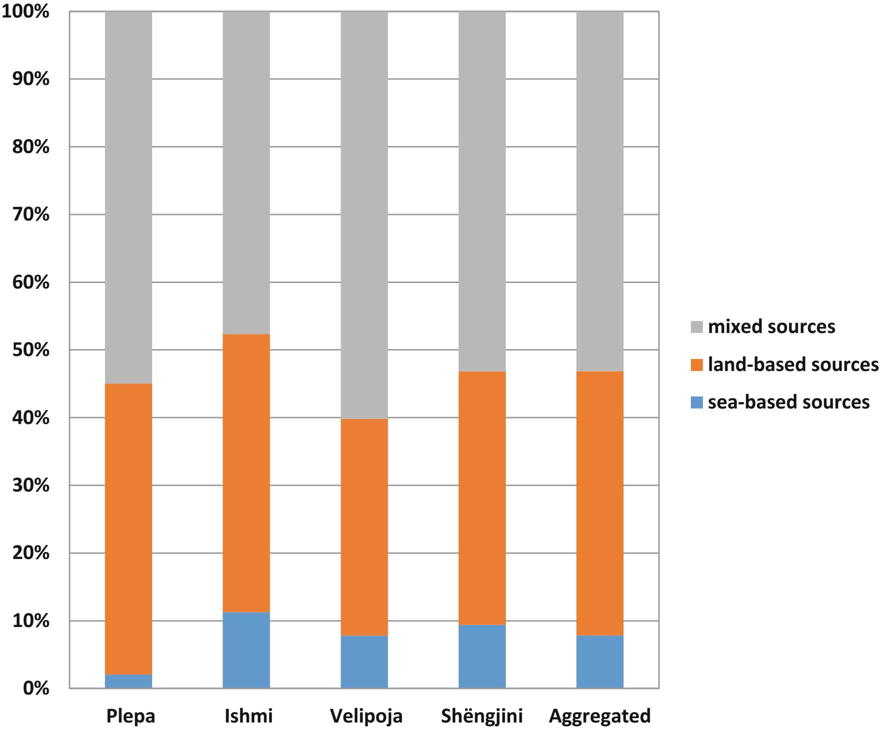


Fig. 11 Percentages of three different sources categories: land-based, sea-based, and mixed sources recorded in beaches: Plepa, Ishmi, Velipoja, Shëngjini, and the aggregated level

Herzegovina (1.9% vs. 82.8%) [26]. For the Mediterranean Sea, PNUE/PAM/MEDPOL [43] reported that most of the marine litter comes from land-based rather than sea-based sources.

At the individual beach level (Fig. 11), the inputs of litter from sea-based sources range from 2.1 to 11.3%. In contrast, litter from land-based sources accounts for 32–43%. The highest contribution of sea-based sources vs. land-based sources was observed at Ishmi (11.3% vs. 41.1%), followed by Shëngjini (9.4% vs. 37.4%), Velipoja (7.8% vs. 32%), and Plepa (2.1% vs. 43%). On five north-western Adriatic beaches (Italy), the majority of marine litter also comes from land-based sources [27].

3.4 Options to Manage the Litter of Beaches

The present study shows that shoreline sources such as tourism and recreational activities, in addition to the poor waste management practices, are the main sources of beach litter deposited on surveyed beaches. We recommend the following priority management options based on the beach litter outline of each of the surveyed beaches:

– Measures to deal with cartons/ tetra pack (others) paper cardboard items

In all studied beaches, with exception of Plepa, cartons/tetra pack (others) (G151) is the first or second most frequently found type of litter.

There are serious problems in recycling of composite beverage cartons that completed their lifetime and became waste. This packaging waste is disposed in landfills. It is important to recycle and recover Tetra Pak's due to the different types of recyclable materials (includes 75% paper, 20% polyethylene, and 5% aluminum. Tetra Pak films were cut into over 40 mm pieces and charged to the reactor with stirring and chloroform. Thus paper, aluminum, and polyethylene dissolve in chloroform [44]. Tetra Pak packaging is considered an important source of high-quality waste paper for the pulp industry. Processing of UTPC (used Tetra Pak cartons) is rising rapidly and driven by stricter regulations in terms of waste disposal, as well as by decreasing supply of waste paper in digital era [45].

– Measures to tackle plastics, including single-use plastic items

The same percentage of single-use plastics was found for non-single-use plastics (29%) at the aggregated level. On the aggregated level of studied beaches, plastics (artificial polymer materials) were the first source items found. Non-single-use plastic other bottles and containers (drums) (G13) were the first most often litter items found (in Plepa and Ishmi), and beach use-related cosmetic bottles and containers, e.g., sunblocks (G11), were the fifth most often litter items found (in Ishmi).

Whereas single-use plastics at the aggregated level were the third most often litter items found, cigarette butts and filters (G27) dominated especially Plepa beach. The fourth most often litter items were ranked shopping bags, including pieces (G3), and found in Shëngjini and Plepa as the most frequented beaches, whereas the sixth most often litter items were plastic caps/lids drinks (G21) on Ishmi and Velipoja beaches.

A public awareness-raising campaign to highlight citizens' role, especially targeted to tourists, local residents, and other coastal and marine users toward a behavioral change to reduce consumption of single-use plastics, is imperative in combatting plastic pollution and marine litter.

The European Commission has begun implementing regulations [40] on the reduction of plastic production, especially single-use plastic products. By 2021, the legislative actions for banning certain items as also foreseen under the EU Single-Use Plastics Directive that includes bans on single-use plastic cutlery, single-use plastic plates, plastic straws, cotton bud sticks made of plastic, and plastic balloon sticks as well as oxodegradable plastics, food containers, and expanded polystyrene cups [40]. More details are found in "Moving away from single-Use" [46]. Plastic pollution from plastic bags and their respective pieces has been accepted by the Albanian decision-makers, and thus, the Council of Ministers of Albania decided in 2018 [47] to introduce a ban on lightweight plastic bags, making it illegal to import, manufacture, use, sell, or bring such bags into the country. Since the beginning of July 2018, plastic bags in Albania must have a minimum thickness of 35 μm in order to be reusable and have a recycling value.

- More investments to build other landfills

At the moment, there are only three sanitary landfills available in Albania in Tirana, Elbasan, and Fier [48]. The State of the Environment Report of the European Environment Agency [49] highlights that the Albanian national legal framework related to the integrated management of waste has been completed in accordance with the EU Directive on waste; however much work is needed to fully implement this, while raising public awareness and making more investments in building other landfills according to EU standards are imperative.

- Contaminated Ishmi River Rehabilitation by Dredging Soil Treatment and Water Purification

The largest abundance in terms of items per 100 m stretch was observed in Ishmi with 313.3 items/100 m. Highest abundance of marine litter is found on Shen Pjetri beach in Lalzi Bay (located in the vicinity of Ishmi River and Erzeni River) with 510 items/100 m [16]. It seems that riverine inputs from Ishmi located in the vicinity might be the prevalent pathway of litter deposits on Ishmi and Shen Pjetri beaches. According to Cullaj et al. [50] Ishmi River shows that high levels of nitrites were detected in rivers including Lana and Tirana tributaries; they exceed the EC guide values for high-quality fresh water (<0.01 mg/L for Salmonid waters and <0.03 mg/L for Cyprinid waters). According to ARCADIS ECOLAS/IEEP/Metroeconomica/Enviro-L Content 06/11347 [51], Ishmi River and Gjanica River are the most polluted rivers in Albania. Problems are caused by micropollutants (BTEX), BOD5, COD, phenols, etc. According to Baumann [52] among all Albanian rivers, it is particularly Ishmi River that causes great concern: Around one third of Albania's 2.8 million inhabitants live along this river which is the most polluted river of the country. It flows through Tirana District, including the capital city Tirana, Vora, and Kamza, and collects almost all sewage waters of the region as well as significant amounts of solid waste of industries and inhabitants. There is scientific evidence for this visible pollution of the Ishmi River Basin and its tributaries, particularly the Lana but also the Zeza. These rivers show the highest pollution of all rivers in Albania, with harmful substances exceeding by far EU standards and increasing levels of pollution. The pollution threatens the health of people and has extinguished an important sea turtle breeding ground as well as fishes and other life under water. Furthermore, the touristic potential of the area is minimized, which otherwise would be very attractive due to a high biodiversity and some of the few historic sites that have survived communist times. According to Baumann [52], cleaning Ishmi River is needed: (1) awareness raising among stakeholders; (2) municipality cooperation to improve waste collection in the Ishmi River Basin; (3) reduction of Cd and Pb in the river to harmless levels; (4) establishment of "Constructed Wetlands" as a traditional sewage treatment method in a rural hotspot community; and (5) wastewater treatment in the municipalities of Tirana, Kamza, and Vora.

4 Conclusions

The present study provides the assessment of marine litter pollution on four beaches along the southeastern Adriatic coast. In this study about the Bay of Durrës and the Gulf of Drin, which include Rodoni Bay and Shëngjini Bay, the average density of beach litter amounted to 0.219 items/m^2 or 219 items/100 m ranging from 152.3 to 313.3 items/100 m. The largest abundant beach in terms of items per 100 m stretch was observed in Ishmi with 313.3 items/100 m.

The studied beaches were in human-induced pressures and were classified either as urban, semi-urban, or rural. Plastics or artificial polymer materials were the majority of marine litter items on beaches, with an average of 58%. The most abundant item was G13 (other bottles and containers (drums)) with 6%, followed by G151 (cartons/ tetra pack (others)) with 5.7%, G27 (cigarette butts and filters) with 3.7%, and G3 (shopping bags, incl. pieces) and G11 (beach use-related cosmetic bottles and containers, e.g., sunblocks), respectively, with 3.5% and 3.2%. At the aggregated level, single-use plastics accounted for 29% of the items recorded. The highest abundance of single-use plastic items was recorded at Plepa beach with 58.7%. Plepa as semi-urban beach is classified as the most frequented beach from visitors comparing to other studied beaches, as we also find the second most items G27 (cigarette butts and filters). Litter from shoreline sources such as tourism and recreational activities, including poor waste management practices, accounted for 37.5% of litter collected, while the shipping-related items accounted for 4.8%. Sea-based sources of litter (fisheries and aquaculture, shipping) accounted for 8% of total litter items on all beach locations, contrary to 39% of items attributed to land-based sources (shoreline, tourism and recreational activities, agricultural-related items, medical-related items). It seems that riverine inputs from Ishmi Basin located in the vicinity of Ishmi beach might be the prevalent pathway of litter deposits on this beach with the highest abundance of items.

Our findings may give insights into human nature behavior to manage marine litter deposition on the southeastern Adriatic coast. The litter that was deposited in situ, especially on Ishmi beach (36%), suggests mitigation actions that may substantially help to address the problem such as: (1) awareness raising campaigns to promote concept of “Leave No Trace” to tourists, local residents, and other beach users; (2) increasing targeted clean-up, particularly in summer months when the number of tourists is very high; (3) expanding direct intervention through signage and patrols; (4) legislative actions prohibiting the dumping of marine litter in rivers, as the Ishmi river is heavily polluted by Tirana County; (5) legislative actions for banning certain items as foreseen also under the EU Single-Use Plastics Directive that includes bans on single-use plastic cutlery, plastic plates, plastic straws, cotton bud sticks made of plastic and plastic balloon sticks, as well as oxodegradable plastics, food containers, and expanded polystyrene cups; and (6) fostering broader education and awareness in youth and students about marine litter consequences in the ocean.

Moreover, these results emphasize the key role of coastal areas in the Mediterranean Basin, which also include the Albanian coastline and sustainability of the Integrated Coastal Zone Management (ICZM) at a national level.

Acknowledgments The authors would like to thank the European Union that financed the IPA – DeFishGear project. Albania was included in the international project which aims to facilitate efforts for integrated planning to reduce the environmental impacts of litter-generating (including plastic) activities and ensure the sustainable management of the marine and coastal environment of the Adriatic Sea. This is the second project for marine litter surveillance in Albania. The first surveillance was done for the demersal stock and marine litter in the Eastern Southern Adriatic Sea – supported by the FAO – AdriaMed Project.

References

1. Suaria G, Avio CG, Mineo A, Lattin GL, Magaldi MG, Belmonte G, Moore CJ, Regoli F, Aliani S (2016) The Mediterranean plastic soup: synthetic polymers in Mediterranean surface waters. Nat Sci Rep 6:37551. https://doi.org/10.1038/srep37551
2. Arcangeli A, Campana I, Angeletti D, Atzori F, Azzolin M, Carosso L, Di Miccoli V, Giacoletti A, Gregorietti M, Luperini C, Paraboschi M, Pellegrino G, Ramazio M, Sarà G, Crosti R (2018) Amount, composition, and spatial distribution of floating macro litter along fixed trans-border transects in the Mediterranean basin. Mar Pollut Bull 129:545–554. https://doi.org/10.1016/j.marpolbul.2017.10.028
3. Fortibuoni T, Ronchi F, Mačić V, Mandić M, Mazziotti C, Peterlin M, Prevenios M, Prvan M, Somarakis S, Tutman P, Varezi DB, Virsek MK, Vlachogianni T, Zeri C (2019) A harmonized and coordinated assessment of the abundance and composition of sea floor litter in the Adriatic-Ionian macroregion (Mediterranean Sea). Mar Pollut Bull 139:412–426. https://doi.org/10.1016/j.marpolbul.2019.01.017

4. Borrelle SB, Rochman CM, Liboiron M, Bond AL, Lusher A, Bradshaw H, Provencher JF (2017) Opinion: why we need an international agreement on marine plastic pollution. Proc Natl Acad Sci 114:9994–9997. https://doi.org/10.1073/pnas.1714450114
5. Liubartseva S, Coppini G, Lecci R, Clementi E (2018) Tracking plastics in the Mediterranean: 2D Lagrangian model. Mar Pollut Bull 129(2018):151–162. https://doi.org/10.1016/j.marpolbul.2018.02.019
6. Suaria G, Alliani S (2014) Floating debris in the Mediterranean Sea. Mar Pollut Bull 86:494–504. https://doi.org/10.1016/j.marpolbul.2014.06.025
7. Mistri M, Infantini V, Scoponi M, Granata T, Moruzzi L, Massara F, De Donati M, Munari C (2017) Small plastic debris in sediments from the Central Adriatic Sea: types, occurrence and distribution. Mar Pollut Bull 124(1):435–440. https://doi.org/10.1016/j.marpolbul.2017.07.063
8. Liubartseva S, Coppini G, Lecci R, Cretì S (2016) Regional approach to modeling the transport of floating plastic debris in the Adriatic Sea. Mar Pollut Bull 103:115–127. https://doi.org/10.1016/j.marpolbul.2015.12.031
9. Environmental Crimes (2012) Marine pollution. Situation and legislation applicable in Albania. http://www.judt.unisalento.it/wp-content/uploads/2012/03/enviromental-crimes-albania11.pdf
10. Gjyli L, Mukli L (2010) Assessment of water microbiologic pollution in Durres's marine harbour basin, ALBANIA. J Life Sci U S A 4(4, 29):32–39. https://doi.org/10.17265/1934-7391/2010.06.007
11. Gjyli L, Bacu A, Kolitari J, Gjyli S (2013a) Dynamics of picophytoplankton and presence of cyanobacteria *Synechococcus* in coastal waters of Durrës Bay (Albania). Albanian J Agric Sci (AJAS) 12(4):585–592. https://sites.google.com/a/ubt.edu.al/rssb/
12. Gjyli L, Bacu A, Kolitari J, Gjyli S (2013b) Primarily results of phytoplankton DNA and variation to environmental factors in Durres's bay coastal waters (Albania). J Microbiol Biotechnol Food Sci (JMBFS) 3(2):132–136. http://www.jmbfs.org/
13. Gjyli L, Bacu A (2014) Possible correlation between the diversity of 16-23S rDNA-ITS diversity of *Synechococcus* populations and quality of the waters at Durres Bay. J Nat Tech Sci (JNTS) 19(1):77–90. http://www.akad.gov.al/ash/pdf/periodike/JNTS36-online.pdf
14. Gjyli L, Bacu A, Kolitari J, Gjyli S, Trifoni A (2016) Estimation of N/P ratios levels in a Coastal Bay, Southern Adriatic Sea. J Agric Ecol Res Int 8(1):1–9. www.sciencedomain.org. https://doi.org/10.9734/JAERI/2016/25052
15. Kolitari J, Gjyli L, Carbonara P (2016) Preliminary results and impact of marine litter in Albanian Adriatic area. J Environ Prot Ecol 17(3):922–931
16. Gjyli L, Vlachogianni T, Kolitari J, Matta G, Metalla O, Gjyli S (2020) Marine litter on the Albanian coastline: baseline information for improved management. Ocean Coast Manage 187 (2020):105108. https://doi.org/10.1016/j.ocecoaman.2020.105108
17. UNEP/MAP IG.21/9. Regional plan on marine litter management in the Mediterranean in the framework of article 15 of the Land based sources protocol. UNEP(DEPI)/MED IG.21/9, ANNEX II – Thematic Decisions, pp 143–173
18. Decentralisation and Local Development Programme (dldp) (2015). http://cdinstitute.eu/web/wp-content/uploads/2015/10/FAP-DURRES.pdf
19. Pano N (2008) Pasuritë ujore të Shqipërisë (in German). Akademia e Shkencave e Shqipërisë, Tirana
20. Miho A, Kashta L, Beqiraj S (2013) Between the Land and the Sea – Ecoguide to discover the transitional waters of Albania. Study of University of Tirana, 1–462
21. Semeoshenkova V, Newton A, Contin A, Greggio N (2017) Development and application of an Integrated Beach quality index (BQI). Ocean Coast Manage 143:74–86
22. Galgani F, Hanke G, Werner S, Oosterbaan L, Nilsson P, Fleet D, Kinsey S, Thompson RC, van Franeker J, Vlachogianni TH, Scoullos M, Veiga JM, Palatinus A, Matiddi M, Maes T, Korpinen S, Budziak A, Leslie H, Gago J, Liebezeit G (2013) Guidance on monitoring of marine litter in European seas. Scientific and technical research series. Publications Office of the European Union, Luxembourg

23. Lippiatt S, Opfer S, Arthur C (2013) Marine debris monitoring and assessment. NOAA technical memorandum NOS-OR&R-46, 2013
24. Alkalay R, Pasternak G, Zask A (2007) Clean-coast index—a new approach for beach cleanliness assessment. Ocean Coast Manage 50(5):352–362
25. UNEP (2018) United Nations Environment Programme, 2018. Single-use plastics: a roadmap for sustainability
26. Vlachogianni T, Fortibuoni T, Ronchi F, Zeri C, Mazziotti C, Tutman P, BojanićVarezić D, Palatinus A, Trdan Š, Peterlin M, Mandić M, Markovic O, Prvan M, Kaberi H, Prevenios M, Kolitari J, Kroqi G, Fusco M, Kalampokis E, Scoullos M (2018) Marine litter on the beaches of the Adriatic and Ionian seas: an assessment of their abundance, composition and sources. Mar Pollut Bull 131(A):745–756
27. Munari C, Corbau C, Simeoni U, Mistri M (2016) Marine litter on Mediterranean shores: analysis of composition, spatial distribution and sources in north-western Adriatic beaches. Waste Manag 49:483–490
28. Vlachogianni Th (2019) Marine litter in Mediterranean coastal and marine protected areas – How bad is it. A snapshot assessment report on the amounts, composition and sources of marine litter found on beaches, Interreg Med ACT4LITTER & MIO-ECSDE
29. Giovacchini A, Merlino S, Locritani M, Stroobant M (2018) Spatial distribution of marine litter along Italian coastal areas in the Pelagos sanctuary (Ligurian Sea – NW Mediterranean Sea): a focus on natural and urban beaches. Mar Pollut Bull 130:140–152. https://doi.org/10.1016/j.marpolbul.2018.02.042
30. Topçu EN, Tonay AM, Dede A, Öztürk AA, Öztürk B (2013) Origin and abundance of marine litter along sandy beaches of the Turkish Western Black Sea coast. Mar Environ Res 85:21–28. https://doi.org/10.1016/j.marenvres.2012.12.006
31. Laglbauer BJL, Melo Franco-Santos R, Andreu-Cazenave M, Brunelli L, Papadatou M, Palatinus A, Grego M, Deprez T (2014) Macrodebris and microplastics from beaches in Slovenia. Mar Pollut Bull 89:356–366
32. Gaibor N, Condo-Espinel V, Cornejo-Rodríguez M, Darquea J, Pernia B, Domínguez G, Briz M, Márquez L, Laaz E, Alemán-Dyer C, Avendaño U, Guerrero J, Preciado M, Honorato-Zimmer D, Thiel M (2020) Composition, abundance and sources of anthropogenic marine debris on the beaches from Ecuador – a volunteer-supported study. Mar Pollut Bull 2020:154. https://doi.org/10.1016/j.marpolbul.2020.111068
33. Jayasiri HB, Purushothaman CS, Vennila A (2013) Quantitative analysis of plastic debris on recreational beaches in Mumbai, India. Mar Pollut Bull 77:107–112. https://doi.org/10.1016/j.marpolbul.2013.10.024
34. Asensio-Montesinos F, Anfuso G, Randerson P, Williams AT (2019) Seasonal comparison of beach litter on Mediterranean coastal sites (Alicante, SE Spain). Ocean Coast Manag 181:104914. https://doi.org/10.1016/j.ocecoaman.2019.104914
35. Schernewski G, Balciunas A, Gräwe D, Gräwe U, Klesse K, Schulz M, Wesnigk S, Fleet D, Haseler M, Möllman N, Werner S (2018) Beach macro-litter monitoring on southern Baltic beaches, results, experiences and recommendations. J Coast Conserv 22:5–25. https://doi.org/10.1007/s11852-016-0489-x
36. Kusui T, Noda M (2003) International survey on the distribution of stranded and buried litter on beaches along the sea of Japan. Mar Pollut Bull 47:175–179. https://doi.org/10.1016/S0025-326X(02)00478-2
37. Pasternak G, Zviely D, Ribic CA, Ariel A, Spanier E (2017) Sources, composition and spatial distribution of marine debris along the Mediterranean coast of Israel. Mar Pollut Bull 114:1036–1045. https://doi.org/10.1016/j.marpolbul.2016.11.023
38. Poeta G, Battisti C, Acosta ATR (2014) Marine litter in Mediterranean sandy littorals: spatial distribution patterns along Central Italy coastal dunes. Mar Pollut Bull 89:168–173. https://doi.org/10.1016/j.marpolbul.2014.10.011
39. Addamo AM, Laroche P, Hanke G (2017) Top Marine Beach Litter Items in Europe. EUR 29249 EN, Publications Office of the European Union, Luxembourg

40. DIRECTIVE (EU) 2019/904. DIRECTIVE (EU) 2019/904 OF THE EUROPEAN PARLIAMENT AND OF THE COUNCIL of 5 June 2019 on the reduction of the impact of certain plastic products on the environment. Official Journal of the European Union. L 155/1-15. 12.6.2019. https://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32019L0904
41. Kurtela A, Antolović N (2019) The problem of plastic waste and microplastics in the seas and oceans: impact on marine organisms. Croatian J Fish 77:51–56. https://doi.org/10.2478/cjf-2019-0005
42. Ocean Conservacy (2010) A rising tide of ocean debris. 2009 report. Washington, 64 p
43. PNUE/PAM/MEDPOL (2009) Results of the assessment of the status of marine litter in the Mediterranean. Meeting of MED POL Focal Points No. 334, 91 p
44. Karaboyaci M, Elbek GG, Kilic M, Sencan A (2017) Process design for the recycling of tetra pak components. Eur J Eng Nat Sci 2(1):126–129
45. Zawadiak J, Wojciechowski S, Piotrowski T, Krypa A (2017) Tetra pak recycling – current trends and new developments. Am J Chem Eng 5(3):37–42. https://doi.org/10.11648/j.ajche.20170503.12
46. Moving Away from Single-Use (2019) Guide for national decision makers to implement the single-use plastics directive. https://rethinkplasticalliance.eu/wp-content/uploads/2019/10/2019_10_10_rpa_bffp_sup_guide.pdf
47. Decision on Packaging and Waste, Nr. 232/2018. Vendim Nr. 232/2018, datë 26.04.2018. Për disa ndryshime dhe shtesa në vendimin Nr. 177, datë 06.03.2012, të Këshillit të Ministrave, “Për ambalazhet dhe mbetjet e tyre”. http://www.mjedisi.gov.al/menaxhimi-i-mbetjeve/
48. Plumm H (2017) Climate-friendly integrated solid waste management and circular economy in Albania. Deutsche Gesellschaft für Internationale Zusammenarbeit (GIZ) GmbH
49. EEA (2015) The European environment — state and outlook 2015: an integrated assessment of the European environment. European Environment Agency
50. Cullaj A, Hasko A, Miho A, Achanzd F, Brandle H, Bachofenf R (2005) The quality of Albanian natural waters and the human impact. Review article. Environ Int 31 (2005):133–146. https://doi.org/10.1016/j.envint.2004.06.008
51. ARCADIS ECOLAS/IEEP/Metroeconomica/Enviro-L Content 06/11347 (2007) Albania – Benefits of Compliance with environmental acquis – final report. https://ec.europa.eu/environment/archives/international_issues/pdf/report_albania.pdf
52. Baumann L (2019) Feasibility study: stopping the pollution of Ishmi River Basin in Albania. Feasibility study. Technical University of Berlin. https://doi.org/10.13140/RG.2.2.29953.61281

Plastic Pollution in East Asia: Macroplastics and Microplastics in the Aquatic Environment and Mitigation Efforts by Various Actors

Bruno Andreas Walther, Takashi Kusui, Ning Yen, Chieh-Shen Hu, and Hyemi Lee

Contents

1 Introduction 354
2 Methods 355
3 Results 357
  3.1 China 357
  3.2 Japan 365
  3.3 South Korea 371
  3.4 Taiwan 380
4 Discussion and Conclusion 386
References 389

Abstract Plastic pollution has become an increasingly worrying threat to the aquatic environment. The oceans and seas in East Asia are among the world's most polluted. Therefore, East Asian societies should make concerted efforts to

---

B. A. Walther (✉)
Department of Biological Sciences, National Sun Yat-sen University, Gushan District, Kaohsiung City, Taiwan
e-mail: bawalther2009@gmail.com

T. Kusui
Toyama Prefectural University, Toyama, Japan
e-mail: kusui@pu-toyama.ac.jp

N. Yen and C.-S. Hu
IndigoWaters Institute, Kaohsiung City, Taiwan
e-mail: ning@indigowaters.org; jason@indigowaters.org

H. Lee
Medipeace, Guro-gu, Seoul, Republic of Korea
e-mail: annieleehyemi@gmail.com

Friederike Stock, Georg Reifferscheid, Nicole Brennholt, and Evgeniia Kostianaia (eds.),
Plastics in the Aquatic Environment - Part I: Current Status and Challenges,
Hdb Env Chem (2022) 111: 353–404, DOI 10.1007/698_2020_508,
© Springer Nature Switzerland AG 2020, Published online: 16 April 2020

tackle the problem. In this review, we summarize the current state of scientific research about macro- and microplastic contamination of the aquatic environment, including biota, consecutively for four East Asian countries (China, Japan, South Korea, and Taiwan). For the same four countries, we also summarize mitigation efforts to decrease the plastic pollution in these four countries, which includes government policies and waste management; education, media, monitoring, and outreach campaigns by NGOs; and inventors and businesses developing alternative products and methods of production and recycling. This review aims to give an overview which will hopefully inspire a more concerted effort by East Asian governments to support the relevant science but also to tackle the plastic pollution problem with much needed policies and management solutions.

Keywords Coastal pollution, East Asian seas, Microplastic contamination of food, Plastic pollution, Plastic waste management, Recycling

1 Introduction

Plastic pollution is a rapidly worsening environmental problem in terrestrial habitats [1] but even more in aquatic habitats such as freshwater, coastal, and oceanic ones [2–6]. Since global plastic production and waste generation have been growing exponentially, with production at approximately 335 million metric tons (MT) in 2016 [7–9], plastic pollution will continue to worsen unless emissions are seriously curtailed. Between 4.8 and 12.7 million MT of plastics are estimated to enter the oceans annually [10], while the remainder is either recycled, incinerated, or landfilled or enters other ecosystems [7].

Once in the environment, plastic objects and fragments (1) damage and endanger ships; (2) cause the injury and death of animals through entanglement and ingestion; (3) visually and structurally damage oceanic, coastal, and freshwater ecosystems; (4) spread invasive species and diseases; and (5) degrade to meso-, micro-, and nanoplastic particles which can either enter the food chain directly or contaminate it via chemical leaching [2, 5, 11–16]. Possible human health impacts are (1) accidents; (2) the direct ingestion of microplastics and the possible resulting internal injury [17, 18]; (3) the indirect contamination of air, food, and water with unhealthy chemicals [1, 19]; and (4) microplastics serving as pathogen vectors [20].

Concerns about plastic pollution should be especially relevant to East Asian societies because man-made debris pollution made up predominantly by plastic materials has reached pervasive and catastrophic proportions in East Asian rivers, oceans, and coastlines, with some of the world's highest levels of plastic pollution reported [10, 21–29]. Moreover, a relatively high proportion of people's diet comes from seafood [29–31].

Therefore, East Asian societies should make concerted efforts to tackle the growing plastic pollution. While cleanup efforts certainly help alleviate the problem at least locally, given the scale and speed of the problem, any serious solutions to decrease and finally eliminate plastic emissions into the environment must (1) introduce source reduction policies (including bans, charges, deposits, fees, fines, incentives, penalties, refunds, and taxes); (2) improve waste management and recycling with the ultimate goal of a completely circular materials economy, education, and behavioral change [32–47]; (3) replace plastic packaging with biodegradable materials (e.g., [48, 49]); and (4) clean up affected areas such as lakes, rivers, beaches, and the oceans themselves [29, 50, 51]. Furthermore, new or improved international and national instruments and treaties including effective enforcement are needed [35].

From our review, it is obvious that such much-needed solutions are only beginning to be implemented. What is encouraging is that research of the problem of plastic pollution as well as possible solutions has been increasing in recent years. In order to survey the problem of macro- and microplastic pollution in the aquatic environments of East Asia as well as mitigation efforts by various actors, we reviewed the available scientific literature of four countries (namely, China, Japan, South Korea, and Taiwan).

2 Methods

This study is a literature review of macro- and microplastic pollution research of the aquatic environments of East Asia and the ensuing mitigation efforts by various actors and stakeholders. For ease of communication, we use the shorthand names for the following countries: China for the People's Republic of China, North Korea for the Democratic People's Republic of Korea, South Korea for the Republic of Korea, and Taiwan for the Republic of China. Of the six East Asian countries, we a priori excluded two, namely, Mongolia and North Korea, because of the scarcity of any relevant research. Furthermore, Mongolia is a landlocked country, which thus precludes research on coastal and oceanic pollution.

All the authors have worked on this topic for several years. Therefore, a lot of the literature which we base this review on was already known to us, and some more was supplied by other experts in the field (see Acknowledgements). Additionally, we performed a standard literature search for English-language sources by searching Google Scholar and Web of Science using appropriate keywords or keyword combinations (e.g., "macroplastic," "microplastic," "plastic pollution," "recycling" in various combinations with the country names China, Japan, Korea, and Taiwan). The literature searches and writing process lasted from July to October 2019.

Currencies are given in United States dollars (USD), Japanese Yen (1,000 JPY = 9.21 USD on 1 October 2019), South Korean won (1,000 won = 0.84 USD on 1 October 2019), and New Taiwan dollars (1,000 NTD = 32.68 USD on 1 October 2019). All abbreviations are given in Table 1. Taiwan's Environmental

Table 1 Abbreviations used in main text

Full name	Abbreviation
Asia-Pacific Economic Cooperation	APEC
Coordinating Body on the Seas of East Asia	COBSEA
Commonwealth Scientific and Industrial Research Organisation, Australia	CSIRO
Environmental non-governmental organization	ENGO
Environmental Protection Law (China)	EPL
Expanded polystyrene (colloquially called “Styrofoam” in Canada and the USA)	EPS
Extended Producer Responsibility	EPR
Fourier transform infrared spectroscopy	FTIR
Greenpeace	GP
International Coastal Cleanup	ICC
International Convention for the Prevention of Pollution from Ships	MARPOL
International Convention on Civil Liability for Oil Pollution Damage	CLC
International Convention on Oil Pollution Preparedness, Response and Co-operation	OPRC
Japan Environmental Action Network	JEAN
Japanese Ministry of the Environment	MOE
Japanese Yen	JPY
Korea Marine Environment Management Corporation	KOEM
Korean Women’s Environmental Network	KWEN
Kuroshio Ocean Education Foundation	KOEF
Metric ton	MT
Ministry of Ecology and Environment, China	MEE
Ministry of Environment, South Korea	ME
Ministry of Ocean and Fisheries, South Korea	MOF
New Taiwan dollars	NTD
Non-governmental organization	NGO
Northwest Pacific Action Plan	NOWPAP
Northwest Pacific Region Environmental Cooperation Center	NPEC
Our Sea of East Asia Network	OSEAN
Polyester	PES
Polyethylene	PE
Polyethylene terephthalate	PET
Polypropylene	PP
Polystyrene	PS
Society of Wilderness	SOW
State Oceanic Administration, China	SOA
Taiwan Environmental Information Association	TEIA
Taiwan’s Environmental Protection Administration	TEPA
United Nations Environment Programme	UNEP
United States of America	USA
United States dollars	USD
Wild at Heart Legal Defense Association	WHLDA
World Wide Fund for Nature (formerly World Wildlife Fund)	WWF

Protection Administration (TEPA) defines single-use as those products which are produced for single-use and almost always disposed after one use. The term "disposable" is also often used for these kinds of products, but we use the term "single-use" throughout this manuscript because of the TEPA's definition.

3 Results

3.1 China

3.1.1 Macro- and Microplastic Contamination of the Aquatic Environment

Chinese scientists began research on microplastics in 2013, covering topics such as the microplastic abundance of various habitats, analytical methods of microplastic detection and estimation, ecotoxicology, ecological risk assessment, microbial degradation, and pollution control and management of plastics and microplastics.

Microplastic pollution of China's inland water systems was recently reviewed by Wu et al. [52] and Zhang et al. [53], while Wang et al. [24] reviewed research and management of plastic pollution in China's coastal environments. We therefore relied on these two reviews (and references therein) for some parts of our review but also attempted to add to it by reviewing publications and other information not included in them.

As Wang et al. [24] emphasized, China is the world's biggest consumer of plastic products and biggest contributor of plastic waste; consequently, most of China's aquatic environments suffer from plastic pollution at various levels, but often catastrophic ones. For example, 81% of China's coastal regions are heavily polluted with plastic debris, damaging ecosystems but also local economies because of lower real estate and tourism value, continuous cleanup costs, and damage to ships and business sectors which use the polluted water.

Wang et al. [24] reviewed 30 studies on plastic pollution in China's coastal environment which had found plastic debris (both macro- and microplastic) in a wide variety of environments: in surface waters and underwater sediments of estuaries, mudflats, rivers, and seas and on the surface as well as in the sediments of beaches (see also [53–68]). Other studies have found microplastics in coral reefs [69–71], dams [53], deep-sea submarine canyons [72, 73], lakes [53, 74–79], mangroves [80, 81], reservoirs [53, 82], and rice-fish co-culture systems [83]. Plastics were also recovered from zooplankton, sea cucumbers, bivalves, clams, mussels, oysters, fishes, Asian finless porpoises, and birds [24, 31, 53, 84–90] as well as table salts [91]. The most common polymer types were cellophane, polyester (PES), polyethylene (PE), polyethylene terephthalate (PET), polypropylene (PP), and polystyrene (PS) [24, 53].

Sources of plastic pollution are the usual suspects: mismanaged waste disposal and fishing gear, tourism-related activities, construction sites, agriculture, manufacturing, wastewater treatment plants, laundry effluent, primary microplastics from personal care products and resin pellets, rubber tire abrasion, etc. [24, 53, 92]. For example, it was estimated that about 39 MT of primary microplastics are released annually into the Chinese environment from shower gel products alone [93], and Cheung and Fok [94] estimated that 209.7 trillion microbeads (or 306.8 MT) were annually released into China's environment. Bai et al. [95] estimated the annual input of plastic waste into the sea from China in the 2010s. In 2011, 0.5–0.8 million MT of plastic waste entered the seas in China, with an annual growth rate of 4.6% until 2017. Wang et al. [96] reviewed and estimated the emissions of primary microplastics in China.

Many microplastics are contaminated with hydrophobic organic compounds [97], other persistent organic pollutants, phthalates, plasticizers, and trace metals [24, 98]. Microplastics in seawater also accumulate microbial communities which appear to facilitate degradation of the microplastics [99].

In addition to the rapidly increasing scientific activity on the plastic pollution issue, the Chinese government has also begun monitoring and research activities. In 2007, the Chinese government began to monitor marine debris at about 50 coastal sites which include agricultural and fishery areas, tourist spots, and ports [100, 101]. In the 2017 China Marine Environmental Quality Bulletin, which is the annual report of the State Oceanic Administration of the People's Republic of China [102], the density of drifting debris, coastal debris, and sea bottom debris was estimated to be 2,845, 52,123, and 1,434 pieces per km^2, respectively. Among the eight categories of debris (namely, plastic, metal, rubber, glass, cloth, paper, wood, and others), plastic items were predominant (74–87%).

In 2014, the Chinese Ministry of Science and Technology launched research on microplastics with the aim to reveal the impact of microplastics pollution on marine ecosystems; established national standards of analysis and monitoring methods, ecological risk assessment; and began research on the sources and control of the pollution and other key technologies [24]. In 2017, the Marine Debris and Microplastics Research Center was established under the National Marine Environmental Monitoring Center to focus on technologies, methods, and management strategies for pollution prevention and control of marine debris and microplastics. In the same year, the State Oceanic Administration (SOA) sampled microplastics along six transects, each on four offshore seas and six beaches. The most prevalent types of drifting microplastic were pellets, fibers, and fragments, and the predominant polymers were PP and PS. The most prevalent types on beaches were pellets, fibers, and lines, while the predominant polymers were also PP and PS [102]. According to officials from the Ministry of Ecology and Environment (MEE), the microplastic pollution in Chinese ocean waters was lower than the global average and similar to the levels detected in the central western Mediterranean Seas and around the Seto Inland Sea of Japan [103].

3.1.2 Mitigation Efforts to Decrease the Plastic Pollution

Since China is associated with some of the world's most polluted oceans and is itself one of the worst polluters, its efforts of combating marine debris need to be addressed. China has been participating in two regional frameworks which have been developed under the United Nations Environment Programme and one partnership which was developed under the United Nations Development Programme (Table 2).

The first regional framework is the Coordinating Body on the Seas of East Asia (COBSEA), a regional intergovernmental policy forum, with China and eight Southeast Asian countries as participating members. Aiming to protect marine and coastal environments, the Action Plan for the Protection and Development of the Marine Environment and Coastal Areas of the East Asian Seas Region (the East Asian Seas Action Plan) was adopted in 1981 and revised in 1994. In recent meetings in 2018 and 2019, the revised COBSEA Regional Action Plan on Marine Litter outlined efforts in the East Asian region to tackle marine litter. It also supports regional organizations, e.g., ASEAN, and addresses global priorities such as Sustainable Development Goal 14 identified by the UN Environment Assembly.

The second regional framework is the Action Plan for the Protection, Management and Development of the Marine and Coastal Environment of the Northwest Pacific Region (NOWPAP) which was adopted in 1994 in order to protect the marine environment from land-based activities in the Northwest Pacific Region. Since 2005, NOWPAP has responded to the growing threat of marine debris in the Northwest Pacific Region through regional cooperation on scientific research and annual discussion in the Tripartite Environment Ministers Meeting. One tangible outcome is the NOWPAP Regional Action Plan on Marine Litter (RAP MALI) [104]. The prevention of marine litter input into marine and coastal environments has been identified as one of the key elements in the next phase of the RAP MALI [104].

Table 2 List of regional frameworks which deal with plastic pollution

Abbreviated name	Founding year	Member states
APEC	1989	Australia, Brunei, Canada, Chile, China, Hong Kong, Indonesia, Japan, Malaysia, Mexico, New Zealand, Papua New Guinea, Peru, Philippines, Russia, Singapore, South Korea, Taiwan, Thailand, USA, Vietnam
COBSEA	1993	Cambodia, China, Indonesia, Malaysia, Philippines, Singapore, South Korea, Thailand, Vietnam
PEMSEA	1993	Cambodia, China, Indonesia, Japan, Laos, North Korea, Philippines, Singapore, South Korea, Timor-Leste, Vietnam
NOWPAP	1994	China, Japan, Russia, South Korea
ASEAN Plus Three (APT)	1997	10 ASEAN members include Brunei, Cambodia, Indonesia, Laos, Malaysia, Myanmar, Philippines, Singapore, Thailand, and Vietnam, plus 3 cooperation entities: China, Japan, and South Korea

See text for more details

Developed under the United Nations Development Programme, the Partnerships in Environmental Management for the Seas of East Asia (PEMSEA) is an intergovernmental organization operating in East Asia to foster and sustain healthy and resilient oceans, coasts, communities, and economies across the region. PEMSEA's partners include 11 countries, NGOs, scientific institutions, industry, and regional programs. In its implementation plan for 2018–2022, pollution reduction and waste management, including the reduction of marine debris and plastics among PEMSEA countries, is clearly outlined as one of the priorities (p. 14 in PEMSEA [105]).

Additionally, two regional intergovernmental and economic cooperation bodies have also contributed and addressed (1) the management of land and sea-based waste and (2) the study of the impact from marine debris. The Asia-Pacific Economic Cooperation (APEC) recognized the threat from marine debris in 2005 [106]. McIlgorm et al. [107] estimated that marine debris has a direct cost of approximately 1.265 billion USD to the 21 Asia-APEC member economies. Under the APEC framework, there are regular meetings of the Oceans and Fishery Working Group, Oceans Ministerial Meetings, seminars, and roundtable meetings between government officials, academics, NGOs, and industry experts. Furthermore, the Association of Southeast Asian Nations (ASEAN) Plus Three (APT) has offered funding to support several environment fora to enhance the awareness of the issue of marine debris and its impacts [108].

Through bilateral or multilateral agreements and cooperation actions, marine debris has become one of the topics which the Chinese government has embraced in order to work with other countries. Chinese President Xi Jinping visited the United States of America (USA) for the 2015 China-US Strategic and Economic Dialogue, and one of the outcomes was to bring together "Sister Cities" for the prevention and control of marine debris. The first two pairs of "Sister Cities" are New York and Weihai and San Francisco and Xiamen which formed partnerships to implement measures to promote waste collection, management, reuse to reduce, and prevention of mismanaged waste entering the ocean. Another example is the Canada-China Joint Statement on Marine Litter and Plastics in which both countries agreed to forge a partnership to combat marine litter [109]. Through the Joint Statement, both sides acknowledged that plastic pollution resulting from current practices has negative impacts on ocean health, biodiversity, economic sustainability, and potentially human health. Moreover, both leaders recognized the importance of embracing a sustainable lifecycle approach to the management of plastics in order to reduce marine debris.

Laws, regulations, and policies to control marine debris have increased in number over the years (Table 3). The very first Chinese law which dealt with waste management and mitigation of marine debris was implemented as Environmental Protection Law (EPL) in 1989; further waste management laws and regulations were introduced in the 1990s. Plastic waste is considered a type of solid waste and should therefore be managed in accordance with China's solid waste-related legislation under the EPL. Zhang [53] listed further regulations implemented through several amendments of the EPL until 2015. However, one of the crucial measures to curb marine plastic pollution, namely, legislation which regulates source reduction and effective waste recycling, remains weak in China. Compared

Table 3 China's laws, regulations, and policies on plastic waste and marine pollution (information taken from Zhang et al. [53] and Wang et al. [112] and other sources)

Name of law, regulation, or policy	Year issued	Authorized departments
Regulations on the control over dumping of wastes in the ocean	1985	State Council of the People's Republic of China
Environmental Protection Law	1989	Ministry of Environmental Protection
Regulations on the prevention of pollution damage to the marine environment by land-based pollutants	1990	State Council of the People's Republic of China
Implementation measures of regulations on the control over dumping of wastes in the ocean	1990	SOA
Law on the prevention and control of environmental pollution by solid waste	1995	State Council of the People's Republic of China
Marine Environmental Protection Law	1999	State Council of the People's Republic of China
Law on Promoting Clean Production	2003 (revised in 2012)	MEE
Interim provisions on dumping sites management	2007	SOA
Technical specifications on pollution control of plastic waste collection and recycling (trial)	2007	MEE
Law on circular economy promotion	2009 (revised in 2018)	MEE
Measures on the administration of imports of solid waste	2011	MEE, National Development and Reform Commission (NDRC), Ministry of Commerce of the People's Republic of China (MOFCOM), General Administration of Customs of the People's Republic of China (GACC), General Administration of Quality Supervision, Inspection and Quarantine of the People's Republic of China (AQSIQ)
Administrative regulations on the pollution control of plastic waste recycling	2012	MEE
Technical regulations for the monitoring and evaluation of marine litter	2015	SOA
Standard and administrative interim measures on industrial conditions of the comprehensive utilization of plastic waste	2015	MIIT
Latest revision of the law on the prevention and control of environmental pollution by solid waste	2016	MEE
Catalogues of solid waste import management	2017	MEE, MOFCOM, NDRC, GACC, AQSIQ

to other countries who have introduced regulations and bans of plastic bags, primary microbeads in cosmetics, plastic straws, and other single-use items, China has only introduced one plastic bag limitation measure in 2008 [110]. In 2009, the National Development and Reform Commission estimated that supermarkets had reduced plastic bag usage by 66%. However, no reduction of plastic bags in the marine litter was detected, so the ban's effectiveness is doubtful [111]. Furthermore, disposable expanded polystyrene (EPS) food service products were banned in 1999, but the ban was never enforced and then rescinded in 2013 [32].

Since China is a contracting party of relevant international conventions of marine pollution control, the Chinese government has been working to improve national laws, regulations, and policies in order to fulfill the obligations of these conventions (summarized in Wang et al. [112] and CCICED [113]). The Marine Environmental Protection Law issued in 1982 was revised several times in order to incorporate relevant regulations from international pollution conventions, including the Protocol of 1978 Relating to the International Convention for the Prevention of Pollution from Ships 1973 (MARPOL 73/78), the International Convention on Oil Pollution Preparedness, Response and Co-operation (OPRC), and the International Convention on Civil Liability for Oil Pollution Damage (CLC) [114]. This law is China's basic law for the protection of the marine environment, and it provides an overarching framework for pollution mitigation, ecosystem protection, and resource conservation. However, its regulations mainly focus on oil spills and chemical pollution but not on plastic pollution of the marine environment.

Mismanaged plastic waste released into the environment is the main source of marine plastic pollution to the oceans (see Introduction). One effective solution is recycling of plastic waste, but the relevant recycling processes and practices in China developed only slowly in the past two decades. The 1989 EPL already included the concept of waste reduction, recycling, and waste management of household waste [114]. However, local government officials usually care more about economic growth and have lackadaisical attitudes toward supervision and enforcement. Relevant regulation or initiatives on waste management and recycling were introduced slowly and met with a lot of problems and resistance by the public. Furthermore, the limited input of civil society due to the authoritarian structure of the Chinese government means that pressure to enforce environmental laws and regulations is weak and ENGOs, journalists, lawyers, and ordinary citizens are all tightly constrained in what they can do to protect the environment [115, 116].

Since 2000, several local initiatives have tried to improve the separation of recyclable materials from household waste, mostly in big cities such as Beijing, Guangzhou, Nanjing, and Shanghai [117–121]. However, due to the lack of public awareness and weak enforcement and supervision, several researchers and media outlets emphasized that the implementation is not effective [120, 122–126]. Not until 2015 did two critical framework guidelines [127, 128] address the effective implementation of the sorting of household waste and separation of recyclables. Subsequently, the 13th Five-Year Plan [127] and the Implementation Plan of the Household Waste Sorting System [129] clearly outlined the relevant regulations and the sorting system for household waste in 46 key cities. Shanghai was the first city to implement them. The remaining 45 key cities have adopted or are planning action and implementation plans for household waste sorting in the near future [130]. These regulations and actions by the Chinese government sped up the adoption of household waste recycling in these key cities in 2019, with a goal of a recycling rate of 35% by the end of 2020.

As mentioned above, the Chinese government regulated the sale of certain plastic bags in 2008; for instance, since 1 June 2008, the production, sale, and use of plastic bags with a thickness of <0.025 mm is banned [53]. However, there is a great lack of inspection and enforcement. Department stores, retailers, and supermarkets should not offer free plastic bags [130], but many plastic bags are still given out in the food and business industries as well as in private shops and markets [24]. Therefore, some media criticized that this regulation was not effective and that the usage of plastic bags has even boomed after the regulation [131]. The ENGO "China Zero Waste Alliance" also reported that this regulation was not effective when it surveyed 1,101 retailer shops in nine cities throughout China [132].

Since 1992, China has imported a large amount of the world's plastic waste [133]. Since plastic pollution and its impact on oceanic environments have since been widely reported, China's national government and local governments have also been eager to solve the problems of waste recycling [112]. In 2013, the Chinese government launched a 10-month intensive inspection named the "Green Fence Operation" to enforce their import regulations, to crack down on "foreign garbage" smuggling activities, and to seize illegal waste [134]. In 2017, the Chinese government announced its National Sword program in order to crack down on the illegal smuggling of foreign waste into China, especially targeting electronic scrap, industrial waste, and plastics [135] and permanently banned the import of nonindustrial plastic waste [53, 133]. With a short notice to the World Trade Organization, the National Sword program banned the import of 24 types of waste materials from 2018 onward [136]. In 2019, the Chinese government has continued to tighten the regulations on imported waste to more types [137] which caused repercussions around the world [133, 138].

To summarize, Chinese national legislation and policies related to plastics waste were lacking prior to 2008. To respond to the emerging serious environmental problems, the Chinese government has sped up its responses during the last decade. However, public awareness in Chinese society of the impact of plastic pollution remains in its infancy. As Zhang et al. [53] emphasized: "Although many laws and regulations already exist regarding the management and control of plastic waste in China, the implementation of these laws and regulations has been largely ineffective and sometimes difficult." Therefore, there is an urgent need to improve laws, policies, regulations, standards, and enforcement for source control as well as for waste management as well as to better educate the public [112].

Local governments have also increased their efforts to control marine debris. To tackle the marine debris problem, China's coastal provinces and municipalities, including Dalian, Guangzhou, Shanghai, and Xiamen, have been proactively carrying out relevant work to control and dispose sea-based marine litter through an effective control and management system. For example, the Dalian municipal government has collaborated with private environmental protection organizations and has also reinforced the management of port sewage and ship garbage. Each year, they have received >6,000 MT of ship garbage from about 7,800 ships. The Fujian government introduced a special plan of marine environmental sanitation in its harbor city of Xiamen by setting up an offshore ship-based garbage collection system [139].

Furthermore, the Chinese government has recently outlined its determination to eliminate pollution in other kinds of water bodies. In the last few years, the Chinese Ministry of Water Resource and the State Oceanic Administration created the so-called leader systems to enforce the management and protection of rivers [140], bays and beaches [141], and lakes [142]. For instance, there are now about 760,000 so-called River Leaders at different administration levels, and even citizens can join as volunteers to conduct regular patrols along local rivers and lakes. Further actions, such as the inspection of solid waste, protection of riverbanks, removal of garbage, and cracking down on illegal sand dredging, have been implemented since 2017.

Finally, we very briefly reviewed some examples of education, outreach, and media. Founded in 2007, the Shanghai Rendu Ocean NGO Development Center (Rendu) is the biggest Chinese ENGO that focuses on marine debris. As part of its mission to clean the ocean, Rendu has mobilized more than 10,000 volunteers in over 200 beach cleanups over the past 12 years to collect about 26 MT of marine debris from Shanghai's coastline. Moreover, it has become the International Coastal Cleanup (ICC) coordinator in China from 2015 onward and has since invited local communities, NGOs, and private sector participants to join the ICC cleanups in September every year. Together with other NGOs, Rendu has also organized a bimonthly beach monitoring project at 25 coastal spots since 2015, with the findings released in annual reports [143]. One result is that four out of the top five most abundant debris items are made from plastic, the top one being plastic bags [144]. Furthermore, Rendu volunteers presented marine environmental education programs at four primary schools in Shanghai [145].

With regard to the impact from media, local filmmaker Wang Jiuliang and his documentaries "Beijing Besieged by Waste" (2011) and "Plastics China" (2017) were influential. After 4 years of investigation, Wang Jiuliang delivered a set of photographs and the documentary "Beijing Besieged by Waste" to reveal the landfill pollution in Beijing. Later, "Plastics China" traced how plastic waste from across the globe was transported to China. Although Wang achieved fame and various awards, the documentary was never screened in China, and reactions to the documentary soon disappeared from the Internet in China [146]. However, the impact of Wang's work continues to stir up Chinese policy-making behind the scene. For example, Wang later found out that most of the illegal dump sites exposed by him had been closed or turned into legal landfills when he revisited the sites [147]. Within a relatively short time period after Wang had received recognition and awards overseas, the Chinese government announced the import ban for solid waste (the National Sword program; see above). While no official statements confirm the consequences caused by Wang's documentaries, the fact that China has recently moved in the direction of better waste management and improved recycling suggests that his work had significant impacts behind the scenes.

3.2 Japan

3.2.1 Macro- and Microplastic Contamination of the Aquatic Environment

Japan is an island country which is comprised of a stratovolcanic archipelago along East Asia's Pacific coast. It is surrounded by eight ocean currents including the Kuroshio and Tsushima Currents which are part of the North Pacific Ocean gyre. In recent years, marine litter which drifts toward Japan owing to these ocean currents has become a major problem for Japan and a study subject for researchers, ENGOs, and government agencies.

The prominence of plastic materials within the floating marine debris was identified in the late 1990s and early 2000s from surveys of the waters around Japan [148–150] and stranded debris [151–155]. Furthermore, the Fisheries Agency of Japan conducted a Pacific-wide sighting survey of floating marine debris from 1986 through 1991 relevant to the North Pacific Ocean and its adjacent waters [150]. The total debris density in coastal waters was 20–40 objects per square nautical mile, whereas that in the north equatorial current area (5°–15° N, across the central Pacific) was approximately 0.2 objects and that in the subarctic boundary area 1–3 objects. The average marine debris composition was 10% non-petrochemical fishing gear, 60% total petrochemical (including fishing gear, Styrofoam, and other plastic debris), and 30% natural objects (e.g., logs and seaweeds).

Beach litter surveys have been conducted by various organizations in Japan. However, there are few quantitative survey data that can be compared. Since 1996, the Northwest Pacific Region Environmental Cooperation Center (NPEC) in Toyama, Japan, has conducted an international research project on marine litter with municipalities and ENGOs from China, Japan, South Korea, and Russia to comprehend the present situation of marine litter in the Northwest Pacific Region. Specifically, this survey was always carried out at the same time of the year using the same survey method along the coast of the Sea of Japan. To quantitatively evaluate the amount of stranded litter on the beach, 10 × 10 m survey units (100 m^2) were set continuously from the water's edge to the backshore zone of beaches. Generally, two or three lines of survey units were set parallel to the coastal line. In each survey unit, the litter was collected and sorted into categories (namely, plastics, rubber, Styrofoam, paper, cloth, glass/pottery, metals, and other artificial items). For the period from 1996 to 2017, the changes in the amount of stranded debris per unit area in 16 different survey sites located within 9 different locations along the Japanese coastline were investigated [156]. The mean weight of debris was 2,334.6 g per 100 m^2 during the study period (ranging from 1,236.9 g to 4,376.2 g per 100 m^2). "Plastics" made up an average of 62.4% of the total weight of all collected debris (ranging from 54.5 to 71.5%), followed by "other artificial items" at 16.9%. These trends were almost identical over all the years, strongly suggesting that there was no decrease in the amount of marine debris in the study area. Another unique characteristic of the NPEC survey is that it investigated buried litter including plastic debris in the coastal sand. The analytical method was developed by Ogi and Fukumoto [154] who had been concerned about the effects of microplastic on the marine ecosystem, based on the results of a stomach content survey of seabirds [157]. To collect the buried litter, 8 L of sand from a 40 × 40 × 5 (depth) cm space was collected using a box-shaped stainless steel frame (after removing visibly stranded litter on the sand) and placed into a bucket. The sand was then mixed with seawater and stirred, after which the supernatant was filtered with a net (0.3 mm mesh) to collect the floating plastic particles. The plastic particles were put into plastic bags and sent for sorting to the Toyama Prefectural University. The buried litter was identified, classified according to size (from less than 1 × 1 mm to over 10 × 10 mm), counted, and weighed after drying. The mean concentration of buried litter in Japan and Russia in 2000 was 9.03 and 2.70 g per m^2, respectively [155]. The stranded (or non-buried) litter was also quantified, with the mean concentration of stranded litter in Japan and Russia in 2000 being 21.44 and 13.44 g per m^2, respectively. The total weight ratio of buried litter to stranded litter averaged over all 26 sampled beaches was 0.65, indicating the significance of buried litter when evaluating the total amount of litter on beaches. Resin pellets were observed on 12 Japanese beaches, albeit on none of the Russian beaches (such pellets were also detected by Mato et al. [158], Endo et al. [159], and Ogata et al. [160] who measured their toxic chemical contents; see also [161]).

In addition to the above surveys, the ICC is conducted by the Japan Environmental Action Network (JEAN) in Japan. JEAN is a nonprofit ENGO, which works toward environmental preservation of the oceans and rivers by conducting marine litter investigations and cleanup activities. According to the 2017 survey results [162], three (hard plastic fragment, plastic sheet or bag fragment, and PS foam fragment) out of four categories which describe different kinds of fragments were in the top 10 most abundant categories by number (the three categories together added up to 29.1%). Considering the litter sources, the proportion of land-based litter (48.9%) was the highest; the second highest was ocean/river/lake-based litter (19.6%). Moreover, plastic products used for beverages, food, smoking products, etc. accounted for over 90% of the land-based debris. During the 27 years sampled from 1990 to 2017, the proportion of the top 10 items did not alter significantly.

In addition, several coastal surveys [163, 164] were conducted to study the sources and geographical distribution of beach debris. For example, the likely sources of marine debris that drifted onto Japanese beaches were studied using disposable lighters as indicators [163]. From August 2003 to May 2004, 6,609 lighters were collected from 120 beaches by nationwide beach combers. Chinese-made lighters accounted for over half of those collected in coastal areas from Yonaguni Island (Okinawa) in the south to Yaku Island (Kagoshima) in the north. Moreover, they accounted for approximately 10–20% of the lighters observed along the coast of the Sea of Japan from Kyushu in the south to Yamagata in the north. South Korean lighters accounted for about 10% in the coastal areas from Okinawa to west of Kyushu, but accounted for >50% of the lighters observed on the coast of the Sea of Japan from Shimane to Fukui which are geographically closer to South Korea. Japanese lighters accounted for >90% of all lighters along the coasts of the Seto Inland Sea and Tokyo Bay and at the Pacific coast north of Shikoku. Lighters from other countries were observed in the coastal areas of Guangdong and Zhejiang (in China) and across South Korea and Taiwan. By studying these lighters, various connections between discharge and flow could be identified.

The Japanese Ministry of the Environment (MOE) conducted a beach survey to establish measures to reduce and collect the marine debris [164]. Eleven coastal locations were selected from seven prefectures as model sampling areas. The coastal debris was surveyed for about a year from October 2007 to September 2008. Based on the country-wise survey of stranded debris using the language descriptions on plastic bottles as an index, many bottles found on remote islands, such as Iriomotejima (54% foreign, 6% Japanese, unidentified 40%), Ishigakijima (47% foreign, 8% Japanese, unidentified 45%), and Tsushima (62% foreign, 16% Japanese, unidentified 22%), likely originated from foreign countries. However, in other areas, the percentages of bottles from Japan were >50% to almost 100%. Based on the examination of the proportion (by weight) of different materials, plastics accounted for 30–40% on the Sea of Japan side. However, natural objects (driftwood and shrubs) accounted for 70–90% depending on the region. The above surveys revealed the following issues. On remote islands, disposal of collected marine debris may not be feasible owing to the inadequate capacity of incineration facilities. In addition, the disposal costs of collected debris were a significant financial burden on these municipalities. Based on the results of these surveys, a new law was enacted to subsequently combat marine debris (see Sect. 3.2.2).

As shown in the previous studies, the majority of marine debris was comprised of plastic, and many fragments were derived from these plastic products. As research on marine debris issues has progressed worldwide, the impact of smaller plastic fragments (or microplastics) on ecosystems has attracted increased attention. In particular, after microplastics were defined by Arthur et al. [165], microplastics in Japan were detected in the oceans [166], rivers [167], sediment [168], and fish [169, 170].

To investigate the concentrations of pelagic microplastics (<5 mm in size) and mesoplastics (>5 mm) in the East Asian seas around Japan, field surveys using two vessels were conducted in the summer 2014 [166]. The total particle count (pieces per km^2) was computed based on the observed concentrations (pieces per m^3) of small plastic fragments (both micro- and mesoplastics) collected with neuston nets. The total particle count of microplastics within the study area was 1,720,000 pieces per km^2, which was 16 times higher than documented for the North Pacific and 27 times higher than in the oceans worldwide. The proportion of mesoplastics increased upstream of the northeastward ocean currents; therefore, the small plastic fragments collected likely originated in the Yellow Sea and East China Sea southwest of the study area.

The distribution of microplastics in 18 Japanese rivers was investigated by Kudo et al. [167]. The magnitude of the density of microplastics in the rivers (0.0064–2.5 pieces per m^3) was an order of magnitude lower than that in the sea near Japan (0.6–4.2 pieces per m^3). With the decrease in size, the number of microplastics increased. The proportions of microplastics less than or equal to 1 mm and 2 mm accounted for about 50% and 80% of all microplastics collected, respectively. PE and PP accounted for over 70% of all microplastic particles.

Matsuguma et al. [168] extracted microplastics from sediment cores collected in Japan, Malaysia, Thailand, and South Africa and used density separation after hydrogen peroxide treatment to remove biofilms. The microplastics were identified using Fourier transform infrared spectroscopy (FTIR). Most of the microplastics were in the range 315 μm–1 mm. The abundance of microplastics in surface sediments varied from 100 pieces per kg of dry sediment in a core collected in the Gulf of Thailand to 1,900 pieces per kg of dry sediment in a core collected in a canal in Tokyo Bay. The significantly higher numbers of PE and PP microplastics found in sediment samples compared to those found in surface water samples collected in a canal in Tokyo Bay suggested that sediments are an important sink for microplastics. In dated sediment cores from Japan, microplastic pollution started in the 1950s, and microplastic numbers increased markedly toward the surface layer corresponding to the 2000s. In all sediment cores from Japan, Malaysia, Thailand, and South Africa, the abundance of microplastics increased toward the surface which is of course linked to the global increase of oceanic microplastic pollution over time.

Microplastics in the digestive tracts of Japanese anchovies (*Engraulis japonicus*) sampled in Tokyo Bay were detected in 49 out of 64 (77%) individuals, with 2.3 pieces on average and up to 15 pieces per individual [169]. Polymers identified by FTIR were again mostly PE (52.0%) and PP (43.3%). Most microplastics were fragments (86.0%), but 7.3% were beads, a few of which were microbeads similar to those found in facial cleansers (microbeads in coastal waters make up at least 10% of all microplastics; see [171]). 80% of the microplastics ranged in size from 150 to 1,000 μm, which is smaller than the reported size range of floating microplastics on the sea surface. The reason may be that the anchovy forages not near the sea surface, but in subsurface waters where microplastics may have a different size range. Since *Engraulis* spp. are an important food for many humans and other organisms, microplastics and their contaminants could thus enter the food chain.

Ushijima et al. [170] documented microplastics >100 μm in seven fish species from five Japanese bays and Lake Biwa. A total of 140 microplastic particles were observed in the digestive tracts of 37.6% of the investigated 197 fishes. All the species (except *Sardinella zunasi*) had ingested microplastics in all the sampled locations, and the mean number of microplastic particles was 1.89 ± 1.41 per fish. The most abundant polymer types were again PP (40.7%) and PE (35.0%). The median size of microplastic particles was 543 μm. The fish species were divided into filter feeders and others on the basis of their ingestion mode. 54.6% of 97 individuals of filter feeders had ingested microplastics, with the total number of particles being 112 and the mean number of microplastics per fish being 2.11 ± 1.54. In contrast, only 21.0% of 100 individuals of the other (non-filter feeding) group had ingested microplastics, with the total number of particles being 28 and the mean number of microplastics per fish being 1.33 ± 0.80. These differences indicated that the ingestion mode influences a fish's ingestion of microplastics.

Recent surveys indicated that microplastics are universally present in the aquatic environment of Japan, including seas, bays, rivers, sediments, and fishes. Moreover, the amount has undoubtedly increased in recent years. It is therefore pertinent to understand how the future will unfold.

The secular variations in the pelagic microplastic abundance in the Pacific Ocean from 1957 to 2066 were predicted based on a combination of numerical modeling and transoceanic surveys conducted meridionally from Antarctica to Japan [172]. The results of the numerical model incorporating removal processes on a 3-year timescale indicated that the weight concentrations of pelagic microplastics around the subtropical convergence zone would increase approximately twofold and fourfold by 2030 and 2060, respectively. Therefore, extensive and strenuous efforts to reduce plastic emissions are crucial in order to reduce the impact of plastic pollution in the future.

3.2.2 Mitigation Efforts to Decrease the Plastic Pollution

In the past, municipalities and voluntary groups mainly collected stranded debris on beaches, but little effort was made at source reduction. However, as mentioned in Sect. 3.2.1, the presence of marine litter including plastics became a pressing environmental issue for Japanese society. Therefore, the Act on Promoting the Treatment of Marine Debris Affecting the Conservation of Good Coastal Landscapes and Environments to Protect Natural Beauty and Variety was passed in 2009 [173]. Since then, the Japanese government has been working extensively on marine litter issues. The MOE has promoted the following activities: (1) collecting and preventing marine litter on Japanese coasts, (2) monitoring the amount and distribution of marine litter (including microplastics) and the toxic substances in it on Japanese coasts and in the seas around Japan, and (3) collaborating internationally with other Asian countries as well as global international frameworks to address marine litter. The Japanese government provided approximately 16 billion JPY of financial support to local governments from fiscal years 2009 to 2015, and approximately 190,000 MT of beach litter was collected and processed nationwide. From fiscal years 2016 to 2018, approximately three billion JPY were provided annually to support marine litter collection and treatment as well as generation control measures in each region. In June 2018, the Act was partially amended, and efforts to combat the problem of microplastics have now been included into it.

As described in Sect. 3.2.1, it was evident that the majority of marine debris collected around Japan was plastic. Furthermore, it was estimated that 20,000–60,000 MT of plastic waste was released from Japan into the ocean annually [10]. The amount of plastic containers and packaging consumed per capita in Japan is the second highest after that of the USA [7].

Meanwhile, the plastic waste generated in 2013 in Japan was 9.4 million MT, with a recycling rate of only 24.8% (material recycling and chemical recycling) and a heat recovery rate of 56.8%, yielding an effective utilization of 81.6% [174]. However, because some of the 24.8% recycling rate was achieved not by domestic recycling but by exporting the plastic waste, import bans in China (see Sect. 3.1.2) and other countries from 2018 onward compelled the establishment of a domestic resource recycling system.

It is evidently necessary to adopt comprehensive measures to address plastic production and pollution, including generation control. At the 2018 G7 Charlevoix Summit in Canada, the "Ocean Plastic Charter" was proposed. However, the Japanese government did not sign it because "Domestic laws had not been prepared" and was subsequently criticized by Japanese ENGOs [175]. One reason is the complete absence of any national bans on single-use plastics in Japan.

In 2019, the Japanese government formulated the "Resource Circulation Strategy for Plastics" as a comprehensive approach to plastics [176]. The fundamental principle of this strategy is "3R + Renewable" (through the implementation of 3R and replacement with renewable resources). It includes (1) reusing or recycling all used plastics by 2035; if this is challenging from technical and economic perspectives, then realize a 100% effective use which includes heat recovery through collaboration with various national parties, (2) doubling the recycling of plastic (use of recycled materials) by 2030, (3) substantially reducing microbeads in washing and scrubbing products by 2020, and (4) reusing or recycling 60% of plastic containers and packaging by 2030. The quantitative targets in this strategy are similar to those of the G7 "Ocean Plastic Charter."

In September 2019, the MOE set up a subcommittee in order to consider legislation for banning the free distribution of plastic bags so that they would need to be purchased instead in 2020 at the earliest [177]. On a more local level, 19 prefectures have been promoting payments for plastic bags through agreements or registration with business operators, and some municipalities are pushing for charges by ordinance.

In response to these movements, various efforts are being undertaken by industries and local governments. These include improvements in the recycling and reuse of plastic packaging materials for in-house products, non-use/suppression/reduction of one-way plastic products used in the organization, and use of paper/wooden straws rather than plastic straws. In the retail industry, shopping bags are being abolished (or charged), and/or usage is being reduced [174].

Furthermore, the following efforts have been undertaken for reducing microplastics. The Plastics Industry Federation and other plastic-related organizations prepared a resin pellet leakage prevention manual [178] and called on the industry to prevent leakage. Nevertheless, resin pellets have since been detected in domestic surveys [155]. Therefore, more thorough implementation is essential. In March 2016, the Japan Cosmetic Industry Association called on 1,100 member companies to voluntarily regulate microbeads. In a survey conducted in 2016, 150 products of facial cleansers and body soaps were purchased as personal care products. Moreover, it was checked for each product whether it correctly indicated on its label whether it contained microbeads or not [179]. From this analysis and the component labeling, it was ascertained that there were two types of face wash among the 150 products which evidently contained microbeads. Because of this relatively low number, it is concluded that the self-regulation of companies with regard to microplastics was progressing.

As described above, substantial efforts in Japan are based on the actions and self-regulation by industries, local governments, and individuals rather than on legal regulations. Therefore, achieving the goals of the "Plastic Resource Recycling Strategy" will continue to be a significant challenge.

3.3 South Korea

3.3.1 Macro- and Microplastic Contamination of the Aquatic Environment

The first studies investigating macro- and microplastic pollution in South Korea were published by Lee et al. [180] and Lee et al. [181], respectively. Subsequently, researchers from South Korea have reported some of the highest levels of ocean microplastic contamination in the world along the country's southern and western coasts [22] and southern coasts [23], as well as in its sandy beaches [21], thus further establishing that the oceans and seas in East Asia are among the world's most polluted (see Introduction).

We first reviewed marine and river plastic pollution. Lee et al. [180] determined the types, quantities, and distribution of marine litter items (categorized into 14 types) pulled up with bottom trawl nets from the seabed of the East China Sea and the South Sea of Korea during 1996–2005 cruises. Litter densities were higher in coastal seas than in the open sea. Fishing gear items, such as fishing lines, nets, octopus jars, and pots, predominated while the contributions of other items, such as clothing, glass, metal, plastic, rubber, vinyl, and wood, remained mainly below 30%. Floating debris sampled in the southeastern sea of Korea south of the Nakdong River Estuary in 2012 resulted in microplastic particles being found at all 20 sampling points, whereas Styrofoam particles only peaked at a few stations far from the Nakdong River Estuary [182]. The dominant particle types were fibers (PES), hard plastic (PE), paint particles (alkyd), and EPS, while less prevalent types were films, pellets, and other foamed plastic materials. There was large spatial and temporal heterogeneity in the samples. Kim et al. [183] estimated the quantity of discarded fishing traps and gill nets in South Korea's coastal waters to be about 11,436 MT and 38,535 MT, respectively.

Chae et al. [22] sampled ocean waters near Incheon harbor in 2013 and found that the microplastic abundance was greater in the ocean's surface microlayer than in the underlying surface seawater and that most of these microplastics originated from ship paint particles (a result mirroring the findings from Song et al. [184]). Song et al. [23] also sampled ocean waters in Jinhae Bay in southern South Korea in 2013 and also found that fragmented microplastics, which included paint resin particles derived from ship paints, accounted for 75% of all particles, followed by spherules, fibers, EPS, and sheets. Song et al. [185] sampled ocean waters in eight coastal areas along almost all parts of South Korea's coastline in 2016–2017 in order to determine the vertical distribution and composition of microplastics >20 μm. The mean microplastic abundance was 871 particles per m^3, was significantly higher in the surface water (0–0.2 m) than in the underlying water column (3–58 m), and was significantly lower near rural than near urban areas. The predominant polymers were PP and PE.

The microplastics (>20 μm) in the Nakdong River itself were sampled in 2017 at three sampling points each in the upstream, midstream, and downstream parts of the river in order to determine their spatiotemporal distribution [186]. The mean microplastic abundance ranged from 293 ± 83 (mean ± S.D.) particles per m^3 in water in the upstream part to 4,760 ± 5,242 in the downstream part. PP and PES accounted for 42% and 23% of all particles in the water, respectively, followed by 28 other polymer types all with <5%. PP and PE accounted for 25% each of all the particles in the sediment, respectively, followed by 20 other polymer types all with <6%. Microplastic particles >300 μm accounted for 74% and 81% in the water and sediment samples, respectively, and the distribution peaked in the 50–150 μm size range. The authors estimated that the annual load of microplastic particles in the river in 2017 was between 5.4 and 11.0 trillion particles weighing between 53.3 and 118.0 MT. Finally, most particles were detected in the wet season, making up 71% in number and 81% in weight.

As shown in numerous studies, the ocean-based pollution can then enter the marine food web. Jang et al. [187] showed that some of the Styrofoam microplastics found in oceans and coastal areas originate from polychaete worms burrowing into Styrofoam debris, especially Styrofoam buoys. These findings suggest that microplastic formation from larger plastic items is due not only to physical or chemical processes [188] but also to biological activities (see also Davidson [189]). Another source of marine microplastics was illuminated by Lee et al.'s [190] study of the percentage of microplastics released by three different kinds of sewage treatment facilities. While they all had treatment efficiencies of about 98% or more due to the large amount of effluent, more than four billion microplastic pieces were released annually from each facility into marine environments. However, many sources and pathways of marine plastic pollution remain unclear. Using a mass balance approach, Kim et al. [191] estimated that the total unaccounted mass of high- and low-density PE in the marine environment from 1995 to 2012 was 28 MT and that the corresponding contribution to marine plastic debris would be approximately 25,000 MT.

South Korean researchers also studied the effects on the biota. Hong et al. [192] found that 21 marine species had been affected by marine debris, including birds, mammals, and one crustacean. To assess the potential impact of microplastics on zooplanktivores, Kang et al. [193] measured the abundance ratio of neustonic microplastics to zooplankton in Geoje eastern Bay and Jinhae Bay, both in the southern sea of Korea, in 2012–2013. The mean microplastics to zooplankton ratios were higher during the earlier dry than during the later rainy season in both years. The authors suggested that zooplanktivores could confuse microplastics with prey items and that this risk is higher in the dry season. Another study by Cho et al. [30] demonstrated unequivocally that certain marine organisms take up microplastics from their environment. Since bivalves are known to accumulate microplastics when they filter large volumes of seawater, the authors examined four popularly consumed bivalve species bought in markets in three major cities in 2017. The four species, namely, Manila clam (*Tapes philippinarum*), mussel (*Mytilus edulis*), oyster (*Crassostrea gigas*), and scallop (*Patinopecten yessoensis*), account for ~80% of total shellfish consumption on South Korea. They found about one microplastic particle per examined individual, whereby particles smaller than 300 μm were the dominant size. The dominant polymers were PE, PP, PS, and PES. This level of contamination was estimated to lead to a mean annual microplastic intake of 212 particles per year for the average South Korean person.

We then reviewed coastal plastic pollution. Marine debris sampled by volunteers on 20 beaches along all parts of South Korea's coastline in 2008–2009 was used to assess the levels of debris pollution and to identify its main sources [194]. The number of items, weight, and volume of marine debris per 100 m of beach was estimated to be 480.9 ± 267.7 (mean ± S.D.) items, 86.5 ± 78.6 kg, and 0.48 ± 0.38 m^3, respectively (cf. estimates for Japan and Taiwan in Walther et al. [29]). Plastics and Styrofoam made up most of the debris composition both in terms of number (66.7%) and volume (62.3%). The main debris sources were assumed to be fishing activities and marine aquaculture, followed by recreational activities along the shoreline. Less than 6% was related to other sources such as smoking, illegal dumping, and medical and hygiene products.

Heo et al. [195] investigated the spatial distribution of small plastic debris on Heungnam beach in 2011. They determined the abundances of small plastic debris items (>2 mm) along the high strandline and the cross-sectional line of the beach. The mean item abundances were 976 ± 405 (mean ± S.D.) items per m^2 at the high strandline and 473 ± 866 particles per m^2 at the cross-sectional line. Styrofoam items accounted for 91% of the total abundance at the high strandline and 96% at the cross-sectional line, while less prevalent types were plastic fragments, pellets, and intact items. Furthermore, there was large spatial heterogeneity among the sampled high strandline and cross-sectional quadrats.

Plastic debris sampled on six beaches near the Nakdong River Estuary in 2012 was placed into three size categories, namely, macroplastics (>25 mm), mesoplastics (5–25 mm), and large microplastics (1–5 mm) [181]. In 1 m^2, the researchers found on average 1 macroplastic, 238 mesoplastic, and 17,906 microplastic particles, with Styrofoam being the most abundant meso- and microplastic debris item, while intact plastic items were most common in the macroplastic debris. All three size categories exhibited significant and positive correlations with each other. Microplastic particles sampled on three beaches on Soya Island west of Seoul in 2013 determined a very high microplastic abundance of 46,334 ± 71,291 (mean ± S.D., range 56–285,673) particles/m^2 [196]; at the time, it was one of the highest levels reported globally. The most prevalent polymer type was PS. Jang et al. [197] sampled 752 plastic debris items (>25 mm) from six beaches along almost all parts of South Korea's coastline in 2013 in order to determine the debris' sources. The items were mostly made of fiber and fabric (55%) but also hard plastic (16%), Styrofoam (12%), film (11%), foamed plastic other than Styrofoam (3%), and other polymers (3%). 56% of all the collected items appeared to be ocean-based while the remainder was land-based.

Plastic debris sampled on 12 beaches along almost all parts of South Korea's coastline in 2013–2014 showed the increasing abundance of particles as particle size decreases. In 1 m^2, Lee et al. [21] found on average 1 macroplastic (>25 mm), 37.7 mesoplastic (5–25 mm), and 880.4 microplastic (1–5 mm) particles, with Styrofoam and fibers being the most abundant types. Unlike Bancin et al. [28] in Taiwan, Lee et al. [21] detected no significant differences between the particle abundances in the high strandline and the backshore for any of the three size groups. Plastic debris sampled on 20 sandy beaches along all parts of South Korea's coastline in 2016 demonstrated the highly heterogeneous distribution of microplastic abundance between beaches [198], with abundances of large microplastics (1–5 mm) ranging from 0 to 2088 particles per m^2 and small microplastics (0.02–1 mm) ranging from 1,400 to 62,800 particles per m^2. Again, abundance increased with decreasing particle size. The main polymers were EPS, PE, and PP. Some of the plastic abundances showed positive relationships with human population, precipitation, abundance of macroplastic debris on the beach, and proximity to a river mouth (this last result was mirrored in Taiwan by Bancin et al. [28]).

Mesoplastic marine debris (5–25 mm) sampled on 20 sandy beaches along all parts of South Korea's coastline from March to May 2016 (Won Joon Shim, in litt. 2019) determined that the mean mesoplastic abundance was 13.2 items per m^2 and the mean weight was 1.5 g per m^2 [199]. Hard plastic (32%) and Styrofoam (49%) were the dominant types by number, but their proportions were highly variable among the beaches. Furthermore, there was large spatial heterogeneity among the beaches both in terms of the number and weight of mesoplastic particles.

Relatively little is still known about how the influx of plastic debris may enhance the spread of toxic chemicals into the aquatic environments of East Asia. Therefore, Jang et al. [200] examined the levels of a flame retardant, namely, hexabromocyclododecane, in EPS which is the predominant marine debris originating mainly from fishing and aquaculture buoys. Marine debris samples of EPS were obtained from buoys and microplastics collected along the South Korean coast from 2013 to 2015 as well as from 12 other countries in the Asia-Pacific region. Hexabromocyclododecane was detected extensively in the examined samples which suggest that this hazardous flame retardant may contaminate aquatic environments and food webs worldwide via the marine debris route.

3.3.2 Mitigation Efforts to Decrease the Plastic Pollution

We first reviewed relevant governmental policies and waste management (see also Chen [35] who reviewed South Korea's initiatives on marine litter which began in 1999).

A new branch of the Ministry of Ocean and Fisheries (MOF) was created in 2000, the Korea Marine Environment Management Corporation (KOEM), which is categorized as a public sector organization. Its goals are to efficiently promote the conservation, management, and improvement of the marine environment, as well as marine pollution control, in order to ensure a clean and rich marine environment in the future [201]. In 2013, KOEM estimated the annual inflow and existing volume of marine waste in South Korea in its "2nd Framework Plan for Marine Waste Management" [202]. Jang et al. [203] estimated that 91,195 MT of marine debris (which includes plastic debris) enters the marine environment annually (of which 36% is from land-based sources and 64% from ocean-based sources). The total stock of marine debris on all South Korean coasts in 2012 was estimated to be 152,241 MT (8% on all coastlines, 90% on the seabed, and 2% in the water column). KOEM also estimated that 44% of the total marine waste was collected (probably through cleanup activities) but that the remaining 56% leaked into the marine environment where it may decompose if the waste is biodegradable [202].

To monitor the ongoing situation, the MOF organized 40 local governments and KOEM to monitor marine debris in the ocean waters along South Korea's coastlines every 2 months [202]. During the six surveys conducted in 2016, 68,421 items were collected, which weighed 11,836 kg and had a volume of 65,404 L [202, 204]. These items were collected in 40,100-m sections distributed all along the South Korean coastline; each section was then subdivided into 20 5-m-wide transects whereby each transect begins at the water's edge at low tide and ends at the first barrier at the back of the shoreline, and 4 out of the 20 transects were then randomly chosen for sampling according to the methods outlined by Opfer et al. [205]. Therefore, the total sampled length was 4 km × 0.2 = 0.8 km. 56.5% of the 68,421 items were made from plastic and 14.4% from Styrofoam; the remaining 29.1% were other types [202]. Plastic and Styrofoam items were also the two greatest types by weight and volume [202]. Naturally, some of that waste does not originate from South Korea but from other neighboring countries and even some Southeastern countries [202].

A number of studies have addressed possible policy and technological solutions. Cho [206] described the generation of sea-based marine debris in South Korean coastal waters and some of the resulting environmental and economic problems. Even though the South Korean government continuously removed marine debris, the generation of marine debris needed to be prevented. Therefore, the government initiated an incentive program for fishermen to collect fishing gear or other marine debris while fishing. The program paid 9.3 million USD for 11,000 MT of collected marine debris from 2003 to 2006. Jung et al. [207] described practical engineering approaches and infrastructure which address the problem of marine debris in South Korea. These changes began in 1999 with a nationwide 10-year project called "A Practical Integrated System for Marine Debris" which developed fundamental changes to the infrastructure and consisted of four linked types of technology: prevention, deep-water survey, removal, and treatment (recycling). Together, they reduced the generation and improved the retrieval of marine debris pollution. Hong et al. [208] evaluated the cost efficiency of three management measures to reduce the pollution from derelict fishing gears and suggested that the current management measures need to be reorganized to improve preventive measures.

The increasing scientific knowledge about the plastic pollution problem as well as the resulting media coverage led to rising public awareness. Choi and Lee [209] enumerated the willingness of Seoul residents to pay for removing the microplastics in the ocean. According to their survey, most South Korean people voiced their concern about microplastic pollution, favored the implementation of progressive government policies to control it, and would be willing to pay some money for it. Another study conducted at the request of Greenpeace (GP) interviewed 1,000 South Korean adults [210]. 86% of the respondents agreed that the self-regulation of companies was lacking and therefore was not working. As a result, this study emphasized the need for compulsory regulations by the government as well as the expansion of environmental risk studies. It also outlined possible ways to manage plastic pollution and separated them into the following strategies: risk management standards setting, expansion of plastic recycling policies, stricter legal regulations and reinforcement of governance, and collaborations with private organizations or research institutes.

With the public increasingly favorable toward decisive action, actual action by the government was then triggered, as is often the case, by a crisis moment. The significant event in this case was the "waste crisis" in the Seoul Metropolitan area in April 2018 which occurred because the private recycling companies declined to collect waste plastics from residential districts, the reason being that they could not make a profit anymore [211–213] because of China's import ban (Sect. 3.1.2) [214]. This "waste crisis" forced the government to come up with solutions which would prevent this problem from happening again [212].

The South Korean government had already enacted the "Framework Act on Resource Circulation" (which is a set of laws that promote sustainable development and proper waste disposal) in 2016 which was enforced in January 2018 [212, 215]. Under this general framework and in response to the waste crisis, the "Basic Plan on Resource Circulation (2018–2027)" was subsequently established in order to set up the mid- to long-term policy goals and strategies [212], and the government also set up a "Comprehensive Measure of Waste Recycling" which aims to reduce plastic waste by 50% and raise the plastics recycling rate to 70% by 2030 from the current 34% rate [211, 216]. With these policies and regulations, the government aimed to establish a "comprehensive system of resource cycling" all the way from production to consumption to management and recycling, thus reducing waste generation, promoting the recycling of high quality waste materials, and optimizing community-based waste management by participatory governance [212]. In contrast to current government policies centered on recycling existing waste, these new laws and regulations aim to reduce waste throughout production, consumption, management, and recycling. Furthermore, the government wants to completely eliminate consumer use of disposable products and restrict excessive packaging [215]. All these new measures are attempts by the current government to aggressively address environmental sustainability because domestic waste generation has continued to grow at alarming rates; in 2016, South Korea produced 156 MT of waste which constituted a 30% rise over 2006 [215].

Since the government introduced these measures, several central administrative agencies (Ministry of Environment (ME); MOF; South Korean Coast Guard; Ministry for Food, Agriculture, Forestry and Fisheries; Ministry of Food and Drug Safety) have begun to manage plastic pollution [210]. The government also announced it would raise financial support for recycling firms by 1.7 billion won [217]. Another part of these efforts is some recently introduced bans on single-use plastics (see below).

We now specify the work of three different government ministries and their efforts to introduce policies to prevent plastic pollution.

Ministry of Environment (ME) Since 2003, the ME has implemented a "producer liability recycling system" by supplementing and improving the waste deposit system that has been in operation since 1992 [202]. This system commits producers to a recycling obligation and levies a non-compliance charge onto producers who do not oblige [202]. According to the standards set by the ME, the mandatory recycling rate for marine products of fish farming (much of it Styrofoam buoys) was 28.1% in 2015 [202], but even this relatively low rate was not achieved in 2015 when only a 23% recycling rate was achieved. Specifically, the fish farming industry used 2026 MT of products, but only 465 MT (23%) were recycled [202]; the remaining 1,561 MT were assumed to have leaked into the ocean [202]. This rate is certainly much lower than the average recycling rate of 61.8% for other plastic items [202].

In May 2018, the ME announced its long-term plan to reduce plastic waste by 50% by 2030 and also that it partnered up with 21 of South Korea's largest cafe and fast-food franchises which promised to make disposable cup material more recycle-friendly and to encourage reusable cup usage by offering 10% discounts for customers who bring their own cups [214, 218] (Taiwan implemented such a discount system in 2011). Action was partially triggered by a comprehensive report found that the number of single-use cups disposed annually had jumped from 19.1 billion in 2009 to 25.7 billion in 2015 [214].

Consequently, the ME banned single-use plastic cups in coffee shops and fast-food shops for in-shop diners (but not take-out diners) in 2018 [214, 217] and plastic bags (with some exceptions for frozen products and wet products, such as fish, meat, tofu, some fruits, and vegetables) at bakeries, department and discount stores, and large (but not small) shopping malls and supermarkets in 2019 [219–221]. Approximately 13,000 supermarkets are affected and are now required to offer customers reusable or recyclable cloth or paper bags. Smaller-sized stores, traditional markets, and bakeries can still provide single-use plastic bags but must charge for them. The cup ban led to a 72% decrease in usage by May 2019, although some shops simply replaced them with single-use paper cups, while other shops actually switched to multi-use cups; plastic bottles, lids, and straws were not banned [214]. Therefore, the ME has recently reviewed the possibility of extending the ban to also apply to single-use paper cups and plastic straws [214]. The ME also announced in July 2019 that all government offices would stop using single-use cups [218]. In 2020, the government will enforce that all plastic beverage bottles must be colorless and transparent for better recycling and that plastic straws will be included into the products which must then be recycled [211].

Ministry of Food and Drug Safety [202] On 29 September 2016, this ministry revised the "Regulations on Safety Standards for Cosmetics" which redefined the term "microplastic" and banned it as an ingredient of cosmetics. Therefore, cosmetics which contain microplastics (defined by the regulation as solid primary plastic particles of <5 mm length, which thus includes microbeads which are <1 mm length) have been banned from being manufactured or imported since July 2017. In July 2018, their sale was also banned. However, this regulation only applies to cleanser or scrubbing products which only account for 0.56% of total cosmetic sales. All other cosmetic products (e.g., makeup products) which include microplastics actually make up 24.5% of total cosmetic sales. Therefore, only 2.3% of cosmetic products which contain microplastics are currently regulated. In January 2017, the "Regulations on Permit, Report and Review of Medical Supplies" was also partially revised which banned medical supplies that include microplastics from being manufactured or imported; in July 2018, their sale was also banned.

Ministry of Ocean and Fisheries (MOF) The MOF, the Korea Institute of Ocean Science and Technology and the Korea Institute of Marine Science and Technology Promotion have been conducting research about microplastics and their impacts since 2011; many conclusions and recommendations from this research were summarized in "The Second National Marine Litter Management Plan (2014–2018)" [202] and "The Third National Marine Litter Management Plan (2019–2023)" [222].

The aims of the Second National Marine Litter Management Plan (2014–2018) were to minimize the occurrence of marine waste, to strengthen public projects of collecting marine waste, and to establish a scientific infrastructure and policies which deal with marine waste [202]. The specific strategies were (1) intensive management of sources of marine waste (68.5 billion won); (2) strengthening marine waste collection projects which focus on the daily lives of people (2385.3 billion won); (3) building a basic management system for marine waste (199.7 billion won); and (4) education and promotion for various targets (e.g., education of fishermen, plastic product companies, students, household recycling, etc.) (49.3 billion won) [202]. Each strategy was then subdivided into four to six initiatives [202].

The MOF planned to establish a basic plan of fishery management regulation every 5 years which includes regulations relevant to marine plastic pollution [202]. For instance, "The Third Fisheries Management Basic Plan (2017–2021)" was published in 2017 which aimed to make it mandatory to only use eco-friendly buoys not made from Styrofoam for fish farms, ensure sustainable yields from ocean fishing, and introduce new certification standards for improved fishery materials and equipment, e.g., to reduce and regulate the use of toxic materials [202] (see also how participatory workshops were held to develop policy ideas and solutions to the Styrofoam buoy debris problem in Lee et al. [223]). "The Third Fisheries Management Basic Plan" contains 3 major initiatives, 9 main tasks, and 25 detailed tasks and has a total budget of 172.4 billion won [202].

We next reviewed some examples of education, media, monitoring, and outreach campaigns by ENGOs. On 14 July 2016, GP Seoul released a joint statement with six other ENGOs (Citizens' Institute for Environmental Studies, Environmental

Justice Foundation, KFEM Ocean Committee, KWEN, OSEAN, WWF) [224] which urged the government to come up with a bill banning microplastic "microbeads" in cosmetic and household goods [224]. Below, we reviewed the campaigns for a microbead ban of three important ENGOs (which was implemented in 2017; see above). Park et al. [224] suggested that ENGOs should consider expanding their topics and campaigns to improve people's awareness of plastic pollution and to push companies and the government to reduce plastic pollution.

The *Korean Women's Environmental Network (KWEN)* [224] is the first ENGO in South Korea to launch activities about microplastic pollution. KWEN is a member of the "Beat the Microbeads Campaign of Plastic Soup Foundation." This is an ENGO of women activists founded in 1999 which cares about the environment in order to achieve an equal and sustainable society. Under the theme of "Eco Cosmetics," KWEN headed a campaign called "Plastic Ocean: Face to Fish" to ban microbeads from cosmetics and personal hygiene products; it also released a list of microplastic-containing cosmetics on its website based on a survey of cosmetics sold in South Korea.

Greenpeace (GP) [224] is a global ENGO, is a member of the "Break Free From Plastic" movement and has participated in a campaign called the "International Plastic Bag Free Day." "Break Free From Plastic" is one of the most active movements campaigning against plastic pollution. Since its launch in September 2016, "Break Free From Plastic" has been joined by more than 1,000 NGOs from around the world, including GP, with the aim of reducing disposable plastic debris and ultimately resolving the plastic pollution problem. The GP East Asia Office published a report about the use of microbeads in cosmetics and personal hygiene products produced by different companies, and it has led consumer movements to pressure these companies. The GP Seoul branch has also led many consumer campaigns, including the aforementioned joint statement with six other ENGOs, and it has three plastic-related activities among its 12 main activities.

The *Our Sea of East Asia Network (OSEAN)* is also a member of "Beat the Microbeads Campaign of Plastic Soup Foundation" [202] and registered under MOF [225]. OSEAN focuses on solutions to marine environmental problems, with a special focus on the marine waste problem [225]. Their three main activities are research, education and promotion, and cooperation with other groups (e.g., with international groups which campaign against marine debris, groups working for ocean protection, citizen groups, marine environmental groups, etc.) [225].

Finally, we reviewed the efforts of some South Korean inventors and businesses who have developed alternative products and methods of production and recycling. Producers can alleviate plastic pollution in two ways: either by reducing the use of plastics or by producing alternative products, e.g., made from biodegradable materials [224]. Since the 1990s, research institutes, major established companies, and venture companies have been researching biodegradable plastics; however, the market scale remains small in South Korea because these alternative materials remain too expensive [226]. As a result, the South Korean domestic market for these products is $<2\%$ of the total global market for biodegradable plastics [226]. Therefore, the development of the South Korean bioplastic technology is underdeveloped compared to that of many other developed countries. Most South Korean companies do not have sufficient technological know-how or investment to be able to fulfill the rather strict standards which the government has imposed on biodegradable products; and even if the companies can fulfill the standards, profits remain poor. To improve profits, the government has recently eased their standards but still profits are not good, so bioplastic products are not being commercialized.

The *Samyang Genex Corporation* [227] was the first company in South Korea to produce a bioplastic made from corn called “isosorbide” in 2014. It has high biodegradability, good transparency, excellent surface hardness, and non-toxicity. Therefore, it will be used for electronic products such as mobile devices, television sets, smartphone displays, car dashboards, food containers, and eco-friendly house building materials.

SK [227] will soon commercialize bioplastics made from CO_2. When this bioplastic is incinerated, it decomposes into CO and CO_2 and produces no toxic fumes. It also has a high transparency and good blockage of oxygen and moisture.

Samsung [227] produced the “Eco-friendly Cellular Phone (SCH-W510)” which has a battery cover which contains 40% of cornstarch-based bioplastic. It was the first South Korean cellular phone which received an eco-friendly mark. Its packaging also used biodegradable craft paper with no use of any plastic packaging. For the packaging of some TV accessories (e.g., remote controls), Samsung used sugarcane-based bioplastics.

3.4 Taiwan

3.4.1 Macro- and Microplastic Contamination of the Aquatic Environment

Given that Taiwan is an island nation and thus completely surrounded by some of the world’s most plastic-polluted waters (see Introduction), that much of its coastline is ravaged by extremely high levels of marine debris pollution, and that a relatively high proportion of Taiwanese people’s diet comes from seafood, it is somewhat surprising that less than ten scientific publications have focused on this topic.

In Sect. 3.4.2, we outlined how Taiwanese ENGOs began to monitor marine debris along Taiwan’s coastlines and to publish reports about it in the 2000s, which was a decade before the first scientific publication appeared. So far, only four publications have investigated the levels of large marine debris on Taiwan’s coastlines. The first publication surveyed four beaches on a small island in southern Taiwan from August 2009 to October 2011 and documented the types and proportions of debris types [228]. 78.3% of the items were made from plastic materials, with other types being glass, metal, and paper. A very high percentage of items was assumed to originate from shoreline and recreational sources and the second highest percentage from ocean and waterway sources.

Later, Kuo and Huang [229] surveyed six sites in northern Taiwan from June 2012 to May 2013 and documented the types, proportions, categories, and sources of marine debris types. The percentage of items made from plastic materials was even higher (85.5%), with the other types again being glass, metal, and paper. Levels of pollution were higher on rocky shores than sandy beaches and fishing ports. Again, most of the debris items originated from recreational sources and the second highest percentage from ocean and waterway sources.

Most recently, Walther et al. [29] used a 12-year dataset collected by volunteers (or citizen scientists) and collated by the Society of Wilderness (SOW) to estimate overall pollution levels for the entire coastline of Taiwan. In total, data from 541 coastal cleanup events held between October 2004 and December 2016 were analyzed. During each event, volunteers sorted and weighed 19 categories of large coastal debris items. The volunteers collected 904,302 items weighing 131,358.3 kg. The five most common debris categories were plastic shopping bags, plastic bottle caps, disposable tablewares, fishing equipment, and plastic drinking straws. 63.6% and 27.2% of items were made of either plastic or plastic mixed with other materials, respectively, and most of these items were made for single-use (e.g., 60% of the items originated from the single-use food and drinks packaging industry, and 15% were plastic bags). One estimate based on multiple linear regression analysis yielded a mean pollution level of 5,937 debris items and 831 kg of debris per km of coastline. Extrapolated to the length of 1,339 km for the entire coastline, it means that, on average during the 12-year period, about 7.9 million items weighing 1,110 MT polluted Taiwan's coastline. Walther et al. [29] concluded by making seven recommendations how to improve the data gathering and verification during cleanup events.

Another study which tracked the changes of large marine debris over several years (2012–2016) was conducted in a remote Taiwanese island in the northern South China Sea [230]. The amount and weight of debris varied greatly between months and years, with Styrofoam and plastic bottles being the most abundant, followed by fishing gear and other plastic products. About half of the debris originated from China and Vietnam.

The study of the levels of microplastic pollution on Taiwan's coastlines is even more recent. The first evidence of this problem was actually provided by an investigation conducted by SOW [231] which measured the density of microplastics in the range of 0.1–2.5 cm on three beaches in New Taipei City, Tainan City, and Kaohsiung City (see also Sect. 3.4.2). This investigation showed microplastic pollution in all locations, with a maximum of 787 microplastics per m^2 of which 72% were Styrofoam particles but also primary microplastics such as pellets.

The first scientific publication was conducted by Kunz et al. [232] and used synchrotron-based FTIR to positively identify microplastic particles collected on four beaches in northern Taiwan in 2015. The polymer types of the 1,097 particles were PE (44%), PP (43%), PS (12%), and ABS (1%).

While Kunz et al. [232] only took one sample per beach, Bancin [28] took 80 samples from one beach in northern Taiwan in 2017 in a systematic manner with 4 transects beginning at the intertidal and ending in the dunes and covering the entire length of the beach. This systematic sampling scheme plus the use of resampling curves allowed a very accurate estimation of the mean pollution level of the beach which was 96.8 microplastic particles per m^2 or approximately 6.8 million particles with an estimated weight of 250.4 kg for the entire 70,130 m^2 beach. The approximate percentages of the polymer types were PE (51%), PP (34%), and PS (15%). The sampling also revealed a high heterogeneity among the samples, and therefore the resampling curves indicated samples sizes of $n \leq 10$ are very unreliable and that sample sizes of at least 10–20 samples are required at a minimum, but to reach truly reliable estimates, sample sizes of $n \geq 50$ are required.

Microplastics were detected in table salts from Taiwan [233, 234]; clams, mussels, oysters, and scallops [235, 236]; and coral fishes and turtles [237].

3.4.2 Mitigation Efforts to Decrease the Plastic Pollution

In response to the growing awareness of the plastic pollution problem, Taiwan's government, ENGOs, and other actors and stakeholders have made efforts to alleviate the problem of waste in general and of single-use plastics in particular since the 1990s, but especially in the 2000s. Below we describe briefly the timeline of the policies of Taiwan's government, followed by the actions of ENGOs to influence public opinion and the government. A more detailed description will be published separately (Walther et al., unpublished manuscript). It should also be interesting to the general reader because it is widely acknowledged that Taiwan has been comparatively successful in tackling some aspects of the problem of plastic pollution, such as one of the world's highest recycling rates.

However, things were quite different only 30 years ago, as economic growth, rising living standards, and soaring consumption had created so much waste that Taiwan earned the nickname "garbage island" in the 1990s; almost no waste was recycled, and two thirds of landfills had reached capacity [238]. The first response of Taiwan's government was to build 24 incinerator plants. It also incentivized companies and consumers to waste less and recycle more beginning with an Extended Producer Responsibility (EPR) policy in 1997, followed by other policy and waste management initiatives (see below). One key part of this system was the compulsory nationwide garbage sorting program introduced in 2006 which forces people to separate different kinds of waste [35, 239–241]. As a result, the average waste produced per capita was reduced by about 30% over the last two decades [238], while the recycling rate increased to approximately 52% of household waste and 77% of industrial waste [238–241] which is a >100% increase using 2002 as baseline [35]. These policies also created thousands of new jobs in about 1,600 recycling companies with an annual revenue of >2 billion NTD [238]. In 2017, about 10% of the total weight of all recycled materials in Taiwan was plastic materials, amounting to 426,345 MT [242].

Despite these successes, a large amount of waste pollution has been and still is being released into Taiwan's environment for a number of reasons. First, a lot of people still throw their garbage into the outside environment (such as ditches, rivers, roadsides, etc.) for various reasons (e.g., they simply do not care or do not want to pay garbage charges, lack of garbage bins in public spaces, etc.). Second, some recycling businesses also fly-tip some waste for various reasons, but often because some waste is uneconomical to recycle. Third, some waste deposit sites are leaking, e.g., during typhoons. Fourth, a lot of waste is also continuously entering the oceans surrounding Taiwan [25, 27, 41, 166, 243, 244], and some of that waste is continuously deposited along Taiwan's coastlines (see Sect. 3.4.1).

To decrease these plastic emissions, Taiwan's government has implemented ten policies which were aimed specifically at the reduction of single-use plastics. The first-ever policy to encourage the recycling of single-use plastics was the "4-in-1 Recycling Program" introduced in 1997. This policy can be classified as an EPR. For each kg of single-use plastic item produced, the plastic producer has to pay a certain recycling fee which in turn is used to support two parts of the recycling industry: the collectors and the certified recycling factories. The collectors receive money from the recycling factories when they sell their collected recyclables to the factory. This program has helped to establish a nationwide recycling industry (see also above).

The second policy was introduced in 2002 and was the first-ever source reduction policy which aimed to reduce the use of plastic bags and single-use tablewares made from plastic [245]. Specifically, the policy banned the handing out of shopping plastic bags thinner than 0.06 mm, and customers had to pay for shopping plastic bags thicker than 0.06 mm. The government promoted the ban with about 1,375 promotional activities [245]. According to a survey of retailers conducted by the TEPA in 2006, the number and weight of plastic shopping bags actually declined by 58% (or 2 billion bags annually) and 68%, respectively, from 2003 to 2006 [35, 246], and the number of people using their own bags had increased by over 60% [245].

In 2005, the third policy introduced regulations against overpackaging, which established rules for the packaging of various goods, e.g., cosmetics, pastries, processed foods, wines, etc., and specifically targeted overpackaged gift boxes which are very popular in Taiwan [247]. In 2006 and 2007, these bans were expanded [248, 249].

In 2011, the fourth policy aimed to reduce the use of single-use cups by encouraging customers to bring their own cups through a reward system (a discount price or a coupon) [250].

In 2012, the government further amended the 2007 policy to decrease the use of single-use plastic trays and package boxes [248]. In the same year, the Tainan city government banned Styrofoam cups [251]. In 2016, the Taipei city government announced that schools, government buildings, and governmental sections must stop selling bottled water; however, this ban did not extend to any other kind of drink sold in single-use plastic or other type of container. Furthermore, schools and governmental departments had to replace their single-use tablewares with multiple-use stainless steel bowls, chopsticks, plates, and other utensils [252].

The eighth policy was a ban of microbeads in personal care and cosmetic products introduced in 2018. Originally, the ban was to be introduced in 2020, but pressure from four Taiwanese ENGOs and a public petition caused the government to advance the ban to 2018 [249].

In 2018, the 2002 plastic bag ban was extended to include another seven businesses which were banned to provide plastic bags for free [249]. The TEPA [253] estimated that this ban would lead to the annual reduction of 1.5 billion plastic bags.

The tenth policy was introduced in 2019 when the TEPA banned single-use plastic straws and expanded its 2006 ban on single-use utensils for eat-in consumers in many restaurants [254, 255]. After the ban on plastic straws was announced, new products and companies quickly sprang up to fill the void of plastic products [254, 256–258].

While Taiwan's government deserves credit for initiating one of the world's best recycling system and, more recently, announced a ban for most single-use plastics during the 2020s, much credit also needs to go to Taiwanese ENGOs who began monitoring plastic pollution levels long before scientists did, educated and then involved the public in cleanup events in order to increase awareness of and information about coastal pollution, and pressured the government through various education, media, and outreach campaigns. Again, a more detailed description will be published separately (Walther et al., unpublished manuscript).

Taiwan's government has financed cleanups of the coastal environment since 1997 [35], which have been augmented by volunteer cleanups, often but not exclusively organized by ENGOs [29, 259]. However, the government did not monitor coastal or marine debris, which eventually caused ENGOs to begin monitoring efforts. When the increasing pollution of Taiwan's coastline with debris (most of it plastic debris) became evident in the early 2000s, Taiwanese ENGOs began (1) to interact with international allies, learning methods from them and adopting them in Taiwan; (2) to organize conferences and workshops, education and outreach campaigns, and coastal cleanup events including data gathering of coastal debris using citizen scientists; and (3) to interact with stakeholders and decision-makers in order to promote source reduction and other relevant policies. For example, SOW began in 2008 to organize coastal cleanup events in order to (1) decrease coastal pollution, (2) educate and actively involve Taiwan's public and media, and (3) document the types, weights, and numbers of debris items. One of the aims of collecting these data was to influence the government's policies by documenting what types of wastes accumulate on Taiwan's coasts.

Another ENGO which played a critically important role was the Kuroshio Ocean Education Foundation (KOEF) because it introduced the ICC method from the USA and adopted it for Taiwan's needs. Through further international cooperation, Taiwanese ENGOs learned how to survey macro- and microplastic pollution from scientists. Through these interactions with international experts and academies, Taiwanese ENGOs not only recognized this issue much earlier than the government but were able to organize international conferences before any public sectors or research institutions got involved in this topic.

In 2010, the National Museum of Marine Science and Technology and four Taiwanese ENGOs, namely, KOEF, SOW, the Taiwan Environmental Information Association (TEIA), and the Tainan City Community College, formed an alliance called Taiwan Ocean Cleanup Alliance (TOCA) and agreed to pool all data generated during the coastal cleanup events. These data then became an important reference for policy-making because reports were published which detailed the amount of coastal debris collected. The release of each report was accompanied by a press conference, with successful media coverage in newspapers and on television. Later, the entire dataset was analyzed in Walther et al. [29] (see Sect. 3.4.1 for details) which again generated media coverage (e.g., [260, 261]).

Moreover, Taiwanese ENGOs learned to use analysis of monitoring data to foster policy changes and therefore pursued several other data collection projects. For example, SOW [231] demonstrated the presence of microplastics on three Taiwanese beaches (see Sect. 3.4.1 for details). This was the first published evidence of microplastic pollution for Taiwan, which also generated media coverage (e.g., [262, 263]), and was then followed by two peer-reviewed publications (see Sect. 3.4.1).

In 2017, SOW worked with CSIRO to collect samples of microplastics from sea water, coastal and terrestrial areas, and rivers in southern Taiwan [264]. Furthermore, GP used manta nets to trawl the ocean surface waters in southern Taiwan, which was then followed up by a similar study from KOEF which trawled the ocean waters around all of Taiwan's main island as well as most important islands totaling 51 sampling points in 2018 [265]. In 2018, GP and SOW used a rapid assessment method for monitoring the existing volume of marine debris pollution around Taiwan's coastline. Counting black bags of waste at 121 different sites, it was estimated that approximately 12.66 million liters of debris are found around Taiwan's coastline. Derelict fishing equipment, plastic bottles, and foamed plastics (such as Styrofoam) were the top three most abundant debris types [266]. Highlighting these results, GP then suggested to the government (1) to focus cleanup activities on such pollution hotspots, (2) to tailor management and recycling methods for different types of fishery waste as soon as possible, (3) to develop alternative fishing gears to replace PS foam floats (also called Styrofoam buoys) used in oyster farming [243, 244], and (4) to set up recycling centers for discarded fishing gear.

In 2018, TEIA and the Wild at Heart Legal Defense Association (WHLDA) conducted a brand audit using the labels on PET bottles to identify the brand and the country of production. The results were then used to call on the Taiwanese government and consumers to promote source reduction and better recycling rates.

The media began to cover the issue of marine debris pollution in 2010 and more intensively beginning in 2017 and certainly also played a large part in moving the current government to adopt more aggressive source reduction policies, such as the planned ban on single-use plastics (the role of media will be covered more extensively in Walther et al., unpublished manuscript).

All the education, monitoring, and media work by Taiwanese ENGOs finally paid off when the election of President Tsai Ing-wen in 2016 ushered in a government with a much friendlier ear toward environmental policies. The starting point for a more collaborative approach between ENGOs and the government began in July 2017 when eight Taiwanese ENGOs (GP, HiiN Studio, KOEF, Tse-Xin Organic Agriculture Foundation, Sea Citizens Foundation, SOW, TEIA, and WHLDA) formed an alliance called the Marine Debris Governance Platform (1) to lobby the government and the public, (2) to work with the TEPA to launch a large-scale quantitative beach debris monitoring program, and (3) to educate the public and reduce the use of plastic products that cause marine pollution [267, 268].

This alliance then worked together with the TEPA to publish the "Action Plan of Marine Debris Governance in Taiwan" [249, 269] in February 2018 which includes a timeline of phasing out four single-use plastic items. The four pillars of the Action Plan are source reduction, prevention and removal, monitoring and surveying (including research), and outreach and public participation. To achieve source reduction, the Action Plan calls for a reduction of single-use plastic items, including a phased ban of most single-use plastic items by 2030, but many additional measures are also planned [249, 267, 270, 271]. Such close collaboration between ENGOs and the government in forming and implementing policies and measures is certainly unprecedented in Taiwan and probably even in East Asia.

4 Discussion and Conclusion

Only a few years ago, Chen [35] wrote "A comprehensive national program to assess or remediate marine litter is currently not available in Taiwan, although marine litter is pervasive along its coastline. No clear integral mechanism exists for solving marine litter problems." The recent successful collaboration between TEPA and several ENGOs to shape and then implement the ambitious "Action Plan of Marine Debris Governance in Taiwan" [249] demonstrates that progress on the plastic pollution problem is possible when top-down and bottom-up approaches coalescence and governments decide to take action while using the accumulated expertise of scientific experts and ENGO representatives to help them formulate effective policies and strategies for source reduction, recycling, and waste management.

However, what our review also reveals is the great disparity between countries due to their different socioeconomic-political systems. While intensive scientific research on plastic pollution has been pursued in the four reviewed countries for about a decade (although with different starting points and much less in Taiwan than in the other three reviewed countries), almost no research is being conducted in Mongolia and North Korea because of these countries' completely different situation. Likewise, the mitigation efforts to decrease the plastic pollution differ greatly between each of the four reviewed countries; the reason again is their different systems of economy and government.

Even though marine plastic pollution is becoming a regional problem which all countries share (e.g., the plastic pollution in the East China Sea and Yellow Sea affects all the surrounding countries), the economic and political decisions to deal with the problem differ wildly between these countries. It is true that scientific collaborations and other exchanges of expertise, e.g., between ENGOs of these countries, have increased in recent decades and that there are now some regional frameworks and other collaborations which deal with plastic pollution (Table 2 and Sects. 3.1.2, 3.2.1, 3.2.2, and 3.4.2). However, when we consider the specific mitigation efforts of each country, they differ greatly in their ambition, scope, and means.

For example, let us consider bans of plastic products. China has banned only one plastic product, namely, some kinds of plastic bags in 2008, and that ban is probably not effective. Japan has no bans on a national level whatsoever. South Korea has only banned some kinds of plastic bags, coffee cups, and microplastics in some products, although admittedly have now proposed some ambitious measures, including bans, for the next decade. Therefore, Taiwan is by far the most ambitious country with its intended (although not yet implemented) ban of many single-use items in 2030 and a host of other policies and measures to promote source reduction, prevention and removal, monitoring and surveying, and outreach and public participation, as outlined in its Action Plan. To begin to understand the reasons for these disparities would go far beyond this review, as we would have to consider the peculiar cultures, economies, histories, and political systems of each country. Thus, it remains true that most environmental policies are shaped within nations by national governments, some are shaped also by regions or municipalities, and very few are shaped by supranational or global organizations or treaties. While a global treaty on plastic pollution would be a very important tool to alleviate the problem [33, 272, 273], current efforts will have to focus on influencing national governments. Therefore, we suggest some lessons can be learned from this review of the efforts of the East Asian governments and ENGOs to tackle the plastic pollution problem.

First, progress on pressing environmental issues often moves along the following steps. First, scientists and ENGOs raise an issue, and eventually the media takes it up (more so in democratic countries, but even in dictatorships, the media covers environmental issues as soon as the government decides that they need to be tackled, e.g., [116]). This is usually followed by calls for voluntary action, such as recycling plastics, which almost invariably proves to have a negligible impact for various reasons. Given enough pressure, governments can then up the ante by taxing undesirable products (e.g., Taiwan's EPR policy) or subsidizing desirable products or systems (e.g., subsidies for certified recycling factories). However, such measures often also fall short of dealing with the problem as plastic emissions continue to rise [9]. Finally, governments can limit or completely ban plastic products (see examples in Part 3).

Many environmentalists would argue that, for many uses of plastic, especially single-use plastics, only banning them will prove to be sufficient to avert further plastic pollution of the biosphere [33, 261], similar to the global bans of ozone-destroying chemicals [273, 274] and persistent organic pollutants [272] and the proposed global ban of burning fossil fuels [275]. However, given that a global ban is likely a long way off, ENGOs and concerned citizens should focus their attention on lobbying national governments for greatly expanded bans of plastic products but also much better plastic recycling and, very importantly, much more research and development of alternative, biodegradable, and non-toxic products [262, 276] which can then quickly substitute the traditional products (see a few examples in Part 3).

Second, even though we appear to be moving toward a post-truth era in many areas of politics [277], many people (and perhaps a majority) can still be influenced by scientific data and its proper analysis. Even better results are achieved if the people themselves are involved in collecting data as citizen scientists [29, 259, 278–280] because this involves education and training which broadens the public's understanding of the problem but also often leads to more people joining activist movements. Therefore, it is important for ENGOs to engage in data collection, analysis, and publication (in popular media and social media but also scientific journals) and to work with scientists because it increases their credibility and the issue's credibility.

Third, we observed in Taiwan that the "plastic reduction wave" really caught on in recent years because of the emerging citizen power and the increasing role of ENGOs in the people's democratic dialogue with its government. We therefore recommend that ENGOs in neighboring Asian countries use Taiwan's example to go beyond their usual focus of mainly promoting environmental education and beach cleanups and embrace policy advocacy and collaboration with governments as much as possible.

Fourth, we note that, to our knowledge, no research on nanoplastic particles [281–286] has been carried out in East Asia. Therefore, governments should fund this important new emerging research field.

Finally, we hope that this review will hopefully inspire a more concerted effort by East Asian governments to support the relevant science but also to tackle the plastic pollution problem with much needed policies and management solutions. For that to happen, we need more of everything which we described above (research, education, campaigns, government actions, etc.), but we would also advocate for much more interregional collaborations between scientists, ENGOs, and governments.

Acknowledgments We thank Bien Chung-Pei, Simona Grano, Alexander Kunz, Po-Hsiu Kuo, Liu Yong-Long, Won Joon Shim, and Joanna Vince for providing references and additional information and Yu-Wen Emily Dai for lots of translation. HL thanks Lee Bokman and Lim Heesuk for logistical support during this study.

References

1. Thompson RC, Moore CJ, vom Saal FS, Swan SH (2009) Plastics, the environment and human health: current consensus and future trends. Philos Trans R Soc Lond B 364(1526):2153–2166
2. Zeng EY (2018) Microplastic contamination in aquatic environments: an emerging matter of environmental urgency. Elsevier, Amsterdam
3. Wagner M, Lambert S (2018) Freshwater microplastics: emerging environmental contaminants? Springer, Berlin
4. Barboza LGA, Gimenez BCG (2015) Microplastics in the marine environment: current trends and future perspectives. Mar Pollut Bull 97(1–2):5–12
5. Bergmann M, Gutow L, Klages M (2015) Marine anthropogenic litter. Springer, Berlin
6. Wang J, Peng J, Tan Z, Gao Y, Zhan Z, Chen Q, Cai L (2017) Microplastics in the surface sediments from the Beijiang River littoral zone: composition, abundance, surface textures and interaction with heavy metals. Chemosphere 171(1):248–258
7. Geyer R, Jambeck JR, Law KL (2017) Production, use, and fate of all plastics ever made. Sci Adv 3(7):e1700782
8. PlasticsEurope (2017) Plastics – the facts 2017: an analysis of European latest plastics production, demand and waste data. PlasticsEurope. http://www.plasticseurope.org/application/files/5715/1717/4180/Plastics_the_facts_2017_FINAL_for_website_one_page.pdf. Accessed 1 Oct 2019
9. Lebreton L, Andrady A (2019) Future scenarios of global plastic waste generation and disposal. Palgrave Commun 5(1):6
10. Jambeck JR, Geyer R, Wilcox C, Siegler TR, Perryman M, Andrady A, Narayan R, Law KL (2015) Plastic waste inputs from land into the ocean. Science 347(6223):768–771
11. Jang YC, Hong S, Lee J, Lee MJ, Shim WJ (2014) Estimation of lost tourism revenue in Geoje Island from the 2011 marine debris pollution event in South Korea. Mar Pollut Bull 81(1):49–54
12. Gregory MR (2009) Environmental implications of plastic debris in marine settings-entanglement, ingestion, smothering, hangers-on, hitch-hiking and alien invasions. Philos Trans R Soc Lond B 364(1526):2013–2025
13. Krelling AP, Williams AT, Turra A (2017) Differences in perception and reaction of tourist groups to beach marine debris that can influence a loss of tourism revenue in coastal areas. Mar Policy 85(1):87–99
14. Koelmans AA, Besseling E, Foekema EM (2014) Leaching of plastic additives to marine organisms. Environ Pollut 187(2):49–54
15. Seltenreich N (2015) New link in the food chain? Marine plastic pollution and seafood safety. Environ Health Perspect 123(2):A34–A41
16. Wilcox C, van Sebille E, Hardesty BD (2015) Threat of plastic pollution to seabirds is global, pervasive, and increasing. Proc Natl Acad Sci U S A 112(38):11899–11904
17. Dehaut A, Cassone A-L, Frère L, Hermabessiere L, Himber C, Rinnert E, Rivière G, Lambert C, Soudant P, Huvet A, Duflos G, Paul-Pont I (2016) Microplastics in seafood: benchmark protocol for their extraction and characterization. Environ Pollut 215(3):223–233
18. Rochman CM, Tahir A, Williams SL, Baxa DV, Lam R, Miller JT, Teh F-C, Werorilangi S, Teh SJ (2015) Anthropogenic debris in seafood: plastic debris and fibers from textiles in fish and bivalves sold for human consumption. Sci Rep 5(1):14340
19. Talsness CE, Andrade AJM, Kuriyama SN, Taylor JA, vom Saal FS (2009) Components of plastic: experimental studies in animals and relevance for human health. Philos Trans R Soc Lond B 364(1526):2079–2096
20. Vethaak AD, Leslie HA (2016) Plastic debris is a human health issue. Environ Sci Technol 50(13):6825–6826

21. Lee J, Lee JS, Jang YC, Hong SY, Shim WJ, Song YK, Hong SH, Jang M, Han GM, Kang D (2015) Distribution and size relationships of plastic marine debris on beaches in South Korea. Arch Environ Contam Toxicol 69(3):288–298
22. Chae D-H, Kim I-S, Kim S-K, Song YK, Shim WJ (2015) Abundance and distribution characteristics of microplastics in surface seawaters of the Incheon/Kyeonggi coastal region. Arch Environ Contam Toxicol 69(3):269–278
23. Song YK, Hong SH, Jang M, Han GM, Shim WJ (2015) Occurrence and distribution of microplastics in the sea surface microlayer in Jinhae Bay, South Korea. Arch Environ Contam Toxicol 69(3):279–287
24. Wang MH, He YD, Sen B (2019) Research and management of plastic pollution in coastal environments of China. Environ Pollut 248(2):898–905
25. Lebreton LC, van der Zwet J, Damsteeg J-W, Slat B, Andrady A, Reisser J (2017) River plastic emissions to the world's oceans. Nat Commun 8(1):15611
26. van Sebille E, Wilcox C, Lebreton L, Maximenko N, Hardesty BD, van Franeker JA, Eriksen M, Siegel D, Galgani F, Law KL (2015) A global inventory of small floating plastic debris. Environ Res Lett 10(2):124006
27. Eriksen M, Lebreton LCM, Carson HS, Thiel M, Moore CJ, Borerro JC, Galgani F, Ryan PG, Reisser J (2014) Plastic pollution in the world's oceans: more than 5 trillion plastic pieces weighing over 250,000 tons afloat at sea. Public Libr Sci One 9(12):e111913
28. Bancin LJ, Walther BA, Lee Y-C, Kunz A (2019) Two-dimensional distribution and abundance of micro- and mesoplastic pollution in the surface sediment of Xialiao Beach, New Taipei City, Taiwan. Mar Pollut Bull 140(1):75–85
29. Walther BA, Kunz A, Hu C-S (2018) Type and quantity of coastal debris pollution in Taiwan: a 12-year nationwide assessment using citizen science data. Mar Pollut Bull 135(2):862–872
30. Cho Y, Shim WJ, Jang M, Han GM, Hong SH (2019) Abundance and characteristics of microplastics in market bivalves from South Korea. Environ Pollut 245(4):1107–1116
31. Teng J, Wang Q, Ran W, Wu D, Liu YF, Sun S, Liu H, Cao RW, Zhao JM (2019) Microplastic in cultured oysters from different coastal areas of China. Sci Total Environ 653(1):1282–1292
32. Niaounakis M (2017) Management of marine plastic debris: prevention, recycling, and waste management. Elsevier, Amsterdam
33. Borrelle SB, Rochman CM, Liboiron M, Bond AL, Lusher A, Bradshaw H, Provencher JF (2017) Opinion: why we need an international agreement on marine plastic pollution. Proc Natl Acad Sci 114(38):9994–9997
34. McKinsey & Company and Ocean Conservancy (2015) Stemming the tide: land-based strategies for a plastic-free ocean. McKinsey & Company and Ocean Conservancy. https://www.marinelittersolutions.com/wp-content/uploads/2015/10/Ocean_Conservatory_report.pdf. Accessed 1 Oct 2019
35. Chen C-L (2015) Regulation and management of marine litter. In: Bergmann M, Gutow L, Klages M (eds) Marine anthropogenic litter. Springer, Berlin, pp 395–428
36. Vince J, Hardesty BD (2017) Plastic pollution challenges in marine and coastal environments: from local to global governance. Restor Ecol 25(1):123–128
37. Xanthos D, Walker TR (2017) International policies to reduce plastic marine pollution from single-use plastics (plastic bags and microbeads): a review. Mar Pollut Bull 118(2):17–26
38. Mendenhall E (2018) Oceans of plastic: a research agenda to propel policy development. Mar Policy 96(3):291–298
39. Schuyler Q, Hardesty BD, Lawson TJ, Opie K, Wilcox C (2018) Economic incentives reduce plastic inputs to the ocean. Mar Policy 96(1):250–255
40. Hartley BL, Thompson RC, Pahl S (2015) Marine litter education boosts children's understanding and self-reported actions. Mar Pollut Bull 90(1–2):209–217
41. Chen CL, Liu TK (2013) Fill the gap: developing management strategies to control garbage pollution from fishing vessels. Mar Policy 40(3):34–40
42. Löhr A, Savelli H, Beunen R, Kalz M, Ragas A, van Belleghem F (2017) Solutions for global marine litter pollution. Curr Opin Environ Sustain 28(2):90–99

43. Bertling J, Bertling R, Hamann L (2018) Kunststoffe in der Umwelt: Mikro- und Makroplastik. Ursachen, Mengen, Umweltschicksale, Wirkungen, Lösungsansätze, Empfehlungen. Fraunhofer-Institut. https://www.umsicht.fraunhofer.de/content/dam/umsicht/de/dokumente/publikationen/2018/kunststoffe-id-umwelt-konsortialstudie-mikroplastik.pdf. Accessed 1 Oct 2019
44. Ten Brink P, Lutchman I, Bassi S, Speck S, Sheavly S, Register K, Woolaway C (2009) Guidelines on the use of market-based instruments to address the problem of marine litter. Institute for European Environmental Policy (IEEP). http://minisites.ieep.eu/assets/477/Economic_Instruments_and_Marine_Litter.pdf. Accessed 1 Oct 2019
45. Jeftic L, Sheavly S, Adler E (2009) Marine litter: a global challenge. United Nations Environment Programme/Intergovernmental Oceanographic Commission. https://wedocs.unep.org/bitstream/handle/20.500.11822/10744/MarineLitterAglobalChallenge.pdf?sequence=1&isAllowed=y. Accessed 1 Oct 2019
46. UNEP (2005) Marine litter: an analytical overview. United Nations Environment Programme. http://wedocs.unep.org/handle/20.500.11822/8348. Accessed 1 Oct 2019
47. UNEP (2016) Marine plastic debris and microplastics – Global lessons and research to inspire action and guide policy change. United Nations Environment Programme. https://wedocs.unep.org/rest/bitstreams/11700/retrieve. Accessed 1 Oct 2019
48. UNEP (2017) Exploring the potential for adopting alternative materials to reduce marine plastic litter. United Nations Environment Programme. http://wedocs.unep.org/bitstream/handle/20.500.11822/25485/plastic_alternative.pdf?sequence=1&isAllowed=. Accessed 1 Oct 2019
49. Ecovative (2019) Packaging: 100% compostable packaging for your products. https://shop.ecovativedesign.com/collections/packaging. Accessed 1 Oct 2019
50. Ocean Conservancy (2018) Fighting for trash free seas. Cleanup reports. https://oceanconservancy.org/trash-free-seas/international-coastal-cleanup/annual-data-release/. Accessed 1 Oct 2019
51. Slat B (2014) How the oceans can clean themselves: a feasibility study. Ocean Cleanup Foundation. https://www.theoceancleanup.com/fileadmin/media-archive/Documents/TOC_Feasibility_study_lowres_V2_0.pdf. Accessed 1 Oct 2019
52. Wu C, Zang K, Xiong X (2018) Microplastic pollution in inland waters focusing on Asia, in Freshwater microplastics: emerging environmental contaminants? In: Wagner M, Lambert S (eds). Springer, Berlin, pp 85–99
53. Zhang K, Shi HH, Peng JP, Wang YH, Xiong X, Wu CX, Lam PKS (2018) Microplastic pollution in China's inland water systems: a review of findings, methods, characteristics, effects, and management. Sci Total Environ 630(3):1641–1653
54. Zhao SY, Wang T, Zhu LX, Xu P, Wang XH, Gao L, Li DJ (2019) Analysis of suspended microplastics in the Changjiang Estuary: implications for riverine plastic load to the ocean. Water Res 161(4):560–569
55. Chen H, Wang S, Guo H, Lin H, Zhang Y, Long Z, Huang H (2019) Study of marine debris around a tourist city in East China: implication for waste management. Sci Total Environ 676(3):278–289
56. Zheng YF, Li JX, Cao W, Liu XH, Jiang FH, Ding JF, Yin XF, Sun CJ (2019) Distribution characteristics of microplastics in the seawater and sediment: a case study in Jiaozhou Bay, China. Sci Total Environ 674(4):27–35
57. Ding L, Mao RF, Guo XT, Yang XM, Zhang Q, Yang C (2019) Microplastics in surface waters and sediments of the Wei River, in the northwest of China. Sci Total Environ 667(2):427–434
58. Xiong X, Wu CX, Elser JJ, Mei ZG, Hao YJ (2019) Occurrence and fate of microplastic debris in middle and lower reaches of the Yangtze River – from inland to the sea. Sci Total Environ 659(1):66–73

59. Zhu JM, Zhang Q, Li YP, Tan SD, Kang ZJ, Yu XY, Lan WL, Cai L, Wang JZ, Shi HH (2019) Microplastic pollution in the Maowei Sea, a typical mariculture bay of China. Sci Total Environ 658(2):62–68
60. Zhao S, Zhu L, Li D (2015) Characterization of small plastic debris on tourism beaches around the South China Sea. Reg Stud Mar Sci 1(2):55–62
61. Zhang B, Wu D, Yang X, Teng J, Liu YL, Zhang C, Zhao JM, Yin XN, You LP, Liu YF, Wang Q (2019) Microplastic pollution in the surface sediments collected from Sishili Bay, North Yellow Sea, China. Mar Pollut Bull 141(4):9–15
62. Li YF, Wolanski E, Dai ZF, Lambrechts J, Tang C, Zhang H (2018) Trapping of plastics in semi-enclosed seas: Insights from the Bohai Sea, China. Mar Pollut Bull 137(1):509–517
63. Wu N, Zhang Y, Zhang XH, Zhao Z, He JH, Li WP, Ma YZ, Niu Z (2019) Occurrence and distribution of microplastics in the surface water and sediment of two typical estuaries in Bohai Bay, China. Environ Sci Processes Impacts 21(7):1143–1152
64. Fan YJ, Zheng K, Zhu ZW, Chen GS, Peng XZ (2019) Distribution, sedimentary record, and persistence of microplastics in the Pearl River catchment, China. Environ Pollut 251(3):862–870
65. Jiang CB, Yin LS, Li ZW, Wen XF, Luo X, Hu SP, Yang HY, Long YN, Deng B, Huang LZ, Liu YZ (2019) Microplastic pollution in the rivers of the Tibet Plateau. Environ Pollut 249(2):91–98
66. Wang T, Zou XQ, Li BJ, Yao YL, Zang Z, Li YL, Yu WW, Wang WZ (2019) Preliminary study of the source apportionment and diversity of microplastics: Taking floating microplastics in the South China Sea as an example. Environ Pollut 245(3):965–974
67. Zhang CF, Zhou HH, Cui YZ, Wang CS, Li YH, Zhang DD (2019) Microplastics in offshore sediment in the Yellow Sea and East China Sea, China. Environ Pollut 244(1):827–833
68. Yan MT, Nie HY, Xu KH, He YH, Hu YT, Huang YM, Wang J (2019) Microplastic abundance, distribution and composition in the Pearl River along Guangzhou city and Pearl River estuary, China. Chemosphere 217(4):879–886
69. Zhang LL, Zhang SP, Wang YH, Yu KF, Li RL (2019) The spatial distribution of microplastic in the sands of a coral reef island in the South China Sea: comparisons of the fringing reef and atoll. Sci Total Environ 688(2):780–786
70. Cheang CC, Ma Y, Fok L (2018) Occurrence and composition of microplastics in the seabed sediments of the coral communities in proximity of a metropolitan area. Int J Environ Res Public Health 15(10):12
71. Ding JF, Jiang FH, Li JX, Wang ZX, Sun CJ, Wang ZY, Fu L, Ding NX, He CF (2019) Microplastics in the coral reef systems from Xisha islands of South China Sea. Environ Sci Technol 53(14):8036–8046
72. Crawford CB, Quinn B (2017) Microplastic pollutants. Elsevier, Amsterdam
73. Peng X, Dasgupta S, Zhong G, Du M, Xu H, Chen M, Chen S, Ta K, Li J (2019) Large debris dumps in the northern South China Sea. Mar Pollut Bull 142(1):164–168
74. Liu SL, Jian MF, Zhou LY, Li WH (2019) Distribution and characteristics of microplastics in the sediments of Poyang Lake, China. Water Sci Technol 79(10):1868–1877
75. Wang WF, Yuan WK, Chen YL, Wang J (2018) Microplastics in surface waters of Dongting Lake and Hong Lake, China. Sci Total Environ 633(4):539–545
76. Wen XF, Du CY, Xu P, Zeng GM, Huang DL, Yin LS, Yin QD, Hu L, Wan J, Zhang JF, Tan SY, Deng R (2018) Microplastic pollution in surface sediments of urban water areas in Changsha, China: abundance, composition, surface textures. Mar Pollut Bull 136(3):414–423
77. Yin LS, Jiang CB, Wen XF, Du CY, Zhong W, Feng ZQ, Long YN, Ma Y (2019) Microplastic pollution in surface water of urban lakes in Changsha, China. Int J Environ Res Public Health 16(9):10
78. Jiang CB, Yin LS, Wen XF, Du CY, Wu LX, Long YN, Liu YZ, Ma Y, Yin QD, Zhou ZY, Pan HM (2018) Microplastics in sediment and surface water of West Dongting Lake and South Dongting Lake: abundance, source and composition. Int J Environ Res Public Health 15 (10):15

79. Yuan WK, Liu XN, Wang WF, Di MX, Wang J (2019) Microplastic abundance, distribution and composition in water, sediments, and wild fish from Poyang Lake, China. Ecotoxicol Environ Saf 170(1):180–187
80. Li J, Zhang H, Zhang KN, Yang RJ, Li RZ, Li YF (2018) Characterization, source, and retention of microplastic in sandy beaches and mangrove wetlands of the Qinzhou Bay, China. Mar Pollut Bull 136(2):401–406
81. Li RL, Zhang LL, Xue BM, Wang YH (2019) Abundance and characteristics of microplastics in the mangrove sediment of the semi-enclosed Maowei Sea of the south China sea: new implications for location, rhizosphere, and sediment compositions. Environ Pollut 244(2):685–692
82. Di MX, Liu XN, Wang WF, Wang J (2019) Manuscript prepared for submission to environmental toxicology and pharmacology pollution in drinking water source areas: microplastics in the Danjiangkou Reservoir, China. Environ Toxicol Pharmacol 65(3):82–89
83. Lv WQ, Zhou WZ, Lub SB, Huang WW, Yuan Q, Tian ML, Lv WG, He DF (2019) Microplastic pollution in rice-fish co-culture system: a report of three farmland stations in Shanghai, China. Sci Total Environ 652(4):1209–1218
84. Zhu L, Wang H, Chen BJ, Sun XM, Qu KM, Xia B (2019) Microplastic ingestion in deep-sea fish from the South China Sea. Sci Total Environ 677(4):493–501
85. Mohsen M, Wang Q, Zhang LB, Sun LN, Lin CG, Yang HS (2019) Heavy metals in sediment, microplastic and sea cucumber *Apostichopus japonicus* from farms in China. Mar Pollut Bull 143(1):42–49
86. Zheng K, Fan YJ, Zhu ZW, Chen GS, Tang CM, Peng XZ (2019) Occurrence and species-specific distribution of plastic debris in wild freshwater fish from the Pearl River Catchment, China. Environ Toxicol Chem 38(7):1504–1513
87. Sun XX, Li QJ, Shi YQ, Zhao YF, Zheng S, Liang JH, Liu T, Tian ZY (2019) Characteristics and retention of microplastics in the digestive tracts of fish from the Yellow Sea. Environ Pollut 249(2):878–885
88. Li H-X, Ma L-S, Lin L, Ni Z-X, Xu X-R, Shi H-H, Yan Y, Zheng G-M, Rittschof D (2018) Microplastics in oysters *Saccostrea cucullata* along the Pearl River Estuary, China. Environ Pollut 236(2):619–625
89. Su L, Deng H, Li BW, Chen QQ, Pettigrove V, Wu CX, Shi HH (2019) The occurrence of microplastic in specific organs in commercially caught fishes from coast and estuary area of east China. J Hazard Mater 365(4):716–724
90. Mohsen M, Wang Q, Zhang LB, Sun L, Lin CG, Yang HS (2019) Microplastic ingestion by the farmed sea cucumber *Apostichopus japonicus* in China. Environ Pollut 245(3):1071–1078
91. Yang D, Shi H, Li L, Li J, Jabeen K, Kolandhasamy P (2015) Microplastic pollution in table salts from China. Environ Sci Technol 49(22):13622–13627
92. Lv XM, Dong Q, Zuo ZQ, Liu YC, Huang X, Wu WM (2019) Microplastics in a municipal wastewater treatment plant: fate, dynamic distribution, removal efficiencies, and control strategies. J Clean Prod 225(2):579–586
93. Lei K, Qiao F, Liu Q, Wei Z, Qi H, Cui S, Yue X, Deng Y, An L (2017) Microplastics releasing from personal care and cosmetic products in China. Mar Pollut Bull 123(1):122–126
94. Cheung PK, Fok L (2017) Characterisation of plastic microbeads in facial scrubs and their estimated emissions in Mainland China. Water Res 122(3):53–61
95. Bai MY, Zhu LX, An LH, Peng GY, Li DJ (2018) Estimation and prediction of plastic waste annual input into the sea from China. Acta Oceanol Sin 37(11):26–39
96. Wang T, Li BJ, Zou XQ, Wang Y, Li YL, Xu YJ, Mao LJ, Zhang CC, Yu WW (2019) Emission of primary microplastics in mainland China: invisible but not negligible. Water Res 162(2):214–224
97. Lo HS, Wong CY, Tam NFY, Cheung SG (2019) Spatial distribution and source identification of hydrophobic organic compounds (HOCs) on sedimentary microplastic in Hong Kong. Chemosphere 219(3):418–426

98. Zhang W, Ma X, Zhang Z, Wang Y, Wang J, Wang J, Ma D (2015) Persistent organic pollutants carried on plastic resin pellets from two beaches in China. Mar Pollut Bull 99(1–2):28–34
99. Xu XY, Wang S, Gao FL, Li JX, Zheng L, Sun CJ, He CF, Wang ZX, Qu LY (2019) Marine microplastic-associated bacterial community succession in response to geography, exposure time, and plastic type in China's coastal seawaters. Mar Pollut Bull 145(4):278–286
100. State Oceanic Administration of the People's Republic of China (2008) 2007 China Marine environmental quality bulletin (in Chinese). State Oceanic Administration of the People's Republic of China. http://www.nmdis.org.cn/c/2011-04-14/56922.shtml. Accessed 1 Oct 2019
101. CBCGDF (2018) How to manage marine microplastic? CBCGDF attended the 2018 APEC workshop on innovative marine debris solutions (in Chinese). China Biodiversity Conservation and Green Development Foundation. http://www.chinadevelopmentbrief.org.cn/org3499/news-7859-1.html. Accessed 1 Oct 2019
102. State Oceanic Administration of the People's Republic of China (2018) 2017 China marine environmental quality bulletin (in Chinese). State Oceanic Administration of the People's Republic of China. http://hys.mee.gov.cn/dtxx/201905/P020190529532197736567.pdf. Accessed 1 Oct 2019
103. Kao M (2018) Ministry of ecology and environment: China's marine microplastic pollution is generally lower than the global average (in Chinese). 31 May: Xinhua Net. http://www.gov.cn/xinwen/2018-05/31/content_5295170.htm. Accessed 1 Oct 2019
104. UNEP Northwest Pacific Action Plan (NOWPAP) (2008) NOWPAP regional action plan on marine litter. NOWPAP Regional Coordinating Unit. http://marinelitternetwork.engr.uga.edu/wp-content/uploads/2017/07/NOWPAP_RAPMALI.pdf. Accessed 1 Oct 2019
105. PEMSEA (2018) SDS-SEA sustainable development strategy for the Seas of East Asia. Implementation plan 2018–2022. Partnerships in Environmental Management for the Seas of East Asia (PEMSEA). http://pemsea.org/sites/default/files/PEMSEA_SDS-SEA_IP_2018-2022_20190606.pdf. Accessed 1 Oct 2019
106. APEC (2005) APEC MRC 05/2005 project: the Seoul oceans declaration implementation across APEC economies. A study for use at the 2nd APEC Ocean Ministerial Meeting (16–17 September 2005). Asia-Pacific Economic Cooperation
107. McIlgorm A, Campbell HF, Rule MJ (2008) Understanding the economic benefits and costs of controlling marine debris in the APEC region (MRC 02/2007). A report to the Asia-Pacific Economic Cooperation Marine resource conservation working group by the National Marine Science Centre (University of New England and Southern Cross University), Coffs Harbour, NSW, Australia, December. National Marine Science Centre
108. ASEAN (2019) Overview of ASEAN plus three cooperation. https://asean.org/storage/2016/01/Overview-of-APT-Cooperation-Jul-2019_For-Web_Rev.pdf. Accessed 1 Oct 2019
109. Minister of Environment and Climate Change (2018) Canada-China joint statement on marine litter and plastics. Minister of Environment and Climate Change. https://www.canada.ca/content/dam/eccc/documents/pdf/international-affairs/compendium/2018/batch-12/canada-china-joint-statement-marine-litter-plastics-2018.pdf. Accessed 1 Oct 2019
110. He HR (2012) Effects of environmental policy on consumption: lessons from the Chinese plastic bag regulation. Environ Dev Econ 17(2):407–431
111. Newman S, Watkins E, Farmer A, Ten Brink P, Schweitzer J-P (2015) The economics of marine litter. In: Bergmann M, Gutow L, Klages M (eds) Marine anthropogenic litter. Springer, Berlin, pp 367–394
112. Wang JJ, Zheng LX, Li JH (2018) A critical review on the sources and instruments of marine microplastics and prospects on the relevant management in China. Waste Manag Res 36(10):898–911
113. CCICED (2019) Special Policy Study on Global Ocean Governance and Ecological Civilization. CCICED task force report. China Council for International Cooperation on Environment and Development. https://www.iisd.org/sites/default/files/publications/cciced/agm/cciced-sps-1-3-oceans.pdf. Accessed 1 Oct 2019

114. Lao H (2000) Implementation of "The Marine Environmental Protection Law of P.R.C., 1999" and related international pollution conventions. (in Chinese). Traffic Environ Protect 6(1):1–6
115. Hart, M. and Cavanagh J., 2012. Environmental standards give the United States an edge over China. 20 April: Center for American Progress. https://www.americanprogress.org/issues/green/news/2012/04/20/11503/environmental-standards-give-the-united-states-an-edge-over-china/. Accessed 1 Oct 2019
116. Walther BA, White A (2018) The emergence of birdwatching in China: history, demographics, activities, motivations, and environmental concerns of Chinese birdwatchers. Bird Conserv Int 28(3):337–349
117. Xinhua Net (2019) Why are the locals still unclear about waste sorting after 19 years? What are the difficulties? (in Chinese). 2 July: Xinhua Net. http://www.xinhuanet.com/politics/2019-07/02/c_1210176190.htm. Accessed 1 Oct 2019
118. Ministry of Construction of the People's Republic of China (2000) Notice on the publication of pilot cities for the collection of domestic waste (in Chinese). Ministry of Construction of the People's Republic of China
119. Nanjing City Government (2013) The ordinary/regulation of managing waste sorting of municipal solid waste in Nanjing City (in Chinese). Nanjing City Government
120. Qiu, Z.Y., 2018. What is the difference between the classification of Chinese and foreign waste? (in Chinese). 26 March: Point Green Network-Green Industry. http://news.inggreen.com/47060.html. Accessed 1 Oct 2019
121. Shanghai City Government (2014) The regulation to promote reduction and waste sorting of municipal solid waste in Shanghai (in Chinese). http://www.shanghai.gov.cn/nw2/nw2314/nw2319/nw2407/nw32093/u26aw38774.html. Accessed 1 Oct 2019
122. RFA (2019) Green Intelligence: Shanghai's waste sorting can't stop the incinerator? 18 July: Radio Free Asia. https://www.rfa.org/mandarin/zhuanlan/luseqingbaoyuan/grn-07182019104910.html. Accessed 1 Oct 2019
123. Wu YX (2019) Shanghai's compulsory waste sorting begins (in Chinese). 2 July: China Dialogue. https://www.chinadialogue.net/article/show/single/ch/11349-Shanghai-starts-separating-waste. Accessed 1 Oct 2019
124. Ren YC, Chang HF, Kao ST (2018) Twenty years of bumpy roads for garbage collection (in Chinese). Oriental Outlook 30(3):18–27
125. Peng Y, Li L, Peng X-Y, Yang P-J, Zhao X-F, Ma Y (2018) A review of municipal solid waste classification in China: development status, problems and countermeasures (in Chinese). China Environ 38(10):3874–3879
126. Qi YM (2017) How do Germany and Switzerland sort their municipal solid waste? (in Chinese). 16 October: Green Shanghai. http://lhsr.sh.gov.cn/sites/lhsr-m/ViewCon.ashx?ctgid=3a222e8f-e1e5-4737-8697-41de8c789e31&infid=793096f8-f3e2-417e-a8dd-bdc5a1703cdb. Accessed 1 Oct 2019
127. State Council of the People's Republic of China (2016) The 13th five-year plan for the protection of the ecological environment (in Chinese). State Council of the People's Republic of China. http://www.gov.cn/2015-06/03/content_2872658.htm. Accessed 1 Oct 2019
128. State Council of the People's Republic of China (2015) Integrated reform plan for promoting ecological progress (Announcement no. 12, 2015) (in Chinese). State Council of the People's Republic of China. http://www.gov.cn/2015-06/03/content_2872658.htm. Accessed 1 Oct 2019
129. Ministry of Housing and Urban-Rural Development of the People's Republic of China (2017) Implementation plan of the household waste sorting system (in Chinese). Ministry of Housing and Urban-Rural Development of the People's Republic of China. http://www.ndrc.gov.cn/zcfb/zcfbqt/201703/W020170331310014947378.pdf. Accessed 1 Oct 2019
130. Lee MJ (2019) Summary and interpretation of the policy of waste sorting at the national level and in key cities in 2019 (in Chinese). http://news.inggreen.com/60195.html. Accessed 1 Oct 2019

131. Chao BJ (2017) The plastic bag limitation order is not a “selling order” (in Chinese). 9 June: People’s Daily http://opinion.people.com.cn/n1/2017/0609/c1003-29327933.html. Accessed 1 Oct 2019
132. China Zero Waste Alliance (2018) The investigative report on the implementation by retailers of the plastic bag limitation order’s tenth anniversary (in Chinese). China Zero Waste Alliance. http://www.lingfeiqi.org/sites/default/files/datadoc/shi_nian_xian_su_ling_shang_jia_zhi_xing_qing_kuang_diao_yan_bao_gao_.pdf. Accessed 1 Oct 2019
133. Brooks AL, Wang S, Jambeck JR (2018) The Chinese import ban and its impact on global plastic waste trade. Sci Adv 4(6):eaat0131
134. State Council Information Office of the People's Republic of China (2014) The general administration of customs interprets the green fence operation, press release 2014/1/10 (in Chinese). State Council of the People’s Republic of China. http://www.scio.gov.cn/ztk/xwfb/2014/gxbxwfbhjs2013nqnjckqk/zcfg30113/Document/1359295/1359295.htm. Accessed 1 Oct 2019
135. Anonymous (2019) China’s new waste and recycling policy. Green Industries South Australia. https://www.greenindustries.sa.gov.au/chinas-new-policy-on-waste-and-recycling. Accessed 1 Oct 2019
136. Ministry of Ecology and Environment of the People’s Republic of China (2017) Announcement of releasing the catalogues of imported waste management (Announcement no. 39, 2017) (in Chinese). Ministry of Ecology and Environment of the People’s Republic of China. http://www.mee.gov.cn/gkml/hbb/bgg/201708/t20170817_419811.htm. Accessed 1 Oct 2019
137. Ministry of Ecology and Environment of the People’s Republic of China (2018) Announcement of revising the catalogues of imported waste management (Announcement no. 68, 2018) (in Chinese). Ministry of Ecology and Environment of the People's Republic of China. http://www.mee.gov.cn/xxgk2018/xxgk/xxgk01/201812/t20181227_687488.html. Accessed 1 Oct 2019
138. McNaughton S, Nowakowski K (2019) How China’s plastic waste ban forced a global recycling reckoning. Natl Geogr 6:1–10
139. NOWPAP (2013) Best practices in dealing with marine litter in fisheries, aquaculture and shipping sectors in the NOWPAP Region. Northwest Pacific Action Plan Marine Environmental Emergency Preparedness and Response Regional Activity Center (NOWPAP MERRAC). http://wedocs.unep.org/bitstream/handle/20.500.11822/26219/best_practice_deal_ML.pdf?sequence=1&isAllowed=y. Accessed 1 Oct 2019
140. State Council of the People’s Republic of China (2016) Opinions on the implementation of the River Leader system in an all-round way (in Chinese). State Council of the People’s Republic of China. http://big5.www.gov.cn/gate/big5/www.gov.cn/gongbao/content/2017/content_5156731.htm. Accessed 1 Oct 2019
141. State Oceanic Administration of the People’s Republic of China (2017) Guiding opinions on launching the pilot work of the "Bayer System" (in Chinese). State Oceanic Administration of the People's Republic of China. http://big5.www.gov.cn/gate/big5/www.gov.cn/xinwen/2017-09/14/content_5224996.htm. Accessed 1 Oct 2019
142. State Council of the People’s Republic of China (2018) Opinions on the implementation of the Lake Leader system (in Chinese). State Council of the People’s Republic of China. http://www.gov.cn/zhengce/2018-01/04/content_5253253.htm. Accessed 1 Oct 2019
143. Shanghai Rendu Ocean NGO Development Center (2016) Monitoring research report about marine debris on some typical Chinese beaches (in Chinese). Shanghai Rendu Ocean NGO Development Center. http://www.renduocean.org/yanjiuchengguo/ccmc_report_2015.pdf. Accessed 1 Oct 2019
144. Liu Q (2018) China promises restrictions on plastic waste. 21 February: Chinadialogue. https://www.chinadialogue.net/article/show/single/en/10412-China-promises-restrictions-on-plastic-waste. Accessed 1 Oct 2019
145. Shanghai Rendu Ocean NGO Development Center (2019) Shanghai Rendu Ocean NGO development center website (in Chinese). Shanghai Rendu Ocean NGO Development Center. http://www.renduocean.org/environmentally1.html. Accessed 1 Oct 2019

146. Zhao K (2017) China's environmental woes, in films that go viral, then vanish. 28 April: New York Times. https://www.nytimes.com/2017/04/28/world/asia/chinas-environmental-woes-in-films-that-go-viral-then-vanish.html. Accessed 1 Oct 2019
147. Chen A (2014) Filmmaker behind Beijing waste exposé turns attention to trash trade. 13 January: South China Morning Post. https://www.scmp.com/lifestyle/article/1402419/filmmaker-behind-beijing-waste-expose-turns-attention-trash-trade. Accessed 1 Oct 2019
148. Yamashita R, Tanimura A (2007) Floating plastic in the Kuroshio Current area, western North Pacific Ocean. Mar Pollut Bull 54(4):485–488
149. Shiomoto A, Kameda T (2005) Distribution of manufactured floating marine debris in near-shore areas around Japan. Mar Pollut Bull 11(50):1430–1432
150. Matsumura S, Nasu K (1997) Distribution of floating debris in the North Pacific Ocean: sighting surveys 1986-1991. In: Coe JM, Rogers DB (eds) Marine debris: sources, impacts, and solutions. Springer-Verlag, pp 19–24
151. Nakashima E, Isobe A, Kako Si, Itai T, Takahashi S (2012) Quantification of toxic metals derived from macroplastic litter on Ookushi Beach, Japan. Environ Sci Technol 46(18):10099–10105
152. Kataoka T, Hinata H, Kato S (2013) Analysis of a beach as a time-invariant linear input/output system of marine litter. Mar Pollut Bull 77(1–2):266–273
153. Shimizu T, Nakai J, Nakajima K, Kozai N, Takahashi G, Matsumoto M, Kikui J (2008) Seasonal variations in coastal debris on Awaji Island, Japan. Mar Pollut Bull 57(1–5):182–186
154. Ogi H, Fukumoto Y (2000) A sorting method for small plastic debris floating on the sea surface and stranded on sandy beaches (in Japanese). Bull Fac Fisher Hokkaido Univ 51(2):71–93
155. Kusui T, Noda M (2003) International survey on the distribution of stranded and buried litter on beaches along the Sea of Japan. Mar Pollut Bull 47(1–6):175–179
156. Northwest Pacific Region Environmental Cooperation Center (2018) NEAR project report of survey of washed-up driftage on beaches (in Japanese). http://www.npec.or.jp/umigomiportal/result/document/result_abst_2017.pdf. Accessed 1 Oct 2019
157. Ogi H (1990) Ingestion of plastic particles by sooty and short-tailed seawaters in the North Pacific. In: Shomura RS, Godfrey ML (eds.) Proceedings of the second international conference of Marine Debris, 2–7 April 1989, Honolulu, Hawaii. NOAA-TM-NMFS-SWFSC-154, Honolulu, Hawaii, USA, pp 635–652. https://repository.library.noaa.gov/view/noaa/6012. Accessed 1 Oct 2019
158. Mato Y, Isobe T, Takada H, Kanehiro H, Ohtake C, Kaminuma T (2001) Plastic resin pellets as a transport medium for toxic chemicals in the marine environment. Environ Sci Technol 35(2):318–324
159. Endo S, Takizawa R, Okuda K, Takada H, Chiba K, Kanehiro H, Ogi H, Yamashita R, Date T (2005) Concentration of polychlorinated biphenyls (PCBs) in beached resin pellets: variability among individual particles and regional differences. Mar Pollut Bull 50(10):1103–1114
160. Ogata Y, Takada H, Mizukawa K, Hirai H, Iwasa S, Endo S, Mato Y, Saha M, Okuda K, Nakashima A (2009) International pellet watch: global monitoring of persistent organic pollutants (POPs) in coastal waters. 1. Initial phase data on PCBs, DDTs, and HCHs. Mar Pollut Bull 58(10):1437–1446
161. Teuten EL, Saquing JM, Knappe DRU, Barlaz MA, Jonsson S, Bjorn A, Rowland SJ, Thompson RC, Galloway TS, Yamashita R, Ochi D, Watanuki Y, Moore C, Pham HV, Tana TS, Prudente M, Boonyatumanond R, Zakaria MP, Akkhavong K, Ogata Y, Hirai H, Iwasa S, Mizukawa K, Hagino Y, Imamura A, Saha M, Takada H (2009) Transport and release of chemicals from plastics to the environment and to wildlife. Philos Trans R Soc Lond B 364(1526):2027–2045
162. JEAN (2018) Summary results of international coastal cleanup 2017 (in Japanese). Japan Environmental Action Network. http://www.jean.jp/activity/result/Jean2017shortR1_a.pdf. Accessed 1 Oct 2019
163. Fujieda S, Kojima A, Kanehiro H (2006) Monitoring marine debris using disposable lighters as an indicator (in Japanese). J Jpn Soc Waste Manage Exp 17(2):117–124

164. MOE (2009) Report on the study group for the model study on domestic reduction measures related to drifting and littering debris (in Japanese). Ministry of the Environment, Government of Japan. http://www.env.go.jp/water/marine_litter/h19_20model_gaiyo.pdf. Accessed 1 Oct 2019
165. Arthur C, Baker J, Bamford H (2009) Proceedings of the international research workshop on the occurrence, effects and fate of microplastic Marine Debris. Sept 9–11, 2008. NOAA Technical Memorandum NOS-OR&R-30. p. marinedebris.noaa.gov/file/2192/download?token=5dvqb-YY. Accessed 1 Oct 2019
166. Isobe A, Uchida K, Tokai T, Iwasaki S (2015) East Asian seas: a hot spot of pelagic microplastics. Mar Pollut Bull 101(2):618–623
167. Kudo K, Kataoka T, Nihei Y, Hinata H, Shimazaki H, Baba H (2017) Basic study for surveying method and distribution of microplastics in Japanese rivers (in Japanese). J Jpn Soc Civ Eng Ser B1 (Hydraul Eng) 7(4):I_1225–I_1230
168. Matsuguma Y, Takada H, Kumata H, Kanke H, Sakurai S, Suzuki T, Itoh M, Okazaki Y, Boonyatumanond R, Zakaria MP, Weerts S, Newman B (2017) Microplastics in sediment cores from Asia and Africa as indicators of temporal trends in plastic pollution. Arch Environ Contam Toxicol 73(2):230–239
169. Tanaka K, Takada H (2016) Microplastic fragments and microbeads in digestive tracts of planktivorous fish from urban coastal waters. Sci Rep 6(8):34351
170. Ushijima T, Tanaka S, Suzuki Y, Yukioka S, Wang M, Nabetani Y, Fujii S, Takada H (2018) Occurrence of microplastics in digestive tracts of fish with different modes of ingestion in Japanese bays and Lake Biwa (in Japanese). J Jpn Soc Water Environ 41(4):107–113
171. Isobe A (2016) Percentage of microbeads in pelagic microplastics within Japanese coastal waters. Mar Pollut Bull 110(1):432–437
172. Isobe A, Iwasaki S, Uchida K, Tokai T (2019) Abundance of non-conservative microplastics in the upper ocean from 1957 to 2066. Nat Commun 10(1):417
173. Matsuzaki Y, Sato K (2018) Efforts to combat marine litter in Japan and future tasks (in Japanese). Mater Cycles Waste Manage Res 29(4):278–285
174. MOE (2019) Document collection of materials about domestic and foreign situations surrounding plastics (in Japanese). Ministry of the Environment, Government of Japan. http://www.env.go.jp/council/03recycle/y0312-01/y031201-s1.pdf. Accessed 1 Oct 2019
175. JEAN (2018) Urge the Japanese Government to sign the G7 Ocean Plastics Charter! https://www.change.org/p/urge-the-japanese-government-to-sign-the-g7-ocean-plastics-charter. Accessed 1 Oct 2019
176. Government of Japan (2019) Resource Recycling Strategy for Plastics (in Japanese). Government of Japan. http://www.env.go.jp/press/files/jp/111747.pdf. Accessed 1 Oct 2019
177. MOE (2019) Outline of the system review for making plastic shopping bags to be charged (draft) (in Japanese). Ministry of the Environment. https://www.env.go.jp/recycle/y0313-01/d1.pdf. Accessed 1 Oct 2019
178. Japan Plastics Industry Federation and Plastic Related Organizations (1993) Resin pellet leakage prevention manual (in Japanese). http://www.jpif.gr.jp/9kankyo/conts/gl_roboshi2_c.pdf. Accessed 1 Oct 2019
179. MOE (2017) 2016 survey report on the distribution of microbeads in Japan and overseas (in Japanese). Ministry of the Environment, Government of Japan. http://www.env.go.jp/water/marirne_litter/microbeads.pdf#page=1&zoom=auto,-16,235. Accessed 1 Oct 2019
180. Lee DI, Cho HS, Jeong SB (2006) Distribution characteristics of marine litter on the sea bed of the East China Sea and the South Sea of Korea. Estuar Coastal Shelf Sci 70(1–2):187–194
181. Lee J, Hong S, Song YK, Hong SH, Jang YC, Jang M, Heo NW, Han GM, Lee MJ, Kang D, Shim WJ (2013) Relationships among the abundances of plastic debris in different size classes on beaches in South Korea. Mar Pollut Bull 77(1–2):349–354
182. Kang JH, Kwon OY, Lee KW, Song YK, Shim WJ (2015) Marine neustonic microplastics around the southeastern coast of Korea. Mar Pollut Bull 96(1–2):304–312
183. Kim S-G, Lee W-I, Yuseok M (2014) The estimation of derelict fishing gear in the coastal waters of South Korea: trap and gill-net fisheries. Mar Policy 46(3):119–122

184. Song YK, Hong SH, Jang M, Kang J-H, Kwon OY, Han GM, Shim WJ (2014) Large accumulation of micro-sized synthetic polymer particles in the sea surface microlayer. Environ Sci Technol 48(16):9014–9021
185. Song YK, Hong SH, Eo S, Jang M, Han GM, Isobe A, Shim WJ (2018) Horizontal and vertical distribution of microplastics in Korean coastal waters. Environ Sci Technol 52(21):12188–12197
186. Eo S, Hong SH, Song YK, Han GM, Shim WJ (2019) Spatiotemporal distribution and annual load of microplastics in the Nakdong River, South Korea. Water Res 160(3):228–237
187. Jang M, Shim WJ, Han GM, Song YK, Hong SH (2018) Formation of microplastics by polychaetes (*Marphysa sanguinea*) inhabiting expanded polystyrene marine debris. Mar Pollut Bull 131(4):365–369
188. Song YK, Hong SH, Jang M, Han GM, Jung SW, Shim WJ (2017) Combined effects of UV exposure duration and mechanical abrasion on microplastic fragmentation by polymer type. Environ Sci Technol 51(8):4368–4376
189. Davidson TM (2012) Boring crustaceans damage polystyrene floats under docks polluting marine waters with microplastic. Mar Pollut Bull 64(9):1821–1828
190. Lee H, Kim Y (2018) Treatment characteristics of microplastics at biological sewage treatment facilities in Korea. Mar Pollut Bull 137(2):1–8
191. Kim M, Hyun S, Kwon JH (2015) Estimation of the environmental load of high- and low-density polyethylene from South Korea using a mass balance approach. Arch Environ Contam Toxicol 69(3):367–373
192. Hong S, Lee J, Jang YC, Kim YJ, Kim HJ, Han D, Hong SH, Kang D, Shim WJ (2013) Impacts of marine debris on wild animals in the coastal area of Korea. Mar Pollut Bull 66(1–2):117–124
193. Kang JH, Kwon OY, Shim WJ (2015) Potential threat of microplastics to zooplanktivores in the surface waters of the Southern Sea of Korea. Arch Environ Contam Toxicol 69(3):340–351
194. Hong S, Lee J, Kang D, Choi H-W, Ko S-H (2014) Quantities, composition, and sources of beach debris in Korea from the results of nationwide monitoring. Mar Pollut Bull 84(4):27–34
195. Heo NW, Hong SH, Han GM, Hong S, Lee J, Song YK, Jang M, Shim WJ (2013) Distribution of small plastic debris in cross-section and high strandline on Heungnam Beach, South Korea. Ocean Sci J 48(2):225–233
196. Kim I-S, Chae D-H, Kim S-K, Choi S, Woo S-B (2015) Factors influencing the spatial variation of microplastics on high-tidal coastal beaches in Korea. Arch Environ Contam Toxicol 69(3):299–309
197. Jang YC, Lee J, Hong S, Lee JS, Shim WJ, Song YK (2014) Sources of plastic marine debris on beaches of Korea: more from the ocean than the land. Ocean Sci J 49(2):151–162
198. Eo S, Hong SH, Song YK, Lee J, Lee J, Shim WJ (2018) Abundance, composition, and distribution of microplastics larger than 20 μm in sand beaches of South Korea. Environ Pollut 238(1):894–902
199. Lee J, Lee J, Hong S, Hong SH, Shim WJ, Eo S (2017) Characteristics of meso-sized plastic marine debris on 20 beaches in Korea. Mar Pollut Bull 123(1–2):92–96
200. Jang M, Shim WJ, Han GM, Rani M, Song YK, Hong SH (2017) Widespread detection of a brominated flame retardant, hexabromocyclododecane, in expanded polystyrene marine debris and microplastics from South Korea and the Asia-Pacific coastal region. Environ Pollut 231(2):785–794
201. KOEM (2019) Purpose of establishment. https://www.koem.or.kr/site/eng/01/10101020000002019071509.jsp. Accessed 1 Oct 2019
202. An DH, Kim JI (2018) Proposing policy for the prevention of marine pollution from microplastics (in Korean). J Environ Policy Adm 26(3):77–102
203. Jang YC, Lee J, Hong S, Mok JY, Kim KS, Lee YJ, Choi HW, Kang H, Lee S (2014) Estimation of the annual flow and stock of marine debris in South Korea for management purposes. Mar Pollut Bull 86(1–2):505–511
204. Ministry of Ocean and Fisheries & Korea Marine Environment Management Corporation (2016) 2016 National Marine Waste Monitoring project management service. Final Report. Ministry of Ocean and Fisheries, Sejong City; Korea Marine Environment Management

Corporation, Seoul, Republic of Korea. https://www.malic.or.kr/file/download.do?no=610. Accessed 1 Oct 2019

205. Opfer S, Arthur C, Lippiatt P (2012) NOAA marine debris shoreline survey field guide. National Oceanic and Atmospheric Administration. https://marinedebris.noaa.gov/sites/default/files/ShorelineFieldGuide2012.pdf. Accessed 1 Oct 2019
206. Cho DO (2009) The incentive program for fishermen to collect marine debris in Korea. Mar Pollut Bull 58(3):415–417
207. Jung RT, Sung HG, Chun T-B, Keel S-I (2010) Practical engineering approaches and infrastructure to address the problem of marine debris in Korea. Mar Pollut Bull 60(9):1523–1532
208. Hong S, Lee J, Kang D (2015) Emergy evaluation of management measures for derelict fishing gears in Korea. Ocean Sci J 50(3):603–613
209. Choi EC, Lee JS (2018) The willingness to pay for removing the microplastics in the ocean – The case of Seoul metropolitan area, South Korea. Mar Policy 93(1):93–100
210. Park JH (2018) A study on the current status of microplastic pollution and its management methods in major countries (in Korean). J Water Policy Econ 31(3):77–88
211. Anonymous (2018) Gov't toughens recycling policy to reduce plastic waste. 10 May: Yonhap News Agency. https://en.yna.co.kr/view/AEN20180510008800315. Accessed 1 Oct 2019
212. Ju MS (2019) Comprehensive measure of waste recycling. Korea Environ Policy Bull 16(1):1–19
213. Lim S-Y, Chon K-P, Chung E (2018) Inside the chaos of Korea's plastic waste crisis. 6 April: Korea Joongang Daily. http://koreajoongangdaily.joins.com/news/article/article.aspx?aid=3046555. Accessed 1 Oct 2019
214. Kim E-J, Bulley J (2018) [Debriefing] Korea's cafe plastic purge. 17 September: Korea Joongang Daily. http://koreajoongangdaily.joins.com/news/article/article.aspx?aid=3053259. Accessed 1 Oct 2019
215. Kim E-J, Yonhap (2018) Gov't plan to cut waste would ban plastic cups. 5 September: Korea Joongang Daily. http://koreajoongangdaily.joins.com/news/article/article.aspx?aid=3052780. Accessed 1 Oct 2019
216. Kim GE (2019) Microplastic & environmental policy (in Korean). In: Korea environmental policy and administration society conference paper 2019(2): 15–17
217. Lee W (2018) South Korea bans disposable coffee cups, plastic bags. 1 August: United Press International (UPI). https://www.upi.com/Top_News/World-News/2018/08/02/South-Korea-bans-disposable-coffee-cups-plastic-bags/6841533181415/. Accessed 1 Oct 2019
218. Kim J-Y, Lee T-Y, Chung E (2018) Gov't begins crackdown on plastic cups. 1 August: Korea Joongang Daily. http://koreajoongangdaily.joins.com/news/article/article.aspx?aid=3051333. Accessed 1 Oct 2019
219. Bahk E-J (2019) Single-use plastic bags banned at stores. 1 April: Korea Times. https://www.koreatimes.co.kr/www/nation/2019/04/371_266418.html. Accessed 1 Oct 2019
220. Anonymous (2019) Ban on disposable plastic bags to take effect April 1. 27 March: Korea Times. https://www.koreatimes.co.kr/www/nation/2019/03/371_266148.html. Accessed 1 Oct 2019
221. Regan H, Kwon J (2018) South Korea bans single-use plastic bags from major supermarkets. 31 December: CNN. https://edition.cnn.com/2018/12/31/asia/south-korea-plastic-bag-ban-intl/index.html. Accessed 1 Oct 2019
222. Suh WR (2018) Progress in addressing marine litter in Korea: recent policies and efforts to protect the marine environment from marine litter. Korea Marine Environment Management Corporation. https://wedocs.unep.org/bitstream/handle/20.500.11822/26499/ML_Korea.pdf?sequence=1&isAllowed=y. Accessed 1 Oct 2019
223. Lee J, Hong S, Jang YC, Lee MJ, Kang D, Shim WJ (2015) Finding solutions for the styrofoam buoy debris problem through participatory workshops. Mar Policy 51(4):182–189
224. Park JG, Suh YW, Jo JH, Jeong D, Gan SY (2018) Microplastic management trends and policy recommendations (in Korean). Korea Environ Inst Res Rep 13(1):1–139
225. OSEAN (2019) Our Sea of East Asia network. http://www.osean.net/main/. Accessed 1 Oct 2019

226. You Y-S, Oh Y-S, Hong S-H, Choi S-W (2015) International trends in development, commercialization and market of bio-plastics (in Korean). Clean Technology 21(3):141–152
227. Korea Environment Corporation (2014) Global trends in plastic recycling: Plastic debris in the ocean and bioplastics. 2014 International Environmental Information Trends Data Base. Korea Environment Corporation. http://webbook.me.go.kr/DLi-File/096/010/001/5584061.pdf. Accessed 1 Oct 2019
228. Liu T-K, Wang M-W, Chen P (2013) Influence of waste management policy on the characteristics of beach litter in Kaohsiung, Taiwan. Mar Pollut Bull 72(1):99–106
229. Kuo F-J, Huang H-W (2014) Strategy for mitigation of marine debris: analysis of sources and composition of marine debris in northern Taiwan. Mar Pollut Bull 83(1):70–78
230. Ko C-Y, Hsin Y-C, Yu T-L, Liu K-L, Shiah F-K, Jeng M-S (2018) Monitoring multi-year macro ocean litter dynamics and backward-tracking simulation of litter origins on a remote island in the South China Sea. Environ Res Lett 13(4):044021
231. SOW (2014) Reduce plastic and avoid the 'age of plastics' (in Chinese). Society of Wilderness. https://www.sow.org.tw/info/news/20141120/3229. Accessed 1 Oct 2019
232. Kunz A, Walther BA, Löwemark L, Lee Y-C (2016) Distribution and quantity of microplastic on sandy beaches along the northern coast of Taiwan. Mar Pollut Bull 111(1–2):126–135
233. Walther B (2019) Table salt with a pinch of plastic. 17 July: Taipei Times. http://www.taipeitimes.com/News/editorials/archives/2019/07/17/2003718804. Accessed 1 Oct 2019
234. Lee H, Kunz A, Shim WJ, Walther BA (2019) Microplastic contamination of table salts from Taiwan, including a global review. Sci Rep 9(1):10145
235. Lin C-N (2018) Microplastics found in seafood, water: survey. 26 September: Taipei Times. http://www.taipeitimes.com/News/taiwan/archives/2018/09/26/2003701186. Accessed 1 Oct 2019
236. Environmental Analysis Laboratory (EPA) (2018) The survey for microplastic in seawater, sand, gravel, and marine life around the coastal areas of Taiwan, Penghu, and Mazu beach (in Chinese). Environmental Analysis Laboratory (EPA)
237. Tsai T-H, Hetherington W (2019) Most fish contain microplastic: study. 14 May: Taipei Times. http://www.taipeitimes.com/News/taiwan/archives/2019/05/14/2003715109. Accessed 1 Oct 2019
238. Piotrowski J (2018) Emerging economies are rapidly adding to the global pile of garbage. 29 September: Ensia. https://www.economist.com/special-report/2018/09/29/emerging-economies-are-rapidly-adding-to-the-global-pile-of-garbage. Accessed 1 Oct 2019
239. Chen K (2016) Taiwan: The world's geniuses of garbage disposal. 17 May: Wall Street Journal. https://www.wsj.com/articles/taiwan-the-worlds-geniuses-of-garbage-disposal-1463519134. Accessed 1 Oct 2019
240. Rossi M (2018) Taiwan has one of the highest recycling rates in the world. Here's how that happened. 18 December: Ensia. https://ensia.com/features/taiwan-recycling-upcycling/. Accessed 1 Oct 2019
241. Maynard N (2018) Taiwan's waste reduction miracle. 24 January: The News Lens. https://international.thenewslens.com/article/88257. Accessed 1 Oct 2019
242. EPA (2018) Yearbook of environmental protection statistics (in Chinese). Environmental Protection Administration. https://www.epa.gov.tw/DisplayFile.aspx?FileID=71D9060F53D70A19&P=5023d7c5-fb4e-4fd9-bc66-7fa724cf2148. Accessed 1 Oct 2019
243. Chen C-L, Kuo P-H, Lee T-C, Liu C-H (2018) Snow lines on shorelines: solving Styrofoam buoy marine debris from oyster culture in Taiwan. Ocean Coast Manag 165(3):346–355
244. Liu T-K, Kao J-C, Chen P (2015) Tragedy of the unwanted commons: governing the marine debris in Taiwan's oyster farming. Mar Policy 53(2):123–130
245. EPA (2019) Taiwan's plastic ban. Environmental Protection Administration. http://www.rachel.org/?q=en/node/149. Accessed 1 Oct 2019
246. Central News Agency (2018) What has changed over the past 16 years because of the implementation of plastic reduction policies? (in Chinese). http://www.cna.com.tw/project/20180730-Plastic/. Accessed 1 Oct 2019
247. EPA (2019) Regulation on over-packaging (in Chinese). https://oaout.epa.gov.tw/law/LawContent.aspx?id=GL006490. Accessed 1 Oct 2019

248. EPA (2012) Restrict the use of plastic trays and boxes (in Chinese). https://oaout.epa.gov.tw/law/LawContent.aspx?id=GL006484. Accessed 1 Oct 2019
249. Marine Debris Governance Platform (2018) Action plan of Marine Debris Governance in Taiwan, 1st edn. Environmental Protection Administration. https://www.epa.gov.tw/DisplayFile.aspx?FileID=C434A8EB6704AFC1&P=7e57ca93-38e7-47fe-896e-a0aa329ba447. Accessed 1 Oct 2019
250. EPA Taiwan (2011) The implementation of source reduction and recycled subsidy of single-use to go cups (in Chinese). https://oaout.epa.gov.tw/law/LawContent.aspx?id=GL006479. Accessed 1 Oct 2019
251. Tainan City Government (2019) Policy of outlawing styrofoam cups (in Chinese). https://web.tainan.gov.tw/epb/cp.aspx?n=15979. Accessed 1 Oct 2019
252. Taipei City Government (2015) The implementation to ban single-use and melamine tableware in Taipei City (in Chinese). https://www-ws.gov.taipei/Download.ashx?u=LzAwMS9VcGxvYWQvMzYzL3JlbGZpbGUvNDExNDMvNzUzNDAxMi9jZjVmYTY2My0xYjVmLTRkNzktOGEyYy02NTRhNGZkYjI2ODkucGRm&n=44CM6Ie65YyX5biC5pS%2f5bqc56aB55So5LiA5qyh5oCn5Y%2bK576O6ICQ55q%2f6aSQ5YW344CN5Z%2b36KGM6KaB6bueLnBkZg%3d%3d&icon=..pdf. Accessed 1 Oct 2019
253. EPA Taiwan (2017) EPA announces amendments to “Restricted use of plastic bags for shopping, implementation methods and implementation dates” (in Chinese). https://enews.epa.gov.tw/enews/fact_Newsdetail.asp?InputTime=1060815165139. Accessed 1 Oct 2019
254. Everington K (2019) Taiwan's ban on plastic straws kicks in today. 1 July: Taiwan News. https://www.taiwannews.com.tw/en/news/3735676. Accessed 1 Oct 2019
255. Huang T-T (2019) Taiwan expands restrictions on single-use cutlery. 8 August: Taiwan News. https://www.taiwannews.com.tw/en/news/3760767. Accessed 1 Oct 2019
256. Lu K-C (2019) Made-in-Taiwan biodegradable plastic, delivering big for Starbucks and Apple. 11 July: Commonwealth Magazine. https://english.cw.com.tw/article/article.action?id=2473. Accessed 1 Oct 2019
257. Anonymous (2018) Eco-straws win at inventors’ fair. 22 October: Taipei Times. http://www.taipeitimes.com/News/taiwan/archives/2018/10/22/2003702822. Accessed 1 Oct 2019
258. Anonymous (2018) Drinking responsibly: non-plastic straws. 19 September: Taipei Times. http://www.taipeitimes.com/News/lang/archives/2018/09/19/2003700675. Accessed 1 Oct 2019
259. Walther BA, Hu C-S (2018) How citizen-scientists helped us to document the type and quantity of coastal debris pollution in Taiwan. Mar Litter News 10(2):24–28
260. Anonymous (2019) EDITORIAL: A Sisyphean task, but worth it. 25 May: Taipei Times. http://www.taipeitimes.com/News/editorials/archives/2019/05/25/2003715743. Accessed 1 Oct 2019
261. Walther B (2019) State must act on plastics problem. 4 June: Taipei Times. http://www.taipeitimes.com/News/editorials/archives/2019/06/04/2003716285. Accessed 1 Oct 2019
262. Walther B (2015) Nation engulfed by plastic tsunami. 9 January: Taipei Times. http://www.taipeitimes.com/News/editorials/archives/2015/01/09/2003608789. Accessed 1 Oct 2019
263. Anonymous (2014) Study finds plastic waste a pervasive problem on beaches. 26 November: Taipei Times. http://www.taipeitimes.com/News/taiwan/archives/2014/11/26/2003605341. Accessed 1 Oct 2019
264. CSIRO (2019) The world’s largest plastic pollution survey. https://www.csiro.au/en/Research/OandA/Areas/Marine-resources-and-industries/Marine-debris/Global-marine-pollution. Accessed 1 Oct 2019
265. Hsiao S (2019) Microplastics found in all waters off Taiwan: report. 5 January: Taipei Times. http://www.taipeitimes.com/News/taiwan/archives/2019/01/05/2003707389. Accessed 1 Oct 2019
266. Greenpeace (2018) Restricting plastic waste and cleaning beaches (in Chinese). https://www.greenpeace.org/archive-taiwan/zh/press/releases/oceans/2018/second-fast-screening/. Accessed 1 Oct 2019

267. EPA (2018) Marine Debris action plan for Taiwan (in Chinese). Environmental Protection Administration. https://enews.epa.gov.tw/enews/fact_Newsdetail.asp?InputTime=1070213135101. Accessed 1 Oct 2019
268. Yang C-H (2018) Forging alliances for plastic-free oceans. 2 March: Taipei Times. http://www.taipeitimes.com/News/editorials/archives/2018/03/02/2003688511. Accessed 1 Oct 2019
269. Marine Debris Governance Platform (2018) Action plan of marine debris governance in Taiwan (in Chinese). Environment Protection Administration. https://enews.epa.gov.tw/enews/enews_ftp/107/0213/135101/1070213新聞附件-臺灣海洋廢棄物治理行動方案.pdf. Accessed 1 Oct 2019
270. Lin C-N (2018) EPA sets timetable to ban plastic use. 14 February: Taipei Times. http://www.taipeitimes.com/News/front/archives/2018/02/14/2003687612. Accessed 1 Oct 2019
271. Yen N (2018) From source to sea – a framework for marine litter management between government and NGOs in Taiwan (poster). In: 6th international marine debris conference, San Diego, California, USA. https://www.researchgate.net/publication/334605229_From_Source_To_Sea_-_A_Framework_for_Marine_Litter_Management_Between_Government_and_NGOs_in_Taiwan_Can_governments_work_with_NGO_A_Platform_of_marine_litter_management_in_Taiwan. Accessed 1 Oct 2019
272. Raubenheimer K, McIlgorm A (2018) Can the Basel and Stockholm Conventions provide a global framework to reduce the impact of marine plastic litter? Mar Policy 96(2):285–290
273. Raubenheimer K, McIlgorm A (2017) Is the Montreal Protocol a model that can help solve the global marine plastic debris problem? Mar Policy 81(4):322–329
274. Hand E (2016) CFC bans pay off as Antarctic ozone layer starts to mend. Science 353(6294):16–17
275. Green F (2018) The logic of fossil fuel bans. Nat Clim Chang 8(2):449–451
276. McDonough W, Braungart M (2008) Cradle to cradle: re-making the way we make things. Vintage Books, New York
277. Lewandowsky S, Ecker UK, Cook J (2017) Beyond misinformation: understanding and coping with the “post-truth” era. J Appl Res Mem Cogn 6(4):353–369
278. Thiel M, Penna-Díaz MA, Luna-Jorquera G, Sala S, Sellanes J, Stotz W (2014) Citizen scientists and marine research: volunteer participants, their contributions and projection for the future. Oceanogr Mar Biol Annu Rev 52(3):257–314
279. Hidalgo-Ruz V, Thiel M (2015) The contribution of citizen scientists to the monitoring of marine litter. In: Bergmann M, Gutow L, Klages M (eds) Marine anthropogenic litter. Springer, Berlin, pp 429–447
280. Cigliano JA, Meyer R, Ballard HL, Freitag A, Phillips TB, Wasser A (2015) Making marine and coastal citizen science matter. Ocean Coastal Manag 115(1):77–87
281. Mattsson K, Jocic S, Doverbratt I, Hansson L-A (2018) Nanoplastics in the aquatic environment. In: Zeng EY (ed) Microplastic contamination in aquatic environments: an emerging matter of environmental urgency. Elsevier, Amsterdam, pp 379–399
282. Rist S, Hartmann NB (2018) Aquatic ecotoxicity of microplastics and nanoplastics: lessons learned from engineered nanomaterials. In: Wagner M, Lambert S (eds) Freshwater microplastics: emerging environmental contaminants? Springer, Berlin, pp 25–49
283. Galloway T (2015) Micro- and nano-plastics and human health. In: Bergmann M, Gutow L, Klages M (eds) Marine anthropogenic litter. Springer, Berlin, pp 343–366
284. Mattsson K, Hansson LA, Cedervall T (2015) Nano-plastics in the aquatic environment. Environ Sci Processes Impacts 17(2):1712–1721
285. EFSA Panel on Contaminants in the Food Chain (CONTAM) (2016) Presence of microplastics and nanoplastics in food, with particular focus on seafood. EFSA J 14(6):e04501
286. Revel M, Châtel A, Mouneyrac C (2018) Micro (nano) plastics: A threat to human health? Curr Opin Environ Sci Health 1(4):17–23

The Microplastics in Metro Manila Rivers: Characteristics, Sources, and Abatement

Maria Antonia N. Tanchuling and Ezra D. Osorio

Contents

1 Introduction 406
2 Microplastics in Metro Manila Rivers 407
  2.1 Study Area 407
  2.2 Abundance of Microplastics in Surface Waters and Sediments 408
  2.3 Characteristics and Possible Sources of the Microplastics 411
3 Solid Waste Management Regulatory Framework 414
  3.1 Republic Act 9003: The Ecological Solid Waste Management Act 414
  3.2 Local Ordinances 416
  3.3 Regulations on Incineration and Open Burning 416
4 Solid Waste Management in Metro Manila 416
  4.1 Generation 417
  4.2 Segregation at Source 418
  4.3 Collection 419
  4.4 Recycling and Diversion Activities 421
  4.5 Disposal 423
5 Moving Forward to Prevent Plastic Pollution 423
References 424

Abstract Microplastics, which are considered as emerging contaminants, have been reported to be leaked to the open environment on a global scale. Few studies have been conducted on the occurrence of microplastics on several water bodies in the country given the fact that the Philippines is considered to be the third largest contributor of plastics in oceans. This chapter described the composition and distribution of plastic wastes and quantified and characterized microplastics in terms of

---

M. A. N. Tanchuling (✉)
Institute of Civil Engineering, University of the Philippines, Diliman, Quezon City, Philippines
e-mail: mntanchuling@up.edu.ph

E. D. Osorio
Environmental Engineering Program, University of the Philippines, Diliman, Quezon City, Philippines
e-mail: edosorio1@up.edu.ph

Friederike Stock, Georg Reifferscheid, Nicole Brennholt, and Evgeniia Kostianaia (eds.),
Plastics in the Aquatic Environment - Part I: Current Status and Challenges,
Hdb Env Chem (2022) 111: 405–426, DOI 10.1007/698_2020_659,
© Springer Nature Switzerland AG 2020, Published online: 2 September 2020

shape and polymer type in several rivers especially within Metro Manila draining to two of the most economically important water bodies, the Manila Bay and the Laguna de Bay. Extracted microplastics in sampling sites are mostly fragments derived from larger plastics (secondary microplastics) which signified the importance of an efficient solid waste management to reduce the leakage of the plastic waste and microplastics to the open environment.

Keywords Metro Manila, Microplastics, Rivers, Solid waste management, Waste analysis and characterization

1 Introduction

According to a previous study [1], it is estimated that the Philippines contributed a total of 1.88 million metric tons (MMT)/year of plastic waste to oceans as of 2010, making the country the third largest contributor of plastics in the ocean globally. The estimation is based on factors such as population, waste collection rates, economic status, and the population's proximity to coastal areas.

Plastic wastes can be fragmented into smaller particles called microplastics (<5 mm). There are two sources of microplastics commonly found in aquatic systems: primary and secondary microplastics [2]. Primary microplastics are those that are generated for use in a broad variety of consumer and industrial applications such as different sizes of microbeads used in cosmetic scrubs as abrasives and exfoliants [3–5] and larger virgin plastics pellets used as raw materials in fabrication [6]. On the other hand, secondary microplastics are derived from larger plastic materials which are fragmented by processes such as UV degradation or machine washing [2]. These secondary microplastics are linked to be the major contributors to the ubiquitous amounts of microplastics present in the environment [7]. The sources of secondary microplastics include fishing nets, household items, and other discarded plastic debris [8].

In recent years, the occurrence of microplastics in several environmental media around the world has been documented. Research on microplastics in the Philippines is scant with only a few studies done. There are a few conducted outside the capital, but most studies focus on the microplastic occurrence in the rivers within Metro Manila.

Metro Manila is a region located in the southwestern portion of the island of Luzon (Fig. 1) and is the center of economic and political activities of the country. It is comprised of 16 highly urbanized cities and 1 municipality occupying a total land area of 619.57 km^2 and is home to approximately 12.8 million people. Straddling though the metropolis is the 25-km-long Pasig River, which drains to the Manila Bay on the east and to the Laguna de Bay on the west during high tide. Manila Bay is the country's main port of maritime trade and travel, and Laguna de Bay is the largest

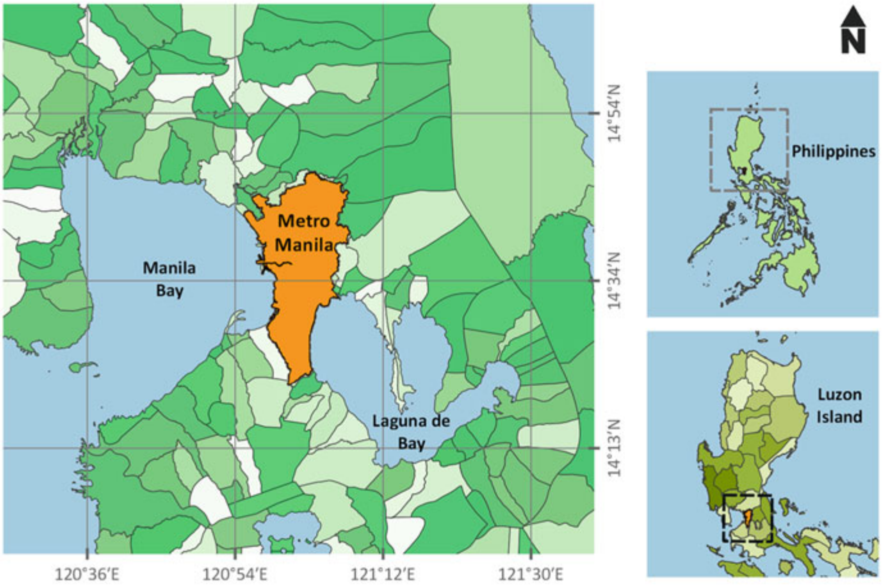


Fig. 1 Geographical map of Metro Manila

freshwater lake in the country. The water from Laguna de Bay flows to Manila Bay during the months of June to December, but it is virtually stagnant from March to May.

2 Microplastics in Metro Manila Rivers

2.1 Study Area

Several initial studies on the microplastic pollution in rivers and creeks in Metro Manila and its neighboring provinces were conducted in 2018–2020 [9–13]. Figure 2 shows the sampling sites and their condition during sampling. The sampling sites in the rivers of Cañas, Pasig, Sapang Baho, Tullahan, and Tunasan are congregated by household communities especially informal settlers wherein direct dumping was observed. Solid wastes can be seen floating on rivers and creeks, as well as along the riverbanks. For the Parañaque River, the sampling area is close to the Bulungan Seafood Market and the Las Piñas-Parañaque Critical Habitat and Ecotourism Area (LPPCHEA), the only natural wetland sanctuary for birds in Metro Manila. Several canoes and bancas were seen beside the seafood market using plastic straws, ropes, and fishing nets to dock. Meanwhile, the sampling site in the Meycauayan River is observed to have few constructed facilities surrounding it; however, abundant solid wastes are accumulated along the banks, indicating that these wastes come from the

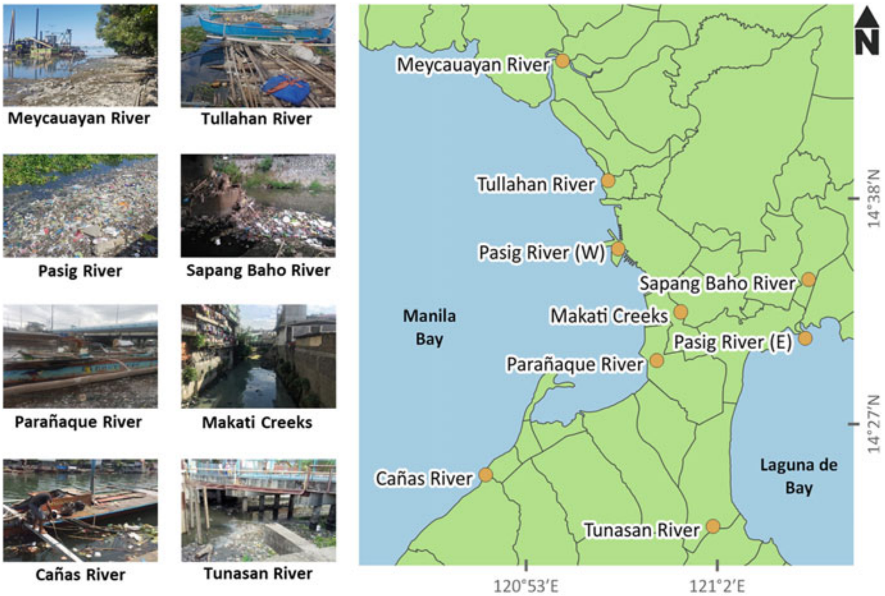


Fig. 2 Sampling sites for waste analysis and characterization and microplastic study

mid and upstream of the river which are heavily populated areas. It should also be noted that most of the plastic and manufacturing industries in the country are situated within the watershed of the Marilao-Meycauayan-Obando (MMO) and Navotas-Malabon-Tenejeros-Tullahan (NMTT) river systems.

2.2 *Abundance of Microplastics in Surface Waters and Sediments*

Microplastics in the surface waters were detected in all study areas with a mean abundance ranging from about 800 particles/m^3 up to 60,000 particles/m^3 (Fig. 3) [9–13]. Among the different water bodies being studied, the highest concentration of microplastics in surface waters was found to be in the Meycauayan River, followed by the Tullahan River, while the lowest concentration was identified in the Makati Creeks. The Makati Creeks had limited access along their banks, and the flow was relatively faster which prevented the plastic wastes from being accumulated along their banks.

Microplastics in the sediments were also detected in the rivers of Cañas, Meycauayan, Parañaque, Pasig (both east and west sides), and the Tullahan River [9, 13]. The highest mean abundance was in the Meycauayan River, while the lowest in the Pasig River (East–Laguna de Bay point) (Fig. 3). As compared to the surface water, the transport of microplastics in sediments is slower than those floating

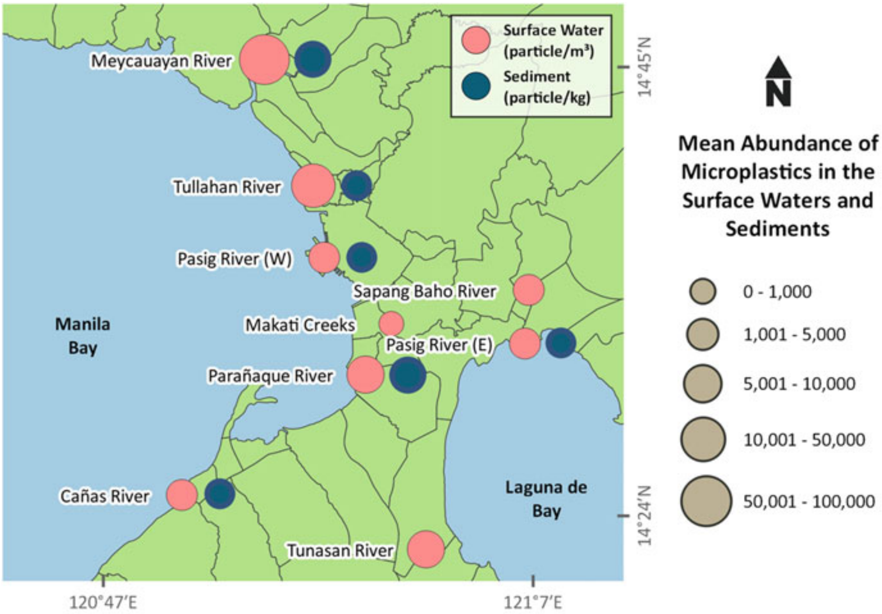


Fig. 3 Mean abundance of microplastics in surface waters and sediments for all sampling sites

microplastics in surface water. Hence, the microplastic pollution levels in sediments are more closely related to the proximity of the source [14]. For example, the microplastics collected in the sediments of the Parañaque River were relatively higher as compared to the other sampling areas. This is more likely because of the established seafood market located adjacent to its river mouth.

Table 1 shows the related studies across the globe regarding the mean abundance of microplastics in the surface waters and sediments. However, it should be noted that only related studies that used similar quantification units (particles/m^3 for surface water; particles/kg of dry weight for sediment) for the abundance of microplastics were gathered.

The mean abundance of microplastics in the surface waters from studies in other countries indicates a wide variation from as low as 11 particles/m^3 in the Deep Bay of Hong Kong [15] to as high as 7,630 particles/m^3 in the Queen Charlotte Sound of Canada [17]. Generally, the microplastics detected in the surface waters of Meycauayan, Tullahan, Tunasan, and Paranaque were relatively higher compared to other studies [15–17]. The abundance of microplastics in the Meycauayan River is at least seven times greater than the observations in Queen Charlotte Sound by Desforges et al. [17].

Concentrations of microplastics in the sediments of sampling areas were higher than in previous studies [18–22, 24] even if the size of the microplastics being studied was smaller than 75 μm. On the other hand, the results were comparable to those detected in the nearshore and tributary in Canada [23] and to the subtidal in

Table 1 Mean abundance of microplastics reported in surface waters and sediments worldwide

Mean abundance	Study area and country (from lowest to highest abundance)	Concerned particle size	Reference
Particle/m^3	Surface water		
0–1,000	Deep Bay (Hong Kong)	<5 mm	Tsang et al. [15]
	East China Sea (China)	0.5–5 mm	Zhao et al. [16]
	Tsang Yi (Hong Kong)	<5 mm	Tsang et al. [15]
	Northeast Pacific Ocean (Canada)	<5 mm	Desforges et al. [17]
	Makati Creeks (Philippines)	0.3–5 mm	Lumongsod et al. [12]
1,001–5,000	Sapang Baho River (Philippines)	0.3–5 mm	Bonifacio et al. [11]
	Cañas River (Philippines)	0.075–5 mm	Osorio [13]
	Pasig River east side (Philippines)	0.3–5 mm	Adricula et al. [9]
	Pasig River west side (Philippines)	0.075–5 mm	Osorio [13]
	Yangtze estuary (China)	0.5–5 mm	Zhao et al. [16]
5,001–10,000	Parañaque River (Philippines)	0.075–5 mm	Osorio [13]
	Tunasan River (Philippines)	0.3–5 mm	Argota et al. [10]
	Queen Charlotte sound (Canada)	<5 mm	Desforges et al. [17]
10,001–50,000	Tullahan River (Philippines)	0.075–5 mm	Osorio [13]
50,001–100,000	Meycauayan River (Philippines)	0.075–5 mm	Osorio [13]
Particle/kg	Sediment		
0–100	Beach (Germany)	<1 mm	Liebezeit et al. [18]
	Beach (Singapore)	1.6 μm–5 mm	Ng and Obbard [19]
	Mangrove (Singapore)	1.6 μm–5 mm	Nor and Obbard [20]
	South Yellow Sea offshore (China)	1 μm–5 mm	Zhao et al. [21]
101–500	North Yellow Sea offshore (China)	1 μm–5 mm	Zhao et al. [21]
	Harbor (Belgium)	38 μm–1 mm	Claessens et al. [22]
	Beach (Canada)	<5 mm	Ballent et al. [23]
	Bohai Sea offshore (China)	1 μm–5 mm	Zhao et al. [21]
	Beach (Slovenia)	0.25–5 mm	Laglbauer et al. [24]
	Pasig River east side (Philippines)	0.3–5 mm	Adricula et al. [9]

(continued)

Table 1 (continued)

Mean abundance	Study area and country (from lowest to highest abundance)	Concerned particle size	Reference
501–1,000	Cañas River (Philippines)	0.075–5 mm	Osorio [13]
	Tributary (Canada)	<5 mm	Ballent et al. [23]
	Pasig River west side (Philippines)	0.075–5 mm	Osorio [13]
	Tullahan River (Philippines)	0.075–5 mm	Osorio [13]
	Nearshore (Canada)	<5 mm	Ballent et al. [23]
1,000–1,500	Parañaque River (Philippines)	0.075–5 mm	Osorio [13]
	Meycauayan River (Philippines)	0.075–5 mm	Osorio [13]
	Subtidal (Italy)	0.7 μm–1 mm	Vianello et al. [25]
>1,500	Tidal flat (Germany)	<1 mm	Liebezeit et al. [18]

Italy [25] and lower than those in the tidal flat in Germany [18]. Consistent with the local studies, sediments collected from beaches, harbors, subtidal, and nearshore areas have relatively higher pollution levels of microplastics compared to the sediments collected offshore.

Although these comparisons might be inaccurate due to the differences in the methodologies used, the concerned particle size being studied, and even the time and date of sampling, these studies present the general overview of the microplastic contamination in different locations worldwide. Areas subjected to intense human activities are more likely to have greater microplastic pollution.

2.3 *Characteristics and Possible Sources of the Microplastics*

Detected microplastics were classified according to their shape. These microplastic particles occurring in a variety of shapes were further sorted as fragment, film, pellet, line/fiber, sheet, and foam, as suggested by Zhang et al. [26].

Microplastics shape distribution in the surface waters and sediments of the sampling sites showed the dominant occurrence of fragments (Fig. 4), as noted elsewhere [27–29]. These fragments could derive from the breakdown of plastic materials which include packaging, bags and containers, household products, and fishing tools [30–32].

Interestingly, plastic pellets which are considered as primary microplastics and utilized as raw materials for manufactured plastic products [33, 34] were found in small amounts in surface waters in most sampling sites and in the sediments of the Meycauayan and Tullahan Rivers. The possible sources are the manufacturing industry or the microbeads found in cleansing agents.

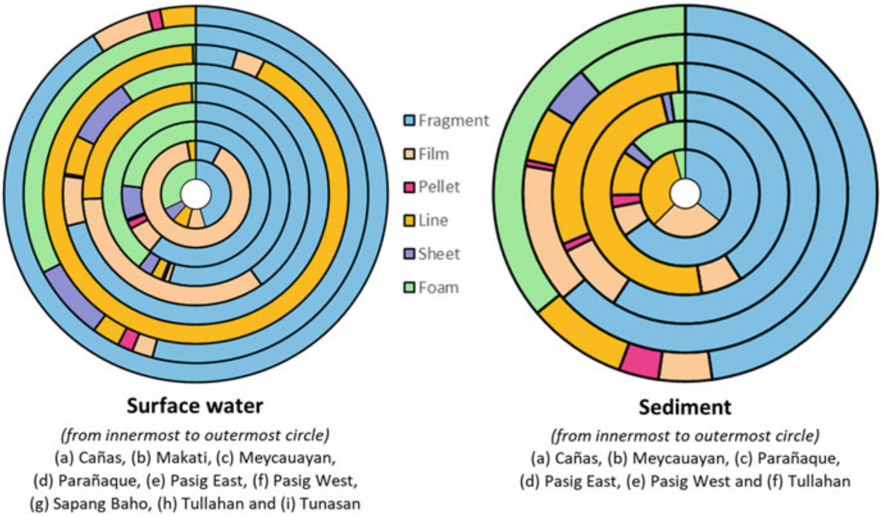


Fig. 4 Shape distribution of microplastics in surface waters and sediments for all sampling sites

The abundance of fragments can be explained by garbage dumping and mismanaged plastic wastes especially for the rivers surrounded by densely populated areas as supported by the amount of larger plastics collected during the conduct of Waste Analysis and Characterization Study (WACS). WACS was conducted by taking samples on the riverbanks and sorting them according to the different waste components. About 25–45% of the waste collected in the rivers and creeks are plastics.

The polymer makeup of microplastics was also identified using the Fourier transform infrared (FTIR) spectroscopy. Although the identified polymer types do not establish the origin of the particles, it still provides useful information to narrow down the possible sources [17]. Figure 5 shows the percentage distribution of polymer types identified to selected microplastics extracted from the surface water and sediment samples. The particles are classified into the following plastic types: polyethylene terephthalate (PET), high-density polyethylene (HDPE), polyvinyl chloride (PVC), low-density polyethylene (LDPE), polypropylene (PP), polystyrene (PS), polyurethane (PU), polyethylene (PE), and others.

The most abundant polymer type across the samples for both surface waters and sediments is PP, followed by PE and PS. These polymer types were also reported to be the most extensively used plastics [35]. Most of the analyzed fragments, lines, and sheets were identified as PP. Polypropylene, the most produced polymer around the world [36], is primarily used as furnishing and for tools which include sportswear, carpets, floor coverings, pipes, nets, and tools for fishing [25, 37]. The extracted microplastics made of PP were likely derived from the breakdown of widely used plastics such as plastic containers, hangers, pails, toys, and fishing nets, which is consistent to the plastic waste characterization conducted on each sampling site (Fig. 6).

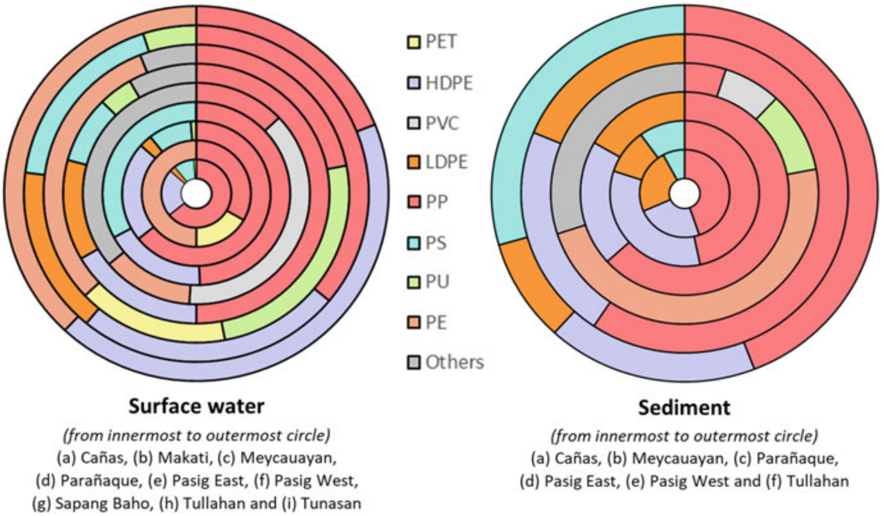


Fig. 5 Polymer type distribution of microplastics in surface waters and sediments for all sampling sites

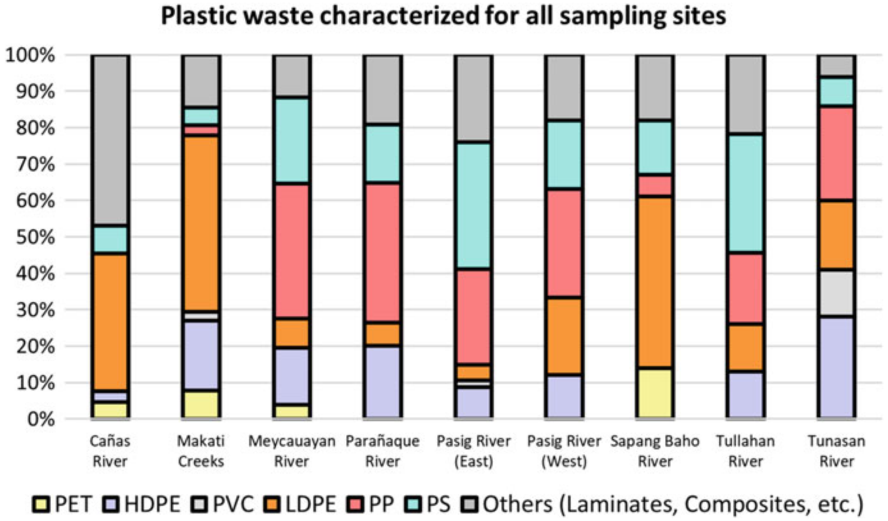


Fig. 6 Percent weight distribution of plastic waste characterized for all sampling sites

Furthermore, polyethylene (both high- and low-density) is from packaging such as films and fragmented plastics [38]. Several plastics made up of PE were collected from both macroplastics and microplastics in the rivers of Cañas, Pasig (East–Laguna de Bay point), and Tunasan. These plastics most likely originated from discarded household items as the rivers are surrounded by large household communities wherein direct littering and garbage dumping are observed. The extracted pellets were identified as HDPE, while most films were identified as LDPE. The concentration of PE films, fragments, and pellets was found to be higher in surface waters as compared to concentrations in the sediments. This finding is consistent with the study of Ng and Obbard [19].

Extracted foams in all samples were mostly comprised of PS which is widely used in plastic manufacturing and packaging industries for disposable items such as cups and food containers [39]. Despite being lightweight, sufficient amounts of foams of both microplastics and macroplastics were collected in the rivers of Meycauayan, Parañaque, and Tullahan. Large quantities of disposable plastic containers produced, specifically food and product containers utilized for delivery services and takeout food and even from the foam fishing floats and rafts used by fishermen, were observed during sampling.

The abundance of microplastics in the sampling sites is mostly related to population density and urban development within watersheds [40]. Domestic and industrial discharge, surface runoff, illegal municipal dumping, and spillage were identified as the main contributors [41] to the microplastic pollution observed in sampling sites.

The results from the study suggested that most of the microplastics extracted are considered as locally derived secondary microplastics, derived from larger plastics.

3 Solid Waste Management Regulatory Framework

To be able to abate the proliferation of microplastics in the rivers and oceans, it is important to evaluate where plastics are coming from. The characterization would point to the fragmented plastics, usually from plastic packaging to be the main source, indicating that the plastic wastes were not properly managed and were leaked to the open environment. This section discusses the regulations that are in place to ensure that wastes are properly managed.

3.1 Republic Act 9003: The Ecological Solid Waste Management Act

The Philippine government enacted the Republic Act 9003 on January 26, 2001, in response to the critical condition of solid waste management problem and the threat it poses to the environment and public health. The Law declares the intention of the state to adopt a systematic, comprehensive, and ecological solid waste management program that will ensure the protection of environment and public health [42].

The National Solid Waste Management Commission (NSWMC) was formed under the Office of the President, in which its main duty is to prescribe policies to achieve the goals of the Law and to oversee the implementation of the solid waste management programs in every local government unit. It is the main government entity in charge of solid waste management policy making and monitoring implementation of law and national and local solid waste management (SWM) plans. The NSWMC is led by the Department of Environment and Natural Resources (DENR) with 14 government sectoral members and 3 private sectoral members. Representatives from the manufacturing and packaging industries, recycling industry, and non-governmental organization (NGO) comprise the private sector.

The local government units (LGUs), particularly cities and municipalities, are the primary responsible units in the implementation of RA 9003. They are tasked to prepare local 10-year SWM plans; manage the collection, diversion, and disposal of various wastes within their jurisdiction; create and maintain materials recovery facilities (MRFs); and adopt revenue-generating measures to support local SWM [43].

The LGUs are mandated to divert 25% of their generated waste through waste diversion strategies such as composting, re-use, and recycling activities within 5 years after the effectivity of the Law. Moreover, the waste diversion rate should be increased every 3 years, and there should be a segregation of solid waste at source.

The creation of MRFs in every barangay[1] or cluster of barangays is mandatory, wherein the barangay is primarily responsible for the segregation and collection of the biodegradable and recyclable wastes. On the other hand, the city or municipality is tasked for the collection of the non-recyclable materials and special wastes. As of 2018, there are 10,730 MRFs all throughout the country, catering only to 33.3% of the barangays.

For the disposal site of wastes, the Law prohibits the existence of both open dumpsites and controlled disposal facilities. Currently, there are now 191 sanitary landfills in the country. However, the current number of landfills is only about 11% of the total required number of landfills nationwide considering that it has been two decades already since the RA 9003 took effect.

The Law also gives incentives to any individuals and public and private entities including LGUs and NGOs that have undertaken projects, technologies, process, and activities to the implementation of the SWM programs. Furthermore, penalties, fines, and sanctions are imposed to any individuals, private entities, and government officials who have violated and failed to comply and enforce the rules and regulations.

Two decades since the passing of RA 9003, the implementation is deficient. Many of the local governments are not equipped with technical skills and resources to implement a good solid waste management system. The problems of solid waste management have continued due to the lack of infrastructures for waste collection, transportation, and disposal. The archipelagic nature of the country makes hauling very costly, making it difficult to increase the waste collection coverage.

---

[1] The smallest political unit comprising a city or municipality.

In many areas not covered by the centralized collection of wastes, the barangays are expected to manage their own wastes. Biodegradable wastes are supposed to be composted, and residuals brought to what is termed as Residual Containment Area (RCA). The recyclable materials should be sorted in MRFs and brought to junk shops. How this is implemented would vary from one barangay to another. While there are some who manage to follow these, there are those who have residents burning their plastics and other residual wastes.

3.2 Local Ordinances

There are also local ordinances related to various aspects of waste management, such as those regulating the use of single-use plastics. As of 2019, 489 cities and municipalities (30% of all cities and municipalities in the country) have some form of policy to regulate the use of plastics, particularly single-carrier plastic bags and plastic straws. Some of these LGUs started implementing plastics regulations since 2011. However, the effectiveness of these initiatives has not yet been assessed. In Metro Manila, 13 out of 17 LGUs have plastic ordinances.

3.3 Regulations on Incineration and Open Burning

There is a general prohibition on the use of incineration and open burning for the disposal of waste according to Section 20 of Republic Act No. 8749, or the Philippine Clean Air Act of 1999. Incineration, as defined by the Law, is prohibited if it emits poisonous and toxic fumes.

Currently, the government is exploring the use and operation of waste-to-energy (WtE) technologies using the guidelines provided by the NSWMC Resolution No. 669, Series of 2016. Furthermore, the DENR also issued Department Administrative Order (DAO) 2019-21 on the Guidelines Governing Waste to Energy Facilities for the Integrated Management of Municipal Solid Waste. There are also pending Senate bills filed in the 18th Congress institutionalizing WtE systems, which aim to establish environmentally sound waste management systems including the facilities that cover reduction, segregation, recycling, re-use, disposal, and conversion of waste into useful resources.

4 Solid Waste Management in Metro Manila

The level of solid waste management infrastructure is highly variable within the country, depending on the level of urbanization of the city or municipality. In Metro Manila, the solid waste management flow can be illustrated in Fig. 7. Collection is

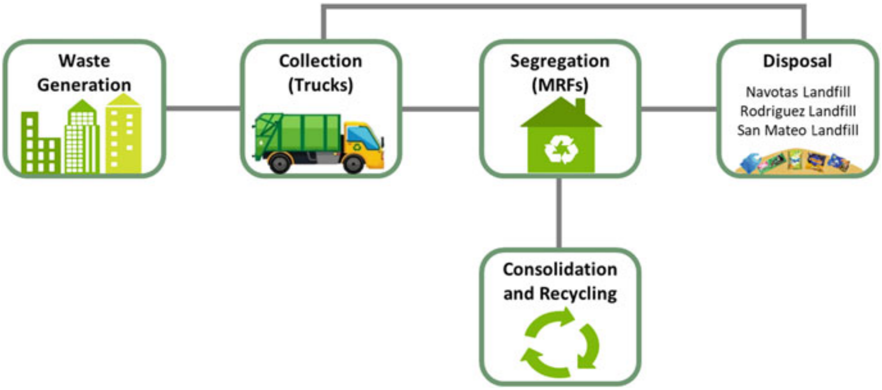


Fig. 7 Solid waste management value chain of Metro Manila

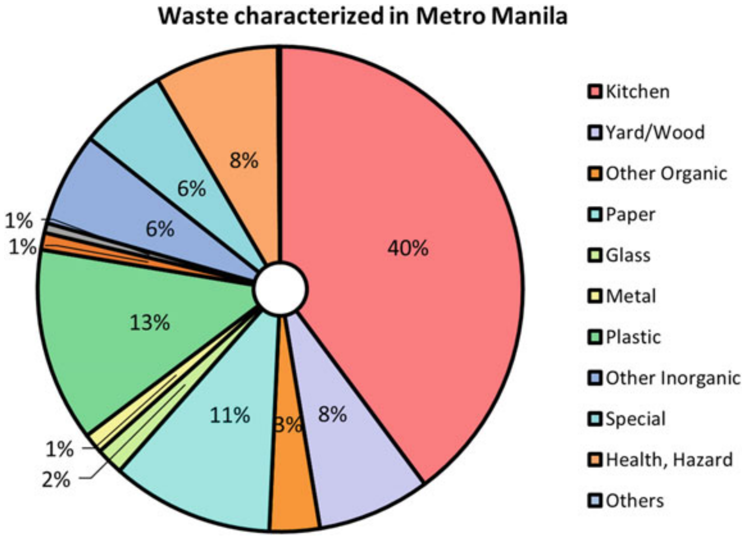


Fig. 8 Percent weight distribution of solid waste characterized in Metro Manila

done by the city government which hauls off the wastes and brings them to either an MRF or directly to the landfill. Recoverable materials are sorted in MRFs and brought to recyclers.

4.1 Generation

The estimated waste generated by Metro Manila is 9,989 tonnes/day. The typical composition by weight is shown in Fig. 8. A huge portion is comprised of kitchen

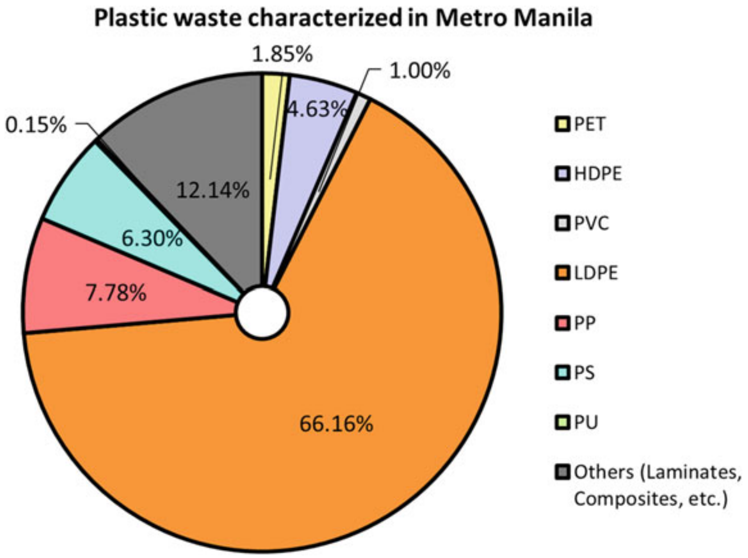


Fig. 9 Percent weight distribution of plastic waste characterized in Metro Manila

waste and other organic wastes. The plastic component in the wastes is 13%. With a population of 12.8 million as of 2015, the waste generation per capita is 0.78 kg/day, which is higher than the national average of 0.40 kg/day [44–49].

The breakdown of the plastic wastes is shown in Fig. 9, based on the plastics that are brought to disposal sites by waste collection trucks. The plastic wastes are mostly composed of LDPE (66.16%), followed by other plastics such as laminates and composites (12.14%). LDPE are usually thin plastic bags used to contain food and are usually contaminated. Laminates are the packaging for sachets of single-use goods such as shampoo, toothpaste, catsup, etc. There is no market for these as recyclables, and they are thus not collected by waste pickers. The plastic waste generation rate per person is computed to be 0.10 kg/day, which is higher than the rest of the country since urbanized areas consume more plastics than those in rural areas.

4.2 *Segregation at Source*

There are local ordinances which require residents to segregate their wastes into degradable and biodegradable components. The enforcement and the compliance of cities vary. There are still a lot of mixed wastes, including recyclable materials, that are being brought to the landfills.

There are many households and other waste generators such as schools, offices, malls, and other institutions who have good waste management practices, such as

Fig. 10 Sorting station in a centralized MRF

segregating recyclable materials such as paper, hard plastics, glass, and metals. These are either brought to their barangays' MRFs or sold to junk shops or to itinerant waste buyers who also sell them to junk shops.

As mandated in RA 9003, LGUs are required to ensure that every barangay or cluster of barangays have its own MRF. The MRF shall receive solid waste for final sorting, segregation, composting, and recycling, with the residual wastes transferred to a sanitary landfill. It should also include a transfer station or sorting station, drop-off center, a composting facility for biodegradable wastes, and a recycling facility. As of 2019, out of the 1,710 barangays in Metro Manila, only 974 MRFs were listed by the NSWMC in the metropolis. Figures 10 and 11 show a centralized MRF in one of the cities.

4.3 Collection

The collection efficiency in Metro Manila is reported to be at 100%. Hauling is undertaken by private contractors paid by the local government. The frequency of collection varies also. Some would have daily collections, while others less frequently.

Collection vehicles used are trucks, some with compactors, but most others are ordinary dump trucks. Garbage bins, or plastic bags with wastes that are left by residents outside their residences, are collected manually by garbage collectors and hauled off to the trucks (Fig. 12). It is quite common that the garbage collectors would pick valuable materials for them to sell to junk shops along the way to the

Fig. 11 Collected recyclable plastics in a centralized MRF

Fig. 12 Collected waste was hauled off to a dump truck

disposal sites. Although highly discouraged by the LGUs, this adds to the diversion rates for recyclable materials.

Although it is said that there is full collection coverage, there is no collection of wastes in areas where there are no roads, like in informal settlements. The uncollected volume of wastes ends up in waterways and although not allowed, is

Fig. 13 Informal settler families residing beside an estero

burned in backyards. There are informal settler families (ISFs) who live by and on the estero[2] itself. There are about 234,738 ISFs residing within the Manila Bay Watershed with 39,635 ISFs in Metro Manila [50]. Due to the close proximity of these ISFs to the water bodies, they dispose their solid wastes and even human wastes directly to the river and to the bay, causing massive pollution in the watershed (Fig. 13).

4.4 Recycling and Diversion Activities

The recycling industry is limited to few materials which can be recycled. The level and extent of recycling is limited in terms of materials that can be recycled and the geographical coverage. The materials that have value and collected by waste pickers and bought by junk shops are paper, clear glass, cardboard, aluminum, and tin. Among the plastics, it is usually the clear PET, PP, and the rigid HDPE which are collected (Fig. 14). The collection infrastructure is limited to the highly urbanized centers only such as in Metro Manila. In many areas such as in islands and other remote areas, there is no collection of recyclable materials.

A major stakeholder in the recycling industry, aside from the consolidators and recycling plants, are the waste pickers and buyers, including the small junk shops, which are largely undocumented. They have positive contribution in increasing diversion rates of recoverable materials.

---

[2] A tidal channel used as a drainage canal.

Fig. 14 Collected recyclable plastics as feed of a recycling facility

Fig. 15 Plastic bricks produced in MRF

Metro Manila in particular has seen an increase in recycling rates (of all recyclables, including non-plastic wastes) from 6% in 1997 up to 33% in 2010 due to the implementation of RA 9003 and the waste diversion strategies by the LGUs and NGOs [51].

There are efforts from the private sector, NGOs, and LGUs to improve recycling rates. Laminates are being collected and cut into strips and contained in PET bottles, in exchange for grocery items or other goods from the LGUs. Commonly called as ecobricks, these are utilized as reusable building blocks. There are also innovations

Fig. 16 Bags and other commodities made up of plastic waste

of using shredded plastics as additive in concrete and producing bricks from melted PS and LDPE with the use of a plastic densifier (Fig. 15). Plastic wastes are also reused creatively and turned into decors and useful products (Fig. 16).

4.5 Disposal

Landfilling is the only option available for the residual wastes. Currently, solid wastes from the various cities in Metro Manila are disposed either in Navotas Sanitary Landfill in the west, in Rodriguez Sanitary Landfill, or in San Mateo, Rizal, both located to the east of Metro Manila.

There are plans to put up a waste-to-energy plant in Quezon City, one of the cities in Metro Manila, but as of now it is not yet certain whether it will push through [52]. There is also a strong lobby against WtE schemes since according to those who oppose it, incineration is banned in the country as stipulated in the Clean Air Act.

5 Moving Forward to Prevent Plastic Pollution

It is evident that microplastics come from the fragmentation of macroplastics, which have leaked to the environment due to poor collection coverage and low recycling activities. To be able to address this, RA 9003 needs to be properly enforced and implemented. Local government units which are primarily tasked to manage solid wastes have to be capacitated technically and logistically to fulfill its mandate.

Capacity building, coupled with good governance, is necessary to improve the implementation of the SMW laws and ordinances.

The Manila Bay Coordinating Office (MBCO) was instituted to rehabilitate Manila Bay. It recognizes that solid wastes are major sources of pollution to the Bay and addresses the problem by monitoring sources of pollution and the compliance of LGUs on solid waste and wastewater management. Plans are under way to relocate ISFs away from the esteros.

The situation that was presented here is that of Metro Manila's only. There are thousands of other rivers outside the capital that need to be studied and protected too. While it is expected that the wastes generated outside the capital are less, the infrastructure for solid waste management is also poorer, so it cannot be safely assumed that the situation is better outside the capital. Instead, this is good evidence that attention and care should be given to all our bodies of water as polluting them with plastic wastes has serious implications to the environment and people's health, well-being, and livelihood.

References

1. Jambeck JR, Geyer R, Wilcox C, Siegler TR, Perryman M, Andrady A, Narayan R, Law KL (2015) Marine pollution. Plastic waste inputs from land into the ocean. Science 347:768–771. https://doi.org/10.1126/science.1260352
2. MSFD Technical Subgroup on Marine Litter (2013) Guidance on monitoring of marine litter in European seas. Publications Office of the European Union, Luxembourg. https://doi.org/10.2788/99475
3. Fendall LS, Sewell MA (2009) Contributing to marine pollution by washing your face: microplastics in facial cleansers. Mar Pollut Bull 58:1225–1228
4. Darling SJ, Green ARS, Veríssimo D (2015) Scientific evidence supports a ban on microbeads. Environ Sci Technol 49(18):10759–10761
5. Leslie H (2015) Plastic in cosmetics. Institute for Environmental Studies
6. Browne MA, Crump P, Niven S, Teuten E, Tonkin A, Galloway T, Thompson R (2011) Accumulation of microplastic on shorelines worldwide: sources and sinks. Environ Sci Technol 45(21):9175–9179. https://doi.org/10.1021/es201811s
7. Eriksen M, Lebreton LC, Carson HS, Thiel M, Moore CJ, Borerro JC, Galgani F, Ryan PG, Reisser J (2014) Plastic pollution in the world's oceans: more than 5 trillion plastic pieces weighing over 250,000 tons afloat at sea. PLoS One 9(12):111913.5
8. Eerkes-Medrano D, Thompson RC, Aldridge DC (2015) Microplastics in freshwater systems: a review of the emerging threats, identification of knowledge gaps and prioritisation of research needs. Water Res 75:63–82
9. Adricula FRM, Pilapil LH (2019) Characterization and analysis of microplastics in surface water and sediments in Pasig River mouths. Undergraduate Thesis, University of the Philippines – Diliman
10. Argota HL, Bajado JA, Diola MD, Tanchuling MN (2018) Macro- and microplastic pollution in Tunasan River, Metro Manila, Philippines. In: Proceedings of the 4th symposium of the asian regional branch of international waste working group (IWWG-ARB) 2019, Bangkok, Thailand, 20–22 February 2019
11. Bonifacio DR, Delos Santos MJ (2019) Characterization and analysis of microplastics in Sapang Baho River, Rizal. Undergraduate Thesis, University of the Philippines – Diliman

12. Lumongsod SLR, Tanchuling MAN (2019) Microplastic characterization and analysis in Three Makati City Creeks. In: Proceedings on the 4th symposium of the international waste working group-Asian regional board (IWWG-ARB) 2019, Bangkok, Thailand, 20–22 February 2019
13. Osorio ED (2019) Ubiquity of microplastics in the surface waters and sediments sampled for five identified river mouths of Manila Bay. Graduate thesis, University of the Philippines-Diliman
14. Su L, Xue Y, Li L, Yang D, Kolandhasamy P, Li D, Shi H (2016) Microplastics in Taihu Lake, China. Environ Pollut 216:711–719. https://doi.org/10.1016/j.envpol.2016.06.036
15. Tsang YY, Mak CW, Liebich C, Lam SW, Sze ETP, Chan KM (2017) Microplastic pollution in the marine waters and sediments of Hong Kong. Mar Pollut Bull 115(1–2):20–28. https://doi.org/10.1016/j.marpolbul.2016.11.003
16. Zhao S, Zhu L, Wang T, Li D (2014) Suspended microplastics in the surface water of the Yangtze estuary system, China: first observations on occurrence, distribution. Mar Pollut Bull 86(1–2):562–568
17. Desforges JPW, Galbraith M, Dangerfield N, Ross PS (2014) Widespread distribution of microplastics in subsurface seawater in the NE Pacific Ocean. Mar Pollut Bull 79:94–99
18. Liebezeit G, Dubaish F (2012) Microplastics in beaches of the east Frisian islands: Spiekeroog and Kachelotplate. Bull Environ Contam Toxicol 89(1):213–217
19. Ng KL, Obbard JP (2006) Prevalence of microplastics in Singapore's coastal marine environment. Mar Pollut Bull 52:761–767. https://doi.org/10.1016/j.marpolbul.2005.11.017
20. Nor NHM, Obbard JP (2014) Microplastics in Singapore's coastal mangrove system. Mar Pollut Bull 79:278–283. https://doi.org/10.1016/j.marpolbul.2013.11.025
21. Zhao J, Ran W, Teng J, Liu Y, Liu H, Yin X, Wang Q (2018) Microplastic pollution in sediments from the Bohai Sea and the yellow. Sci Total Environ 640–641:637–645. https://doi.org/10.1016/j.scitotenv.2018.05.346
22. Claessens M, De Meester S, Van Landuyt L, De Clerck K, Janssen CR (2011) Occurrence and distribution of microplastics in marine sediments along the Belgian coast. Mar Pollut Bull 62:2199–2204. https://doi.org/10.1016/j.marpolbul.2011.06.030
23. Ballent A, Corcoran PL, Madden O, Helm PA, Longstaffe FJ (2016) Sources and sinks of microplastics in Canadian Lake Ontario nearshore, tributary and beach sediments. Mar Pollut Bull 110(1):383–395. https://doi.org/10.1016/j.marpolbul.2016.06.037
24. Laglbauer BJL, Franco-Santos RM, Andreu-Cazenave M, Brunelli L, Papadatou M, Palatinus A, Grego M, Deprez T (2014) Macrodebris and microplastics from beaches in Slovenia. Mar Pollut Bull 89:356–366. https://doi.org/10.1016/j.marpolbul.2014.09.036
25. Vianello A, Boldrin A, Guerriero P, Moschino V, Rella R, Sturaro A, Da Ros L (2013) Microplastic particles in sediments of Lagoon of Venice, Italy: first observations on occurrence, spatial patterns and identification. Estuar Coast Shelf Sci 130:54–61. https://doi.org/10.1016/j.ecss.2013.03.022
26. Zhang K, Shi H, Peng J, Wang Y, Xiong X, Wu C (2018) Microplastic pollution in China's inland water systems: a review of findings, methods, characteristics, effects, and management. Sci Total Environ 630:1641–1653
27. Figueiredo GM, Vianna TMP (2018) Suspended microplastics in a highly pol-luted bay: abundance, size, and availability for mesozooplankton. Mar Pollut Bull 135(6):256–265. https://doi.org/10.1016/j.marpolbul.2018.07.020
28. Moore CJ, Moore SL, Leecaster MK, Weisberg SB (2001) A comparison of plastic and plankton in the North Pacific Central Gyre. Mar Pollut Bull 42:1297–1300
29. Ivar do Sul JA, Spengler A, Costa MF (2009) Here, there and everywhere. Small plastic fragments and pellets on beaches of Fernando de Noronha (equatorial Western Atlantic). Mar Pollut Bull 58(8):1236–1238
30. Derraik JGB (2002) The pollution of the marine environment by plastic debris: a review. Mar Pollut Bull 44:842
31. Thompson RC, Olsen Y, Mitchell RP, Davis A, Rowland SJ, John AWG, McGonigle D, Russell AE (2004) Lost at sea: where is all the plastic? Science 304:838

32. Zhang K, Gong W, Lv JZ, Xiong X, Wu CX (2015) Accumulation of floating microplastics behind the Three Gorges Dam. Environ Pollut 204:117–123
33. Hidalgo-Ruz V, Gutow L, Thompson RC, Thiel M (2012) Microplastics in the marine environment: a review of the methods used for identification and quantification. Environ Sci Technol 46(6):3060–3075
34. McDermid KJ, McMullen TL (2004) Quantitative analysis of small-plastic debris on beaches in the Hawaiian Archipelago. Mar Pollut Bull 48:790–794
35. Zhang K, Su J, Xiong X, Wu X, Wu C, Liu J (2016) Microplastic pollution of lakeshore sediments from remote lakes in Tibet plateau, China. Environ Pollut 219:450–455. https://doi.org/10.1016/j.envpol.2016.05.048
36. PlasticsEurope (2015) Plastics e the facts 2015: an analysis of European plastics production. Demand Waste Data 30. http://www.plasticseurope.org/Document/plasticse-the-facts-2015.aspx
37. Wang W, Ndungu AW, Li Z, Wang J (2017) Microplastics pollution in inland freshwaters of China: a case study in urban surface waters of Wuhan, China. Sci Total Environ 575:1369–1374
38. Sruthy S, Ramasamy EV (2017) Microplastic pollution in Vembanad Lake, Kerala, India: the first report of microplastics in lake and estuarine sediments in India. Environ Pollut 222:315–322. https://doi.org/10.1016/j.envpol.2016.12.038
39. Di M, Wang J (2018) Microplastics in surface waters and sediments of the Three Gorges Reservoir, China. Sci Total Environ 616–617:1620–1627. https://doi.org/10.1016/j.scitotenv.2017.10.150
40. Yonkos LT, Friedel EA, Perez-Reyes AC, Ghosal S, Arthur CD (2014) Microplastics in four estuarine rivers in the Chesapeake Bay, U.S.A. Environ Sci Technol 48:14195
41. Zbyszewski M, Corcoran PL, Hockin A (2014) Comparison of the distribution and degradation of plastic debris along shorelines of the Great Lakes. J Great Lakes Res 40:288–299
42. Republic Act 9003 (2001) An act providing for an ecological solid waste management program, creating the necessary institutional mechanisms and incentives, declaring certain acts prohibited and providing penalties, appropriating funds therefor, and for other purposes. https://www.lawphil.net/statutes/repacts/ra2001/ra_9003_2001.html. Accessed 15 May 2020
43. Atienza V (2008) Review of the waste management system in the Philippines: initiative to promote waste segregation and recycling through good governance. Institute of Developing Economies
44. Local Government of Manila City (2014) 10-year solid waste management plan
45. Local Government of Marikina City (2015) 10-year solid waste management plan
46. Local Government of Navotas City (2014) 10-year solid waste management plan
47. Local Government of Pasig City (2014) 10-year solid waste management plan
48. Local Government of Quezon City (2017) 10-year solid waste management plan
49. Local Government of San Juan City (2014) 10-year solid waste management plan
50. Manila Bay Coordinating Office (2018) Manila Bay clean-up, rehabilitation and preservation program 3rd quarterly report 2018. http://mbco.denr.gov.ph/status-report/. Accessed 23 May 2020
51. NSWMC (2010). Country presentation Republic of the Philippines. In: Second meeting of the regional 3R Forum in Asia, Kuala Lumpur, Malaysia. 4–6 October 2010
52. Fernandez HA (2019) Are waste-to-energy plants bad investments in the Philippines? News article, 17 June 2019. https://www.eco-business.com/news/are-waste-to-energy-plants-bad-investments-in-the-philippines/. Accessed 2 May 2020

Plastic Contamination in Brazilian Freshwater and Coastal Environments: A Source-to-Sea Transboundary Approach

André R. A. Lima, Manuela D. Silva, Fernanda E. Possatto, Guilherme V. B. Ferreira, and Allan P. Krelling

Contents

1 Introduction 428
2 Materials and Methods 429
3 Results 430
  3.1 Sources of Plastic Debris 430
  3.2 Composition and Spatial Distribution of Macroplastics 432
  3.3 Composition and Spatial Distribution of Microplastics 440
  3.4 Interaction Between Plastic Debris and Biota in Freshwater and Estuarine Ecosystems 442
4 Discussion 449
  4.1 Contributions, Lessons Learned and Knowledge Gaps Regarding Plastic Pollution in Brazilian Riverine and Coastal Environments 449
  4.2 Conventions and Regulations Regarding Plastic Pollution in Brazilian Riverine and Coastal Environments 451

A. R. A. Lima
Marine and Environmental Sciences Centre, ISPA – Instituto Universitário, Department of Biosciences, Lisboa, Portugal
e-mail: andre.ricardoaraujolima@gmail.com

M. D. Silva
Federal University of Paraná, Post-Graduation in Environment and Development, Curitiba, Brazil
e-mail: leladreyer@yahoo.com.br

F. E. Possatto
Federal University of Paraná, Centro de Estudos do Mar, Pontal do Paraná, Brazil
e-mail: fernandapossatto@gmail.com

G. V. B. Ferreira
Federal University of Pernambuco, Recife, Brazil
e-mail: guilherme.vbf@gmail.com

A. P. Krelling (✉)
Federal Institute of Paraná (IFPR), Environmental Studies Department, Paranaguá, Brazil
e-mail: allan.krelling@ifpr.edu.br

Friederike Stock, Georg Reifferscheid, Nicole Brennholt, and Evgeniia Kostianaia (eds.),
Plastics in the Aquatic Environment - Part I: Current Status and Challenges,
Hdb Env Chem (2022) 111: 427–460, DOI 10.1007/698_2020_514,

© Springer Nature Switzerland AG 2020, Published online: 5 May 2020

4.3 Problems Still Needing Attention and Recommendations 453
5 Conclusion 455
References 457

Abstract Plastic debris is ubiquitous in aquatic environments. Freshwater and estuarine ecosystems are not different, and plastic contamination is abundant. Despite ecological and socioeconomic importance, previous reviews identified a low number of publications regarding these ecosystems. An organized review to provide a comprehensive qualitative overview of the plastic debris pollution in Brazil was conducted, considering the literature available ($n = 37$) on Brazilian freshwater and estuarine areas. Literature was reviewed analysing the potential sources, distribution and contamination patterns in different environmental compartments aiming at gathering information that will contribute to the understanding of the status of the plastic pollution in these Brazilian systems. Results indicate that research regarding freshwater systems is almost absent, whilst estuaries present studies considering all-sized plastics. Sources of plastic debris varied according to the environmental compartment. Composition and distribution are also dependent of the compartment investigated; however, there is a clear dominance of plastics. Regarding distribution, it is a clear knowledge gap, considering that most studies analysed describe the levels of pollution without concluding about pathways and trajectories. Interaction between plastic debris and biota was highlighted, including benthos, mammals, reptiles, molluscs, and fishes. The source-to-sea approach might be a key approach to comprehensively understand the plastic debris problems within the Brazilian coast.

Keywords Brazil, Plastic contamination, Source-to-sea, Transboundary

1 Introduction

The high global consumption and plastic versatility in a wide range of products, in several formats, types and ways combined with inadequate solid waste management, have become a constant problem in coastal environments [1–6]. Marine debris have affected a wide range of organisms, from planktonic, fishes, birds, turtles to big marine mammals as pinnipeds and cetaceans by ingestion or entanglement [3]. Solid waste management is very important, but to be efficient to combat marine debris, it is important to understand aspects such as sources, pathways and trajectories. Several system-entry sources are identified, including urban areas and associated drainage or shipping routes [5, 7, 8], and marine debris accumulation can be influenced by many factors, such as rainfall [9, 10], hydrological factors [11, 12], coastline geography [13] and estuary-specific circulation processes [2, 5, 12].

A good diagnostic about the marine debris situation is essential to solve the problem (not dependent to take action) preventing it in the source. For that, adopting the source-to-sea approach may be useful. However, what do we know about the different compartments along the source to sea continuum in Brazil? Within this transboundary continuum, two compartments, freshwater and estuarine environments, were considered understudied by previous studies, especially in Latin America [14]. But is it still a nowadays valid pattern? As rivers and coastal environments are key compartments for the adoption of a source-to-sea transboundary approach to deal with plastic pollution, it is crucial to understand what knowledge is available about these systems in Brazil.

Because of that, this chapter aims to show a literature review of the main sources of micro-, meso- and macroplastic debris, the composition and spatial distribution and the interaction between plastic debris and biota in freshwater and estuarine systems in Brazil.

2 Materials and Methods

Among the various methodological procedures that can accompany the mapping and evaluation of the academic production of a specific topic, Araújo [15] cites that the bibliometric analysis allows both the definition of the relevance of a research and also contributes to the analysis of a thematic-trend in an arrangement of scientific works. However, since they currently involve both quantitative and qualitative techniques, the work involving this methodology needs to make clear the procedures and steps adopted in understanding the form, structure or volume of scientific communication [16].

The stages adopted here were:

1. Selection of database for scientific literature (Scopus and Web of Science)
2. Definition of keywords (marine litter or marine debris; microplastic or microlitter or nanoplastic; estuarine or river or bay; and Brazil) following the main analytical categories for this research
3. Test to define the most appropriate keywords for the search of scientific literature through the relative percentage of the corresponding return value of each keyword
4. Combined search in selected databases
5. Organization of data in a reference management software (EndNote): *research corpus*
6. Reading of selected scientific literature for the composition of the final portfolio of systematic study.

Of the 713 scientific papers, a total of 643 papers were discarded by the following criteria: “Is the study area in Brazil?” and “The study covers: estuarine area, bay or river near estuary?”. If both answers were “yes”, the study remained in the analysis portfolio or because they are common among the databases. In this way, 70 papers were selected. However, only 37 papers were sent to the systematic analysis described below, because the others focused on adjacent areas to estuarine environments and they will be the target of a complimentary investigation regarding only such specific environments. It is worth mentioning that repositories of universities and even sites of important scientific events in the area also pointed out important readings in the area. However, this specific literature was not used in the systematic analysis itself, but as a complementary reading for the composition of the discussion of this chapter. The bibliometric analysis considered papers published from 2003 to 2019.

3 Results

3.1 Sources of Plastic Debris

Plastics have been acknowledged to have several sources in South America [17]. Land-based and sea-based activities are the most common sources. They include activities performed during urban waste disposal, along waterways and marine traffic, salmon and mussel farms, transport from rivers and streams and fishing and harbour activities [2, 5, 8]. Considering that source-to-sea approach is an ecological concept, which considers a continuum of environmental compartments, the following analysis regarding sources was conducted focusing on different ecosystems, disregarding the size of the plastics. In addition, wastewater treatment plants are a contemporary source of microbeads and microfibres released by industries of cosmetic/personal care products and textile industries, respectively [18].

3.1.1 Sources of Plastic Debris in Freshwater Systems

Very few studies have addressed the sources of plastic debris in Brazilian freshwater systems (Fig. 1). The contamination of freshwater fishes (*serrasalmid*) inhabiting the Xingu River, Amazon, revealed the origin of the microplastics ingested [19]. Fragments of polyethylene denounced the source associated with fishing gear lost or discarded in the river basin, whilst the polyamide, polyester and polyethylene terephthalate polymers pointed to sources from discarded trash, since they are commonly used to produce plastic bags, bottles, threads and clothes [19]. These observations support the same conclusion reported for the Pajeú River Basin, where the contamination of a freshwater fish (*Hoplosternum littorale*) revealed that urbanization close to water bodies is a potential source of microplastics to this freshwater environment [20].

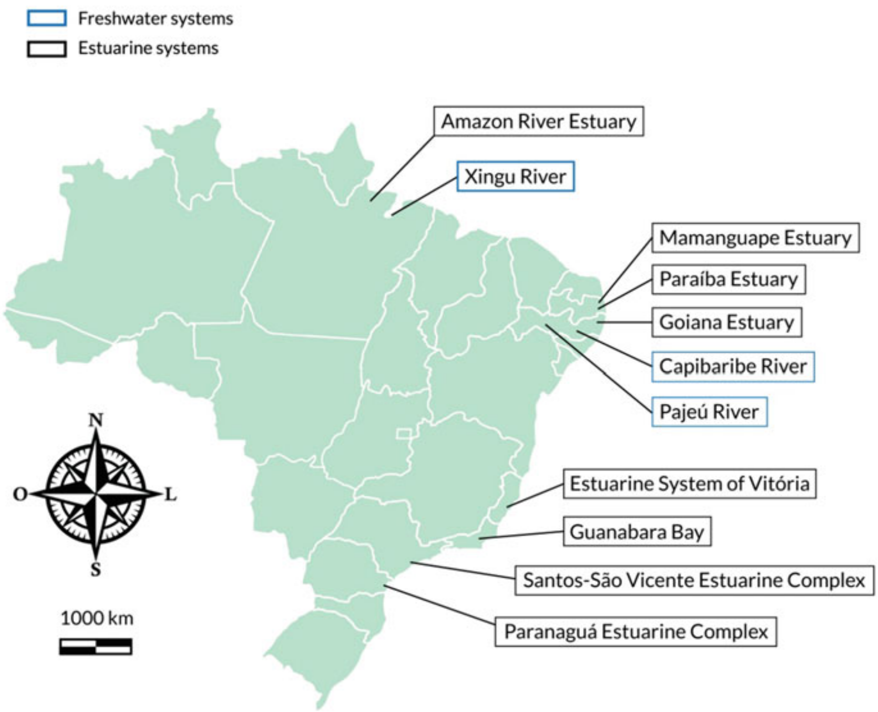


Fig. 1 Map of the Brazilian freshwater and estuarine systems already studied regarding marine debris. Source: André R. A. Lima & Guilherme V. B. Ferreira

3.1.2 Sources of Plastic Debris in Estuarine Systems

Plastics coming from the river basin, inappropriate disposal of communities along the margins and fishery activities (mussel pickers) are the main sources of macroplastics to the mangrove forest of the Goiana estuary (Fig. 1) [21]. Additionally, digging of sediments for mussels by fishers and dredging of the tidal plain are secondary sources of macroplastics to the main channel [21]. Moreover, polystyrene foam buoys, ropes and nets represented 22.3% of all marine debris in an estuarine beach located in the lower Goiana estuary. These items have as sources fishing related activities [10]. Unlabelled plastic bags, PET bottles, caps and soft packaging and rigid containers (33.6%) can have either local or non-local sources, whilst rubber, polyurethane foam and sewage-derived plastics (2%) are related to domestic sources, mainly the fishing villages along the margins of the lower estuary [10].

For the Goiana estuary (Fig. 1), the sources of microplastics were inferred according to their distribution patterns [3]. During the dry season, when the stratification of the water column is more pronounced, microplastics in the middle estuary have the lowest density, whilst in the upper and lower estuaries, the microplastics are abundant. This means that the middle estuary is a physical boundary for microplastics and, therefore, those found in the upper estuary have the river basin as the main source, whilst those found in the lower estuary are associated with coastal villages, harbours and local fishery activities [3]. For this same system, the colour and length of microfilaments ingested by the commercial fish *Cynoscion acoupa* (acoupa weakfish) were also used to infer about their sources [22]. Longer and little weathered filaments, especially white and black ones, were frequently ingested in the upper estuary, suggesting a riverine origin. Contrarily, smaller filaments with signs of weathering, especially red ones, were proportionally more ingested in the lower estuary, suggesting a coastal/oceanic source [22].

Fishing activities are also acknowledged as one of the most important sources of microfibres to the estuarine system of Vitória (SE) (Fig. 1), when fishing nets are unintentionally disposed or abandoned or even during ship loading and unloading [23].

Floating macroplastics entering Guanabara Bay have their main source in the continental runoff [24]. For this same system, the sources of microplastics are not only associated with local rivers and streams but also with fishing and harbour activities [25]. On the other hand, in Jurujuba Cove, a cove with three beaches located within Guanabara Bay, microplastics have their main origins associated with the flush of domestic effluents from a stream discharging into the cove and the fragmentation of blue gallon bottles used for mussel cultivation [26].

In the Santos-Sao Vicente Estuarine Complex (Fig. 1), the sources of plastic debris are acknowledged to be the regional garbage dumps located close to the mangrove, illegal dumping of domestic items, irregular settlements along the estuarine margins and lack of sanitation [27]. The deficiency in the basic sewage system and the deliberate disposal of debris into this estuarine complex are the main causes of macroplastic contamination resulting in a high percentage of domestic (55.41%) and multiple (42.71%) sources, although sources such as tourism, fishery and hospital are also important (1.88%) [28].

For the Paranaguá Estuarine Complex (Fig. 1), the majority (5,620) of the macroplastic items (46.6%) are from a non-identifiable source, followed by beach users (1,996; 16.6%), domestic (1,915; 15.9%), fisheries (1,364; 11.3%), ships and harbour (866; 7.2%) and sewage related items (287; 2.4%), which together represents 53.4% [2]. Another probable source of marine debris is associated with land-based rubbish dumps in the cities of Paranaguá, Antonina and Morretes [5]. This estuary urges attention from the government and citizens because the region is acknowledged as a World Heritage Site due to the presence of one of the last remnants of the Atlantic rainforest [5] (Fig. 2).

3.2 Composition and Spatial Distribution of Macroplastics

Plastics are known to represent the greater part of the items found in marine debris. In the Brazilian freshwater and estuarine systems, such pattern is also observed. Considering the source-to-sea approach and the connectivity between environmental

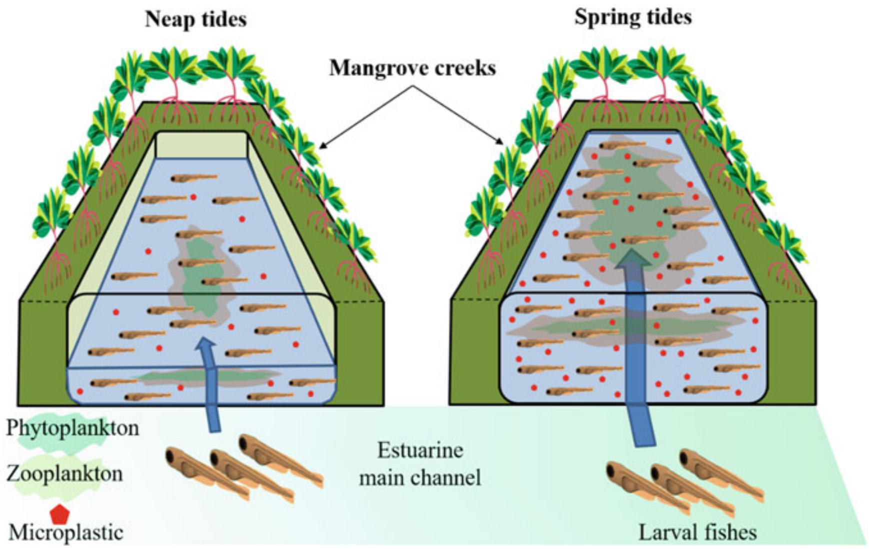


Fig. 2 Patterns of microplastic accumulation in mangrove creeks as ruled by moon phases and tidal cycles according to Lima et al. [31]. Source: André R. A. Lima & Guilherme V. B. Ferreira

compartments, it is expected that these environments work as a temporary sink to these items, being conducted to coastal waters and shorelines and, ultimately, going to the ocean water column and bottom (Fig. 3) [11].

3.2.1 Patterns of Macroplastic Contamination in Freshwater Systems

The only study reporting the composition and spatial distribution of plastic debris in freshwater systems was performed in the Setúbal floodplain lake (Paraná River) [29]. Although this is not a Brazilian system, this lake is located within a river that has its origin in the confluence of two important Brazilian rivers, the Grande and Paranaíba Rivers, which in turn extend through Argentina and Paraguay. There, an alarming plastic contamination was detected in shoreline sediments (Table 1). In total, 217 macroplastic items were collected (1.15 items m^2), among which were food wrappers, bags and disposable foam food containers, the most common type. These plastics were especially composed of polypropylene, polystyrene and polyethylene. Also, the present study was the only one to identify densities of mesoplastics, which showed an average density of 25 items m^2 [29]. Foam plastics (expanded polystyrene) and hard plastics (various polymers) were the most common categories. The highest abundance of macro- and mesoplastics at the downstream site is likely to be a result of the entrapment in a concrete breakwater.

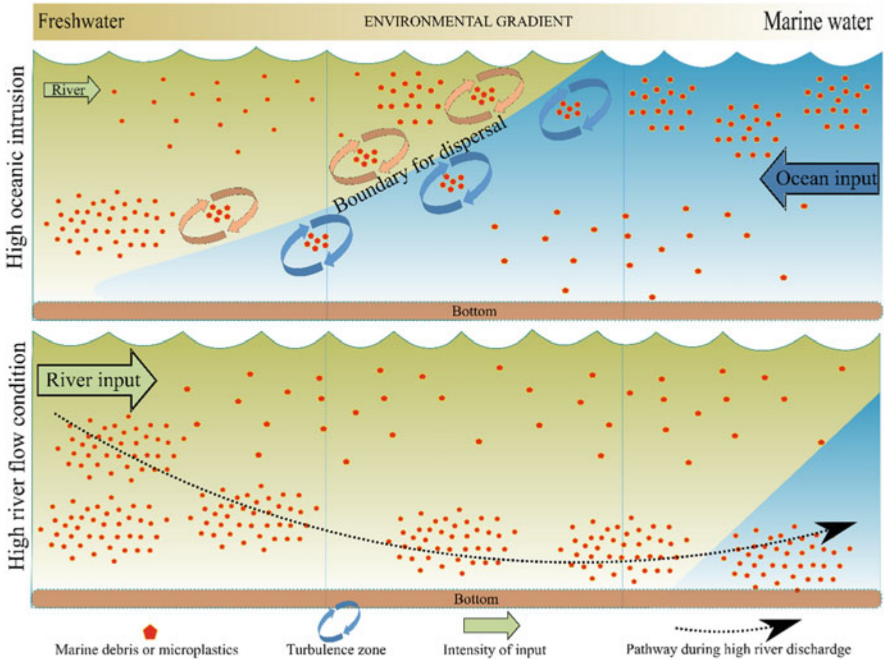


Fig. 3 Conceptual model for the transboundary movement of microplastics along an environmental gradient from freshwater to the ocean according to Krelling and Turra [2], Krelling et al. [11] and Lima et al. [3]. Source: André R. A. Lima & Guilherme V. B. Ferreira

3.2.2 Patterns of Macroplastic Contamination in the Water Column of Estuarine Systems

In Guanabara Bay (Fig. 1), one of the most impacted systems in the tropical western Atlantic, pollution has a positive relationship with the increased urbanization and industrialization [17]. The floating debris is among the major concerns regarding pollution due to the inefficient management of solid wastes by the municipalities of the Rio de Janeiro Macrometropolis [24]. Plastic bag fragments, styrofoam, food packages, clothes, flexible rods, pellets, cigarette butts, straws, cups and gillnets composed the debris in the bay between 2013 and 2015 [24]. Plastics accounted for 71–84% of all items.

Macroplastics were also frequently present in the Santos-Sao Vicente Estuarine Complex, representing 89.64% of the floating debris (2,339 items) observed over 17 months [28]. Ebb spring tide conditions were responsible for entrapping the debris upstream in the U-shaped system due to the confluence of the flows from both channels [28].

In the Paranaguá Estuarine Complex, the density of benthic marine debris (i.e. glass, foam, clothing, metal, plastic), and in particular plastic pieces of bags, wrappers and cups (92.4%), was significantly higher near urbanized locations and

Table 1 Materials and methods applied in surveys to evaluate the contamination of Brazilian freshwater and estuarine systems with macro, meso and microplastics

System	Size	Category	Type of plastics	No. and/or density	Sampling design	Sampler	Sample	References
Goiana Estuary	Macro	Buried plastics	Hard plastics	38 items (5,900 items 100 m^{-3})	30 cores along the mangrove forest, the top and fringe of the tidal plain/Monthly sampling from March 2009 to May 2010	Cylindrical cores	Sediments	Costa et al. [21]
			Soft plastics					
			Nylon polyamide					
	Macro	Deposited debris	Hard plastics	6,944 items (10.8 items 100 m^{-2})	3 transects in a estuarine beach located inside the lower estuary/Monthly sampling from April 2006 to March 2007	Hand collection	Surface sediments	Ivar do Sul and Costa [10]
			Soft plastics					
			Foam					
			Nylon					
			Rubber					
			Cloth					
			Wood					
			Metal					
			Paper					
	Micro	Floating plastics	Hard plastics	14,724 items (26.04 items 100 m^{-3})	3 bottom and 3 surface trawls in the upper, middle and lower estuary/Monthly sampling from April 2012 to March 2013	Plankton net – 300 μm	Surface and bottom waters	Lima et al. [3, 32]
			Soft plastics					
			Threads					
			Paint chips					
	Macro	Floating plastics	Hard plastics	1,662 items (3.4 items 100 m^{-3})	12 mangrove creeks/April and May 2008	Trap net – 500 μm	Water column	Lima et al. [31]

(continued)

Table 1 (continued)

System	Size	Category	Type of plastics	No. and/or density	Sampling design	Sampler	Sample	References
			Soft plastics					
	Micro		Threads	2,710 items (1.4 items 100 m^{-3})				
			Paint chips					
Guanabara Bay	Macro	Floating litter	Plastic fragments	Not available	1 site in the upper bay and 4 sites at the bay entrance/Review from 2013 to 2015	Hand collection	Surface water	Bernardino and Franz [24]
			Styrofoam					
			Food packages					
			Clothes					
			Flexible rods					
			Pellets					
			Cigarette butts					
			Straw					
			Cups					
			Gillnets					
	Micro	Floating plastics	Fragment	1,640 items 100 m^{-3}	3 hauls per each 3 stations in the Jurujuba Cove/Rainy and dry seasons	Plankton net – 150 µm	Water column	Castro et al. [26]
			Sheet					
			Fibres					
			Pellets					
	Micro	Buried plastics	Fibres	8,766 items (1,200 to 130,000 items 100 m^{-2})	34 sites along 17 sandy beaches/cold-dry and warm-rainy seasons	Non informed	Sediments	De Carvalho and Baptista Neto [25]
			Fragments					
			Styrofoam					

			Pellets					
	Micro	Floating plastics	Film	6,300 items 100 m^{-3}	3 sites in the bay/March and September 2016	Plankton net–64 µm and 200 µm	Surface water	Figueiredo and Vianna [34]
			Fragments					
			Fibres					
	Micro	Floating plastics	Fragments	710 items 100 m^{-3}	3 stations in the western portion of the Bay/Summer of 2016	Plankton net – 300 µm	Surface water	Olivatto et al. [33]
			Fibres					
			Styrofoam					
			Rubber					
			Film					
			Granule					
Vitória Bay	Micro	Buried plastics	Fibres	247 items (0 to 38 items $sample^{-1}$)	20 sites along the main channel	Van Veen Grab Sampler	Sediments	Baptista Neto et al. [23]
			Plastic fragments					
Setúbal lake	Macro	Buried Plastics	Food wrappers	11,500 items 100 m^{-2}	2 transects	Hand collection	Sediments	Blettler et al. [29]
			Bags					
			Foam					
	Meso	Buried plastics	Foam	2,500 items 100 m^{-2}		Stainless steel shovel		
			Hard plastic					
	Micro	Buried plastics	Hard plastic	70,400 items 100 m^{-2}				
			Fibres					
Santos-São Vincente Estuarine Complex	Macro	Deposited debris	Plastic bags	2,129 (133 items m^{-2})	2 surveys in 4 upstream and 4 downstream sites along the swamp/August 2006	Hand collection	Surface sediments	Cordeiro and Costa [27]
			Food wrappers					
	Macro	Floating litter	Plastic fragments	2,339 items (121.56 to 173.33		Visual censuses	Surface water	Fernandino et al. [28]

(continued)

Table 1 (continued)

System	Size	Category	Type of plastics	No. and/or density	Sampling design	Sampler	Sample	References
			Cloth	items campaign^{-1})	6 stations along the main channel/ Monthly sampling between July 2010 and January 2012			
			Glass					
			Metal					
			Paper					
			Rubber					
			Wood					
Paranaguá Estuarine Complex	Macro	Deposited debris	Plastic fragments	12,048 items	4 transects along 9 estuarine beaches located in the internal, median and external sectors of the outer estuary/ Periods of high riverine discharges, frontal systems and regular weather conditions between 2015 and 2016	Hand collection	Surface sediments	Krelling and Turra [2]
			Styrofoam					
			Wood					
			Rubber					
			Glass					
			Metal					
			Cloth					
			Cigarette butts					
			Paper					

port areas in the upper and middle sectors, reaching 23.37 items ha^{-1} on average, but no seasonal trends were observed [5].

3.2.3 Patterns of Macroplastic Contamination in Mangrove Areas

Mangrove forests act as a retainer of macroplastics for long periods until they reach the margin of the river, when tidal action flushes the plastics away [30]. In the mangrove forest and the tidal plain of the Goiana estuary, 38 weathered macroplastic fragments were found, with hard polypropylene plastics, soft packaging (cellophane) and nylon polyamide being the most common items, totalling 59 items m^3 [21]. The accumulation of plastics occurred during the dry season, and the mangrove forest was the most contaminated area [21]. Another survey recorded 2,710 macroplastic items (>5–181 mm) in the water column of 12 mangrove creeks, representing 62% of the total plastic debris found [31]. The total density of macroplastics in these creeks is comparable to the density of larvae of the commercial taxa *Cynoscion acoupa* (~1.4 items or larvae 100 m^{-3}). During the full moon, when the spring tide flooded the mangrove forest and flushed more efficiently the mangrove soil, macroplastics presented the highest density, representing the probable pathway of plastic debris from the mangrove forest to the main channel [31].

Among the solid wastes accumulated along mangrove swamps in the Santos-Sao Vicente Estuarine Complex (Fig. 1), 62.81% are composed of macroplastics [27]. In total, 2,129 items (1.33 items m^2) weighing 207.5 kg (129.66 g m^2) were collected (Table 1). The upstream sites are the most contaminated, with plastic bags and food wrappers being the most abundant items [27].

3.2.4 Patterns of Macroplastic Contamination on Estuarine Beaches

A total of 6,944 marine debris were collected in an estuarine beach located within the lower part of the Goiana estuary, with an average density of 10.8 items 100 m^{-2} [10]. Plastics represented >95% of all items. Hard and soft polyethylene plastics were the most frequent, followed by polystyrene foam. A greater amount of marine debris is deposited during the rainy season, when river runoff increases and may carry debris from the upstream area seawards [10].

In total, 12,048 marine debris were collected in nine beaches located along a gradient (i.e. internal, median and external) in the lower portion of the Paranaguá Estuarine Complex (Fig. 1) [2]. Plastics, especially fragments, dominated the items (74.8%), followed by styrofoam (8.7%). The higher abundance and most types of marine debris were observed during periods of high riverine discharge along the entire gradient, with a high dominance of domestic, sewage and fisheries related items (Fig. 3). An intermediate abundance of debris was observed during periods of intense southerly winds associated with frontal systems. Under this condition, the external sector had the lowest abundance of items when compared with the other environmental conditions [2].

This means that rather than bringing ocean-generated items to the coast, the frontal systems act as a generator of longshore drifts that is likely clearing the external sector transporting marine debris offshore. On the other hand, marine debris had the lowest abundance and less variety of types during regular weather conditions (absence of high river discharge and frontal systems). Under such conditions, the internal and median sectors had the lowest abundance of both factors, and domestic and sewage related items had a homogeneous distribution along the gradient. The number of items during a higher river discharge overpassed the amount observed during regular weather conditions, mainly in the external sector. Such result indicates that land-generated items from the innermost parts are transported into the system through river discharge [2].

A simplified hydrodynamic model of dispersion, ground-truthing estimates and regressive vectors revealed that marine debris along the Paranaguá Estuarine Complex gradient are exported after a residence period of 5 days from the inner estuary to the open ocean, which in turn acts as a sinking zone [11]. Once marine debris is exported to the outer estuary, there is no movement upstream anymore (Fig. 3). Therefore, the inner estuary is a ground for generation and release of marine debris, and a transboundary approach must be used to manage marine debris in the land-sea transition zone [11].

3.3 Composition and Spatial Distribution of Microplastics

3.3.1 Patterns of Microplastic Contamination in Freshwater Systems

In freshwater systems, only one study was performed in the Setúbal floodplain lake (Paraná River) regarding microplastics. Microplastics were mainly composed of hard plastics and fibres, totalling 104 items m^{-2}. The highest abundance of microplastics was observed at the upstream site, in contrast to the pattern observed for macro- and mesoplastics in the same area [29].

3.3.2 Patterns of Microplastic Contamination in the Water Column of Estuarine Systems

The distribution patterns of microplastics were assessed in the Goiana estuary, a marine protected area of the type extractive reserve [3]. In total, 14,724 microplastics, representing an average density of 26.04 items 100 m^3, were collected within 12 months (Table 1). Microplastics are found during the entire seasonal cycle and share the same habitats with zooplankton. Their high abundance is comparable to fish eggs and half of fish larvae density in the main channel.

The density of microplastics can overpass the abundance of ichthyoplankton during specific seasons, areas and position in the water column [32]. According to these studies, microplastics are retained within the estuary during the dry season, when the stratification of the water column functions as a physical boundary that does not allow microplastics to cross the middle estuary seaward and even upstream (Fig. 3). This suggests that microplastics from the upper estuary are associated with the river basin and that they have a marine or local origin in the lower estuary. On the other hand, during the end of the rainy season, when precipitation increases and the river runoff is high, microplastics are flushed from the upper estuary to the lower estuary, together with zooplankton (Fig. 3) [3]. During this season, microplastics (<5 mm) and fish larvae have the same density in the lower estuary (~14 items or larvae 100 m^3). Such comparable density increases the chances of microplastic ingestion by organisms that feed on zooplankton [32]. Therefore, the river basin is an important source of microplastics, and the estuary is a route for the exportation of continental microplastics to the open ocean [3, 32].

Mangrove creeks of this same estuary are also contaminated with microplastics, totalling 1,662 items collected within 2 months in 12 creeks [31]. The average density of microplastics is comparable to that of the fourth most abundant fish larvae *Gobionellus oceanicus* (~3.4 items or larvae 100 m^3) inhabiting the creeks. The tidal regimes ruled by changes in the moon phases influence the changes in the composition and abundance of microplastics. This means that during spring tides, the flooding of the creeks is greater and more microplastics are accumulated, whilst the opposite occurs during neap tides (Fig. 2 costa). However, the main concern of the study is that most larvae (80.22%) are in later developmental stages and, thus, are susceptible to ingest microplastics with <2 mm (42.2%).

All surface water samples of the western part of Guanabara Bay are contaminated with microplastics, whose concentration ranged from 1.40 to 21.3 items m^{-3}, with polyethylene and polypropylene being the most abundant polymers (Table 1) [33]. This scenario is consistent with those of highly densely populated coastal regions with the lack of solid waste management, characterizing the bay as one of the most contaminated systems worldwide [33].

A diverse array of small-sized microplastics (<1 mm), especially of blue colour (60%) and composed of polyethylene (72%), were found in surface waters of Jurujuba Cove (Guanabara Bay) [26]. The average density of microplastics was 16.4 items m^3, with higher densities observed on the São Francisco beach during both the rainy and dry seasons. The main concern for the region is that most microplastics are in the dimensions that most MP can be filtered by mussels and the mussel farming in the region may be producing contaminated products for human consumption, since microplastics can carry pathogens or toxic compounds [26].

Another study in the same bay detected that the plankton net with 64 μm mesh collected more microplastics (4.8 items m^3), when compared to the net with 200 μm (1.3 items m^3) [34]. A variety of types, colours and sizes were observed, but blue hard microplastics with a size of 200–300 μm width were the most frequent. Polyethylene and polypropylene were the most common polymers. Comparing the abundance of microplastics and copepods, a prey frequently ingested by fish larvae and chaetognaths, demonstrated that although most microplastics are in the same size ranges of the copepods, they are too diluted to represent risk of ingestion [34].

3.3.3 Patterns of Microplastic Contamination in Estuarine Sediments

Benthic sediments of the estuarine system of Vitória Bay (Fig. 1) were reported to be contaminated by synthetic microfibres (77%) and microfragments (23%) [23]. In total, 247 microplastics were found in 20 samples along the entire main channel, with the lower and uppermost regions being the most contaminated (Table 1). Most microplastics exhibited a plastic-associated microbial community formed by bacteria, fungal filaments and spores, known as plastisphere. This raised questions such as the function of microplastics as microbial vectors, pathogens and transfer of foreign species to non-native habitats [23].

Beach sediments within Guanabara Bay are polluted with small plastic fragments (8,766 particles), including fibres, fragments, styrofoam and pellets [25]. Microplastics are the most abundant category (56%), varying from 740 items m^2 during the cold-dry season in the inner bay to 1,300 items m^2 during the warm-rainy season in the outer bay, being probably the result of the flush of microplastics seawards when rainfall increases [25].

3.4 Interaction Between Plastic Debris and Biota in Freshwater and Estuarine Ecosystems

Several types of interactions between the fauna inhabiting estuaries and marine debris are known very well. They include entanglement [35], ingestion [36], gut blockage [37], transportation of exotic species [38] and even dispersion of pathogenic bacteria (*Escherichia coli* and *Vibrio* spp.) to areas without sewage pollution [39].

3.4.1 Interaction Between Plastic Debris and Benthos of Freshwater Systems

A study performed in the Capibaribe River (Fig. 1) detected that the presence of plastic bags caused changes in the macrobenthic community [40]. Most differences were observed between macrobenthos collected under a plastic bag and macrobenthos collected distant from a plastic bag. Deposit feeders are attracted under plastic bags, since light penetration is limited and microalgae sink and become abundant in the sediment or even because the coverture might serve as a protection against seabird predation. On the other hand, suspension feeders such as the dominant Polychaeta *Streblospio* sp. are excluded due to the feeding limitations imposed by the plastic coverture [40].

3.4.2 Ingestion of Plastic Debris by Mammals, Reptiles and Molluscs in Estuarine Systems

Most studies regarding interaction between plastic debris and estuarine fauna focused on the fates and effects of plastic ingestion. At estuaries (the Mamanguape River Estuary) and coasts (Rio Grande do Norte and Bahia States) of northeast Brazil (Fig. 1), four Antillean manatees (*Trichechus manatus manatus*) were confirmed to have ingested plastic debris [37]. Two were found dead due to ingestion of large amounts of plastic bags and raschel knit fabric or gut blockage. The other two specimens were found debilitated. During rehabilitation, small amounts of plastic debris were eliminated along with faeces [37].

Microplastic contamination has also been confirmed in the commercial mussel *Perna perna* [41, 42] (Table 2). Approximately 75% of the mussels collected in the lower portion of the Santos estuary were contaminated with microplastics with no spatial distribution patterns [41]. At Guanabara Bay, all natural and farmed mussels analysed were highly contaminated with blue and transparent fibres, ranging between 16.6 and 31.2 items per individuals [42]. Such high contamination rates raised questions about human health. Depuration procedures can significantly decrease the quantity of microplastics in mussels, but the quantity that remains in the tissues is still high and may not be adequate for consumption [42].

A high amount of plastic debris was detected in the gut contents of green turtles (*Chelonia mydas*) ($n = 80$) inhabiting the Paranaguá Estuarine Complex [36]. In total, 69.7% of the turtles ingested plastic debris, totalling 3,737 items. From these, three stranded animals died due to debris ingestion. Plastic bags, hard plastics, nylon, polystyrene and rubber were the most frequent ingested debris. Debris ingestion was especially higher during the late rainy season, when the seagrass *Halodule wrightii* was not available for consumption, although debris could have been ingested a long period before and not excreted [36].

3.4.3 Ingestion of Plastic Debris by Fishes in Estuarine Systems

In the lower part of the Amazon River estuary (Fig. 1), 13.8% out of 189 fish specimens representing 46 species captured as bycatch by the shrimp fishery ingested 228 microplastics (polyamide, rayon and polyethylene) [43]. Pellets represented the most frequent item (97.4%), followed by sheets, fragments and threads. The number of microplastics ingested showed a positive correlation with the fish length, but no relationship with the trophic level was detected [43]. However, the main concern of this study is that most species are of commercial importance, which raises human health concerns, since pellets can adsorb chemical pollutants (Fig. 4) [44].

Fishes inhabiting rivers and estuaries are also prone to ingest microplastics, and this seems to have a direct relationship with the degree of anthropogenic activities in the river basin, along estuarine margins and adjacent areas [22, 45]. In the lower

Table 2 Density of meso and microplastics ingested by different taxa inhabiting Brazilian freshwater and estuarine systems

Systems	Trophic category	Species	Size of plastics		References
			Micro (items ind.$^{-1}$)	Meso (items ind.$^{-1}$)	
Xingu River	PIS	*Pristobrycon* cf. *scapularis*	0.21	0.35	Andrade et al. [19]
	PIS	*Pristobrycon eigenmanni*	–	0.33	
	PIS	*Pygocentrus nattereri*	–	0.75	
	PIS	*Serrasalmus manueli*	–	0.14	
	PIS	*Serrasalmus rhombeus*	–	0.33	
	HEB	*Metynnis guaporensis*	–	0.27	
	HEB	*Myloplus rubripinnis*	0.13	0.2	
	HEB	*Myloplus schomburgkii*	0.16	0.16	
	OMN	*Acnodon normani*	–	0.25	
	OMN	*Myloplus rhomboidalis*	–	2	
	OMN	*Ossubtus xinguense*	0.58	1.63	
	OMN	*Tometes ancylorhynchus*	–	0.6	
	OMN	*Tometes kranponhah*	0.14	0.19	
Pajeú River	INS	*Hoplosternum littorale*	3.6	–	Silva-Cavalcanti et al. [20]
Amazon River Estuary	PIS/ ZOB	*Bagre bagre*	12.8	–	Pegado et al. [43]
	PIS/ ZOB	*Bagre marinus*	7.8	–	
	PIS	*Caranx hippos*	30.7	–	
	ZOB	*Selene vomer*	2	–	
	PIS	*Lutjanus analis*	1	–	
	PIS/ ZOB	*Lutjanus synagris*	1	–	
	ZOB	*Narcine brasiliensis*	3	–	
	OMN	*Polydactylus oligodon*	3	–	

(continued)

Table 2 (continued)

Systems	Trophic category	Species	Size of plastics		References
			Micro (items ind.$^{-1}$)	Meso (items ind.$^{-1}$)	
	PIS	*Cynoscion leiarchus*	2	–	
	PIS/ ZOB	*Cynoscion microlepidotus*	1.3	–	
	PIS/ ZOB	*Cynoscion virescens*	3	–	
	PIS/ ZOB	*Macrodon ancylodon*	2	–	
	OPT	*Sphyrna tiburo*	9	–	
	PIS	*Trichiurus lepturus*	2	–	
Goiana River Estuary	PIS	*Cynoscion acoupa*	0.5 to 13	–	Ferreira et al. [22]
	PIS/ ZOB	*Centropomus undecimalis*	0.3 to 9	–	Ferreira et al. [52]
	PIS/ ZOB	*Centropomus mexicanus*	0.5 to 7	–	
	ZOP/ ZOB	*Pomadasys ramosus*	0.2 to 2.3	–	Silva et al. [50]
	ZOP/ ZOB	*Haemulopsis corvinaeformis*	0.3 to 1.4	–	
	ZOB	*Cathorops spixii*	0.15 to 1	–	Possatto et al. [6]
	ZOB	*Cathorops agassizii*	0.4 to 0.9	–	
	ZOB	*Sciades herzbergii*	0.15 to 0.6	–	
	ZOB	*Stellifer brasiliensis*	0.03 to 0.12	–	Dantas et al. [51]
	ZOB	*Stellifer Stellifer*	0.02 to 0.2	–	
	ZOB	*Eugerres brasilianus*	0.16 to 0.78	–	Ramos et al. [49]
	ZOB	*Eucinostomus melanopterus*	0.05 to 0.16	–	
	ZOB	*Diapterus rhombeus*	0.18 to 0.28	–	
Paraíba and Mamanguape estuarine Systems	ZOP	*Opisthonema oglinum*	0.33	–	Vendel et al. [48]
	ZOP	*Rhinosardinia bahiensis*	0.35	–	
	OPT	*Anchoa januaria*	0.13	–	

(continued)

Table 2 (continued)

Systems	Trophic category	Species	Size of plastics		References
			Micro (items ind.$^{-1}$)	Meso (items ind.$^{-1}$)	
	PIS/ ZOP	*Lycengraulis grossidens*	0.17	–	
	OPT	*Atherinella brasiliensis*	0.03	–	
	INS	*Poecilia vivipara*	0.11	–	
	OMN	*Hyporhamphus unifasciatus*	0.15	–	
	PIS	*Oligoplites saurus*	0.17	–	
	ZOB	*Diapterus auratus*	0.97	–	
	ZOB	*Diapterus rhombeus*	0.06	–	
	ZOB	*Eucinostomus argenteus*	0.02	–	
	ZOB	*Eugerres brasilianus*	0.06	–	
	ZOB	*Achirus lineatus*	0.5	–	
	ZOB	*Symphurus tessellatus*	0.25	–	
	ZOB	*Sphoeroides testudineus*	0.09	–	
Santos Estuary	PLK	*Perna perna*	Non-informed	–	Santana et al. [41]
Guanabara Bay	PLK	*Perna perna*	16.6 to 31.2	–	Birnstiel et al. [42]

Trophic categories: *PIS* piscivorous, *HEB* herbivore, *OMN* omnivore, *INS* insectivorous, *PLK* planktivorous, *ZOB* zoobenthivorous, *ZOP* zooplanktivorous, *OPT* opportunist

Xingu River Basin (Fig. 1), 26.7% out of 172 specimens belonging to 16 serrasalmid species were contaminated with plastic fragments [19]. Approximately, 80% of the species were contaminated. Polyethylene, polyvinyl chloride, polyamide, polypropylene and polyethylene terephthalate were the most common polymers. Mesoplastics accounted for most contamination, when compared with microplastics (Table 2). Although the frequency of occurrence and the mass of the plastics did not differ among species or guilds, omnivorous fishes had the highest rates of contamination (25–100%), followed by piscivorous (14.3–75%) and herbivorous (13.3–27.3%) [19].

The diet of the Brazilian silverside *Atherinella brasiliensis*, for example, presented highest microplastic occurrences in the severely impacted Paraíba River estuary and the lowest in the less impacted Mamanguape estuary [4]. Other authors

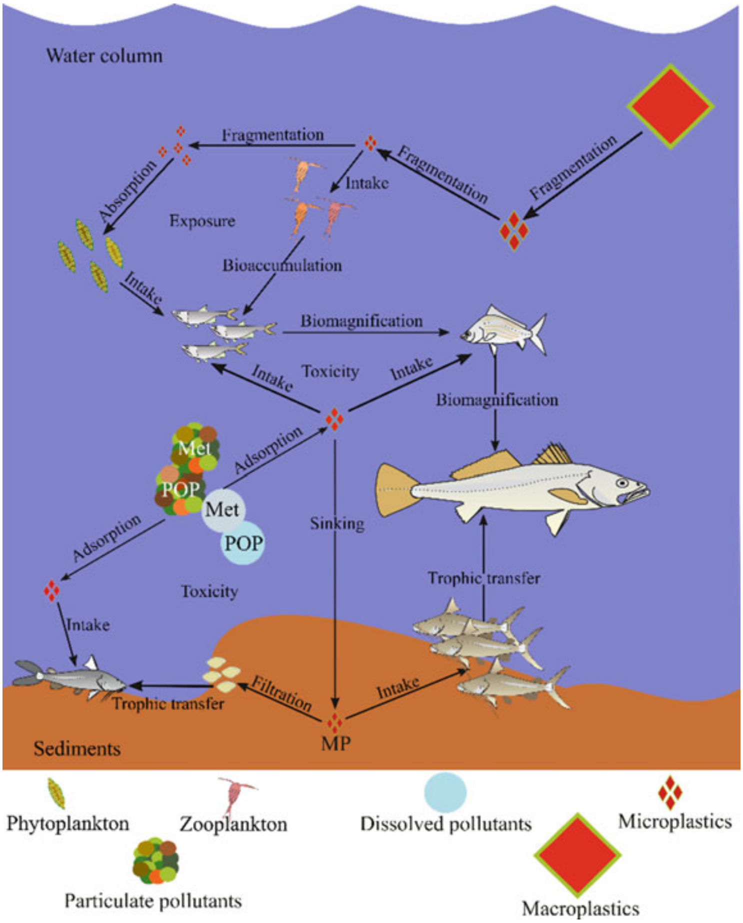


Fig. 4 Some possibilities for the pathways and contamination patterns of plastic debris in the marine environment. Source: André R. A. Lima & Guilherme V. B. Ferreira

[46] proposed another important approach when analysing the diet of *C. acoupa*. For the authors, the ingestion of microplastics is enhanced when the spatio-temporal distribution of the species coincides with periods of high availability of microplastics in the main channel or even in the adjacent coastal area. That contamination varies with the dietary ontogenetic shifts along the life cycle [46]. Another current study asserted that the ingestion of microplastics can have an impact on the health of the individual by diminishing the fish's body condition factor, as revealed in the Guri sea catfish *Genidens genidens* in the Laguna estuarine system [47]. In fact, several conclusions have been generated in recent years and might help to propose managerial action to protect and conserve estuarine resources.

Approximately 9% of the 2,233 fishes inhabiting the Paraíba and Mamanguape estuarine systems ingested microplastics, regardless of length classes, functional guilds and trophic guilds [48]. Fibres were the most frequently ingested type (90%), reaching a maximum of 4 items per individual (Table 2). The widespread contamination on both systems is revealed by the occurrence of microplastics in fish guts along most sampling sites [48].

In the Goiana estuary, at least 11 fish species were evaluated regarding microplastic ingestion (Table 2). There, juveniles, subadults and adults of fishes of commercial and ecological importance were contaminated with microfilaments, especially the blue ones. For the Ariidae catfishes, contamination was recorded in 33% of *Cathorops agassizii* (0.4–0.9 items ind.$^{-1}$), 18% of *C. spixii* (0.15–1 items ind.$^{-1}$) and 18% of *Sciades herzbergii* (0.15–0.6 items ind.$^{-1}$) [6]. The Gerreidae mojarras were also contaminated, being 16% of *Eugerres brasilianus* (0.16–0.78 items ind.$^{-1}$), 9 % of *Eucinostomus melanopterus* (0.05–0.16 items ind.$^{-1}$) and 11% of *Diapterus rhombeus* (0.18–0.28 items ind.$^{-1}$) [49]. For the Haemulidae grunts *Haemulopsis corvinaeformis* and *Pomadasys ramosus*, ingestion of microfilaments varied between 0.3–1.4 items ind.$^{-1}$ and 0.2–2.3 items ind.$^{-1}$, respectively, for the combination of the factors, such as habitats, seasons and ontogenetic phases [50]. In addition, all ontogenetic phases of the Sciaenidae drums were contaminated with blue microfilaments, being 9.2% of *Stellifer stellifer* (0.02–0.2 items ind.$^{-1}$) and 6.9% of *Stellifer brasiliensis* (0.16–0.78 items ind.$^{-1}$) [51]. These contamination rates showed a relationship with the patterns of use of estuarine resources, including habitats and food items.

Regarding *Cynoscion acoupa*, a top predator of commercial importance in the region, microfilaments were more frequently ingested than any natural food item [46]. Among the sampled specimens, only 34% ingested natural prey, whereas 64% of juveniles, 50% of subadults and 100% of adults were contaminated with microfilaments (0.5–13 items ind.$^{-1}$) (Table 2). Moreover, ingestion of microfilaments was detected in all ontogenetic phases of the commercially exploited snooks *Centropomus undecimalis* (0.3–9 items ind.$^{-1}$) and *C. mexicanus* (0.5–7 items ind.$^{-1}$) [52]. More than 50% of the individuals of both species are contaminated. The lower estuary and the coastal zone were the most contaminated sites. The contamination is enhanced with the onset of the piscivory in the adult phase, when peaks of fish ingestion coincide with the peaks of microplastic ingestion [52]. The adult phases of the acoupa weakfish and snooks seem to be more susceptible to contamination through direct ingestion and trophic transfer when they shift their feeding mode to piscivory [22, 52]. Such conclusion is emphasized by the fact that 50% of the fishes ingested by these species were also contaminated, evidencing the likelihood of trophic transfer between prey and predator.

Such behaviour is also reported for the other species assessed in the Goiana estuary, where the contamination with microfilaments was higher in latter phases, when diet and foraging became more complex [49–51]. Additionally, for most species, the highest ingestion rates were observed during the late rainy season in the middle, lower estuaries and coastal zone. This time coincided with the highest availability of microplastics, when river runoff increases and flush plastics seaward [3]. However, to the best of our knowledge, there are few studies addressing the relationship between ontogenetic dietary shifts and microplastic contamination

[22, 46, 49]. These studies asserted that the ecological units are not the species, but their different ontogenetic phases. Ferreira et al. [52] emphasized that the different phases of a species can have multiple and complex uses of essential habitats throughout the year and these behaviours are closely related to microplastic contamination. This means that although fishes may have a great spatial range, the different ontogenetic phases of most species inhabit specific estuarine habitats at least for an entire season (i.e. significant peaks of abundance); and it might coincide with peaks of microplastic availability, when peaks of contamination are commonly recorded [50, 52]. Thus, patterns of estuarine use by fishes can be a good tool to improve management and conservation planning regarding the environmental contamination with plastics.

The contamination of 83% of *H. littorale* specimens was also evaluated in the Pajeú River Basin [20]. Most fragments were microplastics (88.6%), with fibres being the most common type (46.6%). In total, 176 plastics were found in the stomachs of the species, with an average of 3.6 items per fish (Table 2). Microplastics were negatively correlated with food diversity, but positively related to urbanized areas [20].

4 Discussion

4.1 Contributions, Lessons Learned and Knowledge Gaps Regarding Plastic Pollution in Brazilian Riverine and Coastal Environments

The connectivity among river basins, coastal zones and open ocean ruled by environmental gradients is widely discussed in the scientific literature through the so-called source-to-sea continuum or transboundary approach [11, 53, 54]. Such well-established gradient needs to face the intense societal and economic use and occupation. River basins and coastal zones withstand high population density, agricultural/industrial expansion and the improper disposal of wastes and sewages. This significantly alters water quantities and quality, upsetting environmental processes, especially by the contamination of water bodies [17]. The last path for these contaminants is, therefore, the sea.

Among environmental problems, pollution from plastic wastes is a noticeable problem of global concern and acknowledged as one of the world's most pressing environmental issues (Fig. 5). Plastic has reached epidemic proportions with an estimated 100 million tonnes now found in the oceans, being ~90% from land-based sources [55].

Fig. 5 Impacts caused by inappropriate disposal practices. Plastic (**a**) bags in a river margin; (**b**) disposed in a mangrove area; (**c**) along a beach, (**d**) disposed at land, (**e**) interacting with a sea bird, (**f**) in the gut of a dead bird, (**g**, **h**) debris removal along beaches. Source: Wikimages under Creative Commons or Public Domain

4.2 Conventions and Regulations Regarding Plastic Pollution in Brazilian Riverine and Coastal Environments

In May 2019, during the Basel Conference in Geneva, approximately 180 governments pointed plastics as hazardous wastes due to their toxic composition, capacity of adsorbing other pollutants as well as their capacity of fragmentation, which leads to a more dangerous scenario. The Basel Convention on the Control of Transboundary Movements of Hazardous Wastes and their Disposal (the Basel Convention) is thus an international treaty designed to reduce the movements of hazardous waste between nations [56]. The Convention aims to protect humans and the environment against the effects resulting from the generation, transboundary movements and management of hazardous wastes [56].

Marine plastic debris have a clear transboundary nature, emphasizing that this problem has a global scale connected by international impacts [53]. Thus, national measures alone cannot be able to control the problems of marine debris. It urges for international cooperation. The international legislative instruments regarding marine debris are categorized into “hard law” and “soft law” [56]. Hard law agreements are international, intergovernmental and regional conventions describing “legally binding contracts with compulsory requirements or legal operations to the parties” [56]. Examples of hard law agreements are the UN Convention on the Law of the Sea, the Convention on the Control of Transboundary Movements of Hazardous Wastes and their Disposal and the Convention on the Prevention of Marine Pollution by Dumping of Wastes and Other Matter [53]. Soft law agreements describe “nonbinding arrangements between parties” [56]. They include resolutions adopted by conferences, intergovernmental and international organizations, regional strategic action plans, declarations, guidelines and codes of conduct [56].

Despite its importance, the above-mentioned international legislation has not been taken entirely into account by the Brazilian government. Recently, an agreement to limit the global volume of plastic waste was signed by 187 countries – from Norway to Nicaragua. Brazil, as well as the United States and Argentina, opposed to the initiative defined at a meeting in Geneva, Switzerland. The decision was adopted at the end of a 2-week State party conference of three international conventions – Basel, Stockholm and Rotterdam – regarding dangerous products. This allowed the introduction of more effective amendments to the Basel and Stockholm Conventions, known as the Ban Amendment, to better control international traffic and the environmental impact of plastic waste by making it illegal to export hazardous waste from industrialized to least developed countries, even if it is for recycling. The initiative proposed by the UN aims to reduce, starting in 2020, the amount of plastic wastes which are difficult to recycle and sent to poorer nations. It means that countries that export plastic will need the consent of importing countries when it concerns contaminated, mixed or nonrecyclable plastic garbage.

Another relevant aspect regarding especially poorer countries is the lack of basic sanitation. It is acknowledged as the worst issue regarding developing countries in South America [57, 58], and generally the effluents from urban settlements, industries and agriculture are not treated before being discharged [57, 58]. Furthermore, the uncontrolled disposal of sewage and solid wastes is widespread along river basins, estuarine courses, coastal zones and marine waters [59]. It is such a complex situation that during the last 20 years the sanitation project of Rio de Janeiro costed R $ 10 billion (US$2.5 billion), without success. The Plan for the Municipal Environmental Sanitation was created in 2011 and encompasses 15 municipalities of Rio de Janeiro. The plan has the potential to reduce the pollution of the bay with solid wastes and wastewaters; however, all debris produced by the surrounding municipalities keep reaching the estuary. Therefore, the efficiency of these programs is still uncertain and must include social awareness [24].

Also, recycling has gained more attention since 2010, when the National Policy for Solid Wastes (Law 12,305/10) was approved [55]. Therefore, the selective collection and transportation of solid waste have been institutionalized in few municipalities in accordance with Brazilian laws. This law is an instrument of economic and social development to facilitate the collection and return of solid wastes to the corporate sector. However, the difficulties of mobilising citizens for the separation of recyclable materials and the improper surveillance to obligate companies that produce solid wastes to perform proper disposal practices according to legislation are still a concern.

The recent implementation of the law added to the lack of monitoring and surveillance has resulted in the functioning of informal and precarious material recovery facilities. Furthermore, pickers are not contracted or paid for in accordance with legal provisions; incentives under federal law have not been established between municipalities and pickers' cooperatives and associations. Local governments often omit their duties regarding urban solid waste recovery [55]. Approximately 1.835 million tonnes per day of solid waste were collected throughout the country in 2008. Almost 58.3% of the waste collected was sent to sanitary landfills, 19.4% to controlled landfills and 19.8% to dumping sites. Among these, 32% of the collected wastes were recyclable materials, corresponding to 58,527 tonnes per day. However, the recycling of solid wastes was only 13% in 2008.

Other authors recommended some actions aiming at prevention and reduction of marine plastic debris that needs to be addressed by countries in a worldwide perspective [56]. Development of a new international marine plastic debris treaty of the scale and scope of the Montreal Protocol and strengthening and interlinking existing conventions on plastic waste emissions and on marine conservation are urgent tasks needing attention. Some recommendations are urgent and include the ban of disposable plastic food packaging, tableware items and shopping plastic bags or their replacement with biodegradable plastics and/or promotion of reusable packaging systems and ban of microplastics in cosmetics, personal care products and detergents [54]. In addition, development of techniques to reduce the amount of microfibres released by synthetic fabrics during washing, usage of alternative biodegradable materials, or establishment of deposits, return, and restoration systems and introduction of legislation for the dismantling and recycling of fibre-reinforced plastic boats are necessary [53, 56].

Considering the Brazilian territory, it is worth mentioning that the Brazilian government established a voluntary commitment to develop a national strategy to combat marine litter, in the context of the UN Ocean Conference. Within this commitment, there is a goal to develop the first National Plan to Combat Marine Litter. Launched in March 2019, it is still being implemented, and its results might be evaluated in the future to determine its efficiency. It is essential that future efforts within this framework take into account the source-to-sea approach, focusing on environments such as riverine and estuarine compartments.

4.3 Problems Still Needing Attention and Recommendations

Understanding the fate and effects of microplastics in the Brazilian aquatic system is still difficult, since the available studies do not provide a complete picture of the problem. It is notable that few studies have addressed plastic pollution in freshwater and estuarine systems, whilst more beaches are studied. However, semi-enclosed systems, such as rivers and estuaries, have a great capacity of retention and, thus, higher plastic densities when compared to open systems such as the marine environment, where plastics become somewhat "diluted". Despite this, freshwater systems have so far been neglected when compared to marine systems with regard to plastic pollution, which leads to several knowledge gaps and impairs the estimation of river plastic emissions to the world's oceans [60].

Some authors [2, 3] asserted that when the river flow enlarges due to increased rainfall, micro- and macroplastics are flushed seawards. These findings confirm that river basins are the main sources of microplastics transported to the coastal seas [31]. Since Brazil encompasses a variety of aquatic systems experiencing similar problems of pollution, efforts to understand the patterns of microplastic contamination in different settings of a source-to-sea continuum must be prioritized [58, 61].

In Guanabara Bay, for example, the habit of debris disposal in streams or its margins is dated since the seventeenth century [24]. 1847 saw the initiation of the collection of debris from beaches and the implementation of basic sanitation criteria to reduce pollution within the bay. In 1994, the Depollution Program of Guanabara Bay was established aiming to recover the ecosystem and water quality by the construction of sanitation systems. In 2004, the project *eco-barriers* was implemented by the State government to entrap the floating debris flushed by the streams discharging into the bay. In 2005, the project was suspended, but returned 10 years later, when the International Olympic Committee required a better quality of the bay to the State Secretary of the Environment before the beginning of the Olympic Games in 2016.

According to one of the studies [24], some measures for the management of solid wastes need to be urgently addressed by every municipality to follow the Brazilian legislation. They include (a) stimulation, monitoring, inspection and mandatory management of solid wastes in companies; (b) implementation of selective solidarity collection in schools and public bodies; (c) elaboration of public policies for the recognition and valuation of waste pickers; (d) implementation of eco-points and containers for the collection of solid debris; (e) preparation of a waste recycling program and expansion of existing recycling programs; (f) collection of technological waste; (g) environmental education program; (h) implementation and improvements in landfills and hazardous waste landfills.

Furthermore, international alternatives and technologies are available to prevent and mitigate plastic pollution [53, 62]. The technologies for the collection and removal of waterborne debris before it reaches the open sea have been divided into three categories by the Environmental Protection Agency: storm drain inlets, in-line and end-of-pipe debris capture systems and open-water debris capture systems. Moreover, some alternatives have been developed to reduce plastic production and control the management of the generated plastic waste. For example, to prevent the derelict of fishing gears there are useful alternatives to manage plastic pollution, such as fishing gears identification to improve the lost equipments; provision of adequate, affordable, and accessible onshore port reception/collection facilities; and tackling illegal, unreported, and unregulated fishing. In addition, the use of water-soluble polymers, oxidegradable and biodegradable polymers and cellulose acetate can minimize the impacts of plastics in the marine environment. Furthermore, raising public awareness regarding plastic pollution, including training of professionals, education and campaigns in order to positively influence citizens about the problems of plastic pollution, is of great importance and might be provided by national and local authorities for the management of coastal debris [62, 63]. Evaluating the potential economic impacts of plastic debris might also be an important tool to influence positively decision-makers to prevent the input of debris, i.e. beaches and tourist areas [1]. Impacts might be significant; however they are not estimated.

Another approach to understand plastic pollution is the introduction of citizen science projects. These projects involve volunteer participation of citizens, schoolchildren and their teachers who contribute by acquiring information, data and samples to scientific studies [64, 65]. Citizen science encompasses local, national and international scales, with focus on the distribution and composition of marine debris, especially in the intertidal zone, and involves clear protocols, training of volunteers, in situ supervision by professionals and revision of samples and data [64]. In Brazil studies using citizen science are poorly available, but should be a useful alternative to increase the available information on marine debris sources, distribution and ecological impacts [64, 65].

It is possible that other initiatives might not be addressed in the present chapter, e.g. due to several aspects such as methodological limitations and literature available in formats which are not accessible through indexed basis. Considering that, the present chapter shows a systematic review, which is not intended to be a definitive review, but it is a starting point to identify contributions, lessons learned and knowledge gaps regarding plastic pollution in Brazilian riverine and coastal environments. On top of that, it is the first attempt to clearly identify problems still needing attention and recommendations for Brazilian riverine and estuarine environments.

5 Conclusion

The present chapter identified studies developed in Brazilian freshwater and estuarine systems. The literature review indicates a clear knowledge gap especially regarding freshwater systems. The studies focus on different sized debris, including micro-, meso- and macroplastic, and most possess a characteristic of inventory, only indicating the occurrence of plastic debris and suggesting the causes of such scenario. Fewer studies discussed the process by which these items reached the aquatic environments, the pathways and trajectories of the items or environmental factors influencing it.

The main sources of the plastics found in these Brazilian environments varied according to the environmental compartment studied and the size of the items. Most of the items found in freshwater systems were attributed to fisheries, urbanization and improper disposal. In estuarine environments, macroplastics were attributed to a myriad of sources, indicating the complexity of studying debris within this system. The examples of potential sources are runoff and local rivers, beach users, domestic litter, sewage, fisheries, harbours, dredging activities, dumping, improper disposal and the lack of sanitation. The microplastics found in estuaries were mostly associated with fisheries and harbouring activities (e.g. pellets). The relationship between sources and sinks within such environment is a clear knowledge gap, and it is essential to adopt the source-to-sea approach when investigating these areas.

Regarding the composition and spatial distribution of plastic debris, a lack of information is observed regarding freshwater systems. In the only study analysed, macroplastics were commonly composed of plastic bags and food wrapping, and the distribution is affected by physical barriers, such as breakwater, which seems to be determinant in the distribution of this kind of debris by increasing its accumulation. Microplastics are composed of hard plastics and microfibres, especially in the upper part of riverine systems. Stratification along the freshwater system, according to the proximity of the river source, seems to be a relevant factor for the debris distribution.

In the estuarine compartment, macroplastics appeared in the water column and in the bottom. Plastics are dominant, varying from 71% to 92.4%, and the most common items were plastic bags and fragments, food wrapping, cups and other varied items. The proximity to the source (urban areas, harbours, etc.) seems to be a determinant factor for the quality of items of macroplastics within estuarine systems. Microplastics in the estuarine environment were observed in both water column and bottom sediments, especially in the format of pellets, microfragments and microfibres. Especially the presence of polyethylene and polypropylene was noted. There is evidence that physical barriers influence microplastic distribution, especially the floating ones, such as breakwaters and the intermediate sectors of the estuaries, where more accumulation seems to occur. In addition, rainfall and tides seem to be a key environmental process to be investigated regarding microplastic distribution in the Brazilian systems.

In mangrove areas, plastics were investigated especially in the water column. 62% of the samples were plastics, also dominated by plastic bags, wrappings and plastic fragments.

Most debris found in estuarine beaches are plastics (83.5–95%); the most significant sources are domestic waste, sewage and fisheries. Rainfalls and riverine inputs are determinant factors for increasing the amount of plastics, and this compartment might be a temporary fate. Studies regarding mangroves and other intertidal areas, such as estuarine beaches, clearly lack in the literature about the Brazilian coastal areas.

The less studied environment is freshwater systems, and impacts were only observed for benthic species, accounting for a single study identified through this review. On the other hand, in estuarine environments, several studies describe the impacts of plastics on several species, including manatees, green turtles, mussels (*Perna perna*) and several fish species (serrasalmid, *H. litoralle*, *A. brasiliensis*, acoupa, snooks and others). Studies indicate that the level of contamination for fishes in the Brazilian estuaries might be associated with a relationship between microplastic availability and ontogeny phases. It is also suggested that seasons and fish location within the estuarine gradient may influence it as well. Depending on these factors, the studies identified different proportions of interaction of the species with plastic debris, i.e. ingestion rates by fish individuals varied from 6.9% to 100% of the samples. Quantities also varied ranging from 0.13 to 13 items per individual. Microfilaments, microfibres, pellets, nylon, fragments, pieces of plastic bags, rubber, sheets and threads are the most common items.

It is possible to conclude that there is a clear need to develop studies to understand the processes behind such levels of contamination, sources and fates of plastic debris within the different environments along the coastal environmental continuum. Consequently, the source-to-sea approach, which is not only an intellectual framework, but also an ecological continuum, might be a key approach to a comprehensive understanding of the plastic debris problems within the Brazilian coast.

Acknowledgements APK would like to thank Lucas Gottardello Chierigatti for elaborating map. APK kindly thanks the support from the Federal Institute of Paraná (IFPR, Paranaguá Campus). AL acknowledges the European Regional Development Fund (FEDER) through the Lisbon's Regional Operational Programme (LISBOA-01-0145-FEDER-032209) and the Portuguese Foundation for Science & Technology (FCT) through the project PTDC/BIA-BMA/32209/2017 (SARDITEMP). AL also thanks FCT through the project UIDB/04292/2020 granted to MARE (Marine and Environmental Sciences Centre) at the ISPA – University Institute (Department of Biosciences). This study was financed in part by the Coordenação de Aperfeiçoamento de Pessoal de Nível Superior (CAPES) – Brazil – Finance Code 001.

References

1. Krelling AP, Williams AT, Turra A (2017) Differences in perception and reaction of tourist groups to beach marine debris that can influence a loss of tourism revenue in coastal areas. Mar Policy 85:87–99. https://doi.org/10.1016/j.marpol.2017.08.021
2. Krelling AP, Turra A (2019) Influence of oceanographic and meteorological events on the quantity and quality of marine debris along an estuarine gradient. Mar Pollut Bull 139:282–298
3. Lima ARA, Costa MF, Barletta M (2014) Distribution patterns of microplastics within the plankton of a tropical estuary. Environ Res 132:146–155. https://doi.org/10.1016/j.envres.2014.03.031
4. Moreira FT, Prantoni AL, Martini B et al (2016) Small-scale temporal and spatial variability in the abundance of plastic pellets on sandy beaches: methodological considerations for estimating the input of microplastics. Mar Pollut Bull 102:114–121. https://doi.org/10.1016/j.marpolbul.2015.11.051
5. Possatto FE, Spach HL, Cattani AP et al (2015) Marine debris in a World Heritage Listed Brazilian estuary. Mar Pollut Bull 91:548–553. https://doi.org/10.1016/j.marpolbul.2014.09.032
6. Possatto FE, Barletta M, Costa MF et al (2011) Plastic debris ingestion by marine catfish: an unexpected fisheries impact. Mar Pollut Bull 62:1098–1102. https://doi.org/10.1016/j.marpolbul.2011.01.036
7. Santos IR, Friedrich AC, Wallner-Kersanach M, Fillmann G (2005) Influence of socio-economic characteristics of beach users on litter generation. Ocean Coast Manag 48:742–752
8. Leite AS, Santos LL, Costa Y, Hatje V (2014) Influence of proximity to an urban center in the pattern of contamination by marine debris. Mar Pollut Bull 81:242–247. https://doi.org/10.1016/j.marpolbul.2014.01.032
9. de Araújo MCB, Costa MF (2007) An analysis of the riverine contribution to the solid wastes contamination of an isolated beach at the Brazilian Northeast. Manag Environ Qual An Int J 18:6–12. https://doi.org/10.1108/14777830710717677
10. Ivar do Sul JA, Costa MF (2013) Plastic pollution risks in an estuarine conservation unit. J Coast Res 65:48–53. https://doi.org/10.2112/si65-009.1
11. Krelling AP, Souza MM, Williams AT, Turra A (2017) Transboundary movement of marine litter in an estuarine gradient: evaluating sources and sinks using hydrodynamic modelling and ground truthing estimates. Mar Pollut Bull 119:48–63. https://doi.org/10.1016/j.marpolbul.2017.03.034
12. Acha EM, Mianzan HW, Iribarne O et al (2003) The role of the Río de la Plata bottom salinity front in accumulating debris. Mar Pollut Bull 46:197–202
13. Walker TR, Grant J, Archambault MC (2006) Accumulation of marine debris on an intertidal beach in an urban park (Halifax Harbour, Nova Scotia). Water Qual Res J Canada 41:256–262
14. Ivar do Sul JA, Costa MF (2007) Marine debris review for Latin America and the Wider Caribbean region: from the 1970s until now, and where do we go from here? Mar Pollut Bull 54:1087–1104. https://doi.org/10.1016/j.marpolbul.2007.05.004
15. Araújo CA (2006) Bibliometria: evolução histórica e questões atuais. Em questão 12:11–32
16. Araújo RF, Alvarenga L (2011) A bibliometria na pesquisa científica da pós-graduação brasileira de 1987 a 2007. Encontros Bibli Rev eletrônica Bibliotecon e ciência da informação 16:51–70
17. Barletta M, Lima ARA, Costa MF (2019) Distribution, sources and consequences of nutrients, persistent organic pollutants, metals and microplastics in South American estuaries. Sci Total Environ 651:1199–1218
18. Ziajahromi S, Neale PA, Leusch FDL (2016) Wastewater treatment plant effluent as a source of microplastics: review of the fate, chemical interactions and potential risks to aquatic organisms. Water Sci Technol 74:2253–2269
19. Andrade MC, Winemiller KO, Barbosa PS et al (2019) First account of plastic pollution impacting freshwater fishes in the Amazon: ingestion of plastic debris by piranhas and other

serrasalmids with diverse feeding habits. Environ Pollut 244:766–773. https://doi.org/10.1016/j.envpol.2018.10.088

20. Silva-Cavalcanti JS, Silva JDB, de França EJ et al (2017) Microplastics ingestion by a common tropical freshwater fishing resource. Environ Pollut 221:218–226. https://doi.org/10.1016/j.envpol.2016.11.068
21. Costa MF, Silva-Cavalcanti JS, Barbosa CC et al (2011) Plastics buried in the inter-tidal plain of a tropical estuarine ecosystem. J Coast Res 64:339–343
22. Ferreira GVB, Barletta M, Lima ARA et al (2018) High intake rates of microplastics in a Western Atlantic predatory fish, and insights of a direct fishery effect. Environ Pollut 236:706–717. https://doi.org/10.1016/j.envpol.2018.01.095
23. Baptista Neto JA, Gaylarde C, Beech I et al (2019) Microplastics and attached microorganisms in sediments of the Vitória bay estuarine system in SE Brazil. Ocean Coast Manag 169:247–253. https://doi.org/10.1016/j.ocecoaman.2018.12.030
24. Bernardino D, Franz B (2016) Lixo flutuante na Baía de Guanabara: passado, presente e perspectivas para o futuro. Desenvolv e Meio Ambient 38:231–252. https://doi.org/10.5380/dma.v38i0.47024
25. de Carvalho DG, Baptista Neto JA (2016) Microplastic pollution of the beaches of Guanabara Bay, Southeast Brazil. Ocean Coast Manag 128:10–17. https://doi.org/10.1016/j.ocecoaman.2016.04.009
26. Castro RO, Silva ML, Marques MRC, de Araújo FV (2016) Evaluation of microplastics in Jurujuba Cove, Niterói, RJ, Brazil, an area of mussels farming. Mar Pollut Bull 110:555–558. https://doi.org/10.1016/j.marpolbul.2016.05.037
27. Cordeiro CAMM, Costa TM (2010) Evaluation of solid residues removed from a mangrove swamp in the São Vicente Estuary, SP, Brazil. Mar Pollut Bull 60:1762–1767. https://doi.org/10.1016/j.marpolbul.2010.06.010
28. Fernandino G, Elliff CI, Frutuoso GA et al (2016) Considerations on the effects of tidal regimes in the movement of floating litter in an estuarine environment: case study of the estuarine system of Santos-São Vicente, Brazil. Mar Pollut Bull 110:591–595. https://doi.org/10.1016/j.marpolbul.2016.05.080
29. Blettler MCM, Ulla MA, Rabuffetti AP, Garello N (2017) Plastic pollution in freshwater ecosystems: macro-, meso-, and microplastic debris in a floodplain lake. Environ Monit Assess 189(11):581. https://doi.org/10.1007/s10661-017-6305-8
30. Ivar do Sul JA, Costa MF, Silva-Cavalcanti JS, Araújo MCB (2014) Plastic debris retention and exportation by a mangrove forest patch. Mar Pollut Bull 78:252–257. https://doi.org/10.1016/j.marpolbul.2013.11.011
31. Lima ARA, Barletta M, Costa MF et al (2016) Changes in the composition of ichthyoplankton assemblage and plastic debris in mangrove creeks relative to moon phases. J Fish Biol 89:619–640. https://doi.org/10.1111/jfb.12838
32. Lima ARA, Barletta M, Costa MF (2015) Seasonal distribution and interactions between plankton and microplastics in a tropical estuary. Estuar Coast Shelf Sci 165:213–225. https://doi.org/10.1016/j.ecss.2015.05.018
33. Olivatto GP, Martins MCT, Montagner CC et al (2019) Microplastic contamination in surface waters in Guanabara Bay, Rio de Janeiro, Brazil. Mar Pollut Bull 139:157–162. https://doi.org/10.1016/j.marpolbul.2018.12.042
34. Figueiredo GM, Vianna TMP (2018) Suspended microplastics in a highly polluted bay: abundance, size, and availability for mesozooplankton. Mar Pollut Bull 135:256–265. https://doi.org/10.1016/j.marpolbul.2018.07.020
35. Ryan PG (2018) Entanglement of birds in plastics and other synthetic materials. Mar Pollut Bull 135:159–164
36. Guebert-Bartholo FM, Barletta M, Costa MF, Monteiro-Filho ELA (2011) Using gut contents to assess foraging patterns of juvenile green turtles Chelonia mydas in the Paranaguá Estuary, Brazil. Endanger Species Res 13:131–143. https://doi.org/10.3354/esr00320

37. Attademo FLN, Balensiefer DC, da Bôaviagem Freire AC et al (2015) Debris ingestion by the Antillean Manatee (Trichechus manatus manatus). Mar Pollut Bull 101:284–287. https://doi.org/10.1016/j.marpolbul.2015.09.040
38. Rech S, Borrell Y, García-Vazquez E (2016) Marine litter as a vector for non-native species: what we need to know. Mar Pollut Bull 113:40–43. https://doi.org/10.1016/j.marpolbul.2016.08.032
39. Silva MM, Maldonado GC, Castro RO et al (2019) Dispersal of potentially pathogenic bacteria by plastic debris in Guanabara Bay, RJ, Brazil. Mar Pollut Bull 141:561–568
40. Clemente CCC, Paresque K, Santos PJP (2018) The effects of plastic bags presence on a macrobenthic community in a polluted estuary. Mar Pollut Bull 135:630–635. https://doi.org/10.1016/j.marpolbul.2018.07.070
41. Santana MFM, Ascer LG, Custódio MR et al (2016) Microplastic contamination in natural mussel beds from a Brazilian urbanized coastal region: rapid evaluation through bioassessment. Mar Pollut Bull 106:183–189. https://doi.org/10.1016/j.marpolbul.2016.02.074
42. Birnstiel S, Soares-Gomes A, da Gama BAP (2019) Depuration reduces microplastic content in wild and farmed mussels. Mar Pollut Bull 140:241–247. https://doi.org/10.1016/j.marpolbul.2019.01.044
43. De Souza e Silva Pegado T, Schmid K, Winemiller KO et al (2018) First evidence of microplastic ingestion by fishes from the Amazon River estuary. Mar Pollut Bull 133:814–821. https://doi.org/10.1016/j.marpolbul.2018.06.035
44. Fisner M, Taniguchi S, Majer AP et al (2013) Concentration and composition of polycyclic aromatic hydrocarbons (PAHs) in plastic pellets: implications for small-scale diagnostic and environmental monitoring. Mar Pollut Bull 76:349–354. https://doi.org/10.1016/j.marpolbul.2013.09.045
45. Alves VEN, Patrício J, Dolbeth M et al (2016) Do different degrees of human activity affect the diet of Brazilian silverside Atherinella brasiliensis? J Fish Biol 89:1239–1257. https://doi.org/10.1111/jfb.13023
46. Ferreira GVB, Barletta M, Lima ARA et al (2016) Plastic debris contamination in the life cycle of Acoupa weakfish (Cynoscion acoupa) in a tropical estuary. ICES J Mar Sci J du Cons 73:2695–2707. https://doi.org/10.1093/icesjms/fsw108
47. Dantas DV, Ribeiro CIR, Frischknecht CCA et al (2019) Ingestion of plastic fragments by the Guri sea catfish Genidens genidens (Cuvier, 1829) in a subtropical coastal estuarine system. Environ Sci Pollut Res 26:8344–8351. https://doi.org/10.1007/s11356-019-04244-9
48. Vendel AL, Bessa F, Alves VEN et al (2017) Widespread microplastic ingestion by fish assemblages in tropical estuaries subjected to anthropogenic pressures. Mar Pollut Bull 117:448–455. https://doi.org/10.1016/j.marpolbul.2017.01.081
49. Ramos JAA, Barletta M, Costa MF (2012) Ingestion of nylon threads by gerreidae while using a tropical estuary as foraging grounds. Aquat Biol 17:29–34. https://doi.org/10.3354/ab00461
50. Silva MLD, Castro RO, Sales AS, Araújo FVD (2018) Marine debris on beaches of Arraial do Cabo, RJ, Brazil: an important coastal tourist destination. Mar Pollut Bull 130:153–158. https://doi.org/10.1016/j.marpolbul.2018.03.026
51. Dantas DV, Barletta M, da Costa MF (2012) The seasonal and spatial patterns of ingestion of polyfilament nylon fragments by estuarine drums (Sciaenidae). Environ Sci Pollut Res 19:600–606. https://doi.org/10.1007/s11356-011-0579-0
52. Ferreira GVB, Barletta M, Lima ARA (2019) Use of estuarine resources by top predator fishes. How do ecological patterns affect rates of contamination by microplastics? Sci Total Environ 655:292–304. https://doi.org/10.1016/j.scitotenv.2018.11.229
53. Chen C-L (2015) Regulation and management of marine litter. In: Bergmann M, Gutow L, Klages M (eds) Marine anthropogenic litter. Springer, Cham, pp 395–428
54. Schnurr REJ, Alboiu V, Chaudhary M et al (2018) Reducing marine pollution from single-use plastics (SUPs): a review. Mar Pollut Bull 137:157–171. https://doi.org/10.1016/j.marpolbul.2018.10.001

55. Campos HKT (2014) Recycling in Brazil: challenges and prospects. Resour Conserv Recycl 85:130–138
56. Niaounakis M (2017) Regulatory framework. In: Management of marine plastic debris. Elsevier, Cambridge, pp 361–413
57. Huang J, Huang Y, Zhang Z (2014) Coupled effects of natural and anthropogenic controls on seasonal and spatial variations of river water quality during baseflow in a coastal watershed of southeast china. PLoS One 3:e91528
58. Costa MF, Barletta M (2016) Special challenges in the conservation of fishes and aquatic environments of South America. J Fish Biol 89:4–11
59. Pereira LCC, Monteiro MC, Guimarães DO et al (2010) Seasonal effects of wastewater to the water quality of the Caeté river estuary, Brazilian Amazon. An Acad Bras Cienc 82:467–478
60. Lebreton LCM, Van der Zwet J, Damsteeg J-W et al (2017) River plastic emissions to the world's oceans. Nat Commun 8:1–10. https://doi.org/10.1038/ncomms15611
61. Costa MF, Barletta M (2015) Microplastics in coastal and marine environments of the western tropical and sub-tropical Atlantic Ocean. Environ Sci Process Impacts 17:1868–1879
62. Niaounakis M (2017) Prevention and mitigation. In: Management of marine plastic debris. Elsevier, Cambridge, pp 317–359
63. Storrier KL, McGlashan DJ (2006) Development and management of a coastal litter campaign: the voluntary coastal partnership approach. Mar Policy 30:189–196. https://doi.org/10.1016/j.marpol.2005.01.002
64. Hidalgo-Ruz V, Honorato-Zimmer D, Gatta-Rosemary M et al (2018) Spatio-temporal variation of anthropogenic marine debris on Chilean beaches. Mar Pollut Bull 126:516–524. https://doi.org/10.1016/j.marpolbul.2017.11.014
65. Honorato-Zimmer D, Kruse K, Knickmeier K et al (2019) Inter-hemispherical shoreline surveys of anthropogenic marine debris – a binational citizen science project with schoolchildren. Mar Pollut Bull 138:464–473. https://doi.org/10.1016/j.marpolbul.2018.11.048

Marine Litter in the Russian Gulf of Finland and South-East Baltic: Application of Different Methods of Beach Sand Sampling

Alexandra A. Ershova, Tatjana R. Eremina, Irina P. Chubarenko, and Elena E. Esiukova

Contents

1 Introduction 462
2 Marine Litter Study in the Eastern Part of the Gulf of Finland 464
  2.1 Study Area: Neva River Inner Estuary and Outer Part of the Gulf 464
  2.2 Monitoring Methods for Litter on Sandy Beaches 466
  2.3 Marine Litter in the Eastern Part of the Gulf of Finland: Results and Discussion 468
3 South-East Baltic: Kaliningrad Region 471
  3.1 Studies of Beached Marine Litter by Various Methods 471
  3.2 Specific Pollutants: Geosynthetics, Paraffin, Foamed PS 476
  3.3 Three-Dimensional Distribution of Marine Microlitter in the Sand Body of the Beach 479
4 Conclusions 482
References 483

Abstract The chapter combines an overview of several studies of marine litter distribution on beaches and in sediments for two Russian parts of the Baltic Sea: the Gulf of Finland and the South-East Baltic for a period of 2016–2020. Various methods for sand sampling on beaches have been applied, including the OSPAR method, NOAA methodology, and IOW beach litter sampling methods (Frame and Sand Rake methods). The results of field research for the period of 2016–2020 showed both the applicability and some limitations of some methods. Results showed an overall high level of contamination with marine litter and its polymer

---

A. A. Ershova (✉) and T. R. Eremina
Russian State Hydrometeorological University, St. Petersburg, Russia
e-mail: ershova@rshu.ru

I. P. Chubarenko and E. E. Esiukova
P.P. Shirshov Institute of Oceanology RAS, Atlantic Branch, Kaliningrad, Russia

Friederike Stock, Georg Reifferscheid, Nicole Brennholt, and Evgeniia Kostianaia (eds.),
Plastics in the Aquatic Environment - Part I: Current Status and Challenges,
Hdb Env Chem (2022) 111: 461–486, DOI 10.1007/698_2021_746,
© Springer Nature Switzerland AG 2021, Published online: 11 March 2021

components, microplastics in particular, of the Russian Baltic beaches – both regularly cleaned and "wild" isolated beaches. However, in comparison to the Neva Bay and beaches around the world the beaches of the South-East Baltic are quite clean and there is no obvious difference in the contamination of beaches with high (near resort cities) and low (less-visited coastal areas) anthropogenic load. The largest amount of litter of all fractions was found on the beaches of the inner part of the estuary in the Neva Bay. At the same type a high variability of types of litter was shown: the predominant type of litter in the South-East Baltic is foamed plastic (foam/polystyrene foam), together with paraffin, coming from the tank waters from ships, and a specific pollutant-geosynthetic materials that are a new contaminant emerging from coastal engineering protection activities. In the Gulf of Finland region, the most common litter items are plastic pellets, broken glass, cigarette butts, rusty metal, and pieces of building plaster, together with synthetic napkins and cotton bud sticks, that are not retained by the wastewater treatment facilities.

Keywords Gulf of Finland, Marine litter, Methods of monitoring, Microplastics, Sandy beaches, South-East Baltic

1 Introduction

Ecosystems of enclosed seas such as the Baltic Sea with a high anthropogenic load can be especially vulnerable to accumulations of plastic particles along the coast, in lagoons and estuaries. Marine litter is very mobile, especially in the coastal zone: it is carried by currents, deposited on coastal underwater slopes, and accumulates in waters relatively protected from currents and winds, including inland coastal water bodies, like large gulfs and lagoons. Therefore, it is important to assess the anthropogenic pollution of the sea coast based on information on the pollution of all components of this "litter rim."

Despite the numerous studies and monitoring campaigns carried out around the world in the last 10 years there is currently no single unified methodology for sampling and analysis of litter pollution of the coastal and marine environment. One of the problems associated with monitoring of marine litter is the difference in the applied methods and tools in different regions of the world, which complicates the comparison of monitoring results.

The international OSPAR project "Marine Beach Litter" on monitoring marine beach litter (2000–2006) (OSPAR – Convention for the Protection of the Marine Environment of the North-East Atlantic (OSPAR) dated September 22, 1992) became the first in Europe to develop a standardized method for monitoring marine macro-litter on beaches in the North Atlantic region, in which more than 600 surveys were conducted on 51 beaches in eight countries. This project identified the main sources of pollution on European coasts (fishing, waste water, shipping, tourism). The most common items were plastic and polystyrene [1].

The developed marine litter monitoring methodology is reflected in the Marine Strategy Framework Directive Guidance, as well as in the OSPAR Guidelines.

According to the MSFD recommendations, beaches should be selected based on different types of anthropogenic pressure (urban and suburban beaches, etc.) and meet the OSPAR criteria [2]. The survey is recommended to be carried out on at least two 100-m strips on one beach. However, there are still no specific recommendations on the optimal number of such areas for beaches, depending on the level of pollution.

Marine litter is usually classified by the size: "macrolitter" – particles >25 mm in diameter, "mesolitter" – 5 to 25 mm, and "microlitter" – <5 mm [3]. Macrolitter is the most visible for human eye on beaches or floating on the surface. Thus, there is yet much more information on macrolitter campaigns and monitoring. Some data on the amounts of litter on the coasts of the Baltic Sea is available already from the late twentieth century. This information is based on campaigns carried out by various non-governmental organizations or on observations by coastal municipalities. It is not, however, possible to quantitatively compare the results between the campaigns because different methods have been used for collecting litter and estimating their amounts. It also very important to select the most representative beaches for the campaign considering meteorological (storm events frequency), hydrographical, and geomorphological processes.

OSPAR recommendations are widely used in Europe, and on their basis volunteers from different countries participate in beach surveys using standard protocols. In the Baltic region, project MARLIN has united efforts of volunteers and scientists in Sweden, Finland, Estonia, and Latvia, with a total of 120 beach litter assessments throughout 2011–2013 who conducted beach litter surveys in 20 key areas [4]. This project aimed at obtaining new knowledge about marine litter in the Baltic Sea, as well as raising public awareness. For the first time around the Baltic Sea area, all the countries collected and categorized the litter using the same harmonized method based on the protocol of UN Environment Programme on beach litter [5]. The results of this project showed that most of the beach litter in the Gulf of Finland was composed of plastic: 59% on urban beaches, 50% on rural beaches, and 53% on semi-urban beaches, with the highest amount of litter on the Finnish beaches: urban beaches tended to contain more litter than the rural ones. The snow melting period affected the accumulation of litter on beaches as well [4]. Finland and Estonia have continued the monitoring of these beaches in 2014–2015, and Finland has also adopted this protocol into its national monitoring program.

Beach litter accumulation is now the most studied in the Baltic region as compared to plastics distribution in water and on the seafloor; however, the methods for an adequate and harmonized assessment of the distribution and sources of marine litter are still under development. The sources of marine litter here are mainly land-based and are associated with poor waste management including littering, wastewater, and rain drainage management. In European seas over 60% of all marine litter are plastic packaging, predominantly plastic bottles and bags [1]. Previous assessments show that in the Baltic Sea the main sources are considered to include transport, fisheries, household activities, as well as coastal recreation and tourism [6].

The OSPAR method of beach litter survey has also been adopted by HELCOM as a methodology for monitoring beaches in the Baltic Sea, and is described in Recommendation 29/2 in order to obtain comparable results [7]. However, it became obvious that this method is only suitable for wide open beaches of the Atlantic coast, and for closed marine ecosystems, this method must be adapted and refined.

An adaptation of the OSPAR method of beach monitoring was completed by the Leibniz Institute for Baltic Sea Research, Warnemünde, IOW (Leibniz-InstitutfürOstseeforschungWarnemünde, IOW) [8, 9]. Authors have developed a special tool for sieving the sand – "Sand Rake" for open-type Baltic beaches, while using the sand sieve inside a frame for lagoon-type Baltic gulfs and estuaries.

This chapter combines an overview of several studies of marine litter distribution on beaches and in sediments for two Russian parts of the Baltic: the Gulf of Finland, where studies were started in 2018 and the South-East Baltic with longer period of monitoring – since 2016. Various methods for sand sampling that have been applied during the period of 2016–2020 and the results of these studies are discussed briefly.

2 Marine Litter Study in the Eastern Part of the Gulf of Finland

2.1 Study Area: Neva River Inner Estuary and Outer Part of the Gulf

The Gulf of Finland is a water area shared by the three countries (Fig. 1): Finland, Russia, and Estonia and is one of the most unique and fragile ecosystems in the

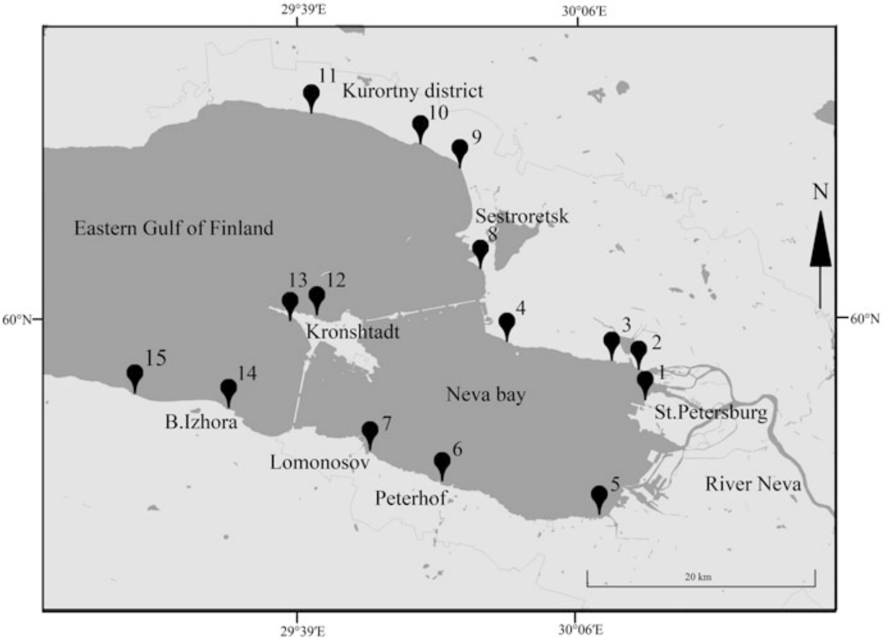


Fig. 1 The Gulf of Finland and Neva Bay, sampling stations in 2018–2019

Baltic Sea due to its special hydrophysical and geomorphological characteristics and pronounced estuarine effects, caused by the inflow of the largest Baltic river – the Neva, with the average annual discharge of 2,500 m^3/s. It is a shallow and brackish ecosystem with a low but unique biodiversity [10]. The intense anthropogenic activity in the highly populated area around the Gulf of Finland is the reason of its main environmental problems: eutrophication, oil and hazardous substances pollution, underwater landscape degradation due to dredging and resources extraction, etc. making this ecosystem very sensitive to the growing human impact.

St. Petersburg is the largest city in the North-West of Russia at the easternmost tip of the Gulf of Finland with over 5,200,000 of permanent residents [11]. Large area of the Russian Gulf of Finland coast is situated in the Leningrad Region with a total number of permanent residents over 1,900,000 people [11]. High population density creates a significant pressure on the Gulf's environment.

The plastic litter problem has never been investigated for the Russian sector of the Gulf of Finland. Thus, during the period of 2018–2019 a pilot study of the problem of marine litter pollution of the coasts of the eastern Gulf of Finland was carried out by the Russian State Hydrometeorological University [12, 13] taking into account the existing experience of marine litter monitoring in the Baltic lagoons and estuaries.

The high population density in the region of the Gulf of Finland together with production of large amounts of plastic wastes poses a high risk of marine litter pollution. The metropolitan area of St. Petersburg together with the Leningrad Oblast produces annually about 112,000 tons of plastic wastes including municipal and industrial wastes [15]. However, due to the absence of any regular monitoring activities here it is not possible to give a quantitative estimate of beach litter pollution levels. An important source of plastic litter in the eastern part of the Gulf of Finland before the construction of the Flood Protection Barrier of St. Petersburg (FPB) was a network of waterways in St. Petersburg and suburbs including over 220 rivers, canals, and streams of various length, as well as reservoirs. From the start of FPB operation there is a constant threat of accumulation of floating litter on the east side of the dam. Entering the water environment, the litter is eventually accumulated and buried on the muddy seafloor of the Neva Bay, but can be released again during the permanent dredging works in the Neva Bay and near harbors of St. Petersburg. This can serve as a secondary source of litter pollution that enters the environment and reaches the coasts after storm events.

The coastline of the Russian part of the Gulf of Finland is very diverse and was formed by subaerial and tectonic processes (skerries), non-marine processes (alluvial plains), by waves (marine erosion, accretion, abrasion coasts) as well as technogenic processes (embankments, hydrotechnical constructions, etc.) [16]. The most active erosion processes occur in the coastal zone of the easternmost part of the Gulf – which is the most valuable recreation area. The easternmost part of the coastline within the Neva River mouth is completely transformed by the technogenic processes.

In terms of the recreation potential the most visited sandy beaches of the Russian part of the Gulf of Finland are located in the Kurortny District (northern coast) and near Peterhof and Lomonosov area (southern coast). Also, long sandy beaches are found further south in the Narva Bay, but due to their remoteness they are not so popular among the local residents. Kurortny District hosts 12 the most popular and visited public beaches in the region that are regularly cleaned by the municipal services (major cleaning before each summer season and then waste is removed twice a day in summer) [11]. However, there are many of the so-called wild beaches in between, that are cleaned randomly throughout the season due to inability of the municipal services to cover the entire 60 km long coastal strip in this district.

For the Russian part of the Gulf of Finland, no deep scientific research on marine litter has been carried out yet, except for several beach cleaning campaigns in 2013–2015 held by the St. Petersburg Administration and local municipalities to increase awareness of marine litter issues amongst the citizens.

2.2 Monitoring Methods for Litter on Sandy Beaches

The Russian coast of the Gulf of Finland in general is characterized by the limited amount of suitable sandy beaches according to recommendations of MSFD and OSPAR (without regular cleaning, exposed sandy beaches without vegetation, little tourism). So, in order to make an assessment of litter pollution in most of the accessible parts of the coastline of the Russian part of the Gulf of Finland an IOW approach [8] was used for different types of beaches in the summers 2018–2019 [12, 14] with application of two beach sand sampling methods – Rake and Frame method. The Rake method and the Frame method focus on large-micro (>2 mm) and mesolitter (5–25 mm) in the 30–50 mm upper sediment layer and were applied at 58 surveys at 15 sandy beaches of the German and Lithuanian Baltic Sea coast between 2014 and 2016. The first rake experiments on the German coast of the Baltic Sea showed that the use of a mesh size of 5 and 2 mm is sufficient. Accordingly, the sand only needs to be sieved twice to the smallest mesh size of 2 mm.

Both methods were developed for sandy beaches, including regularly cleaned ones, and tested during 2014–2016 to monitor 15 beaches in lagoons and estuaries in the southeastern Baltic region (Germany and Lithuania, about 60 samples) [8]. Most of the litter was represented by cigarette butts, plastic, and paraffin wax. These methods have shown the possibility of assessing the “hot spots” accumulation of marine debris on the beaches. An important advantage of these methods is that they do not require elaborated equipment or a laboratory, are low in costs, and can be carried out by volunteers.

The Frame method is used for the coasts of lagoons, closed bays, river estuaries, and always includes the wave wrack line (i.e., the zone of influence of waves and material accumulation). The method was the main one for surveying the coasts of the Neva Bay – the enclosed estuary of the River Neva separated from the outer part of the Gulf by the Flood Protection Barrier (FPB).

The Sand rake method in contrast to the OSPAR method is applied vertically between the water line and the vegetation line along the entire width of the beach from the water line to the beginning of vegetation. The entire transect is divided on 5 m segments that are then sieved individually. For getting reliable results for most of the beaches two or three 0.5 m wide stripes will be sufficient to reach the minimum area of 50 m^2 or the total amount of litter found in all segments not less than 20 items, however at some beaches more stripes are needed. If two or more replicate samples are taken, there must be at least 120 m distance between the samples points to ensure that the rake sampling procedure fits to the 100 m distances as recommended for the selected point approach by OSPAR. (Fig. 2). The Rake method is aimed at surveying large areas of beaches that are regularly cleaned by municipal services from macrolitter. This method was chosen to survey the beaches of the outer part of the Gulf (the Kurortny District, the beaches of Kronstadt, and the southern coast near the FPB).

Fig. 2 Application of Frame and Rake methods on the beaches of the Neva Bay (photo by A. Ershova)

2.3 *Marine Litter in the Eastern Part of the Gulf of Finland: Results and Discussion*

During the summers of 2018–2019, surveys were carried out for seven lagoon-type beaches (Neva Bay) and eight beaches outside the Flood Protection Barrier on the northern and southern coasts of the eastern Gulf of Finland. In general, the number of litter particles of different types differs depending on the type of sampling method, which is primarily associated with the sampling area. The Frame method is aimed at examining the wave wrack-line zone, and the Sand rake method is aimed at the entire width of the beach from the water line to the vegetation line.

In both years the amount and distribution of anthropogenic litter along the coasts varied significantly depending on the location of the beaches, weather conditions, hydrological and morphometric characteristics of the studied coasts.

In 2018 in the Neva Bay, the concentration of litter (of all categories) was: average – 8.3 pcs/m^2, minimum – 1.6 pcs/m^2, maximum – 14 pcs/m^2; the average concentration of microlitter – 3 pcs/m^2, mesolitter – 3.8 pcs/m^2. The main types of litter found on the beaches of the Neva Bay and the eastern part of the Gulf of Finland (both in the inner estuary and in its outer part) were plastic pellets, broken glass, cigarette butts, rusty metal, and pieces of building plaster (Fig. 3). Also, among the mesolitter there were many synthetic napkins, cotton bud sticks, and other polymer products that were being flushed down the drain and are often not caught by treatment facilities and go directly into the waters of the Gulf of Finland.

The largest amount of litter of all fractions is found on the beaches of the inner estuary in the Neva Bay, despite regular beach clean-ups in the area. In the outer part of the estuary, microlitter predominates, while in the Neva Bay meso- and macrolitter prevails, and pollution with microlitter is less pronounced. The prevalence of meso- and macrolitter here can be explained by the fact that most of the beaches are located within the city limits, where large urban areas with a high population density are located and, therefore, with a higher level of anthropogenic pollution. In addition, in general, beaches are not cleaned thoroughly enough, which leads to the accumulation of meso- and macrolitter in the inner estuary in the Neva Bay [17].

In general, the northern and southern coasts of the Neva Bay differed significantly in the amount and composition of litter (particle size, composition, quantity, etc.). In 2018 on the northern coast, the amount of litter was lower, but the most of it was plastic – 50–60% of the total amount of litter of all fractions. In the outer part of the estuary, the predominant type of litter was microplastic, the average amount of which is 0.8 pieces/m^2 in the wrack-line and 0.5 pieces/m^2 – using the Sand Rake method. Thus, the maximum amount of litter, both in mass and in the number of items, was found on the beaches of the southern coast of the Neva Bay (Fig. 4a) The southern coast of the Neva Bay is also significantly different in the composition of the collected waste – more than 50% is broken glass, plaster, and pieces of rusty metal. Plastic here accounted on average up to 10–12% of the total litter amount [14].

Fig. 3 Types of macro-, meso-, and microlitter collected on the sandy beaches of the Gulf of Finland in 2018 and 2019 (plastic pellets, glass, metal pieces, cotton bud sticks, etc.). Photo by A. Ershova

In the outer part of the Gulf in 2018 the concentration of microparticles in general, and plastic in particular, on the northern coast in the Kurortny area, as well as on the northern beach of Kotlin Island, was 5–6 times higher than on the southern coast of the Gulf of Finland (B. Izhora and Lebyazhye), showing accumulation of microplastics on beaches mainly in the outer estuary, while in the inner estuary – in the Neva Bay microplastics are contained in small amounts (Fig. 4b).

Our studies have shown that the selected methods are applicable on the coasts of the Gulf of Finland (both the outer part of the estuary and the Neva Bay). At the same time, these methods should be applied in parallel, and the research results can complement each other, since these methods are aimed at examining different functional zones of the coast: the wave wrack-line zone for analyzing the litter

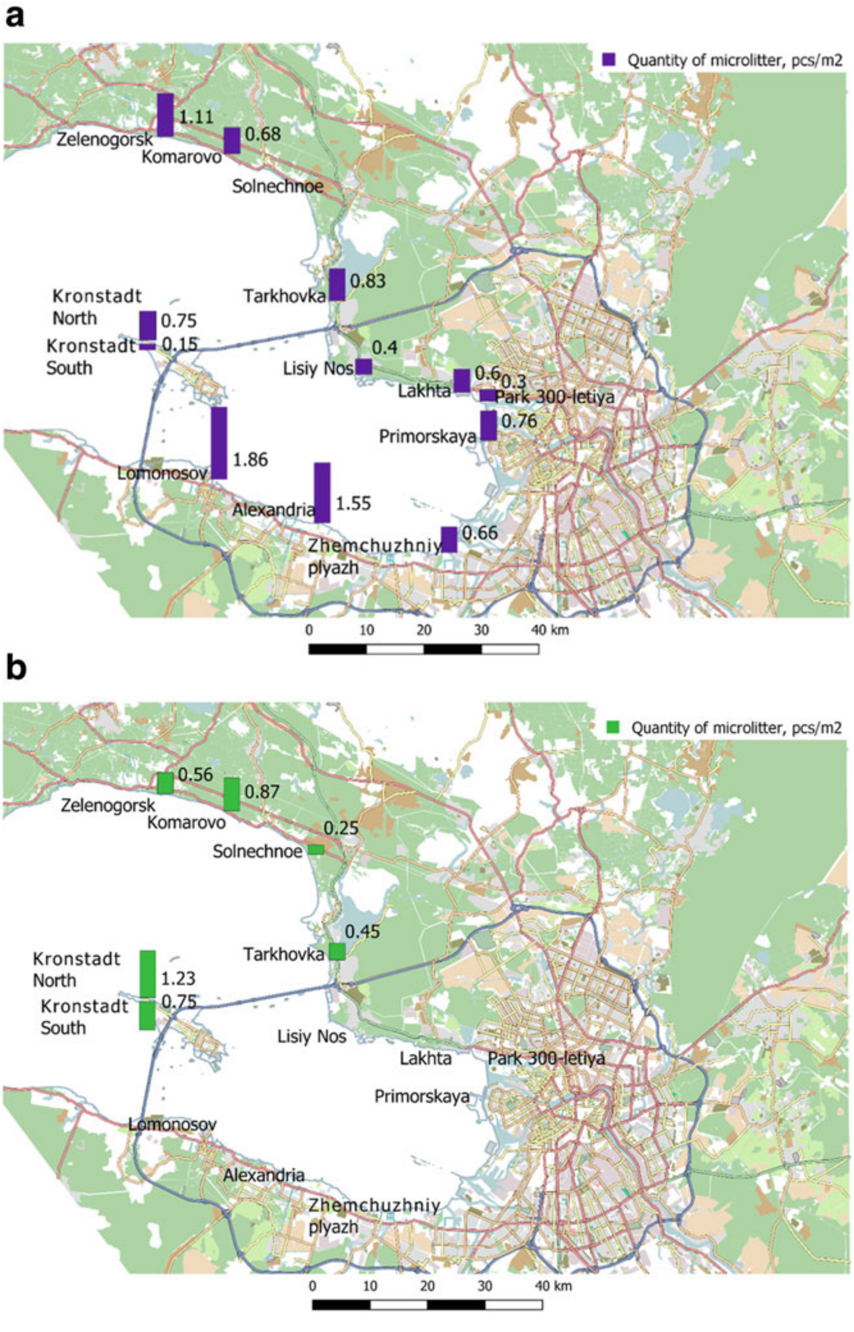


Fig. 4 (**a**) Concentration of marine microlitter, pcs/m^2 in the wave wrack-line zone on the coasts of the Neva Bay and the outer part of the Gulf of Finland (Frame method) in 2018. (**b**) Concentration of marine microlitter, pcs/m^2 on the beaches of the eastern part of the Gulf of Finland (Sand Rake method) in 2018

carried to the beach by sea waves, and on the other hand, the total accumulation of litter on the entire beach width (taking into account seasonal accumulation, the effect of storms, snow melting, etc.) and their sources (the sea, tourists, etc.).

3 South-East Baltic: Kaliningrad Region

The southeastern part of the Baltic Sea is a populated area with developed ship traffic, fishing industry, and a rather high recreation activity. Wide sandy beaches attract lots of visitors in all the seasons, and, at the same time, allow for application of various methods of monitoring of marine litter, e.g. OSPAR method [2], Rake and Frame methods [8], etc. In this section we focus on the results of assessment of beach pollution by anthropogenic litter collected by the Rake and Frame methods for large litter items (macro-, meso-, and large microparticles – $>$ 2 mm), and the modified NOAA method for microplastics (MP, 0.5–5 mm) during the summers of 2015–2020.

3.1 Studies of Beached Marine Litter by Various Methods

Marine litter on the beaches of the South-East Baltic was studied along all the (potentially different by contamination) shores of the Kaliningrad region (see map on Fig. 5): the western shore, open to the prevailing winds in the region, the northern shore, prone to the most severe N/NW storms, and unpopulated shores of the UNESCO National Park at the Curonian Spit.

3.1.1 Beaches of the Curonian Spit National Park

Quantitative estimates of marine anthropogenic litter and MPs in the Curonian Spit National Park and two adjacent popular urban beaches, Klaipeda (Lithuania) and Zelenogradsk (Russia) (Fig. 5) were performed in spring 2018 [18, 19]. The 100-km-long sandy Curonian Spit on the eastern coast of the Baltic Sea is shared by Russia and Lithuania, and both parts of the spit are National Park, with sea beaches 20–80 m wide [21]. The timeframe of the screening (early May, i.e. after the windy winter and well before the beginning of the tourist season) ensures that the vast majority of plastic waste found during sampling on the beaches has been brought ashore by the sea.

In order to understand the "instantaneous" picture of beach contamination, the sampling was carried out as a single effort, during May 1–2, 2018. Macro- (>25 mm), meso- (5–25 mm), and microlitter (2–5 mm) was studied at 5 points, while sand samples were collected at 6 points along the coast for further analyses of large MPs (L-MP, 2–5 mm) and small MPs (S-MP, 0.5–2 mm) particles. Since

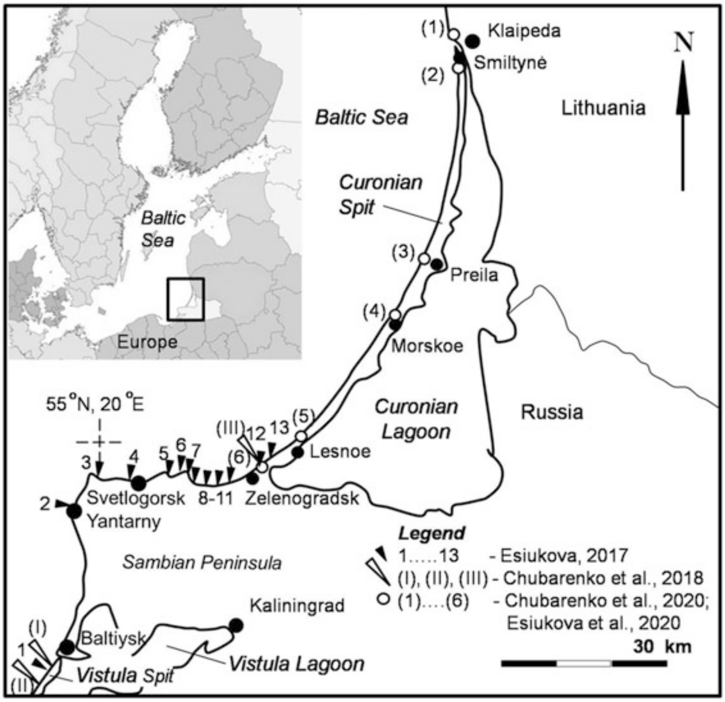


Fig. 5 Sampling locations in the southeastern part of the Baltic Sea in 2015–2020

significantly different fractions of plastic litter were of interest, two sampling methods were applied simultaneously: (1) the Sand Rake method for litter larger than 2 mm [8] and (2) the frame sampling method on the MP (0.5–5 mm) (see details in [19, 20, 22]) (a total of 50 sand samples were taken). 432 anthropogenic particles of litter were collected from a sample area of 135 m^2, resulting in an average surface concentration of litter of 3.2 items/m^2. It turned out that there is a 17-fold difference in the concentration of litter between the Klaipeda beach located in the industrial zone (249 objects of litter found on 10 m^2) and the beaches of the National Park (183 objects collected from 125 m^2), which gives the average surface concentration of litter (and per 1 m of shore length) of 24.9 items/m^2 (498 items/m) and 1.46 items/m^2 (77 items/m), respectively (Table 1). Artificial polymers accounted for 84% of all types of litter (363 items). The distribution of litter types (including paper, metal, glass, and other non-plastic anthropogenic litter) of various sizes was obtained [19, 20]. The Klaipeda beach was the most polluted, where microlitter (2–5 mm) makes up 60% of all items of anthropogenic litter (14.9 items/m^2 out of 24.9 items/m^2), and the abundance of meso- and macrolitter is about an order of magnitude higher than in other sampling points.

Western and northern shores showed the following contamination pattern. During 12 surveys (600 m^2) the amount of litter found was 1,164 items (mean 1.94

Table 1 Abundance of anthropogenic macro-, meso-, and microlitter (items/m^2 and items/m), collected by the Sand Rake method

Station no.	Macrolitter >25 mm	Mesolitter 5–25 mm	Microlitter 2–5 mm	Mean items/m^2/items/m	Sampled area, m^2	Beach width, m
(1)	15.1	6.6	3.2	24.90/498	10	20
(2)	1.1	0.3	0.4	1.82/100	27.5	55
(3)	0.4	0.5	0.3	1.17/35	30	35
(4)	0.9	1.0	0.5	2.34/152	32.5	65
(5)	0.1	0.3	0.1	0.63/22	35	35
Mean ± SD	3.5 ± 4.6	1.7 ± 1.9	0.9 ± 0.9			
Median	0.9	0.5	0.4			

items/m^2 ± 0.94; median 2.48 items/m^2), with a minimum of 0.58 items/m^2 and a maximum of 3.26 items/m^2. The size distribution of the litter was as follows: 370 microlitter items (0.62 items/m^2–31.8%), 497 mesolitter items (0.83 items/m^2–42.7%), and 297 macrolitter items (0.50 items/m^2–25.5%). On 6 beach areas with 2 surveys each, the small-scale spatial variation of litter per beach area was between 0.18 and 1.72 items/m^2 (mean 0.61 ± 0.52 items/m^2; median 0.44 items/m^2). Along the northern shore, the top contamination (3.26 items/m^2 and 2.76 items/m^2) was somewhat higher than along the southern shore (2.78 items/m^2 and 1.10 items/m^2). This was mostly due to high amounts of paraffin (mean 1.03 ± 0.81 items/m^2; median 0.68 items/m^2), contributing along the northern shore versus lower amounts of paraffin (mean 0.07 ± 0.03 items/m^2; median 0.08 items/m^2) along the southern shore. The lowest number of litter items along the southern shore was 0.58 items/m^2 and along the northern shore – 2.38 items/m^2. Mesoplastic items, paraffin, and industrial pellets were predominant, and artificial polymers accounted for 54% of all the litter items [14].

Sand samples for analysis for MPs content (0.5–5 mm) were additionally taken using a wooden frame 18 cm × 18 cm to a depth of 2 cm in four beach zones, with two replicates (about 5 m apart) in each zone: (I) the shore face, (II) the current wrack line, (III) the middle of the winter berm, and (IV) the storm wrack line (i.e., the line left after a storm of the past winter season). Briefly, the stages of extraction and identification of MPs (the modified NOAA method [23] based on the NOAA recommendations [24]) are as follows: sample drying → sieving (cascade of sieves 0.5 mm, 1 mm, 2 mm, 5 mm) → selection of MPs from a fraction of 2–5 mm (optical microscope + UV lamp) → density separation (400 g sample + artificial reference particles, $ZnCl_2$ solution, $\rho = 1.6$ g/cm^3) → filtration (mesh size 174 μm) → liquid oxidation (H_2O_2, 30% + Fe (II) at 75°C) → removal of the calcite fraction (HCl solution) → filtration (mesh 174 μm) → flotation in a separator ($ZnCl_2$, $\rho = 1.6$ g/cm^3) → filtration (mesh 174 μm) → filter drying (Petri dish) → MPs selection (optical microscope × 10 – × 40) → identification (Raman spectrometry). To allow for comparison, the results are reported in (1) items/kg of sediment dry weight (further in the text *items/kg DW*), (2) items/m^2, and (3) items/m of the coastline length [19, 20].

For MPs 0.5–5 mm, variations in abundance were studied both as a whole and with refinement for two size classes (S-MPs (0.5–2 mm) and L-MPs (2–5 mm)) for 4 beach zones. MPs (0.5–5 mm) were found in all 50 beach sand samples from 6 stations, with variations both between and within the station locality. On different beaches, the mean (for the beach zone) values for MPs (0.5–5 mm) range from 1,038 to 7,070 items/m^2 (both stations are located within the National Park area), with the global mean for all stations of 3,155 ± 1,308 items/m^2 ($n = 50$). The MPs distribution does not correlate with those of meso- and macrolitter. Most of the detected particles (74.3%) are various fibers and threads. Fragments and films were 23.1% and 2.6%, respectively. The global mean MPs concentration (0.5–5 mm) per sediment mass is 115 ± 61 items/kg DW ($n = 50$), which is well comparable to the average value of 108 items/kg DW found in [18] for the beach sands of the neighboring Vistula Spit. A deeper analysis of the distribution between MPs size fractions and beach zones at different stations showed that out of the total number of 5,127 particles detected in the size range from 0.5 to 5 mm, 5,102 items (99.5%) belong to the range of 0.5–2 mm (S-MPs).

L-MPs (2–5 mm) were found exclusively within wave wrack lines, while they are absent in the upper sands and on the berm. The difference between stations and beach zones in the range of S-MPs (0.5–2 mm) is obvious, and the analysis shows that the outliers (identified by quartile analysis) – i.e., the areas with extremely high contamination – are found in all the beach zones except of the wet beach face. The 95% confidence intervals of the means are 119 ± 86.8 items/kg DW for the wrack line, 57.5 ± 51.2 items/kg DW for the berm, 30.2 ± 15.6 items/kg DW for the current wrack line, and 52.7 ± 35.7 items/kg DW for the shore face. With (and without) statistically confirmed outliers, the median abundance for the entire dataset is 45.5 ± 22.4 (41.6 ± 22.2) items/kg DW, and the average is 114.8 ± 61.4 (73.9 ± 22.6) items/kg DW.

Thus, the contamination by S-MPs is larger on the dry part of the beach, and is heterogeneous there, with a large standard deviation and the presence of outliers. The largest contamination is associated with the wrack line, which is consistent with other studies for this area (e.g., [18, 22, 24]). The closer to the waterline, the smaller the difference between the mean and median values, and at the wet beach face they become equal: at all stations and in all replicates, the contamination by S-MPs is the same. Moreover, the content of S-MPs in sands of the beach face (52.7 ± 35.7 items/kg DW) is close to the median value 45.5 ± 22.4/41.6 ± 22.2 items/kg DW for the entire S-MPs dataset covering all the sampled locations and all the beach zones. The general picture of contamination in the MPs range (0.5–5 mm) is quite similar for the Vistula Spit area (about 100 km to the southwest), the values are very close [18]: the peaks are about several hundred of items/kg DW, while the mean for all samples (excluding the peak values/outliers) is about 56 and 74 items/kg DW, respectively.

Thus, it was found that, while contamination by the MPs as a whole (0.5–5 mm) is variable both along the shore and across the beach, the MPs concentration at the wet beach face (the shoreline) is practically the same in all the areas and in all the replicates. The concentration of S-MPs (0.5–2 mm) at the beach face can potentially be used as an indicator for monitoring purposes: it provides (1) a benchmark for plastic contamination of the marine environment in a relatively large region (with more or less similar environmental conditions) and (2) an indicator to monitor rising levels of plastic pollution.

3.1.2 Beach Sediment Studies

An assessment of the degree of contamination by MPs (as well as macro/mesoplastic and paraffin) of beach sediments in the Russian sector of the South-East Baltic was carried out in June 2015 to January 2016 with sampling (60 samples) from the upper layer of the beach (up to a depth of 2 cm) in 13 areas of the Kaliningrad coast with different anthropogenic load (see map on Fig. 5). It was performed using a wooden frame with an area of $S = 0.15\ m^2$ and a metal scoop. In each of the 13 areas, 2–7 samples were taken from different points of the storm wrack line at some distance from each other [22].

The samples were dried, weighed, and sieved through a cascade of sieves (5, 1, and 0.5 mm). Fragments of macro- and mesoscopic litter (>25 mm and > 5 mm, respectively) were collected manually using tweezers, counted and weighed. Particles of large MPs (>1 mm – 5 mm) were sampled using an optical microscope and a UV lamp. A more detailed analysis method, the NOAA method, was additionally applied to 20% of the samples [25]. In short: density separation of a 400 g sample ($ZnCl_2$, $\rho = 1.6\ g/cm^3$) → filtration (174 μm mesh size) → liquid oxidation (H_2O_2, 30% + Fe (II) at 75°C) → removal of the calcite fraction (HCl solution) → filtration (174 μm mesh) → flotation in a separator ($ZnCl_2$, $\rho = 1.6\ g/cm^3$) → filtration (174 μm mesh) → filter drying (Petri dish) → MPs selection (optical microscope). The analysis was carried out on a stereomicroscope (Micromed MC2 Zoom Digital) with a magnification from 10 × to 40 × on the filter surface in accordance with the recommendations for determining the MPs [26]: (1) the cell structure and other organic forms are absent in the particles, (2) the fibers must be uniform in color and thickness along their entire length, not segmented, (3) colored particles are uniformly colored, (4) particles must have a clean and uniform color. Macro/mesoplastics, MPs, paraffin wax, amber were counted as separate categories (Fig. 6). Analysis has shown that plastic litter is found at all sampling points and in all seasons. The level of contamination of the Kaliningrad beaches with MPs is indicated in Table 2.

A comparative analysis with beaches around the world showed that the beaches of the Kaliningrad region are quite clean and there is no obvious difference in the contamination of beaches with high (near resort cities) and low (in fairly deserted, less-visited coastal areas) anthropogenic load. The predominant type of the detected MPs contamination is foamed plastic (foam/polystyrene foam). Paraffin, widely present in the area, the source of which is mainly the discharge of tank waters from ships, contributes to the accumulation of MPs: it is an effective “accumulator” of various types of contamination.

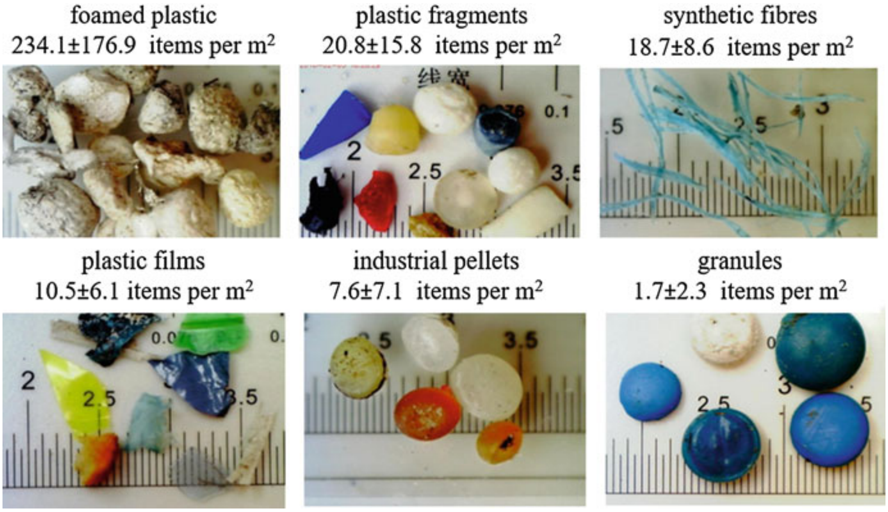


Fig. 6 Different types of MPs in beach sediments in the south-eastern Baltic (items per m^2), photo by E. Esiukova

Table 2 Abundance of MPs in sand samples from various Kaliningrad beaches

	mg/g DW	mg/m^2	items/m^2	items/kg DW
Minimum	$80 \cdot 10^{-3}$	67	7	0.2
Maximum	8.38	16,000	5,560	175.3
Mean	0.05–2.89	370–7,330	42–1,150	1.3–36.3

3.2 *Specific Pollutants: Geosynthetics, Paraffin, Foamed PS*

Wrack of *geosynthetic materials* is a new contaminant emerging from coastal engineering protection activities [27]. There are several main categories of geosynthetics: geotextiles, geogrids, geonets, geomembranes, clay mats (bentonite mats), geopenes, geocells, drainage/infiltration cells, geocomposites [27], as well as woven containers (big-bags). Deformation and destruction of coastal and hydraulic protective structures (gabions, reinforced slopes, retaining and protective walls, protective berms, etc.) containing such components lead to the leakage of remnants of geosynthetic materials into the marine environment. When migrating along the shore, fragments of geosynthetics undergo additional degradation, which leads to their destruction down to macro/meso/microparticles. In water samples collected in the economic zone of the Russian Federation in the Baltic Sea (within the Kaliningrad region), microplastic fibers were found, similar to those obtained during the degradation of geotextile materials such as Dornit [29]. The samples taken from the beach surface also showed the presence of fragments of plastic sheathing from gabions [27].

Fig. 7 Examples of geosynthetic materials found on the beaches: (**a**–**c**) fragments from woven big-bags used in the construction of a promenade in Svetlogorsk and other engineering and coastal protection structures on the northern coast of the Sambian Peninsula, (**d**–**e**) braids from gabions, (**f**–**j**) Dornit-type geotextiles. (Photo by E. Esiukova)

Thanks to the international project of the ERA.Net RUS Plus program "Environmental impact of geosynthetics in aquatic systems" (No RUS_ST2017-212, http://ei-geo.com/), attention was paid to this hot problem. Several local potential sources of beach contamination by geosynthetics (coastal protection structures) were identified in 2018–2020 on the shores of the Sambian Peninsula (Kaliningrad region), and the contamination of sandy beaches by such material was surveyed [27, 28], see Fig. 7. It turned out that the patchy distribution of degraded geosynthetic residues on the beach does not allow the use of well-known methods for assessing the degree of contamination with macro- or MPs (i.e., selected-area methods for their search, such as the OSPAR method, etc.).

A new technique of continuous visual scanning was tested [30, 31] for accounting of the remnants of geosynthetic materials (mega/macro/mesofragments) during the survey of the beach strip. According to the proposed technique of continuous visual scanning, observers must pass along the entire shore, covering the entire width of the beach – from the coastline to the foredune or cliff. The beach is divided into zones (stripes) along the sea, and a group of several people (2–4, depending on the width of the beach and the number of zones) follows along their zones. Each person visually inspects the strip of his/her zone so as to capture the edge of the neighboring zone – for a complete scanning of the entire beach. Each section is being studied (several kilometers – 5–10 km in one effort) with fixing the position (by GPS) and collecting samples of geosynthetics. Participants are equipped by prepared in advance palettes with samples of geosynthetics previously found in local sources (structures) or collected during a preliminary survey of beaches [30, 31]. The main difficulties and problems with such monitoring arise when the sample is not noticed and not taken into account due to: (1) the sample being covered with sand or being in a heap of pebbles/boulders/algae; (2) the sample being smeared or covered with algae or dirt; (3) an unknown type of geosynthetics or modified to the point of impossibility of identification; (4) the sample being located in an inaccessible place (under water, or at a distance from the shore on groins and reinforcement/remains of technical structures, on trunks and branches of trees fallen into the sea, etc.); (5) inattention or fatigue of the person. However, such a technique of continuous visual scanning is the only way (at the moment) to get a sufficiently complete visual picture of not only the level of contamination and distribution of geosynthetic fragments along the coast, but also to identify the features and characteristics of the collected samples, and, if possible, associate this garbage with a local source of beach contamination. Active alongshore transport of fragments of geosynthetics (for example, containers/big-bags) is proved by the observation of the remains of these bags along the entire northern coast of the Sambian Peninsula, and farther north – along the Curonian Spit, including the territory of Lithuania (according to the results of monitoring in 2019). This way, MP particles (fibers) from big-bags were found in sand samples collected on the beaches of the Curonian Spit [20, 21].

Paraffin of various fractions and types is often carried ashore, contaminating the beaches of the Baltic Sea. The exact source of the contamination remains usually unknown, however international experts have established that it comes from washing of tanks and the unauthorized discharges of cargo residues, contaminated ballast water [32, 33]. Experts admit that the reason for the contamination of beaches with paraffin may be emergencies during the development of offshore oil and gas fields and the transportation of oil products by tankers and pipelines, man-made accidents on ships engaged in underwater work near the coast. During winds and storms, paraffin is thrown ashore. Given the system of currents in the Baltic Sea, the source of contamination may be far enough from the point where the paraffin is washed out onto the beach. During the expeditions [22], rather large pieces of paraffin (up to 100–300 g) were found, which can lie everywhere on the beach. However, most of it is dispersed along the wrack lines.

Most often, pieces of paraffin are mixed with sand, particles of organic origin, adhering litter. In order to assess how contaminated the paraffin is with MPs, paraffin samples were taken from 13 areas of the sea coast of the Kaliningrad region [22]. Each piece was weighed, poured in hot water; it melted and floated like a film to the surface of the water, while the adhered sand remained at the bottom. The sample was then cooled at room temperature for 24 h, then the paraffin was collected from the surface, dried and weighed, then examined with an optical microscope and a UV lamp for the presence of plastic particles [22]. In paraffin, microplastic particles were found in 92% of the samples (12), together with organic particles (algae, grass, insect, and zooplankton fragments), amber, and charcoal particles. The sand that fell to the bottom was about 31.0 ± 5.8% of the initial weight of the paraffin sample. MPs were mainly represented by fragments of foamed plastic (expanded polystyrene), synthetic fibers, and plastic films. Analysis shows that the content of MPs in paraffin samples averages at 31.1 ± 18.8 items per sample, or 11,479 ± 10,785 items per kg of a paraffin sample, or 47,628 ± 47,567 items per kilogram of sand from paraffin samples [22]. These values for the MPs content in the paraffin are three orders of magnitude higher than in the surrounding beach sediments. This indicates that the light and rather sticky paraffin (especially aged items that are uneven, in cracks), collected from the wrack line on the beach, is an effective "accumulator" of various types of contamination. The analysis showed that all of the surveyed beaches were contaminated with plastic, and paraffin was a frequent co-component.

One more specific contaminant revealed in sand samples along the sea shore of the Kaliningrad region is *foamed plastic (foamed PS)*. Sampling sands in 13 locations of the beach (in total 60 sand samples from the upper 2-cm layer) using a frame with an area of 0.15 m^2 showed contamination by foamed particles from 0 to 5,380 items/m^2 (mean 234.1 ± 176.9 items/m^2 – from 7 ± 16 to 1,056 ± 1822 items/m^2) [22], mainly in areas with a complex system of currents and eddies along the coast. An increased content of foamed plastic in storm wrack lines was also noted during studies of the three-dimensional distribution of anthropogenic litter in the body of the beach in [18].

3.3 Three-Dimensional Distribution of Marine Microlitter in the Sand Body of the Beach

Level of contamination by anthropogenic microlitter and its distribution in the body of a sandy beach on the shore of the Kaliningrad region was examined on May 5 (St. I), May 6 (St. II), and May 27 (St. III) 2016 [18], see Fig. 5. Beaches with significantly different anthropogenic load were selected for this study: (I) a visited beach near a village on the Vistula Spit, (II) a sandy coast in a remote and deserted area of the Vistula Spit, and (III) a coast near the city beach of the resort town of Zelenogradsk. Sampling was carried out (1) from vertical pits (from 48 to 143 cm in depth) made on a storm wrack line in the middle of the winter berm (8–17 m from the shoreline), (2) from the beach face, and (3) on the beach surface out of wrack lines. Sampling was carried out layer by layer using a cylindrical sampler (metal pipe Ø 15 cm, H = 8 cm) and a sapper blade with a handle installed at an angle of 90° to the shovel handle – from the surface to the level of the appearance of sea water at the bottom of the pit. A long, rigid level bar and a folding wooden building meter were used to measure the depth. At point (Ib), from a ladder on a slope near the waterfront (seven steps were dug up to the sea water level), sand samples (100–170 g each) were taken from the vertical walls of five steps using a metal scoop from visually different layers (sands of different colors and grain size composition). Additionally, sand samples were taken from an area of 0.15 m^2 on the storm wrack lines near the location of the pits (upper layer up to a depth of 2 cm), using a wooden sampling frame and a metal scoop. Moreover, samples were taken from the upper 8 cm layer using a cylindrical sampler at four points with an interval of 10 m along the middle of the beach (16–18 m from the shoreline), regardless of the location of the storm wrack line. All samples were packed in new plastic bags with a string lock and delivered to the laboratory.

In the laboratory, the samples were dried at room temperature, weighed (with the precision 0.1 g), and sieved through a cascade of sieves (5, 1, and 0.5 mm). Those parts of the sample that passed through a 0.5 mm sieve were discarded. The MPs particles, visually identified in sand on a 1 mm sieve, were selected with tweezers, and the remains of sand from 1 and 0.5 mm sieves were combined for further processing. All material collected from 1 and 5 mm sieves was sorted by hand to separate MPs particles from various inclusions (algae, shells, charcoal, paraffin, etc.) using a magnifying glass, optical microscope, and UV lamp. The following types of litter were identified: meso/macroplastic (>5 mm/>25 mm), microparticles (<5 mm), paraffin, amber, organic matter, charcoal, slag, mica, shells, pebbles. Litter of anthropogenic origin (most of which is plastic) was weighed (accuracy 0.1 g); other ingredients were only assessed qualitatively (no/little/much). At this stage, rather large particles were collected, and the putative plastics were visually examined using a UV lamp, mechanical action [34, 35], hot needle testing [36]. The modified NOAA method for extracting MPs from samples (according to [23]), developed on the basis of NOAA recommendations [25], was applied to the part of the sample that remained between the 5 and 0.5 mm sieves (see short description above). Final analysis was performed on a stereomicroscope (Micromed MC2 Zoom Digital) with a magnification of 10× to 40× on the filter surface in accordance with the recommendations for determining MPs [26].

All the collected 52 samples from different horizons of the beach stratum contained MP particles: fragments, films, and flakes, as well as fibers. This classification is based on the general shape of the particles: 3-dimensional (all three main sizes are comparable), 2-dimensional/flat (flexible films and more or less rigid flakes), and 1-dimensional/long (fibers, threads, lines). MP abundancies in the samples (see Table 3) range from 2 to 572 items/kg DW, the average value is 108 items/kg DW, and the average value excluding samples from storm wrack lines and layers with peak concentrations is only 56 items/kg DW.

The maximum MPs abundance in all three areas was found within the range of storm wrack lines on the beach surface – about 400–600 items/kg DW. Two distinct layers with the same level of contamination (about 300 items/kg DW) were found in the thickness of the beach in area (II) at a depth of 40–48 cm and 64–72 cm below the beach surface. These values are followed by the MPs concentration on the bench in the area of the current wrack line – about 150–450 items/kg DW. In the body of the beach sands (excluding obviously peaking points), the average concentrations are an

Table 3 Quantitative distribution of various forms of MP particles in the samples. Locations see on Fig. 5

Location	Sampling from	Fibers, items/kg DW	Films and flakes, items/kg DW	Fragments, items/kg DW
(I)	Wrack line	5–14	3–8	31–365
(II)	Wrack line	307–325	8–17	14–60
(III)	Wrack line	241–335	19–92	124–218
(III)	8 cm surface layer	13–117	0–2	0–1
(Ia)	Pit	8–132	0–14	0–6
(Ib)	Pit/stairs at shore face	117–393	6–19	0–19
(II)	Pit	11–316	1–30	0–27
(III)	Pit	2–15	0–2	0–1

order of magnitude lower (about 10–20, and up to 100 items/kg DW), with similar contamination levels characteristic of the beach surface outside the storm wrack lines (15–118 items/kg DW). The average MP concentrations for the beach strata (with the exception of several peak layers) are of similar order in all three areas: 28 items/kg DW on the beach near the village on the Baltic Spit, 63 items/kg DW in a remote rarely-visited beach, and 7 items/kg DW on a popular city beach (with cleaning of the territory). In particular, in absolute values, the most polluted beach is the most remote and deserted one, while the most visited beach has the lowest contamination. This difference obviously has a non-anthropogenic cause and should be attributed to natural factors.

The distribution of MP particles by depth is uneven and does not show any tendency. It is important that surface contamination does not reflect the MPs content in the sediment stratum: only about 3% (I), 1.5% (II), and 4.5% (III) of the total number of MP particles in the pits was located within the 8-cm thick surface layer. MPs abundance in the surface 2-cm layer of storm wrack lines (i.e., in the most contaminated samples) is also not related to the average concentrations in the beach body: their ratio is about 7,300% for a city beach (III), 400% for a deserted beach (II), and 600% for an ordinary beach (II). That is, a heavily contaminated storm wrack line (mean MPs concentration of 513.5 items/kg DW) rests on the surface of a relatively clean sand (on average 7 items/kg DW) of area (I), while at the remote beach (II), with the mean contamination of 90.5 items/kg DW, a storm wrack line has "only" 365.5 MPs items/kg DW [18].

In the size distribution of MPs particles found in the body of the beach, as many as 98% are in the range of 0.18–3.5 mm, while only 2% are in the range of 3.5–10 mm. This size distribution resembles that reported by Cozár et al. [37] for particles floating at the ocean surface; moreover, there, maximum of the distribution is close ~0.8 mm, while in our case 18% of particles are in the 0.6–0.8 mm range. All our samples contained fibers that were classified as colored and colorless. Colorless translucent fibers dominated. Colored fibers are mainly lustrous pink, blue, red, and green with a range of lengths from ~0.5 mm to several centimeters. Films and flakes were predominantly translucent gray, black, white, and beige, as well as opaque, matte, blue, green, and yellow in the size range from 0.5 mm to 1–3 mm in length. Films were mostly abraded with traces of strong degradation and/or biofouling. The wrack lines in all three regions (I, II, III) show an increased content of foam/expanded polystyrene particles, which is in good agreement with observations at other shores of the Kaliningrad region [22].

Summing up the results of this analysis: (1) The oceanographic reasons for the three-dimensional distribution of MPs in the thickness of the sandy beach are confirmed. (2) Sands from the beach and coastal underwater slope have the same background contamination by MPs. Thus, beach and underwater slope exchange particles of anthropogenic litter, as it happens to natural sediments, especially under the influence of stormy waves. (3) The spots of peak contamination in the body of the beach are associated with influence of stormy events. (4) Coarse-grained sands of dynamically active beach zones/layers are more contaminated. (5) The size distributions of MPs particles in the body of the beach and on the ocean surface are unexpectedly similar.

4 Conclusions

In general, the conducted studies have shown that the methods for beach litter survey (Frame and Sand rake methods) for assessing the composition of marine litter on sandy beaches (from large micro- (2–5 mm), meso- (5–25 mm) to macrolitter (>25 mm)) should be applied in parallel, and the research results can complement each other, since these methods are aimed at examining different functional zones of the coast: on the one hand, the wave wrack-line zone for analyzing the litter carried to the beach with sea waves, and on the other, for analyzing the total accumulation of litter on the entire beach (including seasonal accumulation, storm events, snow melting, etc.) and the sources of their supply (the sea, tourists, etc.). Testing of beach litter methods in the Gulf of Finland and the South-East Baltic revealed certain limitations of the applicability of these methods in changing weather conditions, as well as their dependence on the granulometric composition of beach sand and the level of eutrophication of the water area.

Results of studies held in 2018–2019 showed an overall high level of contamination with marine litter and its polymer components, microplastics in particular, on the Russian beaches – both regularly cleaned and “wild” isolated beaches. The largest amount of litter of all fractions was found on the beaches of the inner part of the estuary in the Neva Bay. The outer part of the estuary is dominated by microlitter, and in general, the coasts of the Neva Bay differ significantly in the amount and composition of marine litter: there is more heavy glass and metal microparticles in the southern shores, and northern shores tend to accumulate lighter microplastic particles. Microplastics averaged up to 10–12% in total amount of marine litter accumulating to a greater extent in the open part of the Gulf of Finland than in the Neva Bay.

As for South-East Baltic, microplastics pollution as a whole (0.5–5 mm) is variable both along the shore and across the beach and microplastics concentration (0.5–2 mm) at the beach face can potentially be used as an indicator for monitoring purposes: it provides (1) a benchmark for plastic contamination of the marine environment in a relatively large region (with more or less similar environmental conditions), and (2) an indicator to monitor rising levels of plastic pollution. A comparative analysis with beaches around the world showed that the beaches of the Kaliningrad region are quite clean and there is no obvious difference in the contamination of beaches with high (near resort cities) and low (in fairly deserted, less-visited coastal areas) anthropogenic load.

However, the predominant types of litter differ significantly among these two Baltic regions: predominant type of litter in the South-East Baltic is foamed plastic (foam/polystyrene foam), together with paraffin, widely present in the area, and the source of which is mainly the discharge of tank waters from ships. There is also a specific pollutant in this area: *geosynthetic materials* that are a new contaminant emerging from coastal engineering protection activities. In the Gulf of Finland region, the most common litter items are plastic pellets, broken glass, cigarette butts, rusty metal, and pieces of building plaster. Also, among mesolitter there were many synthetic napkins, cotton bud sticks, and other polymer products that being flushed down the drain are often not caught by treatment facilities and go directly into the waters of the Gulf of Finland.

The data collected during these studies will be included in the Baltic Sea marine litter database for lagoons and estuaries and urban and suburban beaches. Based on the obtained results general recommendations for the national program of marine litter monitoring will be developed for the Russian coasts of the eastern part of the Gulf of Finland, harmonized with the international monitoring programs in the Baltic region.

References

1. OSPAR Commission (2007) Monitoring of marine litter on beaches in the OSPAR region. OSPAR Pilot Project on Monitoring Marine Beach Litter Report. 75 p
2. OSPAR Commission (2010) Guideline for monitoring marine litter on the beaches in the OSPAR maritime area. https://www.ospar.org/ospar-data/10-02e_beachlitter%20guideline_english%20only.pdf
3. Moore CJ (2008) Synthetic polymers in the marine environment: a rapidly increasing, long-term threat. Environ Res 108:131–139
4. MARLIN (2013) Final report of Baltic marine litter project MARLIN – litter monitoring and raising awareness. http://projects.centralbaltic.eu/project/447-marlin
5. Cheshire AC, Adler E, Barbière J, Cohen Y, Evans S, Jarayabhand S, Jeftic L, Jung RT, Kinsey S, Kusui ET, Lavine I, Manyara P, Oosterbaan L, Pereira MA, Sheavly S, Tkalin A, Varadarajan S, Wenneker B. Westphalen G (2009) UNEP/IOC guidelines on survey and monitoring of marine litter. UNEP regional seas reports and studies no. 186, IOC Technical Series No. 83, 120 p
6. HELCOM (2009) Marine litter in the Baltic Sea region: assessment and priorities for response. Baltic marine environment protection commission. Helsinki, Finland, 20 p
7. HELCOM (2008) HELCOM recommendation 29/2. Marine litter within the Baltic Sea region. http://www.helcom.fi/Recommendations/en_GB/rec29_2/
8. Haseler M, Schernewski G, Balciunas A et al (2018) Monitoring methods for large micro- and meso-litter and applications at Baltic beaches. J Coast Conserv 22:27
9. Schernewski G, Balciunas A, Gräwe D et al (2018) Beach macro-litter monitoring on southern Baltic beaches: results, experiences and recommendations. J Coast Conserv 22:5
10. The Gulf of Finland Assessment. In: Raateoja M, Setala O (eds) Reports of the Finnish Environment Institute 27/2016. Finnish Environment Institute (SYKE). 368 p. ISBN 978-952-11-4578-0 (PDF)
11. Administration of the Federal State Statistics Service for St. Petersburg and the Leningrad Region (Petrostat) http://petrostat.gks.ru/wps/wcm/connect/rosstat_ts/petrostat/ru/statistics/Leningradskaya_area/population/. Retrieved on 24 Jan 2019
12. Eremina T, Ershova A, Martin G, Shilin M (2018) Marine litter monitoring: review for the Gulf of Finland coast. IEEE/OES Baltic international symposium (BALTIC), 8 p. https://doi.org/10.1109/BALTIC.2018.8634860

13. Eremina T, Ershova A, Shilin M (2018) Marine litter monitoring in the Neva Bay (Baltic Sea): first results. MICRO 2018 fate and impact of microplastics: knowledge, actions and solutions. Conference proceedings. 19–23 Nov 2018, Lanzarote, Spain. pp 48–49, ISBN 978-84-09-06477-9
14. Haseler M, Balciunas A, Hauk R, Sabaliauskaite V, Chubarenko I, Ershova A, Schernewski G (2020) Marine litter pollution in Baltic Sea beaches – application of the Sand Rake method. Front Environ Sci 8:599978. https://doi.org/10.3389/fenvs.2020.599978
15. Petov NA (2008) Assessment of polymer wastes generation in Russia and ways of their processing. Polymernyematerialy 4:4–7. (in Russian)
16. Petrov OV, Spiridonov VF (eds) (2010) Atlas of geological and environmental-geological maps of the Russian sector of the Baltic Sea. VSEGEI, St. Petersburg. 78 p. (in Russian)
17. Martyanov SD, Ryabchenko VA, Ershova AA, Eremina TR, Martin G (2019) On the assessment of microplastic distribution in the eastern part of the Gulf of Finland. Fundam Appl Hydrophys 3:32–41. https://doi.org/10.7868/S207366731904004X
18. Chubarenko IP, Esiukova EE, Bagaev AV, Bagaeva MA, Grave AN (2018) Three-dimensional distribution of anthropogenic microparticles in the body of sandy beaches. Sci Total Environ 628–629:1340–1351. https://doi.org/10.1016/j.scitotenv.2018.02.167
19. Chubarenko I, Esiukova E, Khatmullina L, Lobchuk O, Grave A, Kileso K, Haseler M (2020) From macro to micro, from patchy to uniform: analyzing plastic contamination along and across a sandy tide-less coast. Mar Pollut Bull 156:111198. https://doi.org/10.1016/j.marpolbul.2020.111198
20. Esiukova E, Khatmullina L, Lobchuk O, Grave A, Kileso A, Haseler M, Zyubin A, Chubarenko I (2020) From macro to micro: dataset on plastic contamination along and across a sandy tide-less coast (the Curonian Spit, the Baltic Sea). Data Brief 30:105635. https://doi.org/10.1016/j.dib.2020.105635
21. Pupienis D, Buynevich I, Ryabchuk D, Jarmalavicius D, Zilinskas G, Fedorovic J, Kovaleva O, Sergeev A, Cichon-Pupienis A (2017) Spatial patterns in heavy-mineral concentrations along the Curonian Spit coast, southeastern Baltic Sea. Estuar Coast Shelf Sci 195(2017):41–50. https://doi.org/10.1016/j.ecss.2016.08.008
22. Esiukova E (2017) Plastic contamination on the Baltic beaches of Kaliningrad region. Russia Mar Pollut Bull 114(2):1072–1080. https://doi.org/10.1016/j.marpolbul.2016.10.001
23. Zobkov M, Esiukova E (2017) MPs in Baltic bottom sediments: quantification procedures and first results. Mar Pollut Bull 114(2):724–732. https://doi.org/10.1016/j.marpolbul.2016.10.060
24. Chubarenko I, Stepanova N (2017) MPs in sea coastal zone: lessons learned from the Baltic amber. Environ Pollut 224:243–254. https://doi.org/10.1016/j.envpol.2017.01.085
25. Masura J, Baker J, Foster G, Courtney A (2015) Laboratory methods for the analysis of MPs in the marine environment: recommendations for quantifying synthetic particles in waters and sediments. NOAA Technical Memorandum NOS-OR&R-48. 31 p
26. Norén F (2007) Small plastic particles in coastal Swedish waters. KIMO Rep:1–11. http://www.n-research.se/pdf/Small%20plastic%20particles%20in%20Swedish%20West%20Coast%20Waters.pdf
27. Esiukova EE, Chubarenko BV, Burnashov EM (2018) Geosynthetic materials as a source of contamination of the marine environment by plastic litter. Reg Ecol 3(53):15–28. https://doi.org/10.30694/1026-5600-2018-3-15-28. (in Russian)
28. Esiukova E, Chubarenko B, Simon F-G (2018b) Litter of geosynthetic materials on the shore of South-Eastern Baltic (Kaliningrad Oblast, Russian Federation). 2018 IEEE/OES Baltic international symposium (BALTIC), pp 1–6. https://doi.org/10.1109/BALTIC.2018.8634842
29. Zobkov MB, Esiukova EE, Zyubin AY, Samusev IG (2019) Microplastic content variation in water column: the observations with novel sampling tool in stratified Baltic Sea. Mar Pollut Bull 138:193–205. https://doi.org/10.1016/j.marpolbul.2018.11.047
30. Esiukova EE, Chubarenko BV, Chubarenko IP, Kileso AV, Zhelezova EV, Grave AV, Tsukanova ES, Sobaeva DA, Tanurkov AG, Yushmanova AV, Turko NA (2018c) Method of selection and accounting of fragments of geosynthetic materials and its testing at the beaches of

the South-Eastern Baltic. In: Rumyantseva EA (ed) Arctic shores: the path to sustainability: proceedings of the conference. MAGU, Murmansk, pp 76–79 (in Russian)

31. Esiukova EE, Chubarenko BV (2019) Geothyntetic litter – new pollutant as a result of coastal protection. Abstracts of XX international environmental forum “Baltic Sea Day”, March 21–22, St. Petersburg, St. Petersburg: “SvoyeIzdatelstvo” LLC. pp 80–82. http://helcom.ru/baltic_sea_day/bsd_thesis_collection
32. UEG (2014) Contamination of the north and Baltic seas with paraffin. Independent environmental group of experts “Consequences of Contamination Incidents” (UEG) – opinion dated 22 July 2014
33. GESAMP (2007) “Estimates of oil entering the marine environment from sea–based activities”. (IMO/FAO/UNESCO–IOC/UNIDO/WMO/IAEA/UN/UNEP joint Group of Experts on the scientific aspects of marine environmental protection). Rep Stud GESAMP 75. 96 p
34. Qiu Q, Tan Z, Wang J, Peng J, Li M, Zhan Z (2016) Extraction, enumeration and identification methods for monitoring MPs in the environment. Estuar Coast Shelf Sci 176:102–109. https://doi.org/10.1016/j.ecss.2016.04.012
35. Bagaev A, Mizyuk A, Khatmullina L, Isachenko I, Chubarenko I (2017) Anthropogenic fibres in the Baltic Sea water column: field data, laboratory and numerical testing of their motion. Sci Total Environ 599–600:560–571. https://doi.org/10.1016/j.scitotenv.2017.04.185
36. Lusher AL, Welden NA, Sobral P, Cole M (2017) Sampling, isolating and identifying MPs ingested by fish and invertebrates. Anal Methods 9:1346–1360. https://doi.org/10.1039/c6ay02415g
37. Cózar A, Echevarría F, González-Gordillo JI, Irigoien X, Úbeda B, Hernández-León S, Palmae ÁT, Navarrof S, García-de-Lomasa J, Ruizg A, Fernández-de-Puellesh ML, Duarte CM (2014) Plastic litter in the open ocean. Proc Natl Acad Sci U S A 111(28):10239–10244. https://doi.org/10.1073/pnas.1314705111

Role of Environmental Science in Tackling Plastic Pollution

Friederike Stock, Georg Reifferscheid, Nicole Brennholt, and Evgeniia Kostianaia

Contents

1 Introduction 488
2 Overview of This Volume 489
3 Discussion 494
References 495

Abstract The book volume "Plastics in the Aquatic Environment – Part I: Current Status and Challenges" gives an overview about the role of environmental science and provides a sense of the global perspective in dealing with plastic pollution. The volume contains 15 chapters, with two additional chapters written by the editors containing introductory remarks and concluding notes on the role of environmental science in tackling the plastic pollution problem. These 15 chapters present and discuss challenges in research, related, for example, to microplastics analysis, impacts of plastic litter on aquatic environments, plastic waste management, bioplastics; they also review case studies of plastic pollution and contamination in the Philippines, Brazil, Albania, Slovenia, Russia and East Asia, as well as the Mediterranean Sea at large. This chapter provides an overview of the conclusions drawn by the authors of the chapters of this book volume and gives an overall final discussion of the challenges discussed herein.

Keywords Challenges, Open research questions, Plastic, Pollution

---

F. Stock (✉), G. Reifferscheid, and N. Brennholt
Department of Biochemistry, Ecotoxicology, German Federal Institute of Hydrology, Koblenz, Germany
e-mail: stock@bafg.de; reifferscheid@bafg.de; brennholt@bafg.de

E. Kostianaia
P.P. Shirshov Institute of Oceanology, Moscow, Russia
e-mail: evgeniia.kostianaia@ocean.ru

Friederike Stock, Georg Reifferscheid, Nicole Brennholt, and Evgeniia Kostianaia (eds.), *Plastics in the Aquatic Environment - Part I: Current Status and Challenges*, Hdb Env Chem (2022) 111: 487–496, DOI 10.1007/698_2021_793,

© Springer Nature Switzerland AG 2021, Published online: 23 July 2021

1 Introduction

The current book volume – “Plastics in the Aquatic Environment – Part I: Current Status and Challenges” – represents the first volume in the book project “Plastics in the Aquatic Environment”, which also has a second volume “Plastics in the Aquatic Environment – Part II: Stakeholders’ Role against Pollution”. The current book volume focuses on the chemical and biological aspects of plastic pollution, as well as on specific examples of impacts of plastic pollution and associated research in several countries and regions around the world, specifically Philippines, Brazil, Albania, Slovenia, Russia and East Asia, as well as the Mediterranean Sea at large. The second book volume, “Plastics in the Aquatic Environment – Part II: Stakeholders’ Role against Pollution”, considers such aspects of the fight against plastic pollution as environmental policy, law and finance, nature conservation, education and human behaviour.

The authors represent a wide range of research institutions and organizations, such as the German Federal Institute of Hydrology (Koblenz, Germany), Institute of Plastics and Circular Economy (Leibniz University Hannover, Germany), Anglia Ruskin University (Cambridge, UK), Hellenic Centre for Marine Research (Institute of Marine Biological Resources and Inland Waters, Greece), Italian National Institute for Environmental Protection and Research (Bologna, Italy), National Institute of Oceanography and Applied Geophysics (Trieste, Italy), P.P. Shirshov Institute of Oceanology (Moscow, Russia), National University of Science and Technology MISIS (Moscow, Russia), Department of Engineering Science, University of Oxford (UK), Lomonosov Moscow State University (Russia), Skoltech Institute of Science and Technology (Moscow, Russia), Institute of Marine Biology (Odessa, Ukraine), UNESCO, BKV GmbH (Germany), University of Alicante (Spain), University of Ljubljana (Slovenia), Agricultural University of Tirana (Albania), Aleksandër Moisiu University of Durrës (Albania), National Sun Yat-sen University (Taiwan), Toyama Prefectural University (Japan), IndigoWaters Institute (Taiwan), Medipeace (Republic of Korea), University of the Philippines (Quezon city, Philippines), Marine and Environmental Sciences Centre (ISPA – Instituto Universitário, Lisbon, Portugal), Federal University of Paraná (Brazil), Federal University of Pernambuco (Brazil) and Russian State Hydrometeorological University (St. Petersburg, Russia).

Such a diverse spectrum of expertise has allowed, as we as editors hope, for an interesting and engaging discussion of some of the principal aspects of plastic research, such as microplastics analysis, impacts of plastic litter on aquatic environments, plastic waste management, bioplastics, as well as presentation of some specific research in certain countries. Seven chapters discuss plastic pollution research and approaches in such aquatic environments as the Mediterranean Sea at large, with some specific examples of Durrës Bay, Rodoni Bay, the Gulf of Drin and Shëngjini Bay in Albania and research in Slovenia, water bodies of Metro Manila in the Philippines, Brazilian freshwater and estuarine environments, the Russian part of the Baltic Sea, namely the Gulf of Finland and the South-East Baltic, and the aquatic environments of China, Japan, South Korea and Taiwan.

2 Overview of This Volume

The chapter by Stock et al. [1] presents a detailed overview about pitfalls, limitations, advantages and disadvantages in microplastic analyses (sampling, sample preparation and analysis). The authors underline that harmonization and standardization of sampling and analytical methods are still missing and that comparability of data is not yet given. Microplastics are heterogeneously distributed and replicates and repeated measurements are absolutely needed. A harmonized protocol should be implemented so that a better comparability of data is given and data can be used by other researchers. Moreover, critical parameters and limitations should always be reported. The generated data should also be validated and be usable for modelling studies as case studies only cover a small geographical area. In general, the authors point out that microplastics in environmental samples are very challenging as parameters, such as different sizes, shapes, colours, ages of polymers or biofilms, may influence the result of the analysis and the detection of microplastics. Therefore, the authors do not recommend the use of a specific method but to consider the main research question and to use a combination of analytical approaches.

Dierkes et al. [2] summarize analytical methods for analyzing microplastics in environmental samples. Although a diversity of methods exists, harmonization is not yet present. In order to implement measures for reducing microplastic emissions, Dierkes et al. emphasize the need for a reliable, fast and cheap identification method. The authors describe the advantages and differences between the methods. They do not suggest to use a specific method as they generate different information (number and size vs. mass). The size of the analyzed plastics should be considered as for example smaller particles (esp. <10 μm) present a large effort. Another important fact which should be taken into account is sample pretreatment which can be quite time-consuming (density separation, enzymatic or chemical digestion). The authors also point out the need to establish standardized protocols and harmonized quality standards.

In order to maintain the positive features of plastics while overcoming the negative ones, great hope is placed on the development of bioplastics. However, as Endres describes in his chapter “Biodegradable Plastics – End of Life Scenarios” [3], it is important to make a distinction between the biopolymer in its form as a macromolecule and the ready-to-use material. Furthermore, bio-based and biodegradable plastics should be differentiated. Bio-based plastics concern the raw material origin of the polymer feedstock, whereas biodegradability refers to an end of life option. Both features are independent of each other. Although biodegradability defines a material property which depends on the microstructure as well as the chemical structure of the material, in reality, biodegradability is a system feature, because there are many environmental conditions, ranging from industrial compost and sewage treatment plants to soils in a wide range of climatic regions, the beach, seabed and even the human body. Thus it is essential to offer exact data on environmental conditions, as well as points in time when a product or material is considered biodegradable. As regards compostability, test standards in some areas, such as bioplastics and other organic substances, cover well various environmental conditions. On the contrary, test standards in some other areas, for example, degradability in marine systems or soil, are few and are not able to present well complex environmental conditions. Aside from the formation of appropriate standards, future material development requires an advanced knowledge of the relationships between environmental conditions of habitats, microbiology and material parameters, on the one hand, and, on the other hand, ensuing degradation mechanisms and kinetics.

Green [4] points out that only few studies used environmental relevant concentrations of (micro)plastics so that biological and ecological consequences are difficult to decipher. The few studies using prevailing concentrations in the environment revealed mixed results. Therefore, it is necessary to conduct more research by simulating realistic concentrations of (micro)plastics, using mesocosm studies in the natural environment and to conduct experiments on a longer term so that effects can be better understood. Nevertheless, the present studies and their results can be transferred to future impacts as a 50-fold increase of microplastics is well probable in the environment from 2010 to 2100. The authors also show that some effects are already present at current environmental levels. When conducting experiments it is also very important to take into account parameters such as size, shape, chemical composition, abundance of the plastic debris, the type of organism or habitat being polluted, and the existence of other environmental stressors that may potentially affect any impacts [4]. In addition, it should also be noted that biodegradable plastics in natural environments may not decompose very fast and that they may have the same effects as non-biodegradable plastics. Thus, waste management of biodegradable plastics is also of high importance so that these plastics do not enter the environment.

The impact of plastic pollution on marine life (as demonstrated by the Mediterranean Sea) is a widespread phenomenon. Anastasopoulou and Fortibuoni [5] point out that the more research is conducted, the more impacted marine species are described. Different direct and indirect effects such as ingestion (predominantly studied), entanglement or substrates acting as a dispersal for organisms or pathogens prevail. Macroplastics have also been ingested by different species such as fish, birds, turtles and cetaceans. Not much is known about how additives and other contaminants of microplastics affect organisms. With regard to ingestion, the risk is perceived as lower for microplastics than for large plastic parts, as this has not been yet shown by researchers. In contrast, plastic waste leads to ingestion and entanglement and can provoke death and suffering of marine life, e.g. for seabirds, turtles and cetaceans. The published studies always refer to individuals and not to populations as these studies are hampered by different stressors (e.g. environmental and human-induced). Therefore, the role of microplastics may be veiled [5].

Sapozhnikov et al. [6] discuss in detail interactions of plastics with microorganisms and present published work as well as the results of their research of the last years. The main outcome of micro-fouling of different polymers in aquatic environments is that colonization by certain microorganisms occurs (especially diatoms). Microphytes from benthic communities, for instance, may settle on plastics and decompose it. Thereby, different benthic or periphytic species can be present at the same time on different polymers in one habitat. Thus, the presence of colonial microphyte settlements determines the mechanisms of plastic destruction. Research has also shown that certain species of bacteria use plastics as a carbon source. However, this work was done under laboratory conditions. Thus, more research is necessary to study these bacteria under natural conditions. Furthermore, there is some evidence that plastic has toxic effects on the growth and functioning of microorganisms themselves. Moreover, the authors state that it is well probable that a link exists between toxin production of potentially toxic microphytes and their presence on polymers. In addition, microorganisms use polymers as a transport path and thus spread around the world. It is possible that this transport leads to biological invasions. The authors also state that there are still knowledge gaps concerning biodegradation and the interaction with microorganisms.

Zandaryaa [7] from the UNESCO Division of Water Sciences gives an overview of microplastics in freshwater, its sources and pathways and their occurrence. The author summarizes the published studies about microplastics in freshwater environments of the last years and describes the relevance of microplastics with regard to an improved water quality and the UN 2030 Agenda Sustainable Development Goals. Microplastics are found all over the world in different environments. Despite the multitude of publications about microplastics in general, freshwater environments have only been studied for several years. Therefore, knowledge gaps occur. Data for monitoring microplastics in different environments are missing. Ecotoxicologial research has to be intensified in order to estimate risks, accumulation and exposure on organisms and risks of microplastics exposure to humans in drinking water are not known. The author also mentions the need to share knowledge and build research capacities with developing countries. Moreover, harmonization of methods and definitions are needed for better comparing data. Solutions for decreasing the pollution include microplastic reduction at the source along with sustainable consumption and production, replacing and banning plastic products, improving waste management and reducing and recycling plastic waste. This should not only be done with technological advances but also with policy approaches.

Cieplik presents the model “From Land to Sea – Model for the documentation of land-sourced plastic litter” and shows pathways and discharge sources into the North Sea, Baltic Sea and Black Sea [8]. The model aims at estimating origin, quantity and nature of improperly disposed plastic litter (micro- and macroplastics) originating from Germany. In the first step, identification of main discharge pathways and discharge sources took place. Then, a database was set up based on an established data model. In the second step, primary and secondary data were the basis for the calculation of discharge volumes. The results reveal that most plastics transported into the sea are macroplastics. However, the amounts of macroplastic litter differ. The Baltic Sea has the highest discharge as the river basins are characterized by a long coastline. The model also showed that about 80% discharge enters the North Sea, Baltic Sea and Black Sea via the pathways “rivers” and “coastal regions”. For the future, it is possible to include other regions and countries as well as other discharge pathways into the model.

In their chapter Horodytska et al. [9] have a critical look at plastic waste management and describe the current status and its weaknesses. Hereby, they differentiate between developed and developing countries. They show that sustainable waste management along with different collection, sorting and waste treatment systems are more predominant in developed countries. However, it is not clear in which system environmental benefits prevail. Possible recyclable materials may be collected separately. In general, mechanical recycling, chemical recycling, energy recovery and landfilling are waste treatment methods. Some European countries already prohibited landfilling as it is considered as the worst method. In developing countries, municipal collection strategies are lacking or are not efficient and therefore contribute to waste accumulation and environmental pollution. However, in developing countries an informal recycling sector has been established. Valuable and recyclable materials from waste on the streets, houses and landfills are picked up and sold by waste pickers, whose work conditions are horrendous. A circular economy, however, has not even been reached in developed countries although waste management is present and rising recycling rates occur. The study reveals that contamination and degradation reduce the value of possible recyclable material so that these products are downcycled. Therefore, it would be important to significantly improve the quality of recyclates.

The case study about Slovenia by Kalčikova and Gotvajn shows that the country has successfully implemented an environmentally sustainable waste management structure [10]. This has been done by awareness rising, social aspects, fines, education and work of NGOs as well as lowering the amount of waste and at the same time increasing the recycling rate. Despite the many efforts conducted, microplastic pollution is still present along the coasts. Therefore, the authors suggest improving the solid waste management and the wastewater treatment as this seems to be the main source of plastic pollution into the environment.

In their report on marine litter assessment on some beaches along the South-Eastern Adriatic coastline of Albania [11], Kolitari and Gjyli give detailed information about the amount and composition of litter. They found on average 0.219 items/m^2 (219 items/100 m; 152.3–313.3 items/100 m) beach litter. The report shows that shoreline sources (e.g. tourism and recreational activities), in addition to the poor waste management practices, are the main sources of beach litter deposited on surveyed beaches. Therefore, the authors recommend the following options based on the results of their research: actions to tackle cartons/Tetra Pak items, measures to deal with plastics, including single-use plastic items, more investments to set up landfills, and rehabilitation of the polluted Ishmi River by the means of water purification and dredging soil. Moreover, the authors also suggest mitigation measures such as:

(a) carrying out awareness raising campaigns emphasizing the idea of “Leave No Trace” and promoting this concept to locals, tourists and other beach users;
(b) increasing specific clean-up activities, especially in summer during the high touristic season;
(c) intensifying direct intervention by the means of patrols and signs;

(d) securing legislative actions which prohibit litter dumping in rivers, as the Ishmi is significantly polluted by Tirana County;
(e) securing legislative actions for bans of certain items as foreseen also under the EU Single-Use Plastics Directive that includes bans on single-use plastic cutlery, plastic plates, plastic straws, cotton bud sticks made of plastic and plastic balloon sticks, along with oxodegradable plastics, food containers and expanded polystyrene cups;
(f) promoting wider awareness among the youth and students on the consequences of the presence of marine litter in the oceans [11].

Walther et al. provide a thorough overview of plastic pollution in China, Japan, South Korea and Taiwan in East Asia [12]. Although plastic pollution is a common problem and plastic waste ends up in the East China Sea and Yellow Sea, the countries react differently from an economic and political point of view. The authors state that research, interregional cooperation and ENGOs have to be intensified and hope that this chapter could “inspire a more concerted effort” [12] of policies and management solutions by the different governments.

The case study by Tanchuling and Osorio [13] about microplastics in Metro Manila rivers shows that most plastics present in the rivers derive from fragmentation of larger particles (secondary microplastics) which mostly originate from leakages of solid waste from landfills into the environment. In order to prevent plastic pollution, the authors suggest a better implementation of The Ecological Solid Waste Management Act RA9003, especially by helping local governments technically and logistically, and capacity-building coupled with good governance. The Manila Bay Coordinating Office (MBCO) was formed in order to rehabilitate Manila Bay and monitor pollution sources. Moreover, informal settlements close to the rivers where no collection of solid waste exit are planned to be relocated. However, a higher collection and recycling rate still needs to be achieved.

Lima et al. [14] present the situation in Brazilian freshwater and estuarine systems. The results clearly reveal a knowledge gap about composition and distribution of plastics in freshwater systems. Although several studies have been published about estuarine systems with data about the current situation (occurrence, size, number of plastic particles), transport processes, pathways and environmental factors were not studied in detail. Moreover, information is missing about the source-to-sink relationship. The authors suggest to use this key approach of source to sea in order to better understand the plastic pollution along the Brazilian coasts.

The research on beaches in the Baltic Sea shows the necessity of a detailed monitoring on beaches [15]. Although monitoring activities have been conducted since several years all around the world, there are still many areas underrepresented and not studied in detail. Ershova et al. [15] used monitoring methods by OSPAR, NOAA and the IOW beach litter method and combined them as the characteristics of beaches are not always the same and different parts of the beaches are investigated. Moreover, the new generated information will be integrated in a database so that this information is also available for a larger public. Furthermore, recommendations will be made for the national program of marine litter monitoring for the coasts of the

South-East Baltic Sea. The results will also be used on an international level as they will be harmonized with the international monitoring programs in the Baltic region.

3 Discussion

The 15 chapters in this book volume “Plastics in the Aquatic Environment – Part I: Current Status and Challenges” have raised a wide range of important questions and underlined some essential aspects of plastic pollution research. Despite the fact that studies on plastics have been ongoing for quite some time, there still remain many research gaps and open research questions. Some of these are methodological gaps, which reflect the need for harmonization and standardization of analytical methods, protocols and quality standards. For many reasons, it is not possible to use a single method, thus it is recommended to apply a combination of analytical approaches. Another open research question is the necessity to conduct research using realistic concentrations of microplastics and carry out long-term experiments which would provide a better overview of the effects of microplastics.

Development of bioplastics is thought to be very promising. However, still, advanced knowledge is needed to allow for a better understanding of the interaction between environmental conditions of a medium, material characteristics, microbiology and degradation mechanisms and kinetics. A related research niche here is to look into waste management of biodegradable plastics and the period of decomposition in natural environments. Waste management in general still requires a lot of attention and improvement for many regions, because the quality of solid waste management and the wastewater treatment largely impacts plastic pollution in the environment.

More research is also needed to understand the potential impacts of additives and contaminants in microplastics. Moreover, ecotoxicological studies need to be intensified, including a very important aspect of the risks of the presence of microplastics in drinking water. Identification of sources of pollution and establishment of the source-to-sink relationship are essential in order to develop efficient management measures.

The presented chapters have shown that besides research projects in each country, it is also crucial to establish cross-border cooperation for tackling plastic pollution. As this problem does not have borders, and the level of plastic pollution in one country may affect the water quality and plastic contamination in a water body in another country, it is of paramount importance to join forces and help each other where and when needed. Such cooperation should also include knowledge exchange and capacity building.

Most probably, as science advances, we will face new research questions and puzzles and there will be a need for more advanced research. Hopefully, with the development of new technologies and materials, scientists will be able to answer those questions and provide well-supported recommendations for decision- and policymakers. We are fully aware that the aspects of environmental science presented in this book volume are only a tiny part of the myriad of questions that the environmental science is confronted with dealing with plastic pollution. It would have been an impossible task to gather all of them in one book. At the same time, our aim was to highlight some interesting developments and show potentials and achievements, as well as limitations and constraints of the current microplastic analysis, waste management, bioplastics research, capacities of new models and approaches and promising niches for further investigation. We hope that this book volume will be useful for both scientists and policymakers and that the chapters with case studies will provide valuable information and inspiration for other regions. We are positive that this book volume together with its second part, "Plastics in the Aquatic Environment – Part II: Stakeholders' Role against Pollution", has greatly assisted in accumulation and distribution of knowledge and expertise on plastic pollution and we would like to wholeheartedly thank all the authors for their contributions, time and commitment.

Acknowledgements Evgeniia Kostianaia was partially supported in the framework of the Russian Science Foundation Project N 21-17-00191 (2021–2023) "Monitoring of water exchange through the Kerch Strait based on state-of-the-art methods of observations and numerical modeling".

References

1. Stock F et al (2021) Pitfalls and limitations in microplastic analyses. In: Stock F et al (eds) Plastics in the aquatic environment – part I: current status and challenges. Springer, Berlin. https://doi.org/10.1007/698_2020_654
2. Dierkes G, Lauschke T, Földi C (2021) Analytical methods for plastic (microplastic) determination in environmental samples. In: Stock F et al (eds) Plastics in the aquatic environment – part I: current status and challenges. Springer, Berlin. https://doi.org/10.1007/698_2021_744
3. Endres H-J (2021) Biodegradable plastics – end of life scenarios. In: Stock F et al (eds) Plastics in the aquatic environment – part I: current status and challenges. Springer, Berlin. https://doi.org/10.1007/698_2021_745
4. Green DS (2021) Biological and ecological impacts of plastic debris in aquatic ecosystems. In: Stock F et al (eds) Plastics in the aquatic environment – part I: current status and challenges. Springer, Berlin. https://doi.org/10.1007/698_2020_509
5. Anastasopoulou A, Fortibuoni T (2021) Impact of plastic pollution on marine life in the Mediterranean Sea. In: Stock F et al (eds) Plastics in the aquatic environment – part I: current status and challenges. Springer, Berlin. https://doi.org/10.1007/698_2019_421
6. Sapozhnikov P et al (2021) Plastic in the aquatic environment: interactions with microorganisms. In: Stock F et al (eds) Plastics in the aquatic environment – part I: current status and challenges. Springer, Berlin. https://doi.org/10.1007/698_2021_747
7. Zandaryaa S (2021) Freshwater microplastic pollution: the state of knowledge and research. In: Stock F et al (eds) Plastics in the aquatic environment – part I: current status and challenges. Springer, Berlin. https://doi.org/10.1007/698_2021_748
8. Cieplik S (2021) From land to sea – model for the documentation of land-sourced plastic litter. In: Stock F et al (eds) Plastics in the aquatic environment – part I: current status and challenges. Springer, Berlin

9. Horodytska O, Cabanes A, Fullana A (2021) Plastic waste management: current status and weaknesses. In: Stock F et al (eds) Plastics in the aquatic environment – part I: current status and challenges. Springer, Berlin. https://doi.org/10.1007/698_2019_408
10. Kalčíková G, Žgajnar Gotvajn A (2021) Plastic pollution in Slovenia: from plastic waste management to research on microplastics. In: Stock F et al (eds) Plastics in the aquatic environment – part I: current status and challenges. Springer, Berlin. https://doi.org/10.1007/698_2019_402
11. Kolitari J, Gjyli L (2021) Marine litter assessment on some beaches along the southeastern Adriatic coastline (Albania). In: Stock F et al (eds) Plastics in the aquatic environment – part I: current status and challenges. Springer, Berlin. https://doi.org/10.1007/698_2020_627
12. Walther BA et al (2021) Plastic pollution in East Asia: macroplastics and microplastics in the aquatic environment and mitigation efforts by various actors. In: Stock F et al (eds) Plastics in the aquatic environment – part I: current status and challenges. Springer, Berlin. https://doi.org/10.1007/698_2020_508
13. Tanchuling MAN, Osorio ED (2021) The microplastics in metro Manila Rivers: characteristics, sources, and abatement. In: Stock F et al (eds) Plastics in the aquatic environment – part I: current status and challenges. Springer, Berlin. https://doi.org/10.1007/698_2020_659
14. Lima ARA et al (2021) Plastic contamination in Brazilian freshwater and coastal environments: a Source-To-Sea transboundary approach. In: Stock F et al (eds) Plastics in the aquatic environment – part I: current status and challenges. Springer, Berlin. https://doi.org/10.1007/698_2020_514
15. Ershova AA et al (2021) Marine litter in the Russian gulf of Finland and south-East Baltic: application of different methods of beach sand sampling. In: Stock F et al (eds) Plastics in the aquatic environment – part I: current status and challenges. Springer, Berlin. https://doi.org/10.1007/698_2021_746