

Occurrence, Fate and Fluxes of Plastics and Microplastics in Terrestrial and Freshwater Ecosystems



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Abbreviations

ATR	Attenuated total reflectance
ECHA	European Chemicals Agency
EEA	European Environment Agency
ERA	Ecological risk assessment
EVA	Ethylene-vinyl acetate
FTIR	Fourier transform infrared
MaP	Macroplastic
MP	Microplastic
NP	Nanoplastic
PA	Polyamide
PAH	Polycyclic aromatic hydrocarbons
PC	Polycarbonate
PCB	Polychlorinated biphenyl
PE	Polyethylene
PEC	Predicted environmental concentration
PES	Polyester
PET	Polyethylene terephthalate
PMMA	Polymethyl methacrylate
PNEC	Predicted no effect concentration
PP	Polypropylene
PS	Polystyrene
PUR	Polyurethane
PVC	Polyvinylchloride
Pyr-GC/MS	Pyrolysis-gas chromatography/mass spectrometry
SAPEA	Science Advice for Policy by European Academies
SEM	Scanning electron microscopy
WWTP	Wastewater treatment plant
XRF	X-ray fluorescence

1 Introduction

Over the past century, plastic has made the journey from being virtually non-existent to a ubiquitous and integral part of modern life. While plastic has numerous advantages compared to alternative materials, we are facing severe environmental, economic and ethical issues due to the vast plastic waste production and rapid disposal. Up until 2015, the total amount of plastic produced was 8300 million tons, 6300 million tons of which were discarded as waste (Geyer et al. 2017). Much of this waste (79%) is accumulated in landfills or the natural environment, and this amount is expected to increase significantly in the future (up to 12,000 million tons by 2050) if management actions are not immediately taken (Geyer et al. 2017).

Most macroplastics (MaPs) break down due to mechanical and chemical fragmentation into smaller pieces, which are commonly termed microplastics (particles <5 mm; MPs) or nanoplastics (particles <1 μm ; NPs) (Gigault et al. 2018). The breakdown process may take between 50 and 600 years and usually depends on several factors such as the polymer composition and the environmental condition. MPs that are formed due to the breakdown of MaP are commonly referred to as secondary MPs, while MPs intentionally produced in this size range are referred to as primary MPs. Nowadays, MaPs, MPs and NPs can be found floating or in suspension in many water bodies, accumulated in sediments or in terrestrial ecosystems, and even can be transported and deposited in pristine environments due to wind and currents (Dris et al. 2015, 2016; Ballent et al. 2016; Fischer et al. 2016; Hurley and Nizzetto 2018).

The widespread distribution of plastic and its variability in size and shape allow the ingestion by organisms across many trophic levels and habitats (Wright et al. 2013; Kühn et al. 2015). Large plastic debris (MaPs) can cause adverse effects on coastal and marine animals (marine mammals, fish and seabirds) due to ingestion as well as to entanglement which impedes their mobility (Van Franeker et al. 2011; Knowlton et al. 2012; Schuyler et al. 2012; Kühn et al. 2015). Fishing gear, balloons, plastic bags and bottle caps have been identified to be the most harmful type of MaPs to marine organisms (Hardesty et al. 2015). Although most research has focused on the marine environment, freshwater and terrestrial organisms are expected to suffer from the same sort of effects. For example, cattle have been reported to suffocate and die due to the ingestion of plastic bags, which can block airways and stomachs (Ramaswamy and Sharma 2011).

Similar to MaPs, environmental exposure to MPs has raised concerns about their potentially adverse effects in smaller organisms. Ecotoxicological studies with MPs have been primarily conducted using marine organisms (77%), while freshwater organisms have been less researched (23%) (de Sá et al. 2018), and research involving terrestrial organisms is still in its beginnings (Chae and An 2018). MPs may cause physical effects such as internal and external abrasion or blockages of the digestive tract in small invertebrates and fish (Wright et al. 2013; Karami et al. 2016; Jovanović 2017). Research also shows that MPs ingested by freshwater organisms, either on purpose when they are confounded with preys or accidentally due to non-selective feeding strategies (i.e. water filtration or deposit feeding) (de Sá et al. 2018; Windsor et al. 2019), may reduce their feeding efficiency and lower the energy uptake, which often results in reduced growth, reproduction and survival (Foley et al. 2018). In addition, MPs may affect the growth, chlorophyll content, photosynthesis activity and reactive oxygen species of microalgae at high, currently not realistic, concentrations (Prata et al. 2019).

Although several cases evidence deleterious impacts of MaPs on aquatic and terrestrial organisms under laboratory conditions, the capacity of MPs or even NPs to pose a real threat for ecosystems and human health is disputable. This is because the majority of studies showing some impacts of MPs on terrestrial or freshwater organisms have been performed with very high exposure concentrations, while risk at environmentally relevant concentrations has yet to be disclosed (Lenz et al. 2016).

Despite physical effects, the release of additives such as phthalates, chlorinated paraffins and bisphenols present in some MaPs and MPs (Stenmarck et al. 2017) has been reported to induce endocrine-disrupting effects (Rochman et al. 2014). Furthermore, hydrophobic pollutants (e.g. some pesticides, PCBs, PAHs) can be adsorbed to plastics and may be released into the body of the organisms after ingestion, leading to the so-called Trojan Horse effect (Teuten et al. 2009; Koelmans et al. 2016; Crawford and Quinn 2017; Bouhroum et al. 2019). In contrast, it has been suggested that ingested clean MPs may reduce the concentration of bioaccumulated chemicals in the body of contaminated organisms (Lohmann 2017). Furthermore, MPs could not only act as carriers for chemicals but can also transport bacteria or pathogens attached to them (Keswani et al. 2016; Kirstein et al. 2016) across different environmental compartments and regions. In order to provide some responses to the concern produced by the perception of the potential risks for human health and the environment of MPs and of chemicals associated to them, Koelmans et al. (2017a) proposed adverse outcome pathways for assessing and comparing the risk of MaPs, MPs and NPs and highlighted the uncertainties that still exist in both, exposure and effect assessment.

The continuous emission patterns and the breakdown of plastic litter into smaller fractions in the environment may contribute to future concentrations that are orders of magnitude higher than the ones currently monitored (Everaert et al. 2018), thus contributing to a yet uncertain risk scenario. Policies dedicated to control emissions and manage risks of MaPs, MPs and NPs in the environment require a proper understanding of the main emission routes, the current exposure levels and the fluxes among environmental compartments. The available literature describing the exposure and impacts of plastics in the environment, providing a comparative assessment of the global occurrence, transport and fate, has so far mainly focused on the marine environment (see, e.g. GESAMP 2015; Auta et al. 2017). Although the freshwater environment is considered in some recent reviews (see, e.g. SAPEA 2018; Eerkes-Medrano et al. 2015), most studies consider specific emission routes and local monitoring campaigns.

Therefore, this study aimed to assess the state of the knowledge regarding the overall sources of plastic and its occurrence, fate, fluxes and loads into and within different environmental compartments of terrestrial and freshwater ecosystems. This study identifies data gaps that need to be addressed in order to understand the life cycle of the different plastic types in the environment, particularly in the soil-water interface, and provides relevant information to support research into the accumulation and ecotoxicological characterization of plastics to living organisms. Ultimately, this study provides guidance information to derive effective management measures aimed at reducing plastic discharges into the environment and attaining a more sustainable use and consumption of plastics in the nearby future.

2 Environmental Sources of Plastics

Nowadays, Asia is producing 50% of the world's plastic, followed by Europe and North America, producing 19% and 18%, respectively (PlasticsEurope 2018). The majority of plastics can be classified into the two main categories: thermoplastics (pellets that are remelted to manufacture the final product) and thermoset plastics (thermally produced into the commercial shape). Thermoplastics constitute 80% of the total plastic and are the main source of primary MPs. Thermoplastics are mainly formed by polyethylene (PE), polypropylene (PP) or polyvinylchloride (PVC), while thermoset plastics are formed, among others, by polyester (PES), polyurethane (PUR), silicone and polyamide (PA).

Sources of plastics can be classified in terms of the life expectancy of the produced plastics before disposal. Here we classify plastic sources into those with a short-term (single-use or very limited number of times with a useful lifespan up to 1 year), mid-term (up to 10 years), or long-term (more than 10 years) use expectancy.

2.1 *Plastics with Short-Term Use Expectancy*

Single-use items are mainly formed by packaging material, which is the biggest plastic sector worldwide (almost 36% in 2015; Fig. 1) and accounts for almost 50% of the generated plastic waste (Geyer et al. 2017). The vast majority of packaging plastics are PE, PP and polyethylene terephthalate (PET) (Geyer et al. 2017). Except for refillable PET bottles used in some countries, packaging is single-use with a lifespan of less than 6 months. Most foods are wrapped in plastic, and single-use plastic bags have been widely used all over the world due to their convenience, availability and low price. Plastic bags are known to cause severe environmental problems, especially in countries without proper waste management (Adane and Muleta 2009). Thus, many countries have put bans or levies in force to reduce their use or to encourage voluntary reductions (Xanthos and Walker 2017). Many African countries, for instance, have banned single-use plastic bags, while the EU Directive 2015/720 encourages member states to reduce the number of 'lightweight' carrier bags by 2025. Those bans and restrictions have already reduced the plastic bag use drastically in some countries (e.g. Ireland, England, Italy). Moreover, other single-use items like cutlery, plates, cups and straws are planned to be banned in Europe by 2021 (EC 2019).

Another important sector using single-use plastic is agriculture. Plastic films are used for plastic mulching, for the construction of greenhouses and tunnels, or to wrap silage to store animal fodder. The global plastic consumption in agricultural production is estimated to be about 2.5 million tons per year (Hussain and Hanid 2003). A variety of different plastic types are used in agriculture, including PE, PP, ethylene-vinyl acetate copolymer (EVA), PVC and polymethyl methacrylate (PMMA) (Scarascia-Mugnozza et al. 2012).

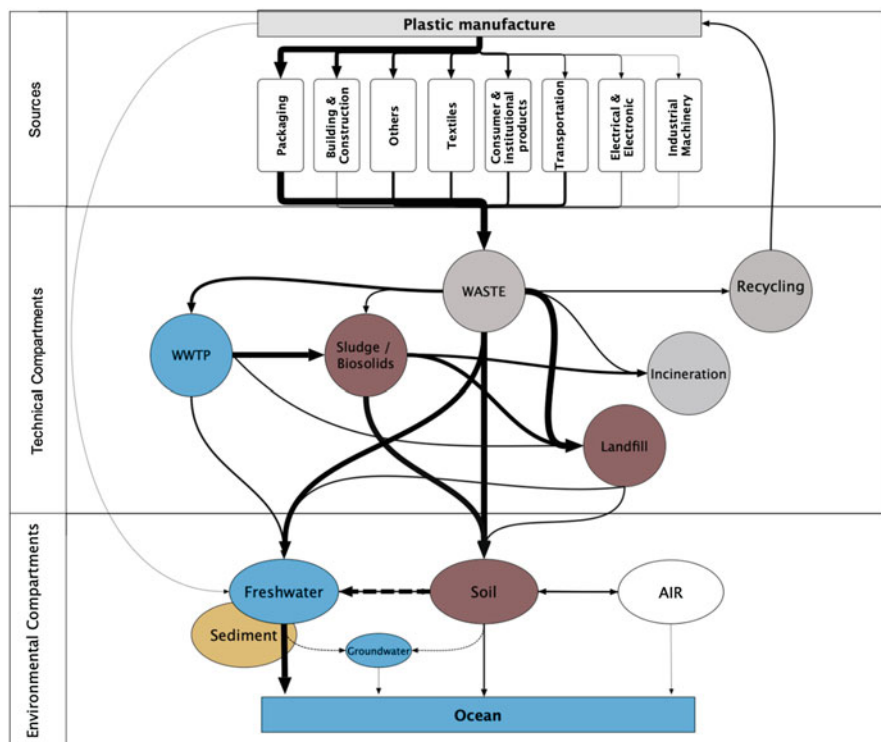


Fig. 1 Production and pathways of plastics into the different environmental compartments. Thickness of the different arrows is related to the quantitative relevance of the different mass flows. The relevance of the different plastic sources mass flows is based on Geyer et al. (2017), while the relevance of the technical compartments environmental flows is based on the reviewed literature or assumptions. Dashed lines indicate yet completely unexplored pathways with unknown relevance

MPs added to consumer products (e.g. as a component of personal care, cosmetic and cleaning products) are specially manufactured to be used once and then washed down the drain. They are often referred to as microbeads, even though they are mostly irregular in shape in order to obtain an abrasive effect (Fendall and Sewell 2009; Napper et al. 2015; Kalčíková et al. 2017). The majority of microbeads in facial and body scrubs are made of PE, with average concentrations of 4.82 g/100 mL body scrub and 0.74 g/100 mL facial scrub (Kalčíková et al. 2017; Gouin et al. 2015). Other plastic polymers used in cosmetic products include polylactic acid, PET, polyethylene isoterephthalate, nylon-12, nylon-6, PMMA, polytetrafluoroethylene and PUR (Leslie 2014; Rochman et al. 2015). Additionally, microbeads are used in industry as abrasives/scrubbers and sand-blasting media as well as in antislip, anti-blocking applications and for medical applications. It has been calculated that more than 4000 tons of PE microbeads were used in cosmetic products all over the EU (including Norway and Switzerland) in 2012 (Gouin et al.

2015), and the USA is emitting 263 tons of PE microbeads per year (2.4 mg per person per day; Gouin et al. 2011). A ban of microplastics intentionally added to products (i.e. microbeads) has been proposed in the EU (EC 2019), while the US Microbead-Free Waters Act of 2015 (US Congress 2015) prohibits the manufacturing, packaging and distribution of rinse-off cosmetics containing plastic microbeads already. This only applies to rinse-off products, while MPs are still permitted as a component in ‘leave-on’ products (e.g. lotions, sunscreens, make-ups and deodorants).

2.2 Plastics with Mid-Term Use Expectancy

Plastics with a mid-term lifespan are mainly found in the sectors of electronics, household, tyres and textiles. The production of electrical and electronic products counts to the fastest growing manufacturing and waste generation sectors (Geyer et al. 2017; Kumar et al. 2017), and as many textiles are made, entirely or to a certain extent, of synthetic plastic fibres (e.g. PA, PES, acrylic), also production rates of synthetic plastic fibres have increased over the last decade. Nowadays, two-thirds of the total fibre production is synthetic plastic fibres, and worldwide 59 tons of plastic textiles were produced in 2015 (Geyer et al. 2017; Gasperi et al. 2018).

Synthetic polymers with rubber-like characteristics are the principal component of vehicle tyres. They are composed of a mixture of natural and synthetic rubbers (styrene-butadiene rubber). While driving, tyre and road wear particles are formed which contain styrene-butadiene rubber in a mix with natural rubber, pavement parts and many other additives (Unice et al. 2013; Sundt et al. 2014). While tyres contain almost 50% of polymers, tyre wear particles, which are a mix of pavement part and polymers, contain only 16–23% of polymers (Kreider et al. 2010).

2.3 Plastics with Long-Term Use Expectancy

Plastics designed for long-term use belong to the following categories: parts of transportation (i.e. vehicle, plane and train parts), building and construction, industrial machinery, consumer products and institutional products. While plastics for the building and construction sector account for the second highest plastic consumption, only a small portion enters the waste stream directly (Fig. 1; Geyer et al. 2017). As these categories do not belong to the items that are usually littered, they are not expected to contribute significantly to the plastic load in the environment. However, their breakdown rate into MPs and NPs (due to exposure to light and weathering), also during their useful lifetime, is not clear.

3 Pathways of Plastic to the Environment

Hereafter plastic waste will refer to all plastic material that is discarded, while litter will include only those items that are not properly discarded. Packaging material is accounting for almost 50% of the generated plastic waste, followed by textiles (almost 14%; Geyer et al. 2017). Most plastic waste is generated in Asia, while America, Japan and the European Union are the world's largest producers of plastic packaging waste per capita.

3.1 *Collected Solid Waste*

Collected plastic waste is either landfilled, incinerated or recycled. In Europe, 27.3% is landfilled, 31.1% is recycled, and 41.6% is incinerated for energy recovery (PlasticsEurope 2018). The percentage of collected plastic waste varies strongly between different countries, depending on the applied waste management plans and policies. While, worldwide, the plastic recycling rate is still low, it has increased by almost 79% within the last 10 years in the EU, including Norway and Switzerland (PlasticsEurope 2018).

Large-scale industrial plastic production began in the 1950s, but plastic recycling was not established until the 1980s. It is estimated that only 9% of the total produced plastic waste up to 2015 has been recycled (Geyer et al. 2017). From this again only a small portion is submitted to primary recycling in which the recycled plastic is used to replace all or a least a proportion of the virgin polymer resins (Hopewell et al. 2009). While high-income countries have sorting and processing facilities, in low-income countries, plastic recycling is not well established. Moreover, certain types of plastic are difficult to recycle. For example, thermoset plastics, including textiles, are usually not recycled.

Plastic that is not recycled but still collected is landfilled or incinerated. In eight EU countries, Norway and Switzerland, a landfill ban for plastic is in force, leading to a very small percentage of plastic being used for landfill applications (PlasticsEurope 2018). On average, 27.3% of the generated plastic waste is landfilled in Europe. In contrast, in low-income countries, waste is mainly stored in open, poorly managed dumps, from where plastic can be transported by wind force. In middle-income countries, some controlled landfills are in place, but open dumping is still common practice. The advantages of combustion of plastic waste are that it can be used for energy recovery and the incinerated plastic cannot enter the environment anymore. At the same time, incineration results in the generation of air pollutants (Verma et al. 2016).

3.2 Wastewater

Both MPs, as well as MaP, enter wastewater either directly if products containing plastic are flushed down the drain (e.g. fibres detached during laundry of textiles, microbeads in consumer products, cotton buds or sanitary products) or in combined sewer systems from street dust and litter. MaPs escape wastewater treatment only on rare occasions and mainly enter the environment with untreated wastewater due to combined sewer overflows, e.g. after heavy rainfall events or snowmelts (Williams and Simmons 1999), or if untreated wastewater enters the environment because wastewater treatment plants (WWTPs) are not in place. Although high-income countries treat on average 70% of the wastewater, yet globally only 20% of the generated wastewater is treated (Sato et al. 2013). For MPs, the situation is different; due to their small size, they can escape the treatment and are also released with treated effluents (Ziajahromi et al. 2016). This pathway for MPs has been increasingly investigated. To date, 24 studies have measured MPs in wastewater (Table S1), from which three have not exclusively assessed MPs but included other litter items in the micro range (microlitter; HELCOM 2014; Michielssen et al. 2016; Talvitie et al. 2017b). Such studies were mainly carried out in northern and western Europe (14 studies), followed by North America (5 studies).

The number of MPs in raw wastewater varies greatly between WWTPs, from a few MPs/L to exceptional maximum values of more than 10,000 MP/L (Fig. 2). The data shown in Fig. 2 are described in more detail in Table S1 where the removal rate, identification method, particle shape and polymer composition are reported. Especially high concentrations have been observed in raw wastewaters in Denmark (Vollertsen and Hansen 2017; Simon et al. 2018). The Danish studies assessed MPs in the smaller size range (i.e. between 10 or 20 and 500 μm), while other studies assessing MPs down to 20 μm found much lower MP concentrations (Talvitie et al. 2015; Leslie et al. 2017).

WWTPs have, in general, a large retention potential for MPs, often higher than 95% (Table S1). However, in treated wastewater the number of MPs varies greatly too, from less than 1 MP/L (Browne et al. 2011; Carr et al. 2016; Murphy et al. 2016; Ziajahromi et al. 2017a) to several hundred (Simon et al. 2018) and up to several thousand MP/L (Vollertsen and Hansen 2017; Fig. 2). Larger MPs are usually better retained during the treatment, so the most frequently observed MPs in treated wastewater are smaller than 300 μm (Dris et al. 2015; Mintenig et al. 2017; Gündođdu et al. 2018; Magni et al. 2019; Talvitie et al. 2017a; Lee and Kim 2018; Wolff et al. 2019; Liu et al. 2019b). For example, Magni et al. (2019) found that 94% of the MPs between 5 and 1 mm were retained by an Italian WWTP, while only 65% of the MPs between 0.1 and 0.01 mm were retained (Magni et al. 2019). Moreover, the number of MPs seems to be increasing with decreasing particle size. Wolff et al. (2019) reported the results of small-sized MPs measured in treated wastewater and indicated that 44% of the measured MPs are between 10 and 30 μm , while 51% are between 30 and 100 μm . Furthermore, current research indicates that the amount of MPs retained by WWTPs is not only influenced by the size but also by the particle shape. Usually, fibres are better retained in WWTPs as compared to

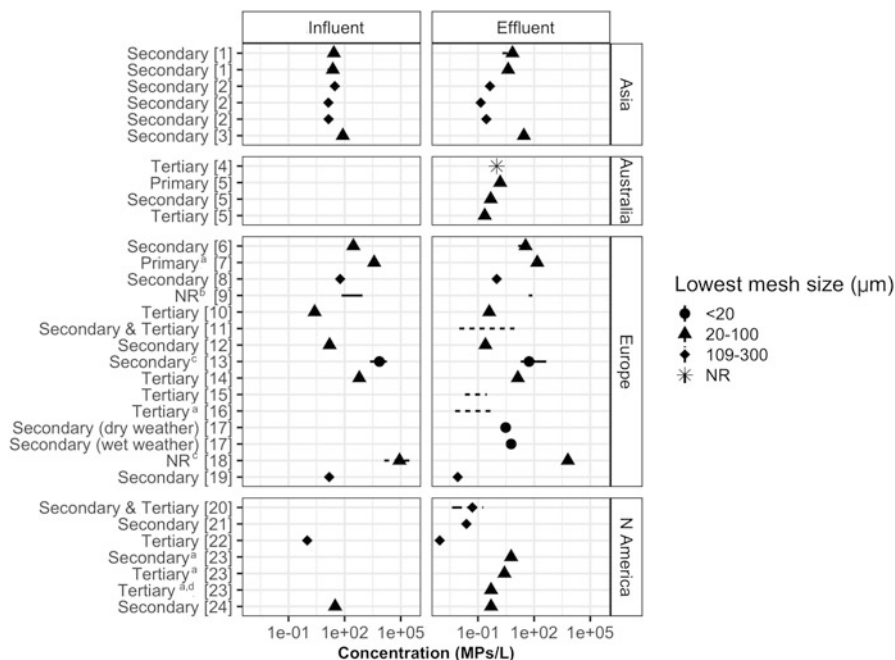


Fig. 2 Mean and/or minimum-maximum MP concentrations (MPs/L) in influent and effluent of municipal WWTPs with different treatment types. NR = not reported. N America = North America. Notes: ^aAll anthropogenic litter in the micro range and not only MPs considered. ^bRange of mean concentrations between seven different WWTPs. ^cUpper size limit of 500 µm. ^dPilot-scale anaerobic membrane bioreactor. References: [1] Gündoğdu et al. (2018), [2] Lee and Kim (2018), [3] Liu et al. (2019b), [4] Browne et al. (2011), [5] Ziajahromi et al. (2017), [6] Dris et al. (2015), [7] HELCOM (2014), [8] Lares et al. (2018), [9] Leslie et al. (2017), [10] Magni et al. (2019), [11] Mintening et al. (2017), [12] Murphy et al. (2016), [13] Simon et al. (2018), [14] Talvitie et al. (2015), [15] Talvitie et al. (2017a), [16] Talvitie et al. (2017b), [17] Wolff et al. (2019), [18] Vollertsen and Hansen (2017), [19] Magnusson and Noren (2014), [20] Mason et al. (2016), [21] Dyachenko et al. (2017), [22] Carr et al. (2016), [23] Michielssen et al. (2016), [24] Gies et al. (2018)

microbeads or other irregular particles (Magnusson and Norén 2014; Talvitie et al. 2017b; Gündoğdu et al. 2018). Fibres and fragments are the most frequently occurring MP types in WWTP effluents (Table S1). Regarding polymer composition, PE particles or PES fibres are the most common plastic types (Table S1). Although a huge amount of tyre debris is suspected to enter WWTPs (Kole et al. 2017), they have not been frequently reported in treated effluents (Table S1). Only Dyachenko et al. (2017) and Lee and Kim (2018) have reported the presence of black particles possibly being tyre fragments.

Concentrations of MPs in wastewaters show some seasonal and diurnal variations related to water consumption rates and human activity (Mintening et al. 2017; Talvitie et al. 2017b; Lares et al. 2018). For instance, Talvitie et al. (2017b) reported that night time concentrations were slightly lower (average concentrations 476.7 and 0.8 µL/L in influent and effluent, respectively) compared to daytime concentrations

(584 and 1.7 $\mu\text{L/L}$ in influent and effluent, respectively). Therefore, MP occurrence seems to be highly variable and depending on a variety of different environmental (weather, season) and behavioural variables but also methodological procedures (i.e. sampling method, including mesh sizes and sample volume), extraction method and determination method. Despite the high retention of MPs by WWTPs, considering the large volumes treated daily, it is considered that more than one million particles can enter the aquatic environment via this pathway per WWTP (Ziajahromi et al. 2017a; Gündoğdu et al. 2018), which constitutes one of the main sources of MPs into the environment.

3.3 *Sludge and Other Agricultural Amendments*

WWTPs retain the majority of MPs during pre- and primary treatment (mechanical treatment and sludge settling processes) and MPs are therefore concentrated in the grease or sludge phase (Murphy et al. 2016; Leslie et al. 2017; Talvitie et al. 2017b). While solids intercepted by grids and grease removal steps are disposed of in landfills, sludge is often reused as fertilizers in agriculture. The amount trapped in the sludge roughly constitutes 50–90% of the MPs present in raw wastewater (Table S2; Magnusson and Norén 2014; Carr et al. 2016; Lee and Kim 2018). MP concentrations measured in sludge range from 650 MPs/kg dw to more than 240,300 MPs/kg dw (Fig. S1, Table S2). Murphy et al. (2016) found significant bigger-sized MPs in the sludge phase compared to MPs in treated wastewater, confirming the differential retention potential of WWTPs regarding MP size. Furthermore, the sludge treatment process (thickening, digestion, drying, stabilization, dewatering) may affect the MP size (Mahon et al. 2017). Similar to wastewater, sludge samples usually show high numbers of fibres, followed by fragments (Table S2), and the main detected polymer is usually PES (particularly when there are many fibres present), followed by PE and PP.

Plastics can end up in compost used as agricultural amendment due to wrong recycling or separation of waste, e.g. if plastic food packaging is disposed of in the organic waste (Mercier et al. 2017; Weithmann et al. 2018). Weithmann et al. (2018) reported that organic fertilizers may contain up to 895 MPs/kg, and Fuller and Gautam (2016) found on average 23,000 mg MP/kg in composted waste materials.

4 Occurrence and Fluxes of Plastics in Environmental Compartments

It is reasonable to hypothesize that in different countries, relevant differences in the emissions of plastic waste and the presence of MaPs and MPs in environmental compartments exist. This may, in particular, originate from differences in waste

management between high-income and low-income countries. In the following sections, some comparisons at the continental level are made. However, as the availability of information is not adequately balanced among countries and continents (e.g. data from Africa and South America are very scarce), the comparison is only partial.

4.1 Air

Studies assessing the occurrence of airborne plastic particles have identified mainly fibres (Dris et al. 2015; Abbasi et al. 2019). Atmospheric fallout of fibres in the area of Paris (France) showed a high variability, with values ranging between 2 and 355 fibres/m²/day; however, half of those were natural (50%; cotton or wool), and only 17% were purely synthetic (mainly PET; Dris et al. 2016). Based on these samples, the same authors estimated that the fibre deposition rate in highly populated urban environments can roughly range between 1.2 and 4 kg/km²/year and concluded that atmospheric fallout might constitute a relevant pathway of MPs. The limited data on atmospheric MPs deposition rates makes it difficult to draw conclusions on the relevance of this pathway for the environmental distribution of MPs. In the study by Dris et al. (2016), suburban fallout was found to be only about 50% of that observed in urban areas (53 particles/m²/day compared to 110 particles/m²/day), and thus it may be assumed that fibre fallout is even lower in natural and agricultural environments.

In addition to fibres, MPs in street dust are also likely to become airborne (Dall'Osto et al. 2014; Gasperi et al. 2018). According to Kole et al. (2017), 12% of the generated tyre dust (1040 tons/year) in the Netherlands ends up in the air. The particles are generated by the interaction of tyres with the road while driving and are generally found along roadside areas (Kreider et al. 2010). Wind and rainfall might influence the atmospheric transport and fallout of MPs, while deposited fibres and street dust in urban environments may be transported via water runoff into sewer systems or directly to terrestrial or aquatic ecosystems. However, studies properly describing such processes are lacking.

4.2 Soil

It has been suggested that agricultural soils could constitute larger MP sinks than marine ecosystems (Hurley and Nizzetto 2018). However, research on the quantification of plastics in soils (for both MaPs and MPs) is still very limited and mostly contracted to the last 4 years. We identified 12 studies reporting plastics in soil, from which 3 considered only a limited number of plastic types (Table S3). The available studies provide first indications of the scale of the pollution and suggest the ubiquitous presence of MPs in terrestrial ecosystems, also beyond agricultural areas. Most

studies report plastic quantities in terms of particles, while some others provide concentrations based on mass measurements, which hampers to some extent direct comparisons among them. The highest MP concentration based on mass has been measured in soils from an industrial area in Australia, which was historically used to produce chlorinated plastic, containing 6700 mg MP/kg dw (Fuller and Gautam 2016). The highest concentration based on the number of MP particles was provided by Vollertsen and Hansen (2017), who described Danish agricultural soils containing about 145,000 MPs/kg, in the size range of 20–500 μm which was based on weight however only 12 mg/kg. Also Chinese farmland soils were found to contain a high MP content, ranging between 70 and 18,760 MPs/kg dw (Fig. S2; Liu et al. 2018; Zhang and Liu 2018; Zhang et al. 2018). In contrast, farmlands in Germany showed a much lower MP occurrence (0.34 MPs/kg dw; Piehl et al. 2018). This might be partly related to differences in the considered MP sizes during the study and due to differences in agricultural practices. While Piehl et al. (2018) assessed MPs of a size between 1 and 5 mm, the study by Vollertsen and Hansen (2017) considered MPs between 20 and 500 μm . However, the different ranges in concentrations may also be attributed to the presence of different input sources.

The application of sewage sludge as agricultural fertilizer (biosolids) is considered to be a major source of MPs to soils. Nizzetto et al. (2016) estimated that between 63,000–430,000 and 44,000–300,000 tons of MPs could be yearly added to agricultural land in Europe and North America, respectively. Corradini et al. (2019) found that an increasing number of sludge applications were positively correlated to increasing MP concentrations in soils. Zubris and Richards (2005) report up to 1210 fibres/kg in soils 5 years after sewage sludge application and detected fibres still 15 years after application, which is another indication for MPs accumulation in soil due to sludge application. On the other hand, almost twice the concentration of MPs was found in Danish fields not treated with sludge compared to treated fields (Vollertsen and Hansen 2017). Additional studies investigating the presence of MPs in soil after application of wastewater sludge are fundamental to estimate the importance of this pathway better.

Irrigation with reclaimed wastewater and the usage of plastic material in agriculture constitute additional sources of plastics in soil ecosystems. Based on studies from China, the latter one seems to be one of the most important plastic sources for elevated MP concentrations in soil in addition to sewage sludge application (Zhang and Liu 2018; Zhang et al. 2018). In contrast to those concentration hotspots, agricultural areas in Germany without plastic mulching or use of sewage sludge as fertilizer, the MP concentration seems much lower (i.e. on average 0.34 MP/kg dw soil; Piehl et al. 2018). As the frequency of the observed MaP polymer types was reflected by the types of MPs, MP particles in this study most likely come from the degradation of (littered) MaP (Piehl et al. 2018). The breakdown of MaP into MPs in terrestrial ecosystems may be dependent on their whereabouts in the soil and on soil cultivation. Williams and Simmons (1996) assessed low-density PE degradation over a period of 4 months in different environments (river beach, in trees at the river bench and buried by soil). They found that MaPs on the soil surface degrade faster as compared to buried plastics and assumed light to be the main influencing

driver (although rainfall and other weathering processes may have affected degradation).

Littering, drift from landfills or spills from industry can also become important sources of plastics into soils. As described above, deposition of MPs from the air can additionally add MPs to soils. This seems, however, more relevant close to urban areas and streets with heavy traffic. Finally, during flood events, plastics from the aquatic environment can be deposited on the shores of rivers (Scheurer and Bigalke 2018). Therefore, based on the data that is available up to now, the primary inputs of MPs into soil seem to come from agricultural practices (sewage sludge, plastic mulching) and the fragmentation of plastic litter.

The most common polymer types reported in soils are PE and PP (Table S3). MaPs reported in terrestrial systems are PE films and bottles (Ramos et al. 2015; Huerta Lwanga et al. 2017b; Piehl et al. 2018). In a more remote place (desert in southern Arizona), plastics that are more mobile due to transportation by wind like plastic bags and balloons have been reported (Zylstra 2013).

The fate of MPs within the soil is not completely clear yet. MPs in soils may be transported along with water runoff and soil erosion into adjacent streams and rivers. So far, there is no knowledge of the importance of this pathway as it has not been experimentally proven. Translocation into deeper soil layers can occur through soil cultivation (Hurley and Nizzetto 2018) or transport by soil organisms. Earthworms and collembolans have been shown to ingest and transport MPs from the soil surface into deeper soil layers (Huerta Lwanga et al. 2017a; Maaß et al. 2017; Rillig et al. 2017). Also other animals, e.g. birds or domestic animals, which have been shown to take up MPs (Zhao et al. 2016; Huerta Lwanga et al. 2017b), can transport MPs over longer distances. To date, it is yet unclear whether low-sized MPs can be transported through soil pores into groundwater, but low concentrations of MPs (0–7 MPs/m³) have been reported in raw drinking waters from groundwater wells (Mintenig et al. 2019). Uptake of plastics by plants is another potential source of mobilization of plastics from soil ecosystems, particularly for NPs; however no studies have investigated this using whole plants (Ng et al. 2018). The only study available in this respect is the one provided by Bandmann et al. (2012), who demonstrated uptake of 20 and 40 nm PS beads by tobacco BY-2 cells in cell culture via endocytosis, while 100 nm beads were excluded.

4.3 *Freshwaters*

Plastic pollution along rivers has been already observed and assessed in the 1990s (Williams and Simmons 1996, 1999). Nevertheless, few studies have reported plastic pollution in freshwaters until the whole environmental movement was initiated a few years ago. Some studies assessing litter in rivers have not exclusively focused on plastic but also included other litter items like glass, paper and wood. Those studies show that about 80% of the litter items are plastics, but do not provide

concentrations or mass estimates (Crosti et al. 2018; González-Fernández et al. 2018; Castro-Jiménez et al. 2019).

Studies focusing on providing concentrations of MaPs in the environment are very limited (Table S4). MaPs concentrations have been reported, for example, for the Los Angeles River, in California (819 MaPs/m³; Moore et al. 2011), the Yangtze River in China (8.74 × 10³ MaPs/km²; Xiong et al. 2019) and lakes (1800 MaPs/km²) and rivers (0.012 MaPs/m³) in Switzerland (Faure et al. 2015). It has been estimated that in the Seine River in France, 28,000 kg of floating plastic are trapped annually by floating debris retention booms (Gasperi et al. 2014), and floating MaPs in the Saigon River in Vietnam were estimated to range between 7500 and 13,700 tons per year (van Emmerik et al. 2018). As only buoyant plastics were considered in those studies, the total loads may be underestimated as plastic is also transported by sub-surface transport (Morritt et al. 2014). The most common MaPs reported in freshwater environments are plastic bottles, food packaging items, plastic bags and sewage-related plastic, like handles from buds of cotton wool and sanitary towels (Table S4). Regarding polymer composition, PP and PE are the plastic types that were omnipresent, and to a lesser extent, PS and PET have been reported (Table S4).

MPs in water have been assessed using different sampling methods and are reported in different units (i.e. particles per water volume or particles per area) (Fig. 3). To be able to compare the results of the different studies, we choose 38 studies which either reported the number of MPs per water volume or gave sufficient information to transform the reported unit. Like in other environmental

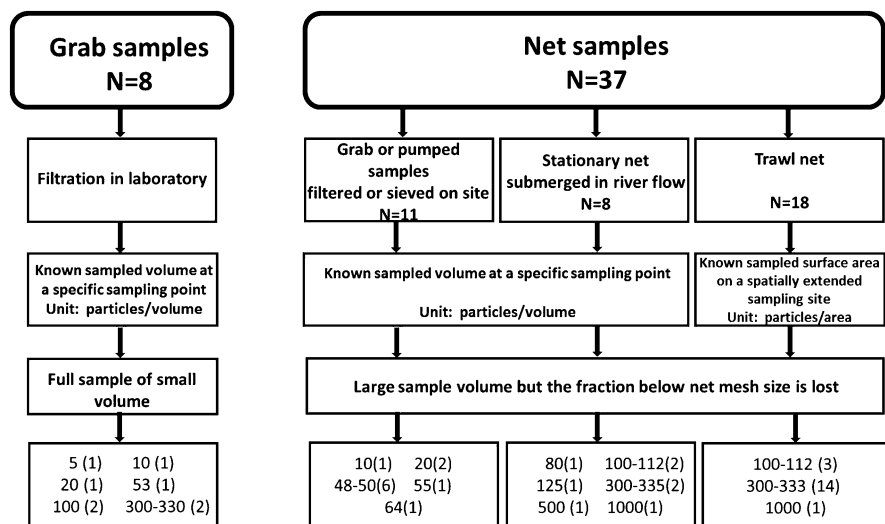


Fig. 3 Overview of the most common sampling methods used for freshwater MPs sampling. N is the number of studies that applied the respective sampling method. This does not correspond to the number of studies listed in Table S5 because some studies used more than one sampling method. In the last row of boxes, the minimum mesh size (µm) used in the different studies is reported (in bracket the number of studies)

compartments, the concentrations varied greatly among studies (Fig. 4). As for Fig. 2, the data shown in Fig. 4 are described in more detail in Table S5 where, besides the identification method, the particle shape, the polymer composition and the sampling methods are reported. Moreover, two additional studies are listed in Table S5, which are not included in the figure because they reported the MP concentration in weight per water volume instead of number per water volume, which hampers a direct comparison. Most studies in Europe found average concentrations of less than 1 to less than 100 MP/m³, while the highest average concentration of 100,000 MP/m³ (with a maximum concentration of 187,000 MP/m³) was measured in the Amsterdam canals (Leslie et al. 2017). Furthermore, Liu et al. (2019a) reported up to 22,849 MP/m³ (average: 1409 MP/m³) in storm water ponds receiving urban runoff in Denmark. The highest peak concentration from all studies was found in the Snake River in North America and was as high as 5,405,000 MP/m³ (average: 91 MP/m²) (Kapp and Yeatman 2018). The second highest peak concentration was reported by Lahens et al. (2018) and corresponds to 519,223 MP/m³ (minimum 17,210 MP/m³) monitored in the Saigon River (Vietnam). Overall, reported concentrations of MPs appear to be higher in Asia, as compared to Europe and North America (Fig. 4). However, most of the studies carried out in Asia were performed in China and focused on assessing lower size classes than those studied in Europe. The only two studies conducted in Europe that considered a very small size (MPs below 20 µm) were the ones by Leslie et al. (2017) and Liu et al. (2019a), who observed by far the highest concentrations. Current research shows that smaller particles (<0.5 mm) are usually the most frequent ones (e.g. Leslie et al. 2017; Yan et al. 2019). Therefore, the higher concentrations found in Asia may be not exclusively related to higher pollution but also to the sampling methods used. The results of this overview are comparable with those reported by Li et al. (2018) on the occurrence of MPs in freshwater.

Studies assessing the concentration of MPs using different net sizes at the same sampling sites found substantial differences in the number of particles intercepted by plankton nets vs trawling nets (Dris et al. 2015; Xiong et al. 2019). Kapp and Yeatman (2018) used different sampling methods to assess the occurrence of particles larger than 100 µm and found that on average there were higher concentrations in grab samples (glass containers were filled with water from the surface) as compared to net samples (Table S5). Also, other differences in study design, such as sample volume, sample depth or sample location in the river, could influence the measured MP concentration. For example, Vermaire et al. (2017) found higher concentrations in grab samples close to the river shore, which were subsequently filtered through a 100 µm net compared to open water samples taken using a 100 µm manta trawl.

Although MPs have been found in remote locations and rural areas, there is evidence that MP concentration increases with proximity to cities (Wang et al. 2017b; Di and Wang 2018; Tibbetts et al. 2018). A modelling study identified the Yangtze River catchment as the catchment transporting the highest plastic loads into the ocean (Schmidt et al. 2017). The four case studies looking at MP concentrations in the Yangtze River found highly variable concentrations but were also among the

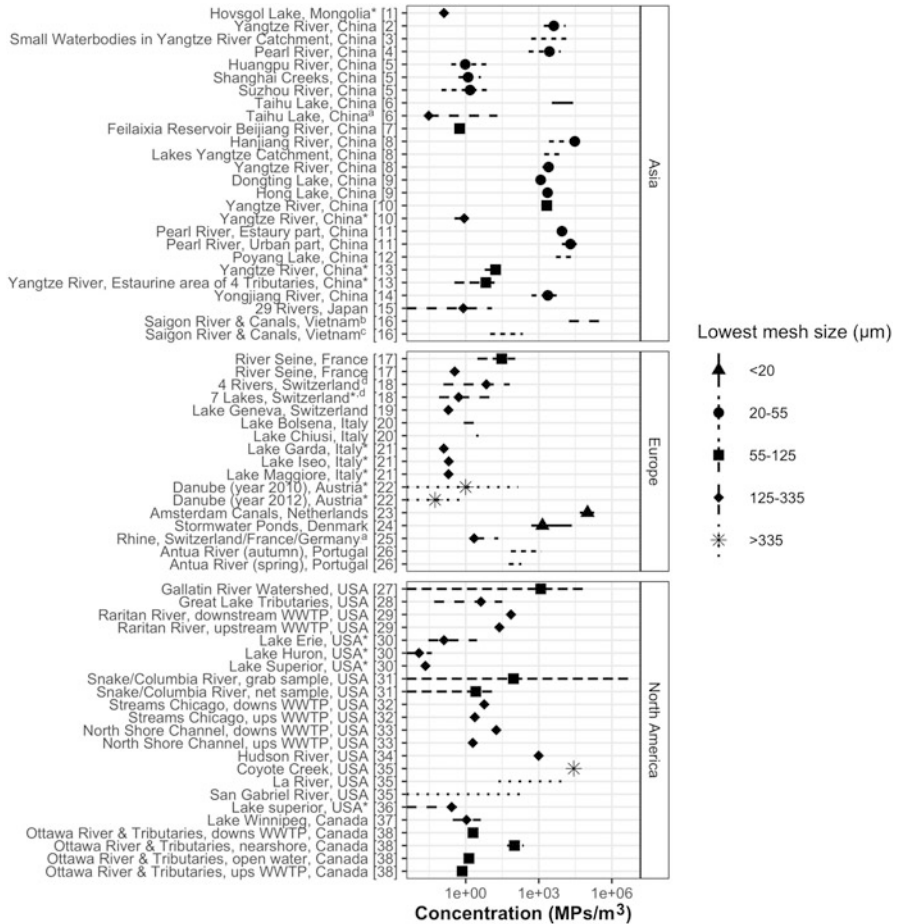


Fig. 4 Mean and/or minimum-maximum MP concentrations (MPs/m^3) in freshwater samples. Downs = downstream. Ups = upstream. Notes: *Concentration in MPs/m^3 was estimated by dividing the reported concentration in particles per area by the height of the net used for sampling. Notes: ^aMinimum concentration not specified. ^bOnly fibres assessed. ^cOnly fragments assessed. ^dMean across all sample sites with minimum and maximum mean concentrations across sample sites. References: [1] Free et al. (2014), [2] Di and Wang (2018), [3] Hu et al. (2018), [4] Lin et al. (2018), [5] Luo et al. (2018), [6] Su et al. (2016), [7] Tan et al. (2019), [8] Wang et al. (2017a), [9] Wang et al. (2018), [10] Xiong et al. (2019), [11] Yan et al. (2019), [12] Yuan et al. (2019), [13] Zhang et al. (2015), [14] Zhang et al. (2019), [15] Kataoka et al. (2019), [16] Lahens et al. (2018), [17] Dris et al. (2015), [18] Faure et al. (2015), [19] Faure et al. (2012), [20] Fischer et al. (2016), [21] Sighicelli et al. (2018), [22] Lechner et al. (2014), [23] Leslie et al. (2017), [24] Liu et al. (2019a), [25] Mani et al. (2016), [26] Rodrigues et al. (2018), [27] Barrows et al. (2018), [28] Baldwin et al. (2016), [29] Estahbanati and Fahrenfeld (2016), [30] Eriksen et al. (2013), [31] Kapp and Yeatman (2018), [32] McCormick et al. (2016), [33] McCormick et al. (2014), [34] Miller et al. (2017), [35] Moore et al. (2011), [36] Hendrickson et al. (2018), [37] Anderson et al. (2017), [38] Vermaire et al. (2017)

highest observed (Zhang et al. 2015; Wang et al. 2017b; Di and Wang 2018; Xiong et al. 2019). However, concentrations in the same order of magnitude were also monitored in other rivers in China such as the Pearl River, which was also ranked under the top ten catchments transporting plastic into the ocean (Schmidt et al. 2017).

Not only spatial hotspots but temporal hotspots based on weather condition may exist in freshwater ecosystems. Storms and rainfall can increase plastic concentration in waters from both lateral (land-based) and sewage effluent discharge points (Fischer et al. 2016), and MPs that had been deposited on river beds can re-enter the water phase after flood events (Hurley et al. 2018a).

Fragments and fibres formed by PE and PP are the most frequently observed particles across all studies evaluating MP pollution in freshwater ecosystems, whereas pellets or beads are only rarely reported as the main occurring plastic types (Table S5). The latter are mainly found in studies along the rivers Rhine and Danube, in the proximity to plastic processing plants, and are thus assumed to be pre-production pellets (Lechner et al. 2014; Lechner and Ramler 2015; Mani et al. 2016). The prevalence of secondary MPs (fragments and fibres) suggests wastewater and runoff as sources for plastic pollution in freshwater ecosystems (Table S5). Several studies confirmed that by demonstrating that MP concentrations are higher downstream of WWTP as compared to sampling sites in upstream areas (McCormick et al. 2014; Estahbanati and Fahrenfeld 2016; Vermaire et al. 2017; Kay et al. 2018). For example, in the Ottawa River (Canada), 0.71 particles/m³ were found upstream of WWTPs compared to 1.99 MPs/m³ downstream. In the Raritan River and the North Shore Channel (USA), 24 MPs/m³ and 1.94 MPs/m³ were found upstream the WWTP, and 71.7 particles and 17.93 MPs/m³ were detected downstream, respectively (McCormick et al. 2014; Estahbanati and Fahrenfeld 2016; Vermaire et al. 2017). As mentioned above, the majority of MPs in wastewater are smaller than 300 µm. Thus it may be presumed that larger MPs enter via different pathway like surface runoff or stem from the breakdown of MaPs directly in the aquatic environment. However, with untreated wastewater, for instance, during sewage overflows, large MPs and MaP can enter river ecosystems. For example, Morrill et al. (2014) identified pollution hotspots in the vicinity of WWTPs that were mainly constituted of sanitary products. MPs hotspots were also detected in areas with low population density but high agricultural use, also pointing to agricultural runoff as an important source (Kapp and Yeatman 2018). Finally, poor waste management likely increases plastic input into aquatic ecosystems (Lahens et al. 2018), where they can break down into smaller particles. Xiong et al. (2019), for example, found that the abundance of MPs is positively related to the presence of MaPs.

4.4 Sediments

Similar to MaP in surface waters, also MaPs in sediments are only rarely assessed, and the way MaP occurrence is reported is highly variable and difficult to compare (Table S6). MaPs along river banks have been observed while assessing buoyant

litter in general (Williams and Simmons 1999; Rech et al. 2014), and river beach sediments in Switzerland contained on average 90 MaPs/m² (Faure et al. 2015). Across different lakeshores, MaPs concentrations have been shown to vary notably (Imhof et al. 2013; Fischer et al. 2016). While high MaPs concentrations have been observed at the south shore of Lake Garda (Italy; with an average concentration of 483 MaP/m²), the occurrence at the north shore was significantly lower (i.e. 0–8.3 MaP/m²; Imhof et al. 2013). Food packaging is among the most frequently observed MaPs, but also bottles, bags and ropes are described by several studies. Regarding the polymer composition, PE and PP, as well as styrofoam (PS), are reported (Table S6).

As for MaPs and the other compartments, the concentration of MPs in freshwater sediments has not been reported in consistent units across all studies. Therefore, we focused on studies that have reported the concentration in MPs/kg sediment. However, studies reporting MPs per sediment area, which gave sufficient information to estimate the concentration in MPs/kg, were also included. Therefore, from the 34 studies that were found during the literature search (Table S7), 30 were chosen for comparisons (Fig. 5). The data shown in Fig. 5 are described in more detail in Table S7 where the type of analysis, particle shapes and polymer composition are reported. The highest sediment concentration of 2071 MPs/kg dw has been found in the urban canals of Amsterdam, where also the highest water concentrations were observed (Leslie et al. 2017). This value is the average of six urban canals with high variability in MP concentration, particularly fibres, where the presence of a hotspot is evident. MP concentrations in river bed sediments seem, in general, higher than in river beach and shore sediments (Fig. 5; Table S7). Most studies on MPs in river bed sediments report concentrations between 100 MP/kg and a few thousands. Studies from Asia were exclusively carried out in China and reported similar concentration ranges as those described in Europe. Interestingly, the study on the Yangtze River (China), which has been estimated to be the highest contributor of plastic to the sea (Schmidt et al. 2017) and is among the highest MP concentrations reported in water (Fig. 4, Table S5), had a comparably low sediment concentration 7–66 MP/kg. The only study carried out in Africa (in a semi-arid South African basin) assessing the concentration of MPs in river sediment reports notable differences between concentrations in summer (1–14.6 MP/kg dw) and winter (13.3–563.8 MP/kg dw), which were related to a reduced flow condition in winter in the studied region (Nel et al. 2018). Subsequently, the hydrological variation shown by many rivers seems to be one of the main factors contributing to MPs deposition and remobilization from river beds. This was also demonstrated by Hurley et al. (2018a), who report that about 70% of the MPs in the sediments of the upper Mersey and Irwell catchments (UK) were exported after a flooding event. Several studies show that, after transportation with the river flows, MPs tend to (re)deposit in low-energy environments, such as meanders, deltas, dams, harbours and coastal lagoons (Claessens et al. 2011; Vianello et al. 2013; Shruti et al. 2019). The deposition of low-density polymers in sediment environments is also related to a density increase by biofouling (e.g. Ye and Andrady 1991; Andrady 2011; Zettler et al. 2013; McCormick et al. 2014).

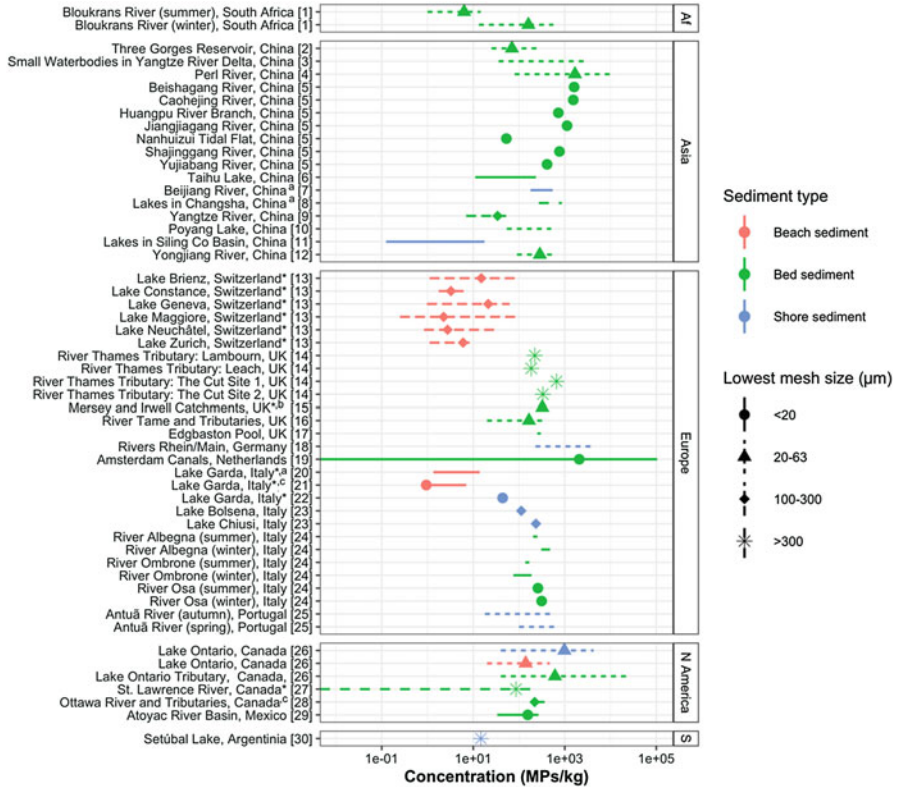


Fig. 5 MP concentrations in different types of sediment samples (MPs/kg). Notes: The sediment type for river sediment was categorized as bed sediment if the type was not clearly stated. ^aConcentration in MPs/kg was estimated by using the sample depth and assuming a density of 1.6 g/cm³ for the sediment. ^aRange of mean concentrations across different sampling sites. ^bMaximum value is shown; ^cno lower value reported. Af = Africa. N America = North America. S = South America. References: [1] Nel et al. (2018), [2] Di and Wang (2018), [3] Hu et al. (2018), [4] Lin et al. (2018), [5] Peng et al. (2018), [6] Su et al. (2016), [7] Wang et al. (2017a), [8] Wen et al. (2018), [9] Xiong et al. (2019), [10] Yuan et al. (2019), [11] Zhang et al. (2016), [12] Zhang et al. (2019), [13] Faure et al. (2015), [14] Horton et al. (2017), [15] Hurlley et al. (2018a), [16] Tibbetts et al. (2018), [17] Vaughan et al. (2017), [18] Klein et al. (2015), [19] Leslie et al. (2017), [20] Imhof et al. (2013), [21] Imhof et al. (2016), [22] Imhof et al. (2018), [23] Fischer et al. (2016), [24] Guerranti et al. (2017), [25] Rodrigues et al. (2018), [26] Ballent et al. (2016), [27] Castañeda et al. (2014), [28] Vermaire et al. (2017), [29] Shruti et al. (2019), [30] Blettler et al. (2017)

For lakes, mainly beach and shore sediment concentrations have been reported. In Europe, average concentrations for beach and shore sediments ranged between 0.94 and 44 MP/kg, while beach and shore sediments from Lake Ontario (Canada) contained much higher concentrations (20–27,830 MPs/kg; Fig. 5, Table S7). Several studies have noted that plastic concentrations differ strongly between different areas of the same lake (Zbyszewski and Corcoran 2011; Imhof et al. 2013;

Zbyszewski et al. 2014; Zhang et al. 2016), suggesting that accumulation is patchy and the formation of contamination hotspots is influenced by winds, waves and/or beach morphology (Imhof et al. 2016, 2018). Similar observations were made at Lake Huron (Canada), in which 94% of all monitored pellets were found to accumulate in one single beach (Zbyszewski and Corcoran 2011). In the Taihu Lake (China), MP concentrations ranged from 11 to 235 MP/kg in different bed areas, and the average MP abundance in sediments in the northwest area was approximately six times higher than the abundance of the south-east area (Su et al. 2016).

Fibres followed by fragments were usually the most common particle types monitored (Table S7). Spheres/beads or pellets were, in rare occasions, reported to be dominant, and mostly in the vicinity to plastic industries (Zbyszewski and Corcoran 2011; Zbyszewski et al. 2014; Corcoran et al. 2015; Hurley et al. 2018a; Peng et al. 2018). Based on polymer type, PE and PP were the most common, despite their buoyant properties, as well as PS (Table S7).

4.5 *Marine*

Rivers are estimated to be the main pathways for plastics entering the oceans. Estimations on the amount of plastic waste entering the ocean through this pathway range between 0.41 and 4×10^6 tons per year (Lebreton et al. 2017; Schmidt et al. 2017). From the top ten river catchments that transport 88–95% of the global plastic load into the oceans, eight are located in Asia (Schmidt et al. 2017). Oceans have been assumed to be the final sink for MaPs and MPs. As this review is focused on terrestrial and freshwater ecosystems, this compartment will not be discussed in detail. A number of articles and reviews have been published on the topic within the last few years which describe plastic occurrence in the oceans and its effects on marine life (see, e.g. Barboza and Gimenez 2015; Jambeck et al. 2015; Auta et al. 2017).

5 Discussion

We fully agree with the statement provided by the SAPEA (2018) report: ‘The number of papers is growing exponentially in this field, but knowledge is not growing at the same rate – there is some redundancy and marginality in the papers’. Furthermore, many papers on plastic pollution do not assess and describe important plastic sources and flows. This review paper made an attempt to describe the available information regarding global environmental loads and the plastic life cycle and to show which further research studies are needed to fully understand specific plastic sources and pathways. This section describes the areas that need further research commitment and development to improve exposure assessments and to evaluate the long-term risks of plastics to terrestrial and freshwater ecosystems.

5.1 *The Need for Advancing and Standardizing Sampling and Analysis Techniques*

As indicated in several parts of this review, the sampling methods reported in the literature are extremely variable and, in many cases, difficult to compare. In marine monitoring studies, the most commonly used method for sampling is the so-called manta trawl, a device similar to a large plankton net with a mesh size usually larger than 300 μm (GESAMP 2015). Using a manta trawl allows to sample a thin layer of surface water, and, therefore, the results are generally reported as MPs (number or weight) per surface area (m^2 or km^2). The same device is frequently used also in freshwater, together with other sampling methods (Fig. 3), that produce results expressed as MPs per volume unit (e.g. L or m^3) and that may consider different size fractions, sometimes down to 20 μm . The results from studies considering the different sampling methods are hardly comparable. Data for surface units may be converted into data for unit volume, by calculating the mouth surface area of the manta trawl. However, this is a rough approximation because the trawl is not always fully immersed. Moreover, with the manta trawl, all particles below 300 μm are lost. This is shown by studies using both sampling methods (Kapp and Yeatman 2018; Lahens et al. 2018; Xiong et al. 2019). Small particles generally represent the largest share of the total amount of particles present in natural waters. Therefore, the manta trawl method largely underestimates the actual MP concentrations, at least in terms of particle numbers.

A recent report (GESAMP 2019) describes and compares methods for sampling MaPs and MPs, with particular focus on the marine environment. The report highlights advantages and disadvantages of the different sampling methods. However, particularly for MPs, precise indications or suggestions of the methods to be used for a better exposure and risk characterization are not provided. It should be noted that the impact of different size fractions may be extremely different on the various components of the aquatic ecosystem (e.g. small fishes, macro-invertebrates, micro-invertebrates, bacteria). Therefore, methods capable to provide quantitative samples of different fractions, including relatively small MPs (e.g. down to 20 μm , achievable with fine-meshed phytoplankton nets), should be used whenever possible.

The available data on soil and sediments is relatively scarce. This may be partly related to the complex and time-consuming procedure required to extract MPs from these matrices (Hurley et al. 2018b). Some studies report MP concentrations as the number of particles per kg, while others provide the weight of MPs per kg. In other cases, data is reported as MP number or weight per surface unit (e.g. mg/m^2). Therefore, the comparison of literature data is not straightforward.

Besides this, existing methods to identify and count MPs are quite variable. Until recently, it was common practice to solely rely on visual detection (using a microscope), which may lead to false positives or false negatives. In more recent studies, visual examination is usually combined with FTIR (Fourier transform infrared) or Raman spectroscopy, which allows polymer identification. This is, however, time-

consuming, and thus frequently only a subsample is subjected to spectroscopic methods. Other studies use different methods like SEM (scanning electron microscopy), XRF (X-ray fluorescence) and Pyr-GC/MS (pyrolysis interfaced with gas chromatography/mass spectrometry) (Klein et al. 2017). It has been observed that MP abundance often varies with the methods used (Song et al. 2015; Mai et al. 2018; Picó et al. 2019), so analytical results may be difficult to compare across studies. The previously mentioned GESAMP report (GESAMP 2019) also compares methods for processing and analysing MPs but does not provide clear suggestions for standardization.

There is an urgent need for standardizing methodologies to be applied to the exposure assessment, which include those related to sample processing, MP extraction, identification and counting, as well as the units to be used for reporting data. The major gap refers to very small-sized MPs (below 20 μm) and NPs. Particularly the latter can currently not be included in monitoring programmes because suitable sampling methods are lacking and analytical methods, such as pyrolysis-GC/MS (Fischer and Scholz-Böttcher 2017), are exploratory (GESAMP 2019).

Most procedures commonly applied to date allow sampling, processing and measuring particles down to a minimum size of 20 μm . Only very few studies measured smaller particles, down to 10 μm (e.g. Leslie et al. 2017; Simon et al. 2018). In theory, very small particles and, especially, NPs should be more abundant in the environment, and their concentrations are expected to increase. The development of methods for the evaluation and quantification of small-sized MPs and NPs is one of the major research needs to assess the potential risks for human and environmental health. In particular, detection technologies to identify nano-sized plastic particles are still lacking (Mai et al. 2018). A promising approach, at least to quantify the mass and the composition (if not the number of particles), could be the use of Pyr-GC/MS (Hendrickson et al. 2018; Mintenig et al. 2018) coupled with methods of small-sized particle separation based on ultrafiltration membrane technologies (Mulder 1998; Judd and Jefferson 2003).

5.2 Towards a Microplastic Mass Balance and Suitable Evaluation of Environmental Fluxes

The difficulties in getting reliable and comparable results for the concentrations of MPs in the different environmental compartments and the limited information regarding some fluxes among compartments make the evaluation of a regional and global mass balance of plastics challenging. However, some first estimates can be made based on the available data, at least to give an approximate order of magnitude of the contribution of different sources to surface waters.

From the data reported in Fig. 2 and Table S1, it can be concluded that the range of particles in effluents from WWTPs that include secondary and tertiary treatments spans from 1 to 5800 MPs/L, with a geometric mean around 29 MPs/L. In

Table 1 Percentage of EU population connected to WWTPs in 2015 (EEA 2019)

	No treatment or no connection with sewerage	Primary treatment	Secondary treatment	Tertiary treatment
Northern	15.1	5.6	2.3	77
Central	3.4	0	16.5	80.1
Southern	23	2.2	21.3	53.4
Eastern	26	0.2	13.6	60.6
South-eastern	40	16.7	22.8	20.6
Weighted average with respect to population	13	2	18	67

non-treated wastewaters, the concentrations range from a few particles/L up to more than 100,000, with a geometric mean of about 242 MPs/L. These data are in reasonable agreement with the percentage of retention by WWTPs reported by several authors, which ranges from 80% to 99% of the number of inflowing particles (see Sect. 4.3).

The approximated per capita consumption of water in Europe is 140 L per day (EEA 2018). Although with some regional differences, it may be estimated that about 85% of the EU population (525 million in the EU plus Norway and Switzerland) is connected to WWTPs with secondary or tertiary treatment, while the remaining population (15%) is connected to WWTPs with only primary treatment or is not connected at all (Table 1).

From these data, it can be estimated that the daily input of MPs (in the range of 20–5000 μm) via wastewater into European surface waters is:

- From treated wastewater: an average value of 1800E+9 particles per day (possible range from 9E+9 to 130E+12 particles/day)
- From untreated wastewater: an average value of 2700E+9 particles per day (possible range from 27E+9 to 1400E+12 particles/day)

Transforming these data on a weight basis is challenging because, in general, only numbers of MPs are reported, while size/weight conversion factors are not readily available. Combined data on numbers and weight are reported in a Danish report (Vollertsen and Hansen 2017) assessing MP occurrence in ten different WWTPs and in the study by Simon et al. (2018). However, both studies took only MPs between 10 or 20 and 500 μm into account. Therefore, estimating the load on a weight basis from the particle numbers is not possible.

Despite their wide range of variability, these estimates give a first approximation of the load of MPs in surface waters from urban wastewater and allow the following observations. First, the load that may be attributed to the relatively small percentage of European untreated wastewaters is much higher than the load deriving from treated wastewater, which points towards a definite need of implementing secondary and tertiary WWTPs in areas that are still not connected to reduce total MPs emission. Taking into account that untreated wastewater is concentrated in south-

eastern Europe, it may be hypothesized that some watersheds (e.g. lower Danube) are subjected to higher contamination than those located in other European regions (Lechner et al. 2014). Unfortunately, data on MP concentrations in surface waters of south-eastern Europe are not available. Siegfried et al. (2017) implemented a modelling approach based on estimations of mass-based per capita consumption rates of personal care products, plastic fibres from textiles, plastic fibres in household dust and tyre debris and concluded that the largest emitted mass from rivers to the sea occurs for tyre debris, followed by textile fibres. Furthermore, they estimated that the majority of plastic particles emitted in Europe flow into the Mediterranean and the Black sea as a result of different socio-economic development and technological status of sewage treatment facilities.

Due to the scarcity of data of water consumption and WWTP implementation, a comparable evaluation cannot be done for other continents. However, it may be hypothesized that the percentage of treated wastewater in Asia and Africa is much lower than in Europe or North America.

The problem is also complicated by the fact that only a relatively small part of the population is connected to sewerage systems. Data from the WHO/UNICEF Joint Monitoring Programme (JMP), referred to 2015, indicate that in Eastern, South-eastern and Central Asia, with a population of more than four billion inhabitants, only 25% of the population is connected with sewerage systems and in sub-Saharan Africa the percentage is lower than 6% (WHO/UNICEF 2019). The high concentrations of MPs in surface waters of Asia (mostly assessed in China), as compared to those measured in Europe (Fig. 4), support the hypothesis regarding the large influence of WWTP on surface water emissions. The dominant shape in WWTP effluents are fibres, followed by fragments. Only in one case a minor amount (<10%) of pellets that may be classified as primary microbeads was observed (Dyachenko et al. 2017).

Wastewater represents only one of the possible pathways of MPs into surface waters, and as discussed in this study, surface runoff from agricultural and urban soils may also represent a major source. Unfortunately, a comparable estimate of MPs emissions from soils due to water runoff is not possible due to field data limitations. On the other hand, this review shows that MP concentrations in WWTP sludge (mainly from Europe) range between $10E+3$ and $10E+5$ particles/kg dw. Nizzetto et al. (2016) estimated that the total yearly input of MPs from sewage sludge to farmland is about 63,000–430,000 tons in Europe and 44,000–300,000 tons in North America. Data on MP concentrations in soil are scarce and scattered (Fig. S2 and Table S3). The majority of data on agricultural soils refer to China and indicate a reduced range of variability (from about 60 to 200 particles/kg dw), except for a couple of higher values (more than 10,000 particles/kg dw) from soils sampled in a greenhouse. Overall this study shows that soil could be considered as a sink as well as a source of MPs to surface water. Therefore, further research is urgently required to assess fluxes of MPs from soils into surface water ecosystems and to assess the fate of MPs in the soil ecosystems, investigating its retention potential and the capacity of MPs to reach groundwater ecosystems. An additional source of MPs to soil and surface water may be

atmospheric fallout (Dris et al. 2016). However, the information available to date does not yet allow a quantitative estimate (Wetherbee et al. 2019). An attempt to perform a quantitative evaluation of emissions to all environmental compartments (air, soil, WWTP, surface waters) has been made for tyre debris indicating that urban and road runoff, as well as atmospheric deposition, may represent relevant contributions (Kole et al. 2017).

The formation or disappearance of MPs within the compartments also has to be considered in an overall mass balance. MaP fragmentation in the different compartments is reasonably one of the major sources of MPs in the environment. However, the patterns of MaP fragmentation, their characterization and quantification in terms of amount produced and time to produce them are still largely unknown. The only fragmentation pattern that is sufficiently documented and quantified is the production of fibres during laundry of synthetic fabrics (Browne et al. 2011; Eerkes-Medrano et al. 2015). Although the amount of fibres may vary depending on the type of clothes (e.g. polymer composition, weave type, age), the type of washing machine and the washing condition, it has been estimated that several thousand fibres are generated per washing cycle (Hartline et al. 2016; Napper and Thompson 2016; Pirc et al. 2016; Carney Almroth et al. 2018).

For any other type of plastic breakdown process, reliable experimental quantitative information is not yet available, although a modelling approach to predict the contribution of MaP breakdown to the MPs bulk in the ocean has been proposed (Koelmans et al. 2017b). Plastic fragmentation in the environment may be extremely variable in function of factors like light intensity, temperature, erosion and other physical impacts. The number and weight of MPs and NPs that may be produced by a MaP item (e.g. a bag or a bottle) in a given time under environmental conditions are still largely unknown. This is an important knowledge gap that must be investigated in depth and may be somewhat inferred based on the amount and type of polymers of MaP litter in the environment and their documented half-lives.

Although plastic polymers are persistent compounds, some polymers can undergo biodegradation (Albertsson et al. 1987). Scientific evidence of biodegradation through bacterial activity and invertebrate digestion mechanisms has increased recently (Briassoulis et al. 2015; Yoshida et al. 2016; Yang et al. 2018). Compared to MaPs, MPs and NPs may be more prone to form complex structures with organic matter particles and be readily attacked by bacteria and invertebrates. Therefore, a real possibility of their complete disappearance exists. Nevertheless, to date, the extent of these degradation processes in environmental compartments, their time scale as well as the patterns and the end products are relatively unknown (SAPEA 2018). Although plastic polymers are practically inert molecules, with low biological and toxicological activity, many monomers, which can be formed during the degradation of plastic, are not. Monovinylchloride (the monomer of PVC), for instance, is a recognized carcinogenic compound (Brandt-Rauf et al. 2012).

5.3 *Microplastics in Environmental Compartments: What Does It Mean in Terms of Risks for Living Organisms?*

As discussed above, information on the presence of MPs in environmental compartments is often biased by the inconsistency of units (e.g. n/L, n/m², mg/L, n/kg, mg/kg), by the variability in size classes sampled and measured and by the complexity in shape and composition that are often not clearly reported. These inconsistencies make the assessment of their possible impact on living organisms rather complex, so the actual environmental risks of different plastics and their associated chemicals remain largely unknown (Koelmans et al. 2017a). It is important to highlight that quantifying the effects of MPs on living organisms by a simple concentration-response relationship of the whole mass of MPs of a certain type found in environmental samples is more complicated than for most chemical contaminants. Their impacts on aquatic organisms depend on a number of factors such as:

- The shape: the physical effect determined by long and thin fibres may be completely different from those determined by microspheres or by irregular fragments (Au et al. 2015; Lambert et al. 2017).
- The size range: the definition of MPs in terms of size is extremely wide (from 5 mm to 1 µm), and the living organisms that may be affected by MPs are also extremely variable in size, for example, in the aquatic environment, from fish to zooplankton; for any type and size of organism, different MP size classes may be ingested, including small sizes (below 20 µm) and NPs, that are practically never measured.
- The composition: for most MP polymers, being the effects mainly physical, it may be hypothesized that the response is not related to the polymer composition; however, for some particular MP particles, such as for tyre debris, the composition is much more complex, and the effects may also be determined by the leaching of non-polymeric chemicals.

Some recent effect studies took these parameters into account, used exposure conditions in relation to the traits of the organisms (i.e. feeding type; substrate preference) or provided dose-response relationships (e.g. Au et al. 2015; Redondo Hasselerharm et al. 2018; Scherer et al. 2017; Ziajahromi et al. 2017b). This allows to conduct an ecological risk assessment with preliminary data for a range of species based on a comparison between an environmental exposure (e.g. a PEC – predicted environmental concentration) and an effect level (e.g. a PNEC – predicted no effect concentration). So far five studies carried out a provisional ecological risk assessment for MPs (Adam et al. 2019; Besseling et al. 2019; Burns and Boxall 2018; Everaert et al. 2018; Zhang et al. 2019). Burns and Boxall (2018) did not identify any ecological risk for measured concentrations in water or sediment. Adam et al. (2019) conducted risk assessment for different continents and found that although no risk is expected for Europe and North America, ‘a risk cannot completely be excluded in Asia’, where the highest MP pollution occurred. These findings are supported by a

case study in the Youngjiang River in south China (Zhang et al. 2019). The authors calculated a risk quotient for each of their sample sites by dividing the measured MP concentrations by PNEC values derived from species sensitivity distribution based on literature toxicity data. They found that for most sites, a risk threshold was not exceeded, except for the two most contaminated sites, which were close to the urban centre of Nanning City (Zhang et al. 2019). Besseling et al. (2019) concluded that hazardous MP concentrations do not occur for freshwaters, while hotspot locations of nearshore marine surface waters may exceed safe concentrations. Similarly, Everaert et al. (2018) derived for the marine environment that at current average concentrations, no risk is expected but that a risk cannot be excluded for heavily polluted sites. As MP concentrations in the environment are usually reported in particle numbers while effect data is based on mass, a transformation was necessary in these studies to be able to compare effect and exposure data. To perform a refined ecological risk assessment of MPs will require much more detailed information on MP exposure with a precise assessment of number (or weight) of particles per size classes, shape and composition. Considering that current methods for the analysis of MPs are complex, expensive and time-consuming, this level of detail is, to date, difficult to achieve. Further research should be devoted to both areas, to refine exposure assessments including areas that are expected to be heavily polluted but on which information is still completely missing such as the Ganges in India or the Amazon in South America (Adam et al. 2019). Moreover, effect assessments should be performed taking into account ecologically relevant combinations of organisms and MP sizes, shapes and types. It is most likely that future risk assessments need to consider MP particle mixtures taking into account different polymer types, shapes and sizes and that exposure and risk indicators are derived taking all these variables into account.

For NPs, risk assessment is currently not feasible as they cannot be detected in environmental samples thus far. Also, regarding the effect assessment, the major unknown issues are related to small and very small particles (Koelmans 2019). NPs are particularly interesting because they may cross cellular membranes and enter into cells if they are below a given size. Within the cells, NPs can possibly interact with the cellular content, structure and function. This represents a substantial difference in comparison with MaPs or MPs. Indeed MPs cannot be accumulated in biological organs and tissues and may produce mainly physical stress on living organisms, although the consequences of that may result in physiological and metabolic alterations. The size threshold below which small plastic particles may enter in the cells is still unknown. Recent studies on NPs performed with reference materials labeled with fluorescent dye demonstrate their capacity to be taken up, enter tissues and accumulate in small organisms (Cui et al. 2017; Lee et al. 2019). However, some authors discuss that this can be an artefact either created by the leaching of those dye paints, which can be taken up into cells or due to the autofluorescence of the evaluated biological tissues (Catarino et al. 2019; Schür et al. 2019).

5.4 How Can MP Inputs into the Environment Be Controlled?

From all the considerations mentioned above, it is evident that the precautionary principles strongly push towards the control of MPs and NPs. From the available literature on MP presence in the environment, it appears that primary MPs represent a relatively small amount of the total bulk of MPs detected, being secondary MPs the largest majority. It is difficult to quantify the percentage of primary MPs in the environment precisely. However, in general, it seems to be never higher than 10%, and in most cases, the percentage is much lower, sometimes almost negligible. For example, in urban wastewater, the majority of MPs is represented by textile fibres (see, e.g. Dris et al., 2015; Vollertsen and Hansen 2017; Wang et al. 2017b), while in runoff water the most abundant particles are fragments from MaP breakdown (see, e.g. Liu et al. 2019a). Therefore, the recent proposal of ECHA (2019) for a ban or restriction of primary MPs may have a limited relevance and effectiveness for the reduction of the presence of MPs in the environment.

Based on the information available to date, the most plausible solution to reduce the environmental emission and exposure to MPs seems to be the control of MaPs. The restrictions on single-use plastic items that will be active in Europe starting from 2021 (EC 2019) seem to be an excellent starting point. Comparable restrictions should be applied in the short term on food and other kinds of packaging, which represent the largest amount of plastic wastes. In addition to restrictions, a more efficient recycling strategy and improvement of the circular economy related to plastic products would be beneficial (Barra and Leonard 2018). However, in some cases, different types of measures should be developed. As shown above, fibres represent the most abundant type of MPs present in wastewater. Since it is almost impossible to ban synthetic fabrics that today make up the majority of our clothing, the solution should be sought in another direction (e.g. by means of retaining fibres in washing machines, water treatment procedures, etc.).

Finally, the substitution of traditional plastic polymers, based on the petrochemical industry, with new-generation polymers, based on biological resources (e.g. PLA, polylactic acid; PHA, polyhydroxyalkanoates) is often proposed as a suitable solution. However, present knowledge on the toxicological properties of these new compounds and of their degradation products must be improved (Lambert and Wagner 2017; Picó and Barceló 2019). Understanding possible biodegradation patterns of traditional and emerging plastic polymers is important for future management and remediation of plastics in the environment.

6 Conclusions

In this study we have described the state of the knowledge regarding the occurrence of MaPs and MPs in different environmental compartments. It has been highlighted that some data gaps still exist in order to better understand their life cycle, to develop a precise mass balance and to quantitatively assess the contribution of the different main sources of MaPs, MPs and NPs in the environment. The emission of MPs from WWTPs into aquatic ecosystems is the environmental pathway that has been most researched. However, there are other pathways that may have similar or even larger contributions and that require further investigation. Those pathways are, for example, the fluxes of plastics from landfills and agricultural soils towards surface and groundwater ecosystems by water runoff or deep-horizon infiltration, or the transport and deposition of plastic particles from the atmosphere. Moreover, quantitative evaluations of the occurrence of large-sized plastics in natural environments need to be performed, and their breakdown rates into MPs and NPs still need to be assessed under different environmental conditions (i.e. temperature and light intensities, water currents).

There is enough experimental evidence demonstrating that the presence of MaPs in aquatic ecosystems represents an environmental risk, particularly for large animals. Regarding MPs, a risk for human and environmental health has not been demonstrated. Available toxicological evidence indicates that some effects on aquatic and terrestrial organisms, vertebrates and invertebrates, have been observed only at concentrations that are orders of magnitude higher than the maximum levels measured in the environment. Other possible effects, such as a potential increase in the bioaccumulation of chemicals due to their transport into the organisms adsorbed on MPs (the ‘Trojan Horse effect’), seem to be context dependent and negligible in comparison with direct accumulation from the surrounding environment (e.g. from water) or from food (Koelmans et al. 2013, 2014; Lohmann 2017; Mohamed Nor and Koelmans 2019). However, research still needs to demonstrate this experimentally. An additional gap is represented by the toxicological risks of NPs that has to be investigated in further detail, taking into account their bioaccumulative and reactive potential in biological tissues, organs and cells.

Current knowledge gaps regarding environmental fluxes and breakdown of MPs and NPs are still too large in order to assess future risks for man and the environment. Furthermore, the bias on sampling and analysis makes a precise quantification challenging. This is particularly difficult for small MPs and NPs, which are probably the more concerning particles from a toxicological point of view. Moreover, although present exposure seems to be far away from levels of concern, it is difficult to predict future emission patterns since they will be closely related to plastic use and management policies. This review shows that the construction of wastewater treatment facilities and the proper management of sludge applications in agriculture are efficient means to reduce MPs emissions. Moreover, the ban of single-use plastics, the substitution of some plastic polymers with biodegradable compounds and the reduction of MPs emission at a source are key to control plastic pollution. From now

onwards, we expect technological solutions to be developed and implemented in this direction. There is no doubt that plastics changed our life in the middle of the last century and the control of plastics will again change our life in the near future.

7 Summary

This study assessed the current knowledge regarding the industrial sources of plastics and MPs, their environmental pathways and load rates and their occurrence and fate in different environmental compartments. Existing data limitations regarding the global environmental cycle and exposure sources of MaPs, MPs and NPs to aquatic and terrestrial ecosystems are highlighted. While the presence of MPs in wastewater and freshwater is relatively well studied, data on sediments and especially soil ecosystems are too limited. Moreover, the overall occurrence of large-sized plastics, the patterns of MP and NP formation from them, the presence and deposition of plastic particles from the atmosphere and the fluxes of all kinds of plastics from soils towards aquatic environments (e.g. by surface water runoff, soil infiltration) are still poorly understood. Finally, this study discussed several research areas that need urgent development in order to better understand the potential ecological risks of plastic pollution and provides some recommendations to improve management and control of plastic and MP inputs into the environment.

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