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REVIEW PAPER

Sources, transport, measurement and impact of nano and microplastics in urban watersheds

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Abstract The growing and pervasive presence of plastic pollution has attracted considerable interest in recent years, especially small (< 5 mm) plastic particles known as 'microplastics' (MPs). Their widespread presence may pose a threat to marine organisms globally. Most of the nano and microplastic (N&MP) pollution in marine environments is assumed to originate from land-based sources, but their sources, transport routes, and transformations are uncertain. Information on freshwater and terrestrial systems is lacking, and data on nanoplastic pollution are particularly sparse. The shortage of systematic studies of freshwater and terrestrial systems is a critical research gap because estimates of plastic release into these systems are much higher than those for oceans. As most plastic pollution originates in urban environments, studies of urban watersheds, particularly those with high population densities and industrial activities, are especially relevant with respect to source apportionment. Released plastic debris is transported in water, soil, and air. It can be exchanged between environmental compartments, adsorb toxic compounds, and ultimately be carried long distances, with potential to cause both physical and chemical harm to a multitude of species. Measurement challenges and a lack of standardized methods has slowed progress in determining the environmental prevalence and impacts of N&MPs. An overall aim of this review is to report the sources and abundances of N&MPs in urban watersheds. We focus on urban watersheds, and summarize monitoring methods and their limitations, knowing that identifying N&MPs and their urban/industrial sources is necessary to reduce their presence in all environments.

Keywords Plastic waste · Polymer degradation · Macroplastic · Microplastic fate · Nanoplastic

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1 Introduction

The growing environmental burden and risks of plastic pollution has been a topic of considerable interest in recent years, especially small plastic particles known as microplastics (> 100 nm and < 5 mm) and nanoplastics (≤ 100 nm). The potential risks of small plastic particles are closely tied to the broader problem of growing plastic production and waste because much of this pollution results from the breakdown of macroplastic items. The total production of plastics from 1950 through 2017 was estimated at 8300 million metric tons (Mt), about 60% of which was discarded as plastic waste (Geyer et al. 2017). Nevertheless, nano and microplastics (N&MPs) are generally perceived as posing greater risks than larger plastic debris because they are often invisible and more likely to be ingested by a much wider range of species (Koelmans et al. 2019b). Numerous studies have shown the widespread presence of MPs in the marine environment, reportedly posing a threat to marine organisms globally [e.g., (Auta et al. 2017; Cole et al. 2011; Hidalgo-Ruz et al. 2012; Song et al. 2014; Wright et al. 2013)]. Widespread contamination of freshwater and terrestrial systems also has been reported (Allen et al. 2019; Dris et al. 2015a, b, 2016, 2017; Gasperi et al. 2018; Horton et al. 2017a, b; Strungaru et al. 2019; Triebskorn et al. 2018; Wagner et al. 2014). More recent studies have indicated the environmental presence of nanoplastics (NPls), which may pose different risks than MPs due to their small size. They have much higher specific surface area for sorption of other contaminants and can translocate to other body tissues after ingestion (Bouwmeester et al. 2015). Effects of N&MPs depend on particle properties that are not well understood. However, smaller size and positive surface charge are associated with higher toxicity [e.g., (Bouwmeester et al. 2015; Shen et al. 2020, Stapleton 2019)]. The potential risks of N&MPs are briefly discussed below and later in this review (Sects. 1.2.3 and 4.1).

Studies of the plastic sources and sinks in urban watersheds, and of N&MP transport and fate, are critical to assessing the environmental impact of plastic pollution at all stages, from freshwater and terrestrial ecosystems to oceans. A large majority (estimated at 80%) of the N&MP pollution in marine environments is assumed to originate from land-based sources, but the actual sources and transport routes are uncertain (Geyer et al. 2017; Jambeck et al. 2015; Rochman 2018). Information on NPl pollution is particularly sparse due to the measurement challenges of nanoscale particles (Lehner et al. 2019). The shortage of systematic studies of freshwater and terrestrial systems is alarming, especially because estimates of plastic release into these systems are much higher than those for oceans (4–23 times larger by mass) (Horton et al. 2017b). As most plastic waste originates in urban environments, studies of urban watersheds, particularly those with high population densities and industrial activities, are especially relevant with respect to source apportionment and transport of plastic pollution. Plastic debris/particles can be transported in water, soil, and air. These pollutants can exchange between environmental compartments, sorb toxic chemicals, and ultimately be carried long distances. An overall aim of this review is to report the sources and abundances of N&MPs in urban watersheds. Their measurement, transport, transformations, and potential impacts also are discussed. We focus on recent findings of N&MPs in urban watersheds and summarize monitoring methods, knowing that identifying N&MPs and their urban/industrial sources is necessary to reduce their presence in all environments.

1.1 Global plastic production and waste management

Large-scale plastic production began in the 1950s and is increasing exponentially (Geyer et al. 2017). In 2017, about 348 Mts were produced globally. This global production represents trillions of dollars in economic returns, with China being the top producer (29.4%) (PlasticsEurope 2018). Based on current trends in production and waste management, another 12–33 billion metric tons of plastic will occupy landfills or pollute the environment by 2050 (Geyer et al. 2017; Rochman et al. 2013a). Of the 8.3 billion metric tons produced between 1950 and 2017, 6.3 billion became plastic waste, only 9% of which was recycled (Geyer et al. 2017). The remainder (79%) has accumulated as plastic litter, with oceans being the final sink (Geyer et al. 2017; Rochman 2018). Polyethylene (PE) (high, low, and linear low density: HDPE, LDPE, LLDPE), polypropylene (PP), polystyrene (PS), polyvinylchloride (PVC), polyethylene terephthalate (PET), and polyurethane (PUR) resins;

Table 1 Estimates[a] of major plastic types and total amounts produced since 1950

Polymer	IUPAC Name	Code	Total produced after 1950 (Mt)[a]	Mass % of total since 1950[a]	Total 2015 primary production (Mt)	% of total produced in 2015	Applications
Nonfiber plastics[b]							
Polyethylene: High density (HDPE), low density (LD), linear low density (LLDPE)	Polyethene or poly(methylene)	HD: 2 LLD & LD: 4	2628	36	64 (LD and LLD) 52 (HDPE)	17 14	Plastic bags, storage containers
Polypropylene (PP)	Poly(1-methylethylene)	5	1533	21	68	18	Rope, bottle caps, gear, strapping
Polyvinylchloride (PVC)	Poly(1-chloroethylene)	3	876	12	38	10	Pipe, film, containers (69% used in building & construction)
Polystyrene: expanded (EPS), extruded (XPS)	Poly(1-phenylethene)	6	< 730	< 10	25	7	EPS: cooler boxes, cups, floats XPS: containers
Polyethylene Terephthalate (PET, PETE)	Poly(ethyl benzene-1,4-dicarboxylate)	1	< 730	< 10	33	9	Bottles, strapping
Polyurethane (PUR)	Butane-1,4-diol		< 730	< 10	27	7	Durable foams (insulation, etc.)
Fiber plastics							
Polyester (PES), polyamide (PA), and acrylic (PMMA)	Poly(methyl 2-methylpropenoate (PMMA)		1000	70% PES, 30% PMMA	59	15	Textiles, nets, ropes

[a]Data from Geyer et al. (2017). Estimated total production in 2015 was 382 million metric tons (Mt) of primary (manufactured from virgin materials) plastics (407 Mt including additives). Generated plastic waste in 2015 estimated at 302 Mt

[b]42% used for packaging (mainly PE, PP, and PET). 'Other' plastics (2015) estimated at 16 Mt (4%) and Additives at 25 Mt (9%)

and polyester (PES), polyamide, and acrylic (PMMA) fibers represent 92% of all plastics ever made (Geyer et al. 2017). The total amount of non-fiber plastics (i.e., those typically not used for textiles) was estimated at 7300 Mt, while PMMA fibers contributed 1000 Mt (Table 1). The largest percentages of the total non-fiber and fiber plastics produced were PE (36%) and PES (70%, mainly PET), respectively. Non-fiber plastics were estimated to be 93% polymer resin and 7% additives [e.g., plasticizers, flame retardants, colorants) by mass. The most common types of plastics reported for environmental studies were PE, PP, PS, PVC, and PET (Koelmans et al. 2019a; Rochman et al. 2013b)].

1.2 Micro and nanoplastic pollutants

Increasing plastic demand and global production of a myriad of plastic types and products is having a cumulative effect on plastic pollution, including a rise in the environmental levels of a diverse set of small plastic contaminants (Rochman et al. 2019). Left unchecked, the environmental burdens of N&MPs are expected to rise sharply due to the persistence of plastics and growing size of environmental reservoirs. Consensus on how to define and categorize plastic debris is lacking (Hartmann et al. 2019). A working definition proposed in 2008 is most common, which defined MPs as plastic particles < 5 mm in diameter,

but different upper and lower size limits have been used. Proposed lower limits for MPs fall in the submicron range. Particles with their largest dimension below these limits are considered NPls. The two fractions are discussed below.

1.2.1 Microplastics

Microplastics are categorized as primary or secondary. Those manufactured at the μm scale are considered primary MPs, which includes PE 'microbeads' in cosmetics, acrylic or PES particles in industrial abrasives, and pre-production plastic pellets called 'nurdles.' In contrast, secondary MPs are formed by the continuous environmental degradation of larger plastic items, through processes that affect polymer structure and reactivity (Andrady 2011, 2015; Gigault et al. 2016; Koelmans et al. 2015; Lambert and Wagner 2016; Lehner et al. 2019). The continued weathering/aging of plastics is expected to yield secondary N&MPs. Polymer breakdown can occur by several processes including hydrolysis, photodegradation (ultraviolet [UV] exposure), mechanical abrasion, temperature fluctuations, and biological and chemical degradation [e.g., (Andrady 2011, 2015; Barnes et al. 2009; Browne et al. 2007, 2011; Imhof et al. 2012; Lehner et al. 2019; Thompson et al. 2004)]. Multiple stressors can act simultaneously. For example, prolonged UV exposure can cause brittleness due to changes in polymer structure (e.g., carbonyl formation), which increases mechanical degradation (Mattsson et al. 2015). Some polymers are more susceptible to certain stressors, e.g., PE and PS are more prone to UV breakdown than other plastics (Feldman 1984; Song et al. 2017). Polymer weathering can produce large amounts of secondary MPs: up to 10^6 MP particles/km^2 were found in subtropical gyres (Law et al. 2010; Law and Thompson 2014). Secondary MPs consist of many polymers, while primary MPs typically consist of PE, PS, and PP (Browne et al. 2007).

1.2.2 Nanoplastics

In addition to MPs, the risks of NPls are a growing concern (Alimi et al. 2018; Andrady 2011; Cozar et al. 2014; Jahnke et al. 2017; Lehner et al. 2019; Mattsson et al. 2015; ter Halle et al. 2016). As mentioned above, different definitions of N&MPs have been proposed. With respect to NPl size, upper limits of $\leq$ 1000 nm (Andrady 2015; Browne et al. 2007; Gigault et al. 2018) and $\leq$ 100 nm (Cole and Galloway 2015; Cole et al. 2015; Crawford 2017; da Costa et al. 2016; Koelmans et al. 2015; Ter Halle et al. 2017) and have been proposed. However, the relevance of a 100-nm cutoff has been questioned. Gigault et al. proposed the following definition of NPls: "plastic particles in a size range of 1–1000 nm that can exhibit colloidal behavior and result from degradation of industrial plastics, as manufacturing byproducts or from use of a plastic object" (Bouwmeester et al. 2015). Like nanomaterials, NPls can be generated by both intentional (manufactured) and unintentional (byproducts) processes, but the proposed definition (Gigault et al. 2018) of NPls would apply only to degradation products, not engineered materials (e.g., polystyrene latex beads).

As with nanomaterials, concerns have been raised about NPls in consumer products, and a lack of regulations on their use/presence (Hansen and Baun 2012; Kessler 2011). Nanoplastics are used in paints, coatings, medicines, electronics and research (Hernandez et al. 2017b; Koelmans et al. 2015). And manufacturing processes such as thermal cutting of PS foam and 3D printing are known to release NPls (Stefaniak et al. 2018; Stephens et al. 2013; Zhang et al. 2012). Over time, environmental concentrations of NPls are expected to increase due to their use in a variety of products (Hernandez et al. 2017b), release as by-products during manufacturing (Stephens et al. 2013; Zhang et al. 2012), and the enormous potential for degradation of macroplastic and MP debris (Besseling et al. 2019; Koelmans et al. 2015; Lee et al. 2013; Song et al. 2017). Hypothetically, degradation of 5-mm MP spheres into 100-nm spheres could produce NPl number concentrations 17 orders of magnitude higher than those of MPs (Besseling et al. 2019).

While studies of MPs in freshwater systems are increasingly being reported (Browne et al. 2010, 2011; Dris et al. 2015a, b, 2016; Eerkes-Medrano et al. 2015; Rillig et al. 2017a, b), environmental studies of NPls are lacking (Alimi et al. 2018; Besseling et al. 2019; Chae and An 2017; Hernandez et al. 2017b; Koelmans et al. 2015; Lehner et al. 2019). No studies of NPls in aquatic or soil systems were reported in recent reviews (Alimi et al. 2018; Lehner et al. 2019). A lack of studies on environmental NPls mainly relates to

difficulties in their recovery and detection in complex matrices (Chae and An 2017; Koelmans et al. 2015; Nguyen et al. 2019). Although data on environmental NPl loads are not yet available, their presence has been widely hypothesized (Chae and An 2017; da Costa et al. 2016; Horton et al. 2017b; Kessler 2011; Mattsson et al. 2015), and several laboratory studies indicate significant environmental releases (Da Costa et al. 2018; Hernandez et al. 2017b; Koelmans et al. 2015). For example, Hernandez et al. (2017b) reported that consumer products could be an important source of NPls in water and soil. Microbeads (PE) were used in the products tested (three facial scrubs) but no NPls were added. Degradation of MPs by high-sheer mixing during product manufacture was hypothesized as the source of the NPls, and/or a broad size distribution of the added bulk MP material (Hernandez et al. 2017b). Analyses confirmed large amounts of PE NPls: at least 300 billion NPls/g.

Products containing microbeads (5- to 1-mm diameter) have been banned in the United States, Canada, and the European Union (Burton 2015; Rochman et al. 2015), but they are widely available elsewhere. Given the continued use, their impact may persist for some time (Burton 2015). The finding of NPls in products with added microbeads (Hernandez et al. 2017b) is noteworthy as it demonstrates, in general, the possibility of NPls in products manufactured by processes that degrade plastics. Other laboratory studies have demonstrated NPl formation by different mechanisms. For example, a study of PS (cup lid) weathering in a simulated marine environment demonstrated NPl and MP (30–2000 nm) releases (Lambert and Wagner 2016). Another possible source may be marine species that ingest MPs. For example, evidence of digestive formation of "NPls" (using the < 1 μm definition) from MPs (31.5 μm) by a planktonic crustacean has been reported (Dawson et al. 2018). If significant uptake by biota occurs (Hermsen et al. 2018), biota may have a major impact on N&MP transport and transformation. The role of ingestion-migration-egestion in the plastic debris burden is unknown. Ultimately, direct releases and degradation of many types of plastics produces diverse N&MP with different environmental impacts. Although environmental levels are currently unknown, NPls may be as pervasive as MPs and larger plastic particles (Alimi et al. 2018). Some studies indicate that the average size of environmental MPs appears to be decreasing (Rocha-Santos and Duarte 2015). The concentrations of small MPs and NPls are expected to increase, which may pose unique risks due to their small size.

1.2.3 Potential risks

Reviews of the ecotoxicological effects of MPs suggest potential for adverse effects in a wide range of aquatic biota (Anbumani and Kakkar 2018; Wang et al. 2019). Fang et al. (2018) examined benthic organisms in Arctic and sub-Arctic regions and reported MP sizes from 0.10 to 1.50 mm, and that fibers were dominant (87%) in trophic transfer. Microplastic ingestion by riverine macroinvertebrates also has been reported (Windsor et al. 2019). Prata et al. (2019) reviewed the impacts of MPs on microalgae, reporting adverse effects at low parts per million (ppm), but that effects were reversible, and no harm is expected at current environmental levels. Besseling et al. (2019) concluded that organisms in near-shore surface waters could be at risk, and that increasing N&MP concentrations may eventually pose more widespread risks, especially for small particles. Most of the laboratory studies have tested concentrations that greatly exceed environmental levels (de Sa et al. 2018; Triebskorn et al. 2018).

Another concern over the past decade is the potential of N&MPs to serve as vectors for associated toxins [e.g., (Curren and Leong 2019; Teuten et al. 2007, 2009; Vethaak and Leslie 2016)]. Ingested plastic has the potential to release associated contaminants into tissues. In contrast, sorption of pollutants also could reduce their bioavailability (Beckingham and Ghosh 2017). In particular, the presence of N&MPs in polluted waters (e.g., urban rivers, wastewater) could result in substantial sequestering of toxins (organics, metals, and/or pathogens), which may be more extensive with NPls due to their high affinity for organic pollutants and high surface area (Besseling et al. 2019; Boerger et al. 2010; Chang et al. 2020; Curren and Leong 2019; Eerkes-Medrano et al. 2015; Eriksson and Burton 2003; Koelmans et al. 2016; Li et al. 2018a, 2019; Murray and Cowie 2011; Rochman 2015; Rochman et al. 2013b, c, 2014; Velzeboer et al. 2014; Wang et al. 2017, 2019; Wright and Kelly 2017; Ziajahromi et al. 2016). Further, their small size allows them to reach other tissues more readily, possibly crossing the blood–brain barrier

(Besseling et al. 2019; Browne et al. 2008; Koelmans et al. 2015). However, in most environments, chemical uptake from N&MPs is negligible relative to other routes (Beckingham and Ghosh 2017; Horton et al. 2017b; Koelmans et al. 2016; Rehse et al. 2018). Nevertheless, in 'hotspot' locations, or if N&MPs concentrations increase significantly, increased bioaccumulation is anticipated (Besseling et al. 2019; Chen et al. 2018; Diepens and Koelmans 2018).

Ingested N&MPs have potential for harm to a wide variety of species due to both direct particle effects (e.g., inflammation) and uptake of associated toxins, and it is important to distinguish the two (Besseling et al. 2017; Koelmans et al. 2017). Inhalation/ingestion of N&MPs may result in inflammation, chemical leaching, and/or particle translocation to other tissues, depending on particle size, shape, and composition. Small particles may accumulate in tissues and release monomers, additives, and sequestered pollutants (e.g., heavy metals, organic pollutants) [e.g., (Chang et al. 2020; Koelmans et al. 2016; Li et al. 2019; Rochman et al. 2014; Wang et al. 2017; Wright and Kelly 2017; Ziajahromi et al. 2016)]. The potential impact of particle shape (fibrous vs. nonfibrous) and size on toxicity is not yet known (Koelmans et al. 2019b). The extent of N&MP contamination in food and beverages and the potential human health implications also are uncertain. These research gaps are discussed in Sect. 4.1.1.

2 Sample collection and preparation

High variability in the MP counts for environmental samples has been observed, likely related to the different locations studied and variety of methods used to collect, process, classify (type, size, number), and identify N&MP particles. Of the studies published in 2016–2017, 43% were reportedly based on visual observation only (unaided and/or under a dissecting microscope) (Burns and Boxall 2018). This approach is relatively simple and low cost, but it has a lower size limit of about 500 μm (Loder and Gerdts 2015) and is subject to interference by natural materials such as cotton and other cellulose fibers, coal/coal fly ash (Eriksen et al. 2013), quartz and calcium carbonate (Ballent et al. 2016), and organic compounds (Ziajahromi et al. 2017). Studies based on visual inspection, without confirmation by an analytical technique, likely overestimated concentrations (Lusher et al. 2017), especially of synthetic fibers (Fischer et al. 2016). Burns and Boxall (2018) reported error rates from 33 to 70%, based on estimates for ten studies (Ballent et al. 2016; Clunies-Ross et al. 2016; Dekiff et al. 2014; Fischer et al. 2016; Hidalgo-Ruz et al. 2012; Horton et al. 2017b; Imhof et al. 2017; Kanhai et al. 2017; Lenz et al. 2015; Lusher 2015).

Standardized methods are key to tracking N&MPs in the environment. Requirements depend on particle properties and sample matrix. Given the pervasiveness of microfibers, steps must be taken to avoid contamination [e.g., (Conley et al. 2019; Dris et al. 2016; Foekema et al. 2013; Talvitie et al. 2015)]. Examples include wearing cotton clothing, rinsing equipment with filtered DI water, using a laminar flow hood, and procedural blanks. Filters should be checked for contamination before use and stored covered. An overview of collection and preparation methods for samples impacted by urban/industrial activities is provided below (Sects. 2.1–2.2). The discussion applies mainly to MPs as reliable methods for NPls in environmental samples are lacking (Lehner et al. 2019). Detailed information on monitoring methods can be found in several reviews [e.g., (Prata et al. 2019; Renner et al. 2018; Schwaferts et al. 2019; Strungaru et al. 2019; Sun et al. 2019; Zarfl 2019)], and in a special issue on this topic (Barcelo and Knepper 2019).

2.1 Sample collection

Sample collection by neuston, manta, and plankton nets (300 or 333-μm) has been widely applied to marine waters. These methods are often not optimal for inland freshwaters [e.g., (Bordós et al. 2019)], subject to particle loss in the lower μm range, and not suitable for WWTPs and other sites in urban watersheds. As reported in Sect. 4, other methods (Table 4) have been used in surveys of WWTPs. Given the anticipated low concentrations (e.g., 1 MP/L in influents of U.S. plants (Carr et al. 2016), large volumes are usually required. And sampling has typically involved a filtration technique due to the complex matrices. For example, Talvitie et al. (2015) used a custom device to capture different particulate size fractions. A pump sampled water from a wastewater stream into the sampler, which consisted of three, clear plastic tubes (60-mm diameter) with screw-on

connectors. Up to three filters cut from plankton nets (200-, 100-, and 20-μm mesh openings) could be used. Filters were inserted at the tube connections and the tubes were screwed tightly together. Wastewater (1 L/min) entering the top of the device passed through the filters (200–20 μm). Volume depended on the number of filters used and sampling location, from 0.3 L for influent to 285 L for effluent. Ziajahromi et al. (2017) also reported a stacked-filter sampler with removable screens (500, 190, 100 and 25 μm stainless steel; 12-cm diameter). It allowed continuous sampling of large wastewater volumes. Sampler efficiency was determined using duplicates of spiked tap water (60 to > 500 μm PS particles) and four size fractions (60–125, 125–250, 250–500 and > 500 μm). Recoveries ranged from 92% (25-μm screen) to 99% (500-μm screen).

2.2 Sample preparation

Prior to analysis, samples from urban watersheds/freshwaters often undergo a two-step procedure to isolate plastic particles from other solids (e.g., wood, sand) and remove any associated organic matter (e.g., biofilms, NOM). Particle isolation has typically been by density separation in saturated salt solutions, wherein particles rise to the surface of the separation medium, passively or with elutriation, based on density. Digestion of organic matter has typically been by (bio)chemical treatments. For density separation, a saturated sodium chloride (NaCl) solution (1.202 g/cm^3) has been used most often as it is inexpensive and non-toxic (Hanvey et al. 2017; Prata et al. 2019; Renner et al. 2018; Van Cauwenberghe et al. 2015). Repeat extractions can improve recovery (Crawford 2017; Nuelle et al. 2014). For example, using a saturated NaCl solution to extract MPs (PE) in sediment, recoveries of 61%, 83%, and 93% were reported for the first, second, and third extractions, respectively (Crawford 2017). Pretreatments to remove organic matter include protocols based on H_2O_2, acids and bases (HNO_3, HCl, NaOH, KOH), and enzymes (Prata et al. 2019; Renner et al. 2018). A key criterion is removal without degradation/loss of plastic particles (Bergmann et al. 2015; Renner et al. 2018). Harsh treatments such as strong acids and bases, and ultrasonic baths, can degrade MPs (e.g., Renner et al. 2018). A wet peroxide oxidation procedure (e.g., 30% H_2O_2 with 0.05 M Fe [II]) removed organic matter with little or no impact (e.g., Eriksen et al. 2013; Tagg et al. 2015).

Density separation and digestion protocols for water and sediment/sludge samples have been reported in numerous publications (e.g., Bayo et al. 2020; Bayo et al. 2016; Crichton et al. 2017; Dehaut et al. 2016; Karami et al. 2017b; Prata et al. 2019; Renner et al. 2018; Song et al. 2015; Sun et al. 2019; Tagg et al. 2015; Wagner et al. 2017). Two reviews (Prata et al. 2019; Renner et al. 2018) reported frequencies of use for collection, separation, digestion, and identification methods. (Renner et al. 2018) reported that a 30% H_2O_2 solution was the most commonly used (41%) digestion method for N&MPs, and NaCl was the most common (53%) salt for density separations, based on 53 (digestion) and 67 (density separation) published papers. Based on 20 articles, Prata et al. (2019) also reported H_2O_2 and NaCl as the most common reagents for digestion and density separation (respectively): 30% H_2O_2 was used for digestion of 25% and 35% of the water and sediment samples (60% used no digestion), while NaCl was used for 65% and 75% (respectively) of the density separations.

Although a saturated NaCl solution is efficient for isolation of lower-density plastics, including PE (0.917–0.965 g/cm^3), PP (0.85–0.94 g/cm^3) and PS (1.04–1.1 g/cm^3) (Crawford 2017; Zobkov and Esiukova 2017), higher-density plastics such as PET (1.4–1.6 g/cm^3) and PVC (1.3–1.7 g/cm^3) have poor extraction efficiencies. Higher-density salt solutions such as sodium iodide (NaI, 1.8 g/cm^3), zinc chloride ($ZnCl_2$, 1.5–1.7 g/cm^3), and sodium polytungstate (SPT, 1.4 g/cm^3) can significantly improve extraction of higher-density plastics, but these reagents are more expensive, and some are environmentally hazardous. To maximize efficiency and minimize pollution, multiple extractions and recycling heavy salt solutions were recommended (Crawford 2017; Dris et al. 2017). A portable, isolation unit used with a ZnCl solution (1.5 g/cm^3) reportedly extracted MPs from sediments in a one-step procedure, with a mean efficiency of 96% (Coppock et al. 2017).

Fuller and Gautam (2016) developed a pressurized fluid extraction (PFE) method for MPs in municipal waste and soil. Benefits include fast, simple, low-cost extractions, with potential for automation. However, mixed extracts give complex spectra and particle sizes and shapes are altered by the conditions. Gies et al.

(2018) applied an oil extraction procedure (OEP) (Crichton et al. 2017) to wastewater matrices that reportedly improved sludge extraction relative to previous methods. An objective was to avoid sieves, unlike Mahon et al. (2017), to prevent contamination and loss of fibers, which can penetrate sieves based on diameter. Sample volumes of 1 L could be processed (without sieving), with a lower particle cutoff of 1 μm. No harsh reagents/high temperatures were required, as used previously [e.g., (Carr et al. 2016; Mintenig et al. 2017)], and a sample mass of 5 g gave good precision. A method described by Talvitie et al. (2017a) also excluded chemical treatment but sample sizes were smaller (0.2–1 g). Depending on the type of sludge and treatment process, MP levels as low as 4 MP/g and as high as 187 MP/g have been reported (Lares et al. 2018; Mahon et al. 2017; Murphy et al. 2016; Talvitie et al. 2017a). Thus, sample volumes may need to be adjusted to ensure extraction of an adequate number of MPs. Based on the sludge masses examined (2.5, 5, and 10 g), the authors concluded that a 5-g sample should provide sufficient accuracy, even at relatively low MP concentrations (Gies et al. 2018).

Dehaut et al. (2016) recommended a KOH method, but peroxide was not tested because the objective was digestion of seafood tissue. Of the methods and plastics (15 common types) tested, five of six protocols showed plastic degradation and/or insufficient tissue digestion, while KOH (10% KOH solution, 24 h, 60 °C) gave efficient digestion without degradation (except cellulose acetate). The authors recommended the method for environmental and seafood quality studies. However, Fenton's reagent was later reported as superior to KOH, NaOH, and H2O2 (Hurley et al. 2018; Tagg et al. 2017), and its use was recommended for organic-rich, complex samples, in conjunction with density separation (Hurley et al. 2018). Temperature, pH, and the ratio of peroxide to ferrous ions were key factors in removal of organic matter. Simon et al. (2018) adjusted the pH to 3 to avoid formation of ferric or ferrous ion complexes, which lowers free iron (II) and slows the reaction. Temperature control (15–30 °C) is essential as the oxidation is exothermic (mixture can boil violently), and a yellow substance precipitated below about 15 °C.

A method using 96% ethanol (EtOH) was reported for isolating N&MPs from vegetal-rich samples (Herrera et al. 2018). Biomaterials such as algal and plant components have lower densities than most MPs and floated on the EtOH surface. Only very low-density particles such as EPS and XPS remained at the surface. After separation, the supernatant was removed and the remaining sample was filtered to recover the plastics. Six plastics (PP, PE, PVC, PUR, PET [fiber], and PS) were tested using five digestion protocols: HCl (3%), NaOH (40%), NaOH (4% in 0.5% surfactant), KOH (10%), and H_2O_2 (30% with 0.05 M iron [Fe(II)] catalyst). The EtOH density separation removed biological matter more efficiently than the digestion methods. The method was reported to be simple, safe, and inexpensive, and it was suggested as a standard protocol for environmental samples (Herrera et al. 2018). No degradation was reported, but larger (mm scale) items (pieces or pellets) were tested. Other plastic types and/or smaller particles could potentially be damaged.

3 Plastic debris in urban watersheds

Land-based sources were estimated to account for up to 80% of the annual plastic released to oceans (12.2 Mt/year), with primary MPs (0.5–1.4 Mt/year) accounting for about 9% of the land-based input (5.4–15.2 Mt/year) (Sherrington 2016). Approximately 94% of the current stock (total) of marine plastic pollution is at the seabed (70 kg/km^2 on average), while about 1% is at the surface (18 kg/km^2), with an average global surface concentration just under 1 kg/km^2. Surface concentrations are higher at specific mid-ocean points, the highest being found in the North Pacific Gyre (18 kg/km^2). Beach contamination is much higher, with an estimated global average of 2000 kg/km^2 (Sherrington 2016).

Land-based sources of N&MPs, especially urban watersheds, also contaminate lakes and rivers globally. The atmosphere and soil are important sources in freshwaters, but relatively little is known about the mass fluxes between these compartments, including the extent to which flooding impacts transport. Studies of freshwater and terrestrial systems in urban/industrialized areas are especially relevant because of their proximity to plastic sources and initial transport media. Major MP sources in urban environments include microfibers from synthetic textiles and tire wear. Wastewater treatment plants (WWTPs) can effectively remove MPs in influents (e.g., 87–99% Rezania et al. 2018), but they release N&MPs to

aquatic and terrestrial ecosystems due to the high discharge volumes, and through application of contaminated sludge on agricultural land (e.g., Bayo et al. 2016; Horton et al. 2017b; Magni et al. 2019; Rillig et al. 2017a, b).

3.1 Microplastic sources and transport

Microplastic particles can be released directly to the environment, and the initial transport medium depends on the source. Major MP sources identified on a global scale (Fig. 1) are: synthetic textiles (34%), tire wear (29%), city dust (24%), road markings/dust (7%), marine coatings (4%), microbeads (2%), and plastic pellets (0.3%) (Boucher and Friot 2017). The category 'city dust' is a composite of common sources in urban environments, including MPs generated through abrasion, weathering, and pouring. Example sources include MPs from of synthetic footwear soles, plastic utensils, infrastructure (e.g., household dust, artificial turf, building coatings), abrasive blasting, and pouring powders.

The many sources of MPs in urban watersheds contaminate household sewage, solid waste, storm water, wastewater and sewage sludge, agricultural runoff (e.g., from sludge-treated soils), industrial effluents, road drainage ditches, and landfill leachates (Bayo et al. 2016; Boucher and Friot 2017; Hoellein et al. 2014; Horton et al. 2017a, b; Murphy 2017). Plastic pollution (aquatic and soil) also occurs through accidental spills (e.g., transportation accidents, leakages) and by intentional dumping. Spills of MP pellets (nurdles) at industrial facilities can enter drains and waterways, and eventually marine environments (Entwistle 2018). The relative contribution of tire wear particles to the total amount of all plastics reaching oceans was estimated at 5–10% (Kole et al. 2017). Their role as sorbents for organic compounds has not been sufficiently investigated but their main components (polymeric rubber and carbon black) favor sorption (Huffer et al. 2017). However, particle properties are expected to change under environmental conditions. Tire wear also contributes to air pollution globally: 3–7% of the particulate matter (PM2.5) in air was attributed to tire wear. As with other MPs, debris from tires (Kole et al. 2017) can enter the food chain, but air pollution is likely the main exposure route.

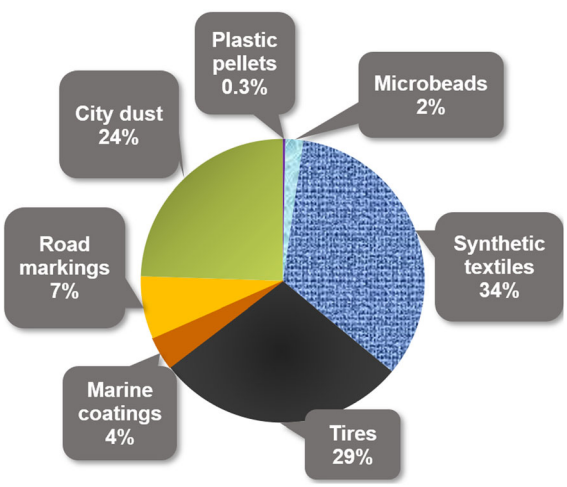

Fig. 1 Contribution of various sources to microplastic pollutants (Boucher and Friot 2017)

Synthetic textile fibers are a major contributor to secondary MPs in aquatic and terrestrial systems (Boucher and Friot 2017; Browne et al. 2011; Carney Almroth et al. 2018; Cesa et al. 2017; Dris et al. 2015a, b, 2016, 2017; Dubaish and Liebezeit 2013; Hartline et al. 2016; Hernandez et al. 2017a; Lin et al. 2018; Napper and Thompson 2016). These microfibers originate from wastewater, especially in highly populated/urbanized areas; diverse non-point sources; and other aquatic and terrestrial systems. Commercial and domestic laundry are major sources (Boucher and Friot 2017; Browne et al. 2011; Cesa et al. 2017; Hartline et al. 2016). Items made of PMMA, spandex, and nylon shed as many as 700,000 MP fibers/kg of laundry (Napper and Thompson 2016) from households and industrial facilities. Smaller amounts (100–300 fibers/L) in washing machine effluents have been reported (Browne et al. 2011). Hernandez et al. (2017a) reported that detergent use had the greatest impact on fiber release of the variables tested: 0.025 and 0.1 mg fibers/g textile, without and with detergent, respectively. Neither the fabric type nor wash conditions affected the overall fiber length distribution, generally 100–800 µm. In another study, PES fleece had the highest fiber loss relative to acrylic and nylon fabrics (Carney Almroth et al. 2018). Based on their sources and relatively small size, aqueous transport of microfibers mainly occurs through wastewater effluents and sludge applications, as with primary MPs (Carr 2017; Horton et al. 2017b). Fiber dispersion and transport in air, with subsequent deposition on land and water also occur (Allen et al.

2019; Dris et al. 2015a, b, 2016, 2017; Gasperi et al. 2018).

Directly released MPs (primary and secondary), including microfibers, tire debris, pellets and microbeads in wastewater, may undergo further degradation in the environment. Information on how rapidly MPs are formed through breakdown of plastic products is limited. Data for several plastics were reported in a recent review (Fotopoulou and Karapanagioti 2017). Biodegradation of some polymers has been reported, but the extent to which microbes degrade N&MPs in different environmental matrices is unknown (Koelmans et al. 2019b). However, microfiber pollution from synthetic textiles may pose a greater problem than degradation of larger plastic items at sea (Boucher and Friot 2017; Henry et al. 2019). According to an IUCN report, between 15 and 31% of all plastics released to oceans (estimated at 4.8–12.7 Mt/year) originate as MPs released directly from households and industrial products, with 35% due to washing of synthetic textiles (Boucher and Friot 2017). Europe and Central Asia alone had annual MP releases into oceans equivalent to the addition of 54 light (e.g., 5 g) grocery plastic bags/person/year. In Europe and North America, regions with adequate waste management, direct release of MPs equaled or exceeded contributions of secondary MPs from mismanaged waste (Boucher and Friot 2017).

Rivers can transport plastic debris over long distances, eventually reaching oceans, and many factors affect transport and availability of N&MPs in aquatic systems. During transport, particles can associate with naturally occurring materials such as refractory natural organic matter (NOM), adhere to and be ingested by a variety of species/organisms, and settle in sediments. Environmental transport and fate depend on particle properties such as polymer type (e.g., hydrophobic, hydrophilic), charge, and size. Transport further depends on particle transformations (oxidation, biofilm growth (Oberbeckmann et al. 2015), adsorbed compounds), which alter their surface chemistry, buoyancy, and aggregation state (e.g., Saavedra et al. 2019). Because N&MPs can have widely different properties due to many particle-specific and environmental factors, is not possible to generalize their transport behavior in aquatic systems.

Air transport of N&MPs (e.g., fibers from clothes dryers and dust dispersion) also occurs, contaminating land and water bodies as atmospheric fallout (Allen et al. 2019; Cai et al. 2017; Dris et al. 2015b, 2016, 2017; Gasperi et al. 2018). Deposition may be driven by wind and precipitation events (Allen et al. 2019; Dris et al. 2017). Data on MP aerosols are currently limited, but several studies have been reported. Dris et al. (2017) investigated fiber contaminants in indoor and outdoor air, finding concentrations of 1.0–60.0 and 0.3–1.5 fibers/m^3, respectively. Indoor deposition rates ranged from 1586 to 11,130 fibers/day/m^2 surface (190–670 fibers/mg settled dust). Most (67%) were natural fibers (mainly cellulose); the remainder (33%) were synthetic, with PP being dominant. Studies of atmospheric fallout in two major cities also were reported. Dris et al. (2015b) found 29–280 particles/m^2-day in total atmospheric fallout (wet and dry deposition) collected on a rooftop in a dense urban area near (11 km from) the center of Paris (France). Most (> 90%) of the MPs were fibers: approximately 50% were > 1000 µm in length, and the remainder were equally distributed in two classes: 100–500 and 501–1000-µm. Cai et al. (2017) reported fiber and non-fiber MPs ranging from 175 to 313 particles/m^2-day in atmospheric fallout in Dongguan (China). Fiber lengths were mainly in the 200–700 µm range. Diverse shapes (e.g., fiber, foam, fragment, film) were found but fibers were dominant. Three polymers types were identified (PE, PP, and PS), and SEM images revealed adhered particles and evidence of particle degradation (e.g., grooves, pits, fractures).

In contrast to cities, where microfibers were dominant, fragments were the primary shape (followed by films and fibers) in a remote mountain catchment (French Pyrenees) (Allen et al. 2019). Contamination was attributed to aerosol deposition (wet and dry) after long-range air transport from populated areas. The average daily MP deposition was 365 items/m^2 catchment surface, with daily counts (items/m^2) of 249 fragments, 73 films, and 44 fibers. Considering only fibers, daily deposition was 36 (± 18) and 28 (± 13)/m^2. The predominant polymer was PS (as fragments), closely followed by PE, both used in many single-use plastic items and packaging materials (Allen et al. 2019). The dominance of these two polymers in a remote area may relate to the relatively low densities of PS foam and PE particles (Allen et al. 2019). The relatively high abundance of PS also may relate to its high susceptibility to mechanical and UV degradation, especially expanded PS (Song et al. 2017).

3.2 Freshwater systems

Most studies of N&MPs have focused on the marine environment (Lambert and Wagner 2018), but studies of freshwater systems are increasing. Abundances of MPs in some freshwaters were reportedly comparable to those in marine waters (Peng et al. 2017) but results have been highly variable (Klein et al. 2018; Rezania et al. 2018) from as few as 10^{-2} pieces m^{-3} (10^{-5}/L) to as many as many as 100 million m^{-3} (10^{5}/L) (Koelmans et al. 2019a). As discussed, variability likely relates to differences in monitoring methods, location (e.g., proximity to urban areas/WWTPs), and natural conditions (e.g., river catchment features, weather) (Eerkes-Medrano et al. 2015; Klein et al. 2018).

Freshwater systems can be sources (e.g., contaminated rivers) and sinks (lakes, sewage sludge) of N&MPs. Particle types and their relative proportions and properties may differ from those for marine waters, depending on matrix, weathering, and other conditions (Klein et al. 2018). For example, MPs in sewage sludge may be larger and contaminated by organic material, while those in clean waters may be relatively small and difficult to see without a microscope (Li et al. 2018b; Tyree and Morrison 2017). Plastic debris has been found in a range of freshwater systems: e.g., 1 MP per 8 gallons (0.03 per/L) of Great Lakes tributary water (Baldwin et al. 2016), an average of 1285 MPs/ft^2 of Saint Lawrence River (Canada) sediment (Castañeda et al. 2014), 112,000 MPs/$mile^2$ of Great Lakes (USA) water (Eriksen et al. 2013), and MPs in 12% of freshwater fish (Sanchez et al. 2014). Sites near highly populated and urbanized areas can have higher abundances (e.g., Eriksen et al. 2013; Peters and Bratton 2016).

Studies of MP contamination in freshwater systems and drinking water have found high variability in the detected concentrations (e.g., Horton et al. 2017b; Koelmans et al. 2019a; Li et al. 2018c; Novotna et al. 2019; Pivokonsky et al. 2018; Triebskorn et al. 2019). Li et al. (2018b) reported surface water concentrations from 10^{-5} to 10^{5} pieces/m^3 and sediment levels of 4–40 x10^4 pieces/m^3 (40–400 pieces/L) for freshwater systems. Koelmans et al. (2019a) reported MP abundances from 10^{-2} to 10^{8} MPs/m^3 in a review of 50 freshwater and drinking water studies. Novotna et al. (2019) reviewed available information on MPs in drinking water sources and potable water. Results of the different studies varied significantly, from zero or few (< 10) to > 4000 MPs/L (Bordós et al. 2019; Luo et al. 2019; Mintenig et al. 2019; Pivokonsky et al. 2018; Su et al. 2018; Wang et al. 2017, 2018). Studies of freshwater systems near urban/populated areas impacted by industrial effluents and WWTPs are summarized in Tables 2 and 3. Results for water are reported in Table 2, while those for sediments are listed in Table 3. For comparison, several studies of drinking water and relatively clean water sources are included (Table 2).

Few studies of water supplies to drinking water treatment plants (DWTPs) have been reported. Pivokonsky et al. (2018) investigated MPs in raw and treated water at three DWTPs with different water sources. Average concentrations ranged from 1473 ± 34 to 3605 ± 497 MPs/L raw water and 338 ± 76 to 628 ± 28 particles/L in treated water. Their study was reportedly one of very few that determined MPs down to 1 µm. Particles < 10 µm accounted for up to 95% of the MPs in both raw and treated water. Fragments were dominant at two plants, while fibers and fragments were most abundant at the third plant. Twelve materials were identified, but the majority (> 70%) of MPs were PET, PP, and PE. Mintenig et al. (2019) investigated a drinking water system with a purified groundwater source. To assess possible MP sources, samples were collected at different locations, from the groundwater supply to household taps. Fourier transform infrared (FTIR) microscopy coupled with a focal plane array (FPA) detector (FTIR-FPA) was used for identification of MPs down to about 20 µm (Loder and Gerdts 2015; Mintenig et al. 2017). The authors reported concentrations from 0 to 7 MPs/m^3, with an overall mean of 0.7 MPs/m^3. No concentration differences were found for different treatment stages, and all detected MPs were fragments (50–150 µm), identified as PE, PA, PES, PVC or epoxy resin. Four of these polymers were used in the system: tanks in the DWTPs were coated with epoxy resin, pipes (in the plants/households) were often PVC or PE, and fittings were usually PA. The MPs were attributed to abrasion of plastic equipment used to purify or transport water (Mintenig et al. 2019).

The reported widespread occurrence of MPs in drinking water has raised concerns for human health, though the reliability of these findings has been questioned due to method limitations. Koelmans

Table 2 Studies of microplastics in freshwater systems: water

Study	Sample type(s)	Collection and particle cut size	Sample preparation	Analysis	MP abundances and types		Comments
					Reported units	Converted[a] units and/or MP types	
Bordós et al. (2019) Hungary Carpathian basin	Surface water of fishponds, rivers and reservoirs	1500 L pumped through connected filters, 2 mm–100 µm	Density separation (1.2 g/mL NaCl) in MPSS unit (Imhof et al. 2012). WPO (30% H_2O_2, 1 h, 80 °C, 450 rpm). Filtered (0.2-µm Al_2O_3 filter)	Visual (microscope) µ-ATR-FTIR. Suspect MPs auto analyzed	3.52–32.05 MPs/m^3 (92% of samples) Mean = 13.79 ± 9.26	0.003–0.03 MP/L Mean = 0.01 PP, PE dominant	Fishponds may be MP deposition area
Di et al. (2019) China Danjiangkou Reservoir (DJKR)	Surface water at 20 sites (drinking water reservoir). Sediments (Table 3)	See Di and Wang (2018). Pumped and sieved. Smallest size 48 µm	Digested in 30% H_2O_2 (24 h). Filtered (0.45-µm glass-fiber filter). Dried filter in Petri dish	Visual (microscope) µ-Raman (140 suspect MPs) and SEM. Confirmed 118/140	Mean (DJKR) = 2594 ± 3875 par/m^3 Range: 467–15,017	Mean = 2.6 par/L (0.5–15) PP (45%), PS (35%), PE (20%). Fibers dominant	Blue dominant (17–85%), then clear Most MPs < 2 mm
Dikareva and Simon (2019) New Zealand (Auckland) Small streams	Water, sediments (Table 3). Streams spanned urbanization gradient	Plankton net (83 mm diameter, 63 µm mesh) 190–4520 L	Rinsed filtered (63 µm) material into beaker, covered with foil, dried (60 °C). Digested (75 °C) in 30% H_2O_2 and 0.05 M Fe(II), H_2SO_4 solution, with repeat additions of H_2O_2 NaCl (1.2 g/mL) density separation. Supernatant filtered (1.2-µm glass-fiber filter). Filter placed in petri dish to dry (60 °C) before analysis	Visual (microscope) 3309 suspect MPs ATR-FTIR on 50. Library search: 10% of suspect MPs were non-MPs (counts adjusted). Mean blank = 16 MPs (correction made)	17–303 items/m^3 Some polymers (e.g., polyamide) may be underestimated due to degradation (digest > 70 °C)	Up to 0.30 par/L Fragments (39%), fibers (34%), and small particles (63–500 µm) dominant. Films also found 70% of particles < 500 µm were fragments	High variability across streams. Local factors likely more important than catchment-scale. No relation between abundance and population or combined stormwater overflows Polymers varied and often not predictable based on size, shape, or color
Hitchcock and Mitrovic (2019) Australia Estuaries	Water from 3 estuaries with different levels of human impact	Plankton nets (45 and 37-µm) Flow meter Stored in 250-mL PE containers in 50% ethanol	Sieved (20 µm). NaCl density separation per Claessens et al. (2013) Digested 12 h in 30% KOH:NaClO solution. Stained with Nile Red	Fluorescence microscopy Raman on particle subset of photo bleached samples	Means (MP/m^3) 98, 246, and 1032	Means (MP/L) 0.098, 0.246, and 1.032 Fragments (< 200 µm) dominant	MPs higher in areas with more human impact (number of townships and WWTPs, and heavy industry). Procedural blanks < 3% of mean MP counts (3.2 particles/blank)

Table 2 continued

Study	Sample type(s)	Collection and particle cut size	Sample preparation	Analysis	MP abundances and types		Comments
					Reported units	Converted[a] units and/or MP types	
Liu et al. (2019) Denmark 7 retention ponds	Water from retention ponds receiving urban and highway stormwater: 3 with residential catchments, 2 industrial, 1 commercial, and 1 highway Sampled Sept. to Oct. 2017	Pumped through filter device with steel mesh (10-µm, 100 mm diameter). Sampling hose put in steel cage (2 cm mesh) to exclude large debris. Sampled until filter clogged (201–454 L). Filters from 3 sampling rounds at each pond were pooled as one sample (752–1139 L)	Soaked filters in SDS to dissociate MPs from solids. Digested in 50% H_2O_2 (2 days). Removed solids in ultrasonic bath. Collected in SDS (200 mL) and digested enzymatically (3 days, 50 °C), then by Fenton's reagent and 0.01 M NaOH. Filtered (10 µm) and extracted solids by $ZnCl_2$ (1.7 g/mL) density separation (twice). Filtered, added 50% ethanol, and applied suspension to Zn selenide transmission window (area restricted by compression cell). Applied and dried in 100-µL increments to obtain proper loading for µ-FTIR	µ-FTIR-FPA imaging (128x128 MCT FPA) 10-mm diameter window scanned in transmission mode Image maps analyzed using MPhunter software (auto MP detection for µ-FTIR image datasets) Size fraction: 10–2000 µm	490–22,894 items/m^3 or an estimated 85–1143 µg/m^3	0.49–22.8 items/L Dominant polymers: PP, PVC, PES, PE, and PS Ponds serving highway and residential areas had the lowest levels, while ponds serving areas with industry and commerce had the highest	PVC tended to be in largest size fraction. Smallest MPs were mixes of less common polymers Residential ponds tended to have the largest MPs and comparable levels, sizes, and composition Study shows land-based sources of MPs are significant, and retention ponds act as pollution hotspots that play a role in transport from land to aquatic systems
Luo et al. (2019) China Yangtze Delta	Surface water (n = 3) at 43 sites. Urban creeks and rivers, estuary, coastal	5 L with pail Used air lift pump from boat to collect estuary/coast samples	Nylon filter (20 µm). Washed into bottles containing KOH solution (10% w/v). Incubated 24–48 h (65 °C, 80 rpm. Filtered with same filter. Stored in Petri dish	Visual (microscope) µ-FTIR (32% of suspect MPs). 20 µm smallest size detected	1.8–2.4 par/L in freshwaters. 0.9 par/L in estuary and coastal waters	Fibers were 88% of MPs in city creeks, and 81% and 85% of those in 2 rivers	No blank correction (blanks 2.6% of sample MPs). MPs levels higher in freshwater
Mintenig et al. (2019) Germany DWTP	Groundwater; inlet, outlet, household water. 9 raw (8 m^3) and 15 drinking water (32 m^3) samples	Filtered (3-µm) tap water at 10 L/min and DWTP water at 5 L/min. Filtered up to 1 m^3 raw and 2.5 m^3 drinking water	Removed $CaCO_3$ and iron in dilute HCl. Rinsed with H_2O-ethanol and filtered (3-µm). WPO (35% H_2O_2). Sample enriched on Al_2O_3 filter (0.2-µm) and dried. Density separation of raw water in $ZnCl_2$ (1.6 g/mL) to remove iron oxide	µ-FTIR-FPA. Analyzed entire filter. 20-µm lower limit CaFl windows held filter	0–7 MPs/m^3 14 samples had no MPs. Results blank corrected (n = 4)	0–0.007 MP/L	Filter unit lids contributed styrene acrylonitrile and PP Large volumes collected (low MPs anticipated)

Table 2 continued

Study	Sample type(s)	Collection and particle cut size	Sample preparation	Analysis	MP abundances and types		Comments
					Reported units	Converted[a] units and/or MP types	
Triebskorn et al. (2019) Germany Elbe River	River water	µ-sieve cascade (5, 20, 100 µm) coupled to submersible pump at 0.5-m depth	Filtered 250–450 L. Extracted in ultrasonic bath, treated with ozone, centrifuged. Density separation in Na polytungstate solution. Filtered on PTFE filter	Visual (microscope) µ-Raman to 4 µm Laser counter for total particles, 1–200 µm	Range (par/m^3): 10^5–9 × 10^5 Total particles = 10^{11}/m^3	100–900 par/L PE (96.5%), PS (1.8%), PP (0.7%), PA (0.5%)	MP loads: 4–30 kg/d ≤ 10 of the 10^6 total particles were plastic
Wiggin and Holland (2019) United States Urban estuary in Long Beach, CA	Water from Los Angeles and San Gabriel rivers, and Long Beach Harbor	Triplicate 20-L grab samples (33 total) using pump and 4-L amber glass jars	4 stainless sieves (500, 124, 63, and 20 µm). Filtered water with 3-µm PC filter (required 3–12 filters). Sieved material rinsed into filter (3-µm PC) units and digested in 15% H_2O_2 (50 °C, 16 h). Rinsed filter units with 20% IPA. Dried filters (covered) Validated WPO protocol	Visual (microscope) 40×–100× Subset of filters in each size class stained with Nile Red and recounted under fluorescent microscope using FITC filter cube	Means, MP/m^3 Without staining Harbor: 8130 LA River: 13,622 San Gabriel: 4161 With staining (respectively) 13,627; 641,292; 63,359	Means, MP/L: Without staining Harbor: 8 LA River: 14 San Gabriel: 4 With staining (respectively) 14, 641, 63	White and clear non-fibers not counted (too similar to background). Small fragments and beads could not be distinguished Procedural blanks and recovery checks included. Counts corrected for controls Fiber estimates likely low due to inability to stain
Yuan et al. (2019) China Poyang Lake (largest freshwater lake in China)	Surface water, sediments, wild crucians 21 sampling sites. Sampled: 11/2017	2, 20-L water (0–1-m depth) with steel sampler. Sieved (50-µm). Residue rinsed into bottle	Water and sediment processed as described previously (Di and Wang 2018). 30% H_2O_2 overnight. Diluted digest and filtered (0.45-µm) onto gridded filter. Air dried, covered	Visual (microscope) µ-Raman. Cleaned sample with alcohol. Used Ag slide to reduce fluorescence	Range (par/L): 5–34 Highest at mid-lake Most < 0.5 mm	Fibers and colored MPs dominant. PE and PP major types No correlation between water and sediment	Domestic sewage is a possible MP source Human and topographic factors may affect distribution
Di and Wang (2018) China Three Gorges Reservoir Region	Surface water and sediments (Table 3) at 29 sites along Yangtze River Aug. 2016	2, 25-L samples pumped at 1-m depth. Sieved (48-µm), rinsed residue into 50-mL jar. Stored in 5% formalin (4 °C)	30% H_2O_2 for 12 h. Filtered with gridded filter (0.45-µm glass-fiber filter). Dried (50 °C) in Petri dish. Kept covered	Visually (microscope) µ-Raman (174 suspect MPs), SEM	Water (par/m^3): Mean = 4703 ± 2816 Range = 1597–12,611 79.8% < 1 mm PS Total types: 38.5% PS, 29.4% PP, and 21% PE	Water (par/L): Mean = 4.7 Range = 1.6–12.6 Fibers 28.6% to 90.5% PP and PE dominant (PS main type in sediments)	MPs higher in urban water than rural. Clear (21–82%), followed by white, and blue. No correlations between sampling sites MP sorption of organics

Table 2 continued

Study	Sample type(s)	Collection and particle cut size	Sample preparation	Analysis	MP abundances and types		Comments
					Reported units	Converted[a] units and/or MP types	
Gray et al. (2018) United States Charleston, South Carolina	Surface μ-layer water. 6 sites in each estuary: Charleston Harbor and Winyah Bay	4-L collected with 2-mm steel mesh and drained into jars. Each dip gave ~ 75 mL	Filtered with nested sieves (500, 150, and 63 μm) Processed solids as described for sediments	Visual (microscope) Subset of particles (> 500 μm) in sediments by ATR-FTIR	Means (par/L): Harbor = 6.6 ± 1.3 Bay = 30.8 ± 12.1	No correction for blanks or FTIR results	Levels comparable to estuaries worldwide. But harbor had many black MPs attributed to tire wear
Kay et al. (2018) England (North) Rivers near 6 WWTPs	Water upstream and downstream of WWTPs in river catchments with different features	Net (300-μm) on frame (250 × 230 mm). Held frame against riverbed, facing upstream. 15-min sample	Filtered by 6 stainless, stacked sieves (5.6 mm to 250 μm). Discarded material on top sieve. Transferred MPs to petri dish for analysis	Visually (microscope) MPs classified as pellets/beads, fibers, and fragments/flakes	Concentrations (not reported) generally higher downstream. In 4 cases, higher upstream	Mean up-to-downstream concentration ratios > 1. For 19/28 cases, ratios were 1–3	Effluents contributed MPs to rivers. Prevalence of fibers indicates textiles. Source apportionment work needed
Lin et al. (2018) China Pearl River, lower course along Guangzhou City	River water and sediments (Table 3) at 14 sites WWTP samples (Table 4)	60-L surface (top 50 cm) with 5-L pumped sampler (Seaward WS-5). Filtered on-site with steel sieve (20 μm). Fixed residues in formaldehyde	Transferred residue to 1-L flask. Added 200 mL 30% H_2O_2, covered with foil and digested 24 h in incubator shaker (65 °C, 80 rpm). Followed by 24-h NaCl density separation (d = 1.2 g/mL). Filtered (5-μm) and covered with petri dish	Visual (optical microscope) μ-FTIR with MCT detector. Subset of suspect MPs Size focus: 0.02–5 mm	Surface water (items/m^3): 379–7924 items/m^3	0.379–7.924 MPs/L PE and PP accounted for 64.3%	Fibers dominant Noted number, size, shape (fibers, fragments, films, pellets) and color
Pivokonsky et al (2018) Czech Republic, Prague DWTP supplied by groundwater	Raw and treated drinking water Sampled: 11/2017–01/2018	3, 1-L samples over 24 h. Total of 27 L each of raw and treated water for SEM analysis. And 9 L each for FTIR, Raman and elemental analyses	WPO (H_2O_2). Vacuum filtered onto successive PTFE filters (5 and 0.2 μm) for SEM. Filtered onto Al_2O_3 filters for qualitative analyses. Dried filters dried in oven (30 °C for 30 min). Stored in Petri dishes in desiccator until analysis	Qualitative analyses (on 25% of Al_2O_3 filter) by μ-FTIR (> 10-μm) and μ-Raman (1–10 μm). Number, size, shape by SEM (3 pieces of PTFE filter). Elements in some particles by SEM–EDS	Means (par/L): Raw:1473 ± 34 to 3605 ± 49 Treated: 338 ± 76 to 628 ± 28 Up to > 4000 8–83% of particles not MPs	Fragments dominant at 2 plants; fibers and fragments most common at one PET, PP, and PE most common (accounted for > 70% of MPs)	One of few studies to determine MPs to 1 μm MPs < 10 μm most common (up to 95%). Higher in raw water than treated

Table 2 continued

Study	Sample type(s)	Collection and particle cut size	Sample preparation	Analysis	MP abundances and types		Comments
					Reported units	Converted[a] units and/or MP types	
Su et al. (2018) China Yangtze River	Water, sediment (Table 3), and Asian Clams in lakes, rivers, and estuaries	5-L water samples (n = 3) with bucket dipped (0–12 cm deep) from boat	Vacuum filtered water through net (20 μm). Washed residue into flask with 100 ml 30% H_2O_2. Digested using oscillation incubator (65 °C, 80 rpm, ≤ 72 h). Filtered and stored filter in Petri dis	Visual (microscope) μ-ATR-FTIR on 150/1303 particles	0.5–3.1 par/L	Fibers dominant MP sizes ranged from 0.021 to 4.83 mm. Most were 0.25–1 mm	Clear and blue particles most common. Clear dominant 0.4–5.0 par/clam (μ-fibers 60–100%)
W. Wang et al. (2018) China Hong and Dongting lakes	Surface water. Inland freshwater systems. 20 sites at Hong Lake and 30 at Dongting Sampled in Sept. 2017	20 L bulk water (0–20 cm depth) collected with pump at each site. Sieved (50-μm). Rinsed residue into bottle. Preserved in 4% formalin solution	30% H_2O_2 (48 h, room temperature). Filtered with glass-fiber filter (0.45-μm). Placed filter in Petri dish for visual examination	Visual (microscope) SEM, μ-Raman of 50 suspect MPs from each lake	par/m^3 Hong Lake: Mean = 2867 ± 989 Range = 1250–4650 Dongting Lake Range = 900–2800 Mean granules: Hong 685 ± 375 Dongting 385 ± 221	par/L: Hong Lake: Mean = 2.9 ± 1.0 Range = 1.2–4.6 Dongting Lake: Range = < 1–2.8 71–78% of MPs colored. Most < 1 mm; > 20% < 330 μm. PE and PP dominant	Fibers 42–92% of MPs, many transparent. Possible sources: fishing nets, sewage, runoff, air Granules irregular or spherical, and colored. Possible sources: domestic effluents and waste from residents. Films (14–16% of MPs) appeared irregular and weathered
Ravit et al. (2017) United States Raritan and Passaic rivers, NJ	River water, adsorbed compounds, toxicity Sampled: 2016 (May-Aug.)	Manta trawl (333 μm net). Triplicates (n = 45 each location). Rinsed residue into jar. Added IPA to 1 sample. Other 2 shipped on ice	Digested 1 of each replicate by Fenton reaction (1:1 0.05 M iron sulfate + 30% H_2O_2). Rinsed any large organic debris and discarded. Recovery verified with blue microbeads (330 μm) spiked samples	Visual (microscope) Pyr-GC–MS on individual particles. HSSPME-GC-ITMS to identify organics on MPs and in water	28,000 to > 3 million particles/km^2	Not reported	Watersheds are among most developed (residential and industrial) in state; (Raritan largest)

Table 2 continued

Study	Sample type(s)	Collection and particle cut size	Sample preparation	Analysis	MP abundances and types		Comments
					Reported units	Converted[a] units and/or MP types	
Leslie et al. (2017) Netherlands Amsterdam canals Rhine and Meuse Rivers (Germany and Netherlands)	Canal water Riverine suspended particulate matter (SPM) WWTPs (Table 4), biota, sediments (Table 3). Sampled 2012–2013	2-L bulk water in glass bottles, at 6 sites in Amsterdam canals	Density separation in saturated NaCl (1.2 g/mL). Filtered subsample on glass-fiber filter (0.7 µm) for visual inspection Al_2O_3 filter (0.2 µm) used for µ-FTIR analysis of sediment and biota	Visual (microscopy) µ-FTIR on particles in sediment and biota (6% of total counted overall)	Amsterdam canal water (par/L): Mean = 100 (± 49) Range = 48–187 River SPM (par/kg, dry wt.) (SD) Meuse = 1400 (520) Rhine (P2) = 4900 (540) Rhine (P3) = 1700 (390)	Fibers, spheres, and foils Fibers dominant Two size categories: > 300 and < 300 µm 10 µm smallest size detected	MPs present in all urban surface water samples. Canal levels similar to effluent from local WWTPs
Miller et al. (2017) United States Hudson River	River water	Top 8–18 cm surface water, with bucket. Top 1 L decanted into jars	Vacuum filtered onto gridded filters (47-mm, 0.47 µm). Stored filters in metal dishes	Visual (microscopy 45x). Counted fibers with lengths ≥ 100 µm Counts background corrected µ-FTIR	Mean microfibers (MPs//L): 1.24 ± 0.14, with lengths 0.33 to 3.59 mm Mean = 0.98 anthropogenic fibers/L (0.625–2.45 fibers/L)	No significant change in fiber abundance from river source to sea µ-FTIR indicated half of fibers were plastic and half non-plastic but anthropogenic	MP fibers: blue dominant, then black, clear, red, and others. Fibers: 43% cotton, 22% PET, 22% fluoropolymer, 7% PP, 7% nitrocellulose/clay Estimate: 34.4% of Hudson River watershed adds 300 million anthropogenic fibers to Atlantic Ocean/day. No relation between counts and population or WWTP location
Anderson et al. (2017) Canada Lake Winnipeg	Water	Manta trawl, 333-µm net. Collected material preserved in 70% ethanol	30% H_2O_2 250-µm sieve separation substrate	SEM–EDS	Mean: 1.93×10^5 par/km^2 (max: 7.48×10^5)	Converted units not available	23% of particles were non-plastic

Table 2 continued

Study	Sample type(s)	Collection and particle cut size	Sample preparation	Analysis	MP abundances and types		Comments
					Reported units	Converted[a] units and/or MP types	
W. Wang et al. (2017) China (Wuhan) Lakes/rivers	Surface waters of urban lakes and rivers. Residents and industries along shore	20-L (0.2 m depth) pumped through 50-µm sieve. Rinsed residue into 50-mL jar. Preserved in 5% formalin at 4 °C	30% H_2O_2 (24 h, in dark, room temperature). Added distilled water and filtered by glass-fiber filter (0.45 µm). Placed filter in Petri dish for visual (microscope) examination	Visual (microscope), particle subset by SEM, FTIR 44 suspect MPs analyzed	1660 ± 639 to 8925 ± 1591 par/m^3	1.7–8.9 par/L PET and PP dominant >80% of MPs < 2 mm Fibers dominant	Wuhan is largest city in central China. Anthropogenic factors greatly affected MP abundances Colored particles 50.4% to 86.9%; 24.7% were transparent
Estahbanati and Fahrenfeld (2016) United States Raritan River, NJ	River water, up and downstream of 4 WWTPs	Plankton net (0.2 m diameter, 0.51 m long) with 153 µm mesh size Vol. = 1.3–13.5 m^3	30% H_2O_2 + Fe(II). NaCl density separation Sieved into 3 quantitative size ranges (500–2000, 250–500, and 125–250 µm) and 1 semi-quantitative (63–125 µm)	Visual (microscope) Classified primary or secondary based on shape and texture	24 ± 11.4 MPs/m^3 upstream, 71.7 ± 60.2 downstream	0.024 MPs/L upstream, 0.072 downstream	Moderate correlation between MP level and distance downstream from WWTP
McCormick et al. (2016) United States 9 streams in Chicago, IL metro area	Upstream and downstream of tertiary WWTPs Also monitored bacteria, O_2, and nutrients Summer, 2014	Neuston net (333-µm). Adapted marine protocol (see McCormick et al. 2014)	See McCormick et al. 2014	Visual (microscope). 5 categories (Eriksen et al. 2013) Subset of particles (2 streams) analyzed by Pyr-GC–MS	Mean (par/m^3): Upstream = 2.355 (± 0.375) Downstream = 5.733 (± 0.850) Higher downstream in 7 of 9 streams	Mean (par/L): Upstream = 0.0024 (± 0.0004) Downstream = 0.0057 (± 0.0008)	Assuming similar levels over seasons, a minimum of 5.6 million and maximum of 1.7 billion MPs discharged annually Pellets, fibers, fragments dominant. Main polymers were PP, PE, and PS
Baldwin et al. (2016) United States Great Lakes	Water Great Lakes and tributaries	333-µm neuston net 0.2–0.35 m depth	125-µm sieve substrate 30% H_2O_2 + Fe	Visual	Range: 0.05–32 par/m^3 Mean = 4.2 par/m^3	0.00005–0.032 par/L	

Table 2 continued

Study	Sample type(s)	Collection and particle cut size	Sample preparation	Analysis	MP abundances and types		Comments
					Reported units	Converted[a] units and/or MP types	
Fischer et al. (2016) Italy Lakes Chiusi and Bolsena	Water and sediments (2b)	6 Manta trawls (0.3 mm, 60 × 18.5 cm) total	Density separation (Thompson et al. 2004) in NaCl solution (d = 1.2 g/mL) in sedimentation column. Removed supernatant and treated with HCl (48 h) at room temperature, then 1 h at 70 °C. Filtered and applied Nile Red	Covered filters with counting grid. Counted under UV-microscope Small subset (20 fibers) by SEM	Range (par/m^3): Chiusi: 2.68–3.36 Bolsena: 0.82–4.42	Range (par/L) Chiusi: 0.0027–0.0034 Bolsena: 0.0008–0.0044	
Su et al. (2016) China Lake Taihu	Water, benthic sediment (Table 3), Asian Clams 3rd largest lake in China (2250 km^2)	Surface water with 333-µm plankton net, 0.3 m deep. Preserved in 5% methylaldehyde Bulk water (5 L)	30% H_2O_2 WPO. Filtration with 100-µm (net samples) and 5-µm PC filter (bulk water)	Visual (microscope) µ-FTIR or SEM–EDS (113/1805 particles) 81/113 confirmed by µ-FTIR	Bulk water (par/L): 3.4–25.8 par/L Net sample max: 6.8 × 10^6 par/km^2	Fibers dominant (48–84%) Main MPs: cellophane, followed by PET, PES, terephthalic acid, PP	Shallow lake with history of pollution. Most MP-polluted freshwater lake worldwide Blue items prevalent in water samples (50–63% of MPs)
Dris et al. (2015a) France Seine and Marne Rivers, Paris	River water (5 sites) upstream and downstream of Paris	0.1–0.35 m depth 80-µm plankton net and 0–0.3 m depth 330-µm manta trawl	Filtered onto glass-fiber filters (1/6 µm). Only particles > 100 considered	Visual (microscope)	3–108 par/m^3 (plankton net) 0.28–0.47 par/m^3 (manta trawl)	0.03–0.108 par/L 0.0028–0.00047 par/L	Plankton net collected mostly fibers. 48% > 1000 µm. Atmospheric fallout may be source of fibers in surface water
Faure et al. (2015) Switzerland Lakes and rivers	Water: 6 largest Swiss lakes, and rivers. Sediments	Manta trawl (300 µm). 320–430 m^3 surface water	Stored at 4 °C in salt-saturated water	Visual (microscope)	Mean (rivers): 7 par/m^3 Range: 0.10–64	Mean (rives): 0.007 par/L Mostly fragments and foam	Pellets small fraction by number but 32–52% by mass. Also examined adsorbed organics, dissected fish, and water birds
Mani et al. (2015) Germany Rhine River	Water	18-cm sampling depth. 300-µm manta net. Mean filtered = 150 m^3	Stored at 5 °C in 10% NaCl. Sieves (separation substrate) Enzyme + H_2O_2. Density separation in 23% NaCl/H_2O (density 1.16 g/mL)	Visual (microscope) FTIR (118 suspect MPs)	Mean = 892,777 par/km^2 (max: 3.9 × 10^6)	Mean[a] = 0.005 par/L Spherules (45.2%), fragments (37.5%), clear spherules (13.2%), fibers (2.5%) and others (1.1%)	MP levels diverse along and across river. PS (29.7%) dominant, then PP (16.9%), other (13.6%), acrylate (9.3%), PES (5.1%) and PVC (1.7%)

Table 2 continued

Study	Sample type(s)	Collection and particle cut size	Sample preparation	Analysis	MP abundances and types		Comments
					Reported units	Converted[a] units and/or MP types	
Zhang et al. (2015) China, Three Gorges Dam	Water	Trawl, 112-μm net	1.6 mm stainless steel sieve. Liquid allowed to settle 1 week. Large particles removed with tweezer	Visual (microscope) ATR-FTIR on 50–100 suspect MPs	Mean = 8.47×10^6 par/km^2 (max: 1.36×10^7)	–	PE, PP, and PS "hotspot" for MP pollution
McCormick et al. (2014) United States N. Shore Channel, Chicago, IL	Water upstream and downstream of WWTP. Also examined microbial assemblages Sampled on 9/13/2013	2 neuston nets (333-μm), behind stationary boat. 20 min samples (n = 4). Rinsed material into 1-L containers. Shipped on ice. Stored at 4 °C	Sieved and stored 0.330–4.75-mm fraction in oven (75 °C) until digestion by WPO (0.05 mol/L Fe(II) + 30% H_2O_2 at ≤ 75 °C). Density separation in NaCl (6 M). Filtered on glass-fiber filter (0.7-μm pore size)	Visual (microscope) Counted 5 random subsamples of filter, each 3% of area 4 procedural blanks. SEM on some particles	Means (± SD) par/m^3: Upstream = 1.94 ± 0.81 Downstream = 17.93 ± 11.05	Means (± SD) par/L: Upstream = 0.002 ± 0.001 Downstream = 0.018 ± 0.011 Samples corrected for fiber background	AS WWTP. Treats domestic wastewater. Ave. flow = 927 million L/day. No effluent disinfection Foam and pellets found only downstream at low levels relative to fragments and fibers
Yonkos et al. (2014) United States 4 estuarine rivers, Chesapeake Bay	Water Papatsco, Magothy, Rhode, and Corsica rivers	Manta net (330 μm) trawl, 15-cm sampling depth	Rinsed samples into 0.3 mm sieves and transferred to pre-weighed glass beakers to dry Weighed and digested (75 °C) by Fenton's reagent. Density separation (300 g/L table salt in DI water)	Visual (microscope) μ-Raman on 10 small (≤ 2 mm) (likely to be misidentified) fragments	Mean, par/km^2 (g/kgsample): Papatsco = 155,374 (102) Magothy = 112,590 (74) Rhode = 67,469 (18) Corsica = 40,852 (9)	Mean[a], par/L: Papatsco = 0.001 Magothy = 0.00075 Rhode = 0.00045 Corsica = 0.00027	Watershed properties[b]: area (km^2), % developed[c], and population: Patapsco: 1637, 54%, 899,000; Magothy: 92, 59%, 32,350; Rhode: 67, 12%, 4300; Corsca: 97, 13.5%, 3500
Free et al. (2014) Mongolia Lake Hovsgol	Water	16-cm sampling depth manta trawl, 333-μm	Tyler sieves (0.355–0.999 mm, 1.00–4.749 mm, > 4.75 mm) 30% H_2O_2, density separation (density = 1.62 g/mL)	Visual (microscope)	20,264 particles/km^2 (max: 44,400)	Mean[a] = 0.00012 par/L Most common: 40% fragment, 38% film, and 20% line/fiber	Range of sizes: 0.333–5 mm. MP density decreased with distance from southwest shore

Table 2 continued

Study	Sample type(s)	Collection and particle cut size	Sample preparation	Analysis	MP abundances and types		Comments
					Reported units	Converted[a] units and/or MP types	
Sadri and Thompson (2014) Southwest England Tamar Estuary	Water	Manta trawl (300 µm)	Sieved (3 mm, 1 mm, and 270 µm)	Classified as: as > 5, 2–5, and < 1 mm FTIR on 50% of counted pieces	Overall mean: 0.028 par/m^3 84% of 204 suspect MPs confirmed	Overall mean: 0.000028 par/L 40% PE, 25% PE, 19% PP, 8.2% PVC and nylon	MPs black and yellow PP only in 1–3 and 3–5 mm size fractions Nylon in < 1 and 1–3 mm fractions Most MPs 1–3 mm
Zhao et al. (2014) China Yangtze Estuary and East China Sea	Water	Collected estuary water (0.2 m deep) with pump and sieved (32 µm). Sea water with neuston net (333 µm). Fixed in formalin	30% H_2O_2 digestion of samples with high organics, followed by density separation in saturated $ZnCl_2$ Floating particles filtered onto 1.2-µm cellulose nitrate filter	Visual (microscope) Counted particles > 500 µm	Means (par/m^3) Estuary: 4137.3 ± 2461.5 (max: 10,200) Sea: 0.167 ± 0.138	Means (par/L) Estuary: 4.1 (max: 10.2) Sea: 0.0002	Most MPs clean and colors. Small fraction black and white Max size was 12.46 mm. > 90% 0.5–5 mm
Lechner et al. (2014) Austria Danube River	Water	Stationary conical driftnets (500 µm), 0.5 m depth	Samples suspended in water and particles removed	Visual sorting by naked eye	Mean (par/1000 m^3) 316.8 ± 4664.6 (max: 141,648)	Mean: 0.00032 par/L	Pellets, flakes, spherules, others
Lima et al. (2014) Brazil Goiana Estuary	Water	Conical plankton net (300-µm)	Floating particles sieved (45-µm)	Visual (microscope)	Mean: 3.1 × 10^{-4}–2.6 × 10^{-3} par/m^3 (max: 0.19 par/m^3)	Mean: 3.1 × 10^{-7}–2.6 × 10^{-6}	
Eriksen et al. (2013) United Sates Great Lakes	Water 21 stations over 1300 km expedition	16-cm sampling depth mesh manta trawl (333 µm)	2 M HCl, Tyler sieves for 3 size classes: 0.355–0.999 mm, 1–4.749 mm, > 4.75 MM)	Visual (microscope). SEM–EDS (counts adjusted based on SEM–EDS)	Mean: 43,157 par/km^2 (max: 466,000)	Mean[a] = 0.00027 par/L (0.000043 par/L)[d] 20% of visually identified MPs < 1 mm were Al silicate	81% of particles in 0.355–0.999 mm range Colored spheres < 1 mm suspected microbeads
Faure et al. (2012) Lake Geneva Mediterranean Sea (MS)	Lake water	Manta trawl with plankton sieve (300 µm)	Sieved to 5 mm	Visual (microscope) Sorted, counted and weighed	par/km^2: 4.81 × 10^4 (Lake Geneva) 1.4–31.5 × 10^4 (MS)	par/L[e] 0.0002 (Lake Geneva) 5.7–126 × 10^{-5} (MS)	

Table 2 continued

Study	Sample type(s)	Collection and particle cut size	Sample preparation	Analysis	MP abundances and types		Comments
					Reported units	Converted[a] units and/or MP types	
Moore et al. (2011) United States 2 California rivers, 1 creek	Water	Hand nets (800 and 500 μm), manta trawl (333 μm)	Tyler sieves (4.75, 2.8, 1.0 mm). Dried samples (65 °C)	Visual 2 size ranges: 1– < 4.75 and ≥ 4.75	Maximum (par/m^3) 1.29×10^4 (Los Angeles River, 1–4.75 mm)	12.9 par/L (max): 71% foams, 14% fragments, 10% pellets, 1% whole items	Sorted, counted, and weighed MPs. By mass: 37% whole items, 29% fragments, 13% pellets, 11% PS foam

CaFl calcium fluoride, *DI* deionized, *FITC* fluorescein isothiocyanate, *FPA* focal plane array (detector), *HCl* hydrochloric acid, H_2O_2 hydrogen peroxide, *HSSPME-GC-ITMS* headspace solid phase micro extraction with GC and ion trap MS, *IPA* isopropyl alcohol, *MCT* mercury-cadmium-telluride, *μ-ATR-FTIR* Fourier transform infrared spectroscopy (FTIR) coupled with an attenuated total reflectance microscope objective (μ-ATR), *NaCl* sodium chloride, *par* particles, *PTFE* polytetrafluoroethylene, *Pyr-GC–MS* pyrolysis gas chromatography with mass spectrometry, *SDS* sodium dodecyl sulfate, *SEM–EDS* scanning electron microscopy with energy dispersive X-ray spectroscopy, *WPO* wet peroxide oxidation, *WWTP* wastewater treatment plant

[a]Values converted from particles/km to particles/L are from Horton et al. 2017a except where indicated. Sample volumes were based on sampling area and depth

[b]2011 values

[c]Urban/industrial and suburban/residential

[d]Value reported by Rezania et al. (2018)

[e]Authors' estimate (based on sampling depth and surface area)

Table 3 Studies of microplastics in freshwater systems: sediments

Study	Sample type	Collection	Sample preparation	Analysis	MP abundances reported/converted[a]	Types/sizes/comments
Bordós et al. (2019) Hungary Carpathian basin	Sediments and water (Table 2)	2–3 kg with Van Veen Grab and spade. Shipped cooled	Weighed amount was added to MPSS for NaCl (1.2 g/mL) density separation. Stirred 4 h and processed by water method (Table 2). 30% H_2O_2	μ-FTIR. Particles selected visually and auto analyzed in ATR mode	Mean: 0.81 ± 0.37 par/kg Range: 0.46–1.62	PP and PS dominant (identified by library match). Fishponds may act as MP sink
Di et al. (2019) China Danjiangkou Reservoir (DJKR)	Sediment at 20 locations	See Di and Wang (2018)	2-step density separation, in NaCl and NaI (Di and Wang 2018). 30% H_2O_2 0.45 μm smallest mesh size	Visual (microscope). μ-Raman of 142 suspect MPs, and subset by SEM	Mean (DJKR) = 24 ± 9 par/kg sediment Range: 15–40 par/kg (wet wt.)	Colors: transparent dominant (12.5–100% for 13 samples), followed by blue Fibers (25–100%) in 19 samples, fragments (14.3–75%) in 13, pellets in just 4. No Styrofoam
Dikareva and Simon (2019) New Zealand (Auckland) Streams	Sediments and surface water (Table 2) in small streams spanning urbanization gradient	Scooped 10–20 samples (5-cm depth) into 150 mL container. Mixed in bucket, sieved (5 mm) into 1 L container, and placed in zip-lock bag for transport to lab	2-step extraction (Claessens et al. 2013). Placed 1-L sediment in elutriation device; added water. Sieved (63 μm) overflow water from device. Transferred filtered solids to centrifuge tube for density separation (NaI, 1.6 g/mL). Centrifuged, filtered (1.2 μm glass-fiber filter) top liquid, dried filter. Repeated density separation on remaining sample. Filtered onto same (dried) filter	See Table 2. Visual (microscope) and physical properties ATR-FTIR	9–80 items/kg (dry wt.) Fragments (79%) and fibers (20%) dominant 88% of fragments < 500 μm. Foam and films rare; pellets absent Smallest fraction (63–500 μm) dominant	See Table 2 Counts blank corrected (Mean procedural blank = 15 particles, n = 6) Polymers varied and often not predictable based on size, shape, and color
Yuan et al. (2019) China Poyang Lake	Sediments, water (Table 2), crucians 21 sites, 11/2017	Collected 2, 500-g surface (0.25 m^2) samples from boat with Van Veen grab. Placed 1 kg in foil bag	Sediments dried (48 h at 50 °C). MPs extracted by density separation 5 size classes: < 0.1 mm, 0.1–0.5 mm, 0.5–1 mm, 1–5 mm, and > 5 mm	Visual (microscope) μ-Raman on 100 particles (on silver slide to reduce fluorescence)	Sediments (par/kg dry wt.): 54–506	Highest MP levels in northern area. Largest freshwater lake in China

Table 3 continued

Study	Sample type	Collection	Sample preparation	Analysis	MP abundances reported/converted[a]	Types/sizes/comments
Di and Wang (2018) China Three Gorges Reservoir	29 sites, sediment and water (Table 2)	2, 1-L surface samples (0.25 m^2 area) per site, by Van Veen Grab. Placed in jar. 0.45 mm smallest mesh size	Stored at 4 °C. 2-step density separation (saturated NaCl then 60% NaI solution). Suspension digested in 30% H_2O_2. Next steps same as water (Table 2)	μ-Raman (see Table 2)	Mean (par/kg wet wt.) = 82 ± 60 Range: 25–300 PS dominant	87% recovery of spiked PE microbeads Fibers: 33.9–100% of MPs. Higher MP levels in countryside. High-density MPs more likely to deposit in sediment. Most transparent, then blue
Gray et al. (2018) United States (South Carolina) Estuaries: Charleston Harbor, Winyah Bay	Intertidal sediments (n = 9–12)	Removed top 2 cm with trowel and weighed in steel buckets	Density separation (800 g NaCl in 4 L seawater). Sieving (500, 150, 63 μm). Digestion in 30% H_2O_2 (1 week). Filtered (38 μm)	Visual (microscope) ATR-FTIR (80 particles) and SEM imaging	Means (par/m^2): Charleston Harbor = 413.8 ± 76.7 Winyah Bay = 221.0 ± 25.6, respectively (high = 441, low = 51)	Fragments dominant (76.2% in harbor and 77.5% in bay). Fragments mainly black (73%). White blue, clear, other colored MPs also present. 18.9% of par in harbor were foam (98% white) First study in Southeastern US estuaries. Variation due to currents, wind, and sources
Imhof et al. (2018) Italy Lake Garda	Sediments at 5 beaches. Three, 20-m 'transect' lines at each	20-m transect (parallel to water), with 10 core samples from each (5-cm depth, 10-cm diameter)	Extraction in MPSS (Imhof et al. 2012) with $ZnCl_2$ (d = 1.6–1.7 g/mL) (Imhof and Laforsch 2016). Filtered on glass-fiber filters	Visual (microscope) μ-Raman down to 1 μm (Imhof and Laforsch 2016)	3508 ± 8855 par/m^2	Sampling approach educed number of samples and improved representativeness (combines quadrate and core sampling approaches)
Lin et al. (2018) China Pearl River, lower course along Guangzhou City	River sediments and water (Table 2) at 14 sites WWTP samples (Table 4)	2 kg sediment (top 5 cm) by Van Veen grab sampler. Shipped in foil bags (storage at − 20 °C)	Dried, crushed, removed large items. Multiple density separations (200-g:1 L sat. NaCl). Filtered (2-μm) supernatants, digested in 10% KOH (65 °C, 80 rpm, 24 h), added NaCl solution, filtered (5-μm)	Visual (microscope) μ-FTIR (MCT) Spectral library match (quality > 80%)	Sediments (items/kg dry wt): 80–9597	Fibers dominant. PE and PP accounted for 73.8% MP abundances varied and may be affected by multiple factors

Table 3 continued

Study	Sample type	Collection	Sample preparation	Analysis	MP abundances reported/converted[a]	Types/sizes/comments
Peng et al. (2018) China Shanghai	Sediment	Shovel to collect upper 5 cm of 0.5 × 0.5 m area (n = 3). 500 g each	Density separation (NaCl). (Masura et al. 2015). No wet sieving or WPO	Visual (microscope) μ-FTIR, ATR Library match (> 70% criterion)	802 ± 594 items/kg sediment (dry wt.) 57% PP, 17% PES, 11% rayon 5.7% cotton + viscose	Spheres dominant (88.98%) in river sediments, followed by fibers (7.55%) and fragments (3.47%). Most (90%) were white spheres. Other MPs (blue, transparent, white, red) relatively minor. Tidal flat sediments had mostly fibers 62.15% of particles 100–500 μm
Su et al. (2018) China Yangtze River	Sediment, water (Table 2), and Asian clams in lakes, rivers, and estuaries	Top 10 cm of sediment by Peterson sampler (n = 3). See Su et al. 2016	Dried sediment. Used 300 g for NaCl (1.2 g/mL) density separation. Supernatant treated same as water samples (H_2O_2 digestion)	Visual (microscope) μ-ATR-FTIR	15–160 par/kg (dry wt.): sediment	Sizes = 0.021–4.83 mm. Most 0.25–1 mm Fibers dominant. Clear and blue particles most common; clear dominant Sediment MPs more similar to those in clams than water. Asian Clam could serve as bioindicator of MPs in freshwater systems, especially sediments
T. Wang et al. (2018) China Yellow Sea wind farm	Sediment and water Sept. 2016	Collected 3-kg sample with a bottom grab. Stored in Al foil bag	Extracted by 2-step process. Fluidization in saturated NaCl. Second extraction in saturated NaI solution. Filtered on cellulose nitrate filter (1.0 μm) and dried	Visual (microscope 80 x). μ-FTIR on 20% of particles. Library search	Sediment: 2.58 ± 1.14 items/g (dry wt.) PET dominant, followed by cellophane and PE	Black, and transparent particles. Most 0.05–5 mm Fibers dominant in sediment (68.7%) and water (75.3%). Cellulose fiber and calcium carbonate pellets major interferents

Table 3 continued

Study	Sample type	Collection	Sample preparation	Analysis	MP abundances reported/converted[a]	Types/sizes/comments
Xiong et al. (2018) China Qinghai Lake	Lakeshore sediment (and others) and water	Collected top 2 cm of 20 cm × 20 cm area with shovel. 3 replicates	Sieved (2 mm mesh size). Transferred particles (on sieve) for analysis. Water passing sieve was treated by density separation in potassium formate (density = 1.54 g/mL). Floating particles digested in 30% H_2O_2 (60 °C). Filtered (1.2 μm) and transferred particles for analysis	Visual (microscope) 3 classes: 0.112–0.5 mm, 0.5–1 mm, 1–5 mm. μ-Raman on all suspect MPs if counts < 100, or 10–15% if > 100	50–1292 MP/m^2 sediment Higher abundances in tourist areas	PE and PP dominant. Sediments dominated by fibers and sheets Raman analyses were on particles > 200 μm as smaller particles could not be manually transferred Wore cotton lab coat and nitrile gloves
Abidli et al. (2017) Northern Tunisia Bizerte Lagoon	Sediment	Removed top 2–3 cm of a 0.25 m × 0.25 m area with stainless spatula	Dried (50 °C, 48 h). High-density NaCl (140 g/L) for density separation. Filtered supernatant (7–10 μm)	Visual (microscope) Fibers dominant: 21–91%, followed by fragments	3000–18,000 items/kg sediment (dry wt.) High MP pollution	Mean sizes: fiber = 1.39 ± 0.27 mm, fragment = 0.51 ± 0.19 mm, No pellets Fibers clear, followed by white, blue, red, green, black. Fragments blue, green, black, and red. MP sources: cities, industry, and fishing
Horton et al. (2017b) UK River Thames Basin (4 sites)	Benthic sediment Sites had range of sewage input 28 Aug.–3 Sept. 2014	4 samples per site. 10-cm depth collected with stainless scoop. Filled 1-L Kilner jar Focus was large MPs (1–4 mm)	Wet sieved into 2 fractions: 1–2 mm and 2.4 mm and dried (80 °C). Used 3 steps to check processing requirements. Visually removed suspect MPs under microscope. Separated particles in remaining material by floatation (1.7–1.8 kg/L $ZnCl_2$). Rinsed and filtered settled material (1.2 μm glass-fiber filter). Inspected under microscope for any suspect MPs that did not float	Visual (microscope) μ-Raman on 20% of particles BioRad KnowItAll® Informatics System–Raman ID Expert software	33.2 ± 16.1 par./100 g sediment (or 332/kg dry wt.) Ave. abundance (par/kg): 185–660, depending on site MPs at all sites, most 1–4 mm	96% fragments in highest-load sample. Site was downstream of storm drain. Fragments (mostly red and yellow) from thermoplastic road paint. Fibers dominant at other sites (from sewage). Site with high sewage input had highest number PP, PES, and polyarylsulphone most common polymers. 'Unidentified' polymer was largest category, inferred by a synthetic dye

Table 3 continued

Study	Sample type	Collection	Sample preparation	Analysis	MP abundances reported/converted[a]	Types/sizes/comments
Leslie et al. (2017) Netherlands Canals and coastal areas	Sediment Amsterdam canals, 12 sites on Dutch North Sea coast, and 3 in Walden Sea	Van Veen grab 2, 1-L samples combined	All sample homogenized before taking sub-sample for analysis. 20 g sediment added to flask with saturated NaCl solution (1.2 g/mL)	Visual (microscope) μ-FTIR (transmission mode) on 6% of suspect MPs. Library search	par/kg sediment (dry wt.): Canal: mean = 2071 (± 4146), range: < 68–10,500 Coast: 100–3600	Results indicate partial MP settling in freshwater and that seabed is a sink Canal sediment particle sizes: 10–300 μm: 0–75%, 300 μm–5 mm: 19–100% MPs present in benthic macroinvertebrates (muscles, oysters): 10–100 MP/g (dry wt.)
Matsuguma et al. (2017) Japan, Thailand, Malaysia, S. Africa	Sediment cores (as trends in plastic pollution)	Gravity corer (8 or 11 cm diam. × 50 or 100 cm length acrylic pipe) 315 μm to 5 mm	10 g freeze-dried sediment. 1 week 30% H_2O_2 to remove biofilms. Density separations in NaI (d = 1.6 g/mL) Tweezers used to transfer suspect MPs for analysis	ATR-FTIR. Carbonyl and vinyl indices used to avoid counting biopolymers Library hit > 60%	100–1900 par/kg sediment (dry wt.)	White (57%), brown (17%) and black (14%) dominant. Main types: PE, PP, PS, PET, PVC, acrylics, polyamides. 79.8% < 1 mm (0.315–1 mm) Increase in MPs towards sediment surface indicates increasing MPs over time
Sruthy and Ramasamy (2017) India Vembanad Lake	Sediment	Van Veen grab (25 cm^2)	Wet samples sieved (< 5 mm), dried, and sieved again (< 5 mm). WPO (30% H_2O_2) Saturated NaCl (d = 1.3 g/mL) density separation of WPO mixture. Filtered supernatant on glass-fiber filter and dried	Visual (microscope) μ-Raman, KnowItAll® (BioRad) Raman library search	96–496 par/m^2 Mean 252.80 ± 25.76 par/m^2 Main types: PE, PS, PP	Transparent and white particles dominant. Film and foam dominant shapes. LDPE dominant Biofilms may make plastics negatively buoyant. May be one of the reasons for PE in the sediments (usually in water column)

Table 3 continued

Study	Sample type	Collection	Sample preparation	Analysis	MP abundances reported/converted[a]	Types/sizes/comments
Vollertsen and Hansen (2017) Denmark Soils, WWTPs, sludge	10 farmland soils: 5 treated with sludge, and 5 untreated WWTP water and sludge (See Table 4)	Cores of about 300 mL	50 g soil sub-sample treated and MPs concentrated in 5 mL ethanol	Aliquot of ethanol suspension transferred to slide for μ-FTIR-FPA analysis Size range: 20–500 μm (See Table 4)	Mean concentrations (dry wt.): Sludge-treated soils: Mean = 82,000 MP/kg (6.2 mg/kg), Median = 71,000 MPs/kg (5.8 mg/kg) Non-treated soils: Mean = 236,000 MP/kg (51 mg/kg), Median = 145,000 MP/kg (12 mg/kg)	Polymer types by mass (13 MPs identified): Sludge-treated soils: 56% PP, 39% PE, 5% nylon Non-treated soils: 89% PE, 10% nylon, 1% PP MP concentrations in soils were considered low (about 10 mg/kg, comparable to heavy metals in Danish soils) Soils without sludge had more MPs (13 in treated, 24 in untreated). But results had high uncertainty due low MP numbers
Wang J, et al. (2017) China Beijiang River	Benthic sediment MPs and associated heavy metals	Collected 3, 20 × 20 cm area (2 cm deep) with shovel. Transferred to foil bag as 1 sample	Dried samples (50 °C) ≥ 48 h. Density separation (sat. NaCl). Cleaned floating particles in ultrasonic bath. Let stand overnight. Filtered supernatant with 47-mm glass-fiber filter (1 μm)	Visual (microscope) SEM–EDS, μ-FTIR (reflectance mode), ICP-MS	Abundances (items/kg dry wt.): 178 ± 69 to 554 ± 107	Brown and blue particles, and PE and PP dominant FTIR and EDS indicated weathering Most associated heavy metals were inherent load Copolymers found at all sites
Ballent et al. (2016) Canada Lake Ontario and tributaries	Benthic and shore sediments	33 nearshore and beach using sediment trap, core and grab sampling	Taylor sieve (5.6, 2.0, and 0.063 mm), except gravity core samples. Half of 0.063 fraction used for density separation Gravity core samples treated by sodium polytungstate (SPT) density separation	5.6 and 2.0 fractions examined visually (microscope). Limit of about 0.25 mm Subset by Raman and X-ray fluorescence	Means (par/kg): 980 lake benthic 140 lake beach 610 tributary benthic Mainly fibers and fragments < 2 mm	Highest (> 1000 par/kg) in Greater Toronto Area. MPs in beach samples decreased with distance from Toronto, and with sediment depth Near-shore means varied with sampler (core, trap, and grab = 2130, 1070, and 730 par/kg, respectively) Both high and low density MPs found

Table 3 continued

Study	Sample type	Collection	Sample preparation	Analysis	MP abundances reported/converted[a]	Types/sizes/comments
Imhof and Laforsch (2016) Italy Lake Garda	Shore sediment from 5 beaches (3 replicates)	See Imhof et al. 2018 (Table 3)	See Imhof et al. 2018 (Table 3)	See Imhof et al. 2018 (Table 3). Metals by ICP-MS	75 particles/m^2 Converted values[a]: 1.2 par/kg	Identified plastic and paint particles
Käppler et al. (2016) Germany (Warnemünde) Gotland Basin	Beach sediment at 6 locations, in triplicate. Marine sediment	Beach: Top 1 cm of 10 × 100 cm area Basin: Sediment traps	MPSS unit (Imhof et al. 2012). Density separation in SPT (d = 1.8 g/mL). Sieved (500-µm). Transferred suspect MPs with tweezers	µ-ATR-FTIR (germanium crystal and MCT detector). µ-Raman	Concentrations not reported	User-generated and commercial spectral libraries used PS, PE, PAN > 500 µm PE, PP, PVC, PC, PS, PTFE, PET, < 400 µm
Fischer et al. (2016) Italy Lakes Chiusi and Bolsena	Shore sediment, 36 samples	Collected top 3 cm of sediment from 0.025 m^2 area. Sieved (5 mm). < 5 mm fraction weighed	Wet sieved: 1–5, 0.5–1, 0.3–0.5, and < 0.3 mm. NaCl density separation, HCl and Nile Red treatments (see Table 3)	See Table 3	Means (par/kg dry wt.) Chiusi: 234 ± 85 Bolsena: 112 ± 32	Fibers and fragments dominant. Up to 40% of suspect synthetic fibers were cotton Nile red stains PE, PP, PETE (best for white or opaque polymers). Not PVC
Su et al. (2016) China Lake Taihu 3rd largest in China (2250 km^2)	Benthic sediment, water (Table 3), Asian Clams	Peterson sampler (3 pooled, 2-kg samples per site)	NaCl density separation of 1 kg wet sediment. Settled overnight and filtered supernatant (5 µm). Processed and analyzed by same method used for water (Table 2)	Visual (microscope) Subset of 113 particles by µ-FTIR or SEM–EDS	11.0–234.6 par/kg sediment (dry wt)	Fibers dominant (48–84%). White and clear particles more common (29–44%) Main MPs: cellophane, followed by PET, PES, terephthalic acid, and PP Most polluted lake in China. Most MP-polluted freshwater lake worldwide
Zhang et al. (2016) China Tibet plateau lake	Sediments High-altitude inland lakes	Collected 20 cm × 20 cm (top 2 cm) with shovel	Sieved (1 mm mesh). Material passed by sieve used for density separation in potassium formate (d = 1.5 g/mL). Filtered on glass-fiber filters (1.2 µm). Dried in petri dish (60 °C)	Visual (microscope) Raman, SEM	8 ± 14 to 563 ± 1219 par/m2 Mostly 1–5 mm PE, PP, PS, PET, PVC	MPs can be high in inland waters without adequate waste management SEM used to examine surface texture for oxidative and mechanical weathering

Table 3 continued

Study	Sample type	Collection	Sample preparation	Analysis	MP abundances reported/converted[a]	Types/sizes/comments
Corcoran et al. (2015) Canada Lake Ontario	Benthic sediment, beach	Beach samples (surface and 5-cm depth) and bottom sediment core samples	Dried, sieved. Added to water. Floating particles removed and remainder used for SPT solution (d = 1.5 g/mL)	Visual (microscope) μ-ATR-FTIR (Ge crystal). Analysis area 80–100 μm diameter, and 1–2 μm depth	Station 403: 26 par/42.2 g Station 208: 9 par/103.2 g Or 616.1 and 87 par/kg	Bottom sediments: PE (74%) dominant. PP (17%) and nitrocellulose (9%). Some MPs were mineral-particle mixtures (from additives or adsorption) Humber River transports pellets into Lake Ontario
Faure et al. (2015) Switzerland multiple lakes	Shore sediment and water (Table 2)	Shores: 5-cm sampling depth, 0.3 m^2 quadrats	Stored in 5-L PP buckets at 4 °C in salt-saturated water	Visual (microscope)	Mean, all beaches: 1300 ± 2000 par/m^2 Converted[a] = 20 par/kg	MPs found in all 33 beach samples PE (62%, mainly films), PP (15%, mainly fragments), PS (12%, mainly foams)
Klein et al. (2015) Germany Rhine, Main Rivers	Shore sediment Main River has industrial influence		Strungaru et al. (2019)	ATR-FTIR	228–3763 par/kg Up to 4000 par/kg	630 μm–5 mm/kg PE, PP, and PS accounted for > 75% of all MPs PET, PVC, EA, EPDM, PA and acrylic-based polymers also present
Talvitie et al. (2015) Finland Helsinki Tertiary WWTPs (see Table 4)	Sediments near WWTPs and reference sediment at sea	Corer sampler. Top 30 cm placed in containers	NaCl density separation (Browne et al. 2010). Supernatant filtered using custom device with 200, 100, and 20-μm filters (see Table S1)	See Table S1	Viikinmäki discharge site (par/kg sediment): 1.7 (± 1.0) fibers, 7.2 (± 4.9) synthetic Suomenoja discharge site: 4.7 (± 3.5) fibers, 10 (± 14) synthetic	Reference site (at sea):1.7 (± 1.3) fibers, 0 synthetic, and 1220 (± 160), 1060 (± 471), and 346 (± 186) black carbon particles at Viikinmäki, Suomenoja, and reference sites, respectively. 70 (± 20), 3.8 (± 2.3), and 1.9 (± 1.5) ring-shaped particles (respectively)

Table 3 continued

Study	Sample type	Collection	Sample preparation	Analysis	MP abundances reported/converted[a]	Types/sizes/comments
Castañeda et al. (2014) Canada St Lawrence River	Benthic sediment 10 sites	Sampling depth 10–15 cm. Petit Ponar grab (225 cm^2 area) and Peterson grab (930 cm^2) samples	Sieved (500 μm mesh) and preserved in ethanol	Visual (microscope) Microbeads identified based on color and shape. Type determined by DSC	Median microbeads across all sites: 13,759 par/m^2 Converted values[a]: 70.6–105.8 par/kg	Converted value reported as a range that depends on assumed sampling depths
Imhof et al. (2013) Italy Lake Garda	Shore sediment collected by random grid sample technique	Three, random-grid samples from a 20 cm grid (0.04^2) at depth of 5 cm (volume = 2 L)	Density separation in $ZnCl_2$ solution (d = 1.6–1.7 g/mL)	μ-Raman SEM Macro and MPs	par/m^2 North shore: 1108 ± 983 South shore: 108 ± 55 Converted[a]: 17 par/kg and 1.7 par/kg	Mainly low-density MPs: PS (45.6%), PE (43.1%), and PP (9.8%). Small fraction (9–500 μm) also had PA and PVC North shore had 483 ± 236 macro plastics/m^2 and South had 8.3. Higher levels in north may be due to south-to-north winds
Vianello et al. (2013) Italy Venice lagoon	Sediments Duplicates at 10 sites	Up to 5 cm taken with box-corer and refrigerated	Density separation in NaCl. Sieved (32 μm) supernatant. Repeated extraction 3 times. Resuspended pooled materials and filtered (0.7 μm glass-fiber filter)	ATR μ-FTIR (MCT detector). 12 unit areas (4.5 mm^2 each) for chemical mapping	2175–672 MP/kg	Of 10 polymer types, PE and PP > 82% of total. 93% of MPs 30–500 μm Fragments at most stations: 87%. Fibers (10%) not detected at 2 sites. Occasional films (2%) and pellets/granules (1%).)

DSC differential scanning calorimetry, *DWTP* drinking water treatment plant, *FPA* focal plane array (detector), H_2O_2 hydrogen peroxide, *ICP-MS* inductively coupled plasma with mass spectrometry, *MBR* membrane bioreactor, *MCT* mercury–cadmium–telluride (detector), *μ-ATR-FTIR* Fourier transform infrared spectroscopy (FTIR) coupled with an attenuated total reflectance microscope objective (μ-ATR), *NaCl* sodium chloride, *par* particles, *SEM–EDS* scanning electron microscopy with energy dispersive X-ray spectroscopy, *WPO* wet peroxide oxidation, *WWTP* wastewater treatment plant, $ZnCl_2$ zinc chloride

[a]Values initially reported as particles/m^2 were converted to particles/kg by Horton et al. 2017a

et al. (2019a) proposed best practices to sample, extract, and detect MPs in water. They further proposed a set of quality criteria to assess the reliability (data quality) of a study and applied them to 50 studies of drinking water and its sources. Data quality for each study was assessed against the criteria and a total accumulated score (TAS) was calculated, with a maximum of 18. The average (min–max) scores for the different water types were: 13.7 (13–14) for bottled water, 11.5 (8–15) for treated tap water, 12.5 (11–14) for DWTP water, 7.9 (4–15) for surface water, and 7.3 (3–13) for wastewater. The authors noted that the average score likely reflects the relative ease of sample preparation (e.g., bottled and tap water required no digestion and were assigned the maximum for this criterion). They also noted that the average scores for DWTPs and tap water (n = 2 for each), and for bottled water (n = 3), are less rigorous because of the relatively few studies on these water types. Of the 50 studies evaluated, 46 (92%) lack reliability due to inadequate quality assurance. On average, the studies were assigned a TAS of 8.4, about half the maximum. The average is comparable to that for studies of MP ingestion by biota (Hermsen et al. 2018). In addition to reporting MP concentrations and quality scores, the authors summarized polymer types and particle shapes. In order of decreasing prevalence, the following polymers were reported for the 50 studies: PE $\sim$ PP $>$ PS $>$ PVC $>$ PET. The abundances reportedly reflect the global plastic demand, and the tendency of PVC and PET particles to settle due to their higher densities. Fragments, fibers, film, foam and pellets were the most frequently reported shapes.

Luo et al. (2019) reported MP abundances in city creeks, rivers, an estuary, and coastal waters (East China Sea) in the Yangtze Delta region. Abundances in freshwater bodies (1.8–2.4 items/L) were higher than those in estuarine and coastal waters (0.9 items/L). Abundances in two rivers tended to be higher downstream, at sites closer to the city center. The authors suggested transport from pollution sources to sinks throughout the river networks. Fibers were highest in city creeks (88%), followed by two rivers (85% and 81%), an estuary (66%), and the sea (37%), with mainly PES in creeks and rivers. Results suggest that the levels and types of MPs vary across the different water bodies, and that small, freshwater bodies are more polluted than estuarine and coastal waters.

Wastewater treatment plants are recognized sources of N&MPs that might affect water quality (e.g., Sun et al. 2019). Kay et al. (2018) investigated MPs in freshwaters upstream and downstream of 6 WWTPs in river catchments across northern England over a 6-week period. All plants were associated with increased MPs in rivers, as reported elsewhere (Estahbanati and Fahrenfeld 2016; McCormick et al. 2014; Morritt et al. 2014). The potential MP sources were diverse and varied within and across catchments, and no patterns were found in the types of MPs detected. The identified sources included atmospheric deposition (fibers), sludge applied to soils, and secondary MPs. Fibers, fragments, and flakes were dominant, consistent with (Ballent et al. 2016; Dris et al. 2015a, 2015b). The prevalence of fibers indicates textile sources, while fragments and flakes suggest secondary MPs. Beads and pellets were dominant only at one site, upstream, as reported by (Mani et al. 2015). The temporal variability was small at some sites and high at others. Concentrations were not reported, but the mean ratio of up-to-downstream MPs was > 1, and between 1 and 3 for 19 of 28 paired samples. On four occasions, MPs were higher upstream, possibly due to concurrent increases upstream and decreases downstream.

Ravit et al. (2017) reported MP surface densities, types, and sizes for 15 locations at two New Jersey (NJ, USA) rivers in heavily urbanized areas: the Raritan and Passaic rivers. Many point sources discharge into the rivers within a 5-mile radius of the sampling sites, including companies that produced personal care products, companies with 'plastic' in their names, and WWTPs. Surface water MPs ranged from 28,000 to $>$ 3 million particles/km^2. The most common forms in order of abundance were: "hard fragment" (38%), foam, line (fiber, filament), film (from bags/wrappers), and pellet (nurdles or microbeads). A hard fragment was identified as PP, a pellet as PE, and black foam as PE and either PVC or a PE-vinyl acetate copolymer. By size, 71% of the MPs ranged from 1 to $>$ 4.5 mm. Analyses of MP and water fractions found similar compounds, indicating transfer between phases and bioavailability to aquatic biota.

Estahbanati and Fahrenfeld (2016) investigated the impact of WWTPs on MP pollution in a recipient river, the Raritan River, in central NJ (USA). The river basin covers 2850 km^2 and provides water for

drinking, irrigation, agriculture, recreation, and industry. The primary land use of the main stem is urban and suburban (51.3%), while the branches have predominantly agricultural and forest lands (61.3%). More than ten municipal WWTPs discharged into the river, five of which are major (> 1 million gallons per day [MGD]). Water samples were collected upstream and downstream of four major plants. Recovered MPs were counted in three, quantitative size categories (500–2000 μm, 250–500 μm, 125–250 μm) and one semi-quantitative category (63–125 μm). Particles were classified under a light microscope as primary or secondary MPs, based on morphology. Concentrations in the 125–250 and 250–500-μm fractions were significantly higher downstream: 72 ± 60 downstream and 24 ± 11 MPs/m^3 upstream. The smaller size classes were in high relative abundance across sites. Primary MPs increased significantly downstream, but secondary MPs were dominant (66–88%) in the quantitative size categories. A moderate correlation between MP concentration and downstream distance was observed.

McCormick et al. (2016) examined MP pollution in surface waters of nine streams in the Chicago (Illinois, USA) metropolitan area. The streams received wastewater from ten facilities with effluent flowrates of 2.16–225 MGD (0.095–9.86 m^3/s) and different treatments. Microplastic particles were counted and classified as fiber, film, fragment, pellet, or foam, as described by Eriksen et al. (2013). Fibers included filament/line-shaped plastic pieces. Films were very thin relative to their surface area and typically had irregular shapes, while pellets were more regular and rounded. Foam particles had sponge-like textures and included PS. Fragments had jagged edges and appeared to be broken from larger pieces of plastic. The average MP flux was about 1.3 million particles/day, but results were highly variable between sites, ranging from 15,000 to 4.7 million particles/day. Pellets, fibers, and fragments were dominant. The main polymers were PP, PE, and PS; film and foam MPs were uncommon. Except for two rivers, MP concentrations were significantly higher downstream. Assuming similar concentrations across seasons, the estimated discharges represent 488 million MPs per river annually, with a minimum of 5.6 million and maximum of 1.7 billion. Further work was recommended for "more robust calculations of annual flux."

Dris et al. (2015a) found MPs (100–5000 μm) in freshwater samples collected in Paris, France, near city WWTPs. Sampling by manta trawl and a plankton net gave different results for the MP types and amounts. Surface concentrations in the Seine River ranged from 3 to 108 particles/m^3 (0.003–0.108 particles/L), with a mean of 30 particles/m^3 (0.03 fibers/L) for samples collected with an 80-μm mesh plankton net, which gave a predominance of fibers relative to a 330-μm mesh manta trawl (Table 2). Greater variety in the shapes and types of MPs was found with the manta trawl, but at much lower concentrations: 0.28–0.47 particles/m^3 ($2.8–4.5 \times 10^{-4}$ particles/L; mean = 3.5×10^{-4}/L). For comparison, raw wastewater from a WWTP contained high levels of fibers, $260–320 \times 10^3$ particles/m^3, (260–320 MPs/L), while the treated effluent contained $14–50 \times 10^3$ particles/m^3 (14–50 MPs/L) (Table 4). The authors suggested that a combined sampling approach may provide more accurate assessments. No upstream–downstream patterns were observed. Levels were higher in April (0.014–0.108 particles/L) than in May (0.003–0.036 particles/L), except at one site. Differences were attributed to local variability, a water flow decrease (20%), and difficulties in visual observations of MPs due to large amounts of natural debris in the May samples. The presence of MPs, > 90% fibers, in atmospheric fallout also was reported, indicating deposition of 29–280 particles/m^2-day.

Yonkos et al. (2014) studied four estuarine rivers in the U.S. Chesapeake Bay, finding high variability in MP types within and between sites. The watersheds had different land uses and a wide range of populations. Surface water was collected monthly using a manta net trawl (MP sizes: 0.3–5.0 mm) during a six-month period. Particles were counted as fibers, thin sheets, hard colored fragments, preproduction pellets, and extruded polystyrene (e.g., styrofoam). Microplastics were found in 59 of 60 samples, at concentrations from < 1.0 to > 560 g/km^2. Small fragments (0.3–2.0 mm) and flexible sheets were most abundant, followed by synthetic fibers, extruded PS, and larger fragments (2.0–5.0 mm). Preproduction pellets were common only in one river (Patapsco). Particles analyzed by optical microscopy coupled with Raman spectroscopy (μ-Raman) all had peaks indicating PE. Two particles had a color and peaks consistent with cobalt phthalocyanine (dye), and a

black particle had two broad peaks indicative of black carbon (possibly burnt plastic). Positive correlation was found between MP concentration and population density, and the extent of urban/suburban development. In three of the four sites, the highest MP levels occurred after major rain events.

3.3 Wastewater treatment plants

Many types of plastic debris in urban watersheds reach WWTP influents. Depending on the plant design, primary, secondary, and tertiary treatments purify influent by physical, chemical, and biological processes. A schematic illustrating this series of treatments is shown in Fig. 2. Incoming floating/suspended solids are removed by screening, sedimentation, flotation, coagulation-flocculation, and filtration in treatment ponds or settling tanks, which capture substantial amounts of solid particles (Stuetz and Stephenson 2009). The capture of common MPs is thought to relate to their relatively low densities, permitting floatation and removal in the grease layer (Murphy et al. 2016) by skimmers (Carr et al. 2016) in the primary treatment. Another possible mechanism is surface fouling, which can cause MPs to sink in a settling tank or associate with flocculants (Carr et al. 2016).

Though just a small fraction of MPs in influent is released in the final effluent, WWTPs can be significant sources of MPs (Browne et al. 2011; Lares et al. 2018; McCormick et al. 2014). Multiple literature reviews on WWTPs have been published (e.g., Enfrin et al. 2019a; Gatidou et al. 2019; Kang et al. 2018; Lares et al. 2018; Prata 2018; Raju et al. 2018; Sun et al. 2019). Studies published since 2017 are listed in Table 4, while earlier studies are reported in Table S1 (Supplementary Information). Several studies are highlighted below.

3.3.1 WWTPs: selected studies

Many studies of WWTPs have focused on N&MPs in final effluent, while others have examined the removal efficiencies of different treatment processes (Carr et al. 2016; Conley et al. 2019; Gies et al. 2018; Lares et al. 2018; Mintenig et al. 2017; Murphy et al. 2016; Talvitie et al. 2017a; Yang et al. 2019). For example, Lares et al. (2018) examined a conventional activated sludge process and an advanced, pilot-scale membrane bioreactor (MBR). The overall removal efficiency was 98.3% (89.8% for fibers and 99.1% for non-fiber MPs). Most MPs were removed before the CAS process, and 99.0% were removed before aeration. The MBR was slightly more efficient: 99.4%. The MBR permeate contained 0.4MP/L while the final effluent of the CAS process was 1.0 MP/L. Lares et al. (2018) also reviewed studies at primary, secondary, and tertiary facilities, representing 15 studies and 52 WWTPs globally (Browne et al. 2011; Carr et al. 2016; Dyachenko et al. 2017; Lares et al. 2018; Leslie et al. 2017; Magnusson and Norén 2014; Mason et al. 2016; Michielssen et al. 2016; Mintenig et al. 2017; Murphy et al. 2016; Talvitie et al. 2015, 2017b; Ziajahromi et al. 2017). Concentrations in final effluents ranged from 0 MP/L (Carr et al. 2016) to 91 MPs/L (Leslie et al. 2017).

As with other studies, different sampling, isolation, and enumeration methods have been used in WWTP studies. Various grab or composite methods have typically been used for sample collection, different digestion/separation procedures (or none) have been used to isolate them, and different size fractions have been collected and classified. Grab sampling of the waste stream is useful to capture samples during peak flows, and detect variability or over short time periods, but the sample may not be representative. Differences in collection methods; lower particle size limit, typically 20–300 μm (Lares et al. 2018); MP types and concentrations; treatment processes (Mahon et al. 2017); and sample preparation and analysis methods (e.g., Underwood et al. 2017) have contributed to the variability, as did the different periods over which the studies were conducted. Most were completed over relatively short periods (one day to 1.5 months). A need for long-term studies to assess temporal variation has been emphasized (Magnusson and Norén 2014; Murphy et al. 2016; Ziajahromi et al. 2017).

In a one-year study, Conley et al. (2019) determined MP loads and removal efficiencies for three WWTPs with different treatment sizes, operations and service compositions (Table 4). The plants discharged into the Charleston Harbor, South Carolina (USA). Overall, MP concentrations at each plant varied within a factor of 2.5 in influent and 4.8 in effluent, with no seasonal trends in concentrations or efficiencies. Mean influent concentrations ranged from 126 to 147 MPs/L, while those for final effluents ranged from 4 to 18 MPs/L. Microfibers were most common (60–70%). The

Table 4 Studies of microplastics in wastewater treatment plants

Study	Sample and plant/ study details	Sample collection	Sample[a] preparation	Analysis[b]	Main findings[c]	Comments
Bayo et al. (2020) Spain Cartagena Urban WWTP AS process with 1° treatment and 2 parallel AS reactors PEq = 210,000 (35,000 m^3/d)	Sampled 4 stages: grit/grease removal (GGR), 1° clarifier (PCL), activated sludge reactor (BRT), and 2° clarifier effluent (EFF) Determined MP and 'microlitter' (ML) particles Survey between Sept. 2016 and April 2018	128 grab samples. Volume depended on treatment stage. Total L: GGR = 60.1, PCL = 59.3, BRT = 103.4, EFF = 143.0 Precise volumes for tests: GGR = 0.5–3.7, PCL = 0.5–3.6, BRT = 1.4–3.7, and EFE = 2.7–17.0 L	Processed 352.6 L of wastewater. EEF sample was directly filtered. All others treated by NaCl density (d) separation (final d = 1.08 g/mL) in 1:3 salt solution to wastewater. Sample filters placed in Petri dishes to dry (100 °C) overnight Clothes of natural fabric were worn	Trinocular microscope. Transferred particle subset for FTIR using diamond anvil compression (DAC) cell Low background but samples blank corrected	Influent maximum: 13.04 MP/L MPs 46.6% of total ML. Overall MP and ML removals: 90.3% and 90.1%, respectively Mean concentrations: MPs/L: 3.20 (± 0.67), 2.59 (± 0.85), 2.13 (± 0.38), 0.31 (± 0.06) for GGR, PCL, BRT, and EFF, respectively ML/L: 12.43 (± 2.70), 9.73 (± 3.04), 3.21 (± 0.50), and 1.23 (± 0.15), respectively Fibers and fragments dominant in effluent, most 400–600 µm. 17 polymers identified. LDPE most prevalent (52.4%) as films (27.7%). Attributed mainly to nearby greenhouses and plastic bags	Biodegradable polymers likely degrade in first treatment stages Non-plastic fraction > MP fraction, except BRT (66.2% MPs) Influents with high suspended solids (SS) had fewer and larger MPs, possibly due to aggregation with SS. PS and PET have surface energies > 25 mN/m and could associate with organic matter
Conley et al. (2019) United States Charleston, South Carolina 3 WWTPs with CAS processes: Plum Island (PI), Rifle Range (RR), Center Street (CS)	Influent and effluent PI treated residential, commercial, and industrial wastes RR and CS treated mainly residential Sampled June, Oct. 2016. Jan., April, July 2017. Further RR samples in June, July 2017	Influent: downstream of headworks and upstream of sludge return Effluent: post-disinfection (before discharge) Duplicate influent and effluent samples	Filtered (43-µm) and transferred solids to Petri dishes. Processed by sonication, 30% H_2O_2 and 1 M HCl. Filtered as 3 size fractions and treated filters with final sonication and rinse. Transferred solids to clean dish or gridded cellulose membrane	Visual (optical microscope) 3 filter fractions: > 418 µm, 178–418 µm, 60–178 µm. Used gridded filter or grid under Petri dish µ-ATR-FTIR (Ge crystal) on select MPs	Means (par/L): PI, RR, CS, respectively (n = 5 or 8): Influent: 147 ± 62, 126 ± 45, 146 ± 57 Effluent: 3.7 ± 1.5, 17.6 ± 6.1, 17.2 ± 10.4 60–70% microfibers Average removals (%) of total, fiber, and non-fiber MPs, respectively: PI = 97.6 ± 1.2, 97.2 ± 1.0, and 98.4 ± 1.3 RR = 85.2 ± 6.0, 80.2 ± 8.0, and 95.4 ± 2.4 CS = 85.5 ± 9.1, 83.7 ± 8.2, and 88.8 ± 9.6 Colors: translucent or white (60%), black (22%), blue/green (13%), red (5%)	No seasonal trends in MP levels or efficiency High potential for MP sorption of toxins in wastewater. High numbers may pose risks Average daily influent MPs per capita: PI = 83,500 ± 29,200 CS = 53,500 ± 28,400 RR = 49,600 ± 15,400

Table 4 continued

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Lv et al. (2019) China Wuxi City WWTP with parallel treatment systems	Influent, effluent pots, mixed liquors and excess sludge of OD and A^2/O-MBR Parallel treatments: oxidation ditch (OD) and MBR Collected February 2018	5 steel sieves (diameter 20 cm): 500, 250, 125, 62.5 and 25 µm in sequence Gravity feed through sampler until sieve clogged (at least 1 L), or to a total volume of 200 L	Digested 3 h in Fenton's reagent (30% H_2O_2 + Fe(II) catalyst, 20 mmol/L) and dried (60 °C). Density separation in NaI solution (1.49 g/mL). Centrifuged (3500 rpm, 5 min). Filtered supernatant (25 µm), rinsed, transferred solids to petri dish	Visual (microscope) ATR-FTIR, and confirmation by library search	Concentrations (MPs/L) Influent: 0.28 ± 0.02 Effluent: OD = 0.13 ± 0.01, MBR = 0.05 ± 0.01 Overall removal (by number): 53.6% OD and 82.1% MBR (systems A and B, respectively) And 99.5% and 97% by mass (respectively) Rotary grit chamber (OD system): 1.32 ± 0.1 Aerated grit chamber (MBR system): 0.22 ± 0.03 MLSS of membrane tank (MBR system): 1.6 ± 0.2 OD system sludge (settling tank): 0.7 ± 0.1 Influent: PET (47%), PS (20%), PE (18%), and PP (15%). Fragments (65%) and fibers (21%) dominant, most PET. Limited films (12%) and foams (2%). No microbeads	Dominant MP sizes in influent: > 500 µm (40%) and 62.5–125 µm (29%) MPs accumulated in sludge. MBR has higher removal efficiency than OD system Results suggest source control (e.g., eliminating MP fibers from laundry effluents) and proper treatment units could significantly reduce MPs in WWTPs No contaminants in negative controls
Magni et al. (2019) Northern Italy Tertiary WWTP PEq = 1.2 million	Inlet, after the settler, effluent, and sludge Sampled 3 days over 1 week, during dry weather, in spring	30 L wastewater collected in steel bucket. Sieved on-site (5 mm, 2 mm, and 63 µm). Sludge in 50-mL beaker	Density separation (NaCl, 1.2 g/ml). Stirred overnight (4 °C). Supernatants filtered (8-µm) and solids rinsed. Samples partially digested (3 days in 15% H_2O_2)	Visual (optical microscope) µ-FTIR	Mean wastewater concentrations (par/L): Influent = 2.5 ± 0.3, After settler: 0.9 ± 0.1 Effluent: 0.4 ± 0.1, Sludge: 113 ± 57 MPs/g sludge (dry wt.) 84% removal efficiency Effluent mainly PES (35%) and polyamide (17%)	Potential discharge of 160 million MPs/day. Sludge traps about 1.1 billion MPs/ton (30 tons daily) Sludge could be an important MP source in agroecosystems

Table 4 continued

Study	Sample and plant/ study details	Sample collection	Sample[a] preparation	Analysis[b]	Main findings[c]	Comments
Wolff et al. (2019) Germany WWTP, mechanical and biological treatments. Service population: 98,500	Effluent from secondary clarifier Surveyed 11/2017 to 01/2018. Wet and dry days	7 grab samples Sampler was a pump, 10-µm cartridge filter and housing (all ss), and silicone hose. Filtered 40–200 L	Oxidation (H_2O_2 [50%] and NaClO) and density separation (ZnCl, 1.9 g/cm^3). Filtered DI water and n-hexane used to rinse small MPs adhered to surfaces	MPs ≥ 10 µm analyzed on sub-area of silicon substrate by µ-Raman (785 nm). Counts based on total deposit area	Non-fiber means (par/m^3) Wet days: 5900, Dry days: 3000, Overall: 3500 Most 30 to < 100 µm. Mainly PET, PP, PE, PS Fiber means (fibers/m^3) Wet days: 1500, Dry days: 730, Overall: 1100 Fiber lengths: 100–1000 µm. Most were PET	Effluent: about 10,000 m^3/d Dry weather samples tended to have fewer MPs than wet weather Hexane rinse removed small, adhered particles (< 20 µm) that could not be removed with water
Yang et al. (2019) China (Beiging) Advanced tertiary STP (Gaobeidian). Serves population of approximately 2.4 million	Influent, effluents, and primary and secondary sludge Largest water reclamation plant in China. Treats mainly domestic effluents Sampled between April and June 2018	30-L samples in glass bottles. Stored at 4 °C	Sieved (5 mm, 50 µm). Rinsed into water, filtered (10 µm), sonicated filters in water, dried, digested (60 °C) by WPO (Fenton's reagent) $ZnCl_2$ density separation (then HCl added to remove $ZnCl_2$). Transferred to separatory funnel, drained solids, filtered solution (10-µm PTFE filter)	Visual (optical microscope) Subset of particles removed for µ-FTIR analysis and count correction	Mean wastewater (MPs/L): Influent: 12.03 ± 1.29 Effluent: 0.59 ± 0.22; > 95% MP removal Sludge (MPs/kg dry wt.): 22.7 ± 12.1 × 10^3 (sludge range: 1.6–56.4 × 10^3) Identified 18 polymers (10 colors), manly PET (42%), PES (19%), and PP (13%) Microfibers dominant (86%) and attributed to domestic washing Average size of 1111 ± 863 µm. Average non-fiber MP was 681 ± 529 µm (most near 300 µm)	Treated mean of 10^6 m^3/day of wastewater. Estimated daily release of 0.59 ± 0.22 × 10^9 MPs 50-µm filter used because smaller size clogged. And MPs < 50 µm could not be manually transferred for µ-FTIR Inspected (visually) solids from separatory funnel. No MPs found

Table 4 continued

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Gies et al. (2018) Canada (Vancouver) Secondary WWTP Plant treats combined wastewater and stormwater PEq = 1.3 million	Influent, primary and secondary effluents and sludges Collected in 2016: 9/16, 9/29, 10/28 (water), and 9/14, 9/27, 10/11 (sludge)	1 L influent in jars. Filtered (0.63 µm) 30 L effluent and rinsed solids into jars. 250-mL jars for sludge 17 wastewater and 12 sludge samples	All samples spiked with MPs. Wastewater allowed to settle 24 h. Extracted water layer with canola oil, filtered (1 µm) oil layer, and digested solids (30% H_2O_2) Same process for sludge (after H_2O added)	Visual (optical microscope) ATR-FTIR (> 3 mm). µ-ATR-FTIR (< 3 mm) 37 suspect MPs (4.7% of total) Dark and white fibers not counted due to high blanks. MP estimates are conservative	Wastewater means (MPs/L): Influent: 31.1 ± 6.7 After primary clarification: 2.6 ± 1.4 After secondary treatment: 0.5 ± 0.2 Sludge (MPs/g): Primary: 14.9 ± 6.3, Secondary: 4.4 ± 2.8 97–99% removal (92% by primary treatment) Fibers (mostly cellulose) dominant (65.6%) MPs in wastewater, then fragments (28.1%) and pellets (5.4%). PES main MP fiber (4 red, 1 blue, 1 green). Counts blank corrected	Treats > 180 billion L/year Influent MP loads (FTIR-corrected) of 1.76 trillion/year Estimates: 1.28 ± 0.54 and 0.36 ± 0.22 trillion MPs trapped in primary and secondary sludge, respectively Estimated discharge: 0.03 ± 0.01 trillion MPs/year Of the 4.8% (37/770) of suspect MPs analyzed by FTIR, only 32.4% were plastic
Simon et al. (2018) Denmark 10 major WWTPs: primary and secondary	Inlet and outlet All plants used AS; 1 had RSF 9 plants managed mainly wastewater from households, and one mainly (75%) from industry	Autosamplers for influent (24 h). 1-L sent to lab Effluent collected in bottles and filtered (10-µm) on-site until 3 filters clogged (4.1–81.5 L)	Influent (+ SDS) sieved (500 µm). 200 mL digested (enzyme, WPO) Effluent solids removed from filters by ultrasound and processed similarly	Focal Plane Array FTIR (FPA-FTIR) Reported MP concentrations based on number and mass Colors not reported	Median (range) (MP/L): Influent = 7216 (2223–10,044) Effluent = 54 (19–447) Removal efficiency = 99.3% (98.3% by mass) Identified 176 MPs in raw wastewater and 222 in treated: 393 non-fiber and 5 PES fibers Main polymer types (by number): Raw wastewater: 27% acrylate, 14% PES, 13% PE-PP, 12% PP, and 10% PE Treated: 27% PE, 25% PES, 14% PE-PP, 12% PP, and 12% acrylate	Estimated 3 t/year of MPs (10–500 µm) released in effluent, or 0.56 g MP/(capita year), for Danish WWTPs. < 0.5% of total MP emissions from Denmark Sludge may be significant source. 188 t/year of MPs removed from wastewater in Denmark and presumed trapped in sludge

Table 4 continued

Study	Sample and plant/ study details	Sample collection	Sample[a] preparation	Analysis[b]	Main findings[c]	Comments
Lares et al. (2018) Finland Municipal WWTP CAS process. Also operated pilot-scale MBR	Influents, effluent, and sludge. Lake water near discharge site Sampled every 2 weeks, between 10/2017 and 01/2018	Collected water in 10-L bucket. Sieved (5 mm and 250 µm) water. Solids on 250-µm sieve rinsed into jars Sludge (150–200 mL) collected in bucket or 0.25-L cup was poured into flasks	Water processed by method of Masura et al. (2015), but no density separation. WPO (Fenton's reagent) of dried samples. Dried influent also treated with cellulase (cellulose fiber removal). Samples filtered onto gridded membrane filters and dried	Visual (microscope) µ-FTIR and µ-Raman 5200 MPs classified as 54 groups. Subset of each group examined visually and by µ-FTIR/ µ-Raman	Means (MPs/L water and MP/g sludge [dry wt.]): Influent = 57.6 (± 12.4) Digested sludge = 170.9 (± 28.7) CAS effluent = 1.0 (± 0.4) MBR sludge: 27.3 (± 4.7) MBR permeate = 0.4 Lake water (near discharge) = 0.3 (± 0.1) 98.3% overall removal. Most MPs removed before CAS process; 99.0% before aeration. MBR slightly more efficient (99.4%) PES most common fiber (79% of total MPs). PE most abundant non-fiber (11%)	Treats 10,000 m^3/day (3.65×10^6 m^3/year) Discharge of 10^7 MPs/day 1.3% and 1.4% of all particles and fibers were examined by FTIR and Raman Automatic composite sampling may be a better approach (Talvitie et al. 2017a). Single monitoring event not generally representative
Li, XW et al. (2018d) China 28 WWTPs in 11 provinces of China	79 dewatered sewage sludge samples Collected in 2014 and 2015	Samples stored at 20 °C. Dried at 105 °C (24 h) to determine total solids. Heated at 600 °C (1 h) to determine volatile solids	Homogenized sample Performed NaCl (1.2 g/mL) density separation on 20-g sub-samples (n = 3). Sieved (37-µm) water layer, rinsed solids with distilled H_2O, and digested (30% H_2O_2) sequentially. Poured digest into distilled H_2O, filtered, rinsed, dried	Visual (microscope). SEM on selected samples (~ 10) Transferred suspect MPs (about 10%) for µ-FTIR. Used polymer, additives and synthetic textile libraries	Concentrations (MPs/kg sludge [dry wt.]) Range = 1.6–56.4 × 10^3 Average (n = 79) = 22.7 ± 12.1 × 10^3 Average MP percentages by shape and color (79 samples): Shape: Fiber (62.5%), shaft (14.9%), film (14.0%), flake (7.3%), sphere (1.3%) Color: white (59.6%), red (9.0%), green (2.3%), black (17.6%), orange (3.3%), blue (1.7%), other (6.5%)	Estimated average release (in China) of sludge-based MPs of 1.56 × 10^{14} MPs/year Total sludge production in 2015 estimated at 40 million tons (moisture 80%). Predicted to increase to > 60 million tons in 2020

Table 4 continued

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Lin et al. (2018) China (Guangzhou) 3 large WWTPs Capacities: W1 = 0.55, W2 = 0.20, and W3 = 1.20 million tons/day	Influent and effluent Plant discharges to Pearl River. River water (Table 2 and sediment (Table 3) also collected A^2/O main process	Collected influent (15 L) after coarse screening and effluent (15 L) after sterilization	Filtered immediately in lab (Leslie et al. 2017) Digested (30% H_2O_2, 65 °C, 80 rpm, 24 h) Density separation 24 h in NaCl (d = 1.2 g/mL). Membrane filtered (5 µm)	Visual (optical microscope) Classified fibers, fragments, films, and pellets µ-FTIR with MCT detector on subset of suspect MPs	Concentrations (MPs/L) at W1, W2, and W3: Influent: 4.2, 0.5, and 1.4; Effluent: 2.7, 0.3, and 0.6 Removals: 35.7%, 40.0%, and 57.1% Main shapes (in influent): foams (57%) and fibers (43%) in W1; fibers (60%), fragments, and pellets (20% each) in W2; fragments (57%) and fibers (43%) in W3. Fibers dominant (67–100%) in all effluents. Fragments 33% in W3	A^2/O process included coarse screening, upgrade pumping, fine screening, sedimentation, AS, second sedimentation, sterilization Blanks (lab, field) had little (< 2%) background (< 2 cotton fibers) Polymer confirmed by library match quality > 80%
Gündoğdu et al. (2018) Turkey (Adana) 2 Secondary WWTPs	Influent and effluent Seyhan and Yüreğir plants supply 1 and 0.5 million people 6 days in August 2017	5 L, 24-h composite samples daily. (automatic collection)	Sieved (55-µm) and digested solids by WPO (30% H_2O_2 and 0.05 M Fe(II)). Density separation in NaI (1.8 g/mL) using centrifuge (5 min, 3500 rpm). Filtered (55-µm) supernatant and rinsed. Stored in Petri dish	Visual (microscope) µ-Raman	Mean ± SD (MPs/L), Seyhan and Yüreğir plants, respectively: Influent: 26.6 ± 3.2 and 23.4 ± 4.1 Effluent: 7.0 ± 0.8 and 4.1 ± 0.3 73–79% removal rates 7 polymers identified. PES most common	Influent: 10^6–6.5 × 10^6 particles/day Effluent: 220,000–1.5 × 10^6 particles/day
Dyachenko et al. (2017) United States Secondary WWTP Serves about 680,000 people. Discharge to San Francisco Bay	Effluent collected after secondary treatment, prior to dechlorination and discharge to SF Bay	Collected effluent 24 h as 2-h composite samples. Collected on 4, stacked sieves: 5, 1, 0.355, and 0.125 mm. 5-mm sieve prevented clogs (its (content discarded). Max. of 1 gallon/min	Rinsed sieves (1, 0.355, and 0.125 mm) into 0.25 L jars with DI water. Stored at 4 °C Extracted by adapted NOAA protocol. Digested by catalytic WPO (0.05 M $FeSO_4$ and 30% H_2O_2, 70 °C, 30 min). Added H_2O_2 as needed. Membrane filtered	Visual (microscope) Suspect MPs (355 µm sieve size) transferred to IR transmitting substrate for µ-FTIR	Significant differences between 2-h (peak flow) and 24-h composite effluent samples: Maximum (5 monthly 24-h samples) = 0.09 MPs/gallon (0.02 MP/L) 2-h sample = 0.64 MPs/gal (0.17 MP/L) Mostly fragments. Pellets or microbeads < 10% Cellulose fibers major interference, followed by undigested fatty acids (in 2° WW) 87% recovery of PS beads (200 µm) in spikes	Discharge (based on 24-h sample): 5 × 10^6 MPs/day Discharges about 57 million gallons wastewater daily (216,000 m^3/day, or 7.88 × 10^7 m^3/y) Cellulose fibers confirmed by Raman. Authors emphasized challenges in quantifying MPs in wastewater (no standard methods)

Table 4 continued

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Kalčíková et al. (2017) Slovenia Ljubljana, Lab-scale WWTP	Single stage, mechanical and biological treatment. (sequencing batch reactor [SBR])	Tested 5 body and facial scrubs with 4.82 and 0.74 g of microbeads per100 mL, respectively	None specified PE in 4 of 5 products. Mostly white particles (2 scrubs also had small amounts of red and blue, and 1 had brown, likely husks and shell powders)	IR, SEM Mean microbead size = 37–96 μm by number (by laser diffraction analysis)	Estimated means: 21 par/m^3 discharged 112,500,000 MPs released daily 52% efficiency for sludge. Overall of 77% (based on estimated efficiency of primary treatment)	Smaller particles (up to 60–70 μm) removed in sludge Authors suggest high affinity of PE for negatively charged AS flocks due to positive surface charge
Leslie et al. (2017) Netherlands 7 municipal WWTPs: designated R1-R7 in paper. R7 was testing an MBR (no longer in use)	Influents and effluents at 4 plants (R3, R4, R5, R7). Effluents and sludge at 3 (R1, R2, R7). Effluent at R6 River suspended particulate matter, canals (Table 2), sediments (Table 3)	2-L of influent, effluent, and sludge collected in jars. Stored in dark (4 °C) Sampled 2012–2013	NaCl (1.2 g/mL) density separations (all samples). Used 100-g aliquots water, and 20-g sludge and sediment subsamples H_2O fractions filtered with 0.7-μm glass-fiber filters. Sediment (and biota) filtered onto Al_2O_3 filters (0.2 μm)	Visual (microscope). Counts, shape, and size (10–300 μm, 300 μm–5 mm) μ-FTIR on subset of particles (6% of total) on Al_2O_3 filter	Mean concentrations: Influent means: 68–238 MP/L (range: 20–910) Effluent means: 51–81 MP/L (range: 9–142) Sludge mean (1 plant): 660 (± 410) MP/kg (wet wt.) (range: 370–950) % of MPs in 10–300-μm fraction: effluent: 49% ± 27, influent: 68% ± 20, and sludge: 62% ± 8 Mean retention (in sludge): 72% (SD 61%) (variability not representative of a given plant) Fibers dominant; spheres and foils also found. Spheres and colored fibers confirmed MPs	Discharge (MPs/day): 7.48×10^8–4.32×10^{10} Efflux: 3.37×10^6–2.63×10^8 m^3/year Procedural blanks had (0–3 fibers). Samples blank corrected Could not obtain spectra of 75% of thin colorless fibers (10% of total MPs) or identify black fibers (37.4%)

Table 4 continued

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Mahon et al. (2017) Ireland 7 WWTPs with PEqs from 6.500 to 2.4 million	Sludge from plants using anaerobic digestion (AD), thermal drying (TD), or lime stabilization (LS) Plants received wastewater from industry, stormwater, and domestic sources	3 replicates (30 g) of treated sludge from each plant. Stored at − 20 °C. Dry matter contents from 24% (AD) to 87% (TD) Wore cotton lab coats and nitrile gloves. Covered samples. Cleaned work surfaces with alcohol LS samples (10 g) too oily to extract. Filtered directly onto glass-fiber filter	TD pellets soaked in water 1 week, placed in water bath (30 °C, 24 h), then on shaker (≥ 12 h). Sieved (250 µm), then filtered (212, 63, and 45 µm) a portion of the washed-through fraction to check for MPs. AD and LS sludges soaked in water and sieved (250, 212, and 45 µm) Extracted AD and TD samples in elutriation column. Filtered (250 µm) extract and rinsed filtered material into separatory funnel for $ZnCl_2$ (1 M) density separation. Drained settled matter and filtered remainder onto glass-fiber filter (1.2-µm)	Visual (microscope) 4 size categories (µm): 250 − 400, 400 − 600, 600 − 1000, 1000 − 4000 About 10% of suspect MPs per filter analyzed by µ-ATR-FTIR 20 samples of LS and TD sludges, and pristine MPs analyzed by SEM	Average concentrations: 4196–15,385 MPs/kg sludge (dry wt.) Recovery for elutriation column based on spiked sediments with known MPs: 90–94% for HDPE and 80% for PVC SEM images showed signs of melting/blistering of TD MPs, and shredding/flaking of LS MPs. Higher numbers of smaller MPs in LS samples may be due to shear during processing. Lower MP abundances in AD samples suggests this process reduces MPs Authors reported bias towards larger particles as they were cut into 2 pieces for FTIR and SEM AD samples were dark and heavy with cellulosic material. TD samples had entrained cellulosic material that was difficult to separate by elutriation/density separation	Visual classification as described by Hidalgo-Ruz et al. (2012) Study highlights potential for treatment process to affect MPs, and possible impact of sludge and biosolids use on terrestrial and aquatic ecosystems. It may have implications for regulations on sludge/biosolids Oily LS samples may be due to breakdown of cellulosic matter by alkaline hydrolysis MP degradation by microorganisms in AD systems should be investigated as a potential remediation method
Mintenig et al. (2017) Germany 12 municipal WWTPs in Lower Saxony PEq = 7.0×10^3–2.1×10^5	Effluents, sludge Primary, secondary and tertiary (4 plants) treatments Sampled between April 22 and 29, 2014	Collected effluent (390–1000 L) with custom mobile pumping device with 10-µm stainless steel cartridge filter and flowmeter Filtered treated water onto 0.2 µm Al_2O_3 filter. Collected 500 g (wet wt.) sludge with shovel	Samples stored at 4 °C Sludge: alkaline treatment, neutralization (HCl), followed by density separation (NaCl) Water: multi-step digestion (enzymatic-oxidative) Samples sieved (500 µm). Retained matter stored in petri dish for ATR-FTIR. Fraction < 500 µm further processed, filtered on Al_2O_3 filters and stored in petri dish	ATR-FTIR (> 500 µm) FPA-based transmission µ-FTIR (< 500 µm down to about 20) Included negative controls	Effluents (MPs/L): 0–0.05 (> 500-µm) 0.01–9 (< 500 µm) PE most common MP, in both size classes, and in water and sludge Synthetic fibers ranged from 0.09–1/L and mainly PES (74%), followed by polyamide (PA) (17%) and PP (9%). Most (61%) 'transparent.' Sewage sludge (variable): 10^3–2.4×10^4 MPs/kg (dry wt.). No MPs in fraction > 500 µm. MP < 500 µm in all sewage samples	Estimated annual discharges: 9–400 × 10^7 MP/plant MPs/day: 4.19×10^4 to 1.24×10^7 Annual efflux: 1.9×10^5 to 1.3×10^7 m^3/year Produces 72–3000 t sludge per year. > 50% of sludge from 46 plants in region used for energy 1.24–5.67 × 10^9 MPs/year in sludge. Fibers not counted as background not assessed

Table 4 continued

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Talvitie et al. (2017a) Finland Advanced tertiary WWTP PEquation (800,000) Largest WWTP in Finland	Influent, after pretreatment and after AS process, plant effluent, excess sludge, reject water, and dried sludge AS process had aeration tanks and secondary clarifiers Sampled during a 1-week period (10/2015)	Grab samples (vol. depended on water quality), and automated 24-h composite and sequential (n = 3) samples Except influent, samples filtered onto filters (300, 100, and 20 µm) in custom filtering device (Talvitie et al. 2015) Collected influent, excess sludge, and reject water in a beaker. Dry sludge hand collected	Subsamples of wet sludge, reject water, and dry sludge mixed with 1 L tap water. Filtered with device used for wastewater. All filters stored in Petri dishes until analysis	Visual (microscope) Counted natural and synthetic microliter and color noted Imaging FTIR (FTIRi). Custom textile fiber library Transferred 752 particles from 3 effluent filters (for each day) to ZnSe windows for analysis	Mean microlitter concentrations (par/L): Grab samples: Influent: 380 (± 52.2)–686.7 (± 155.0) Effluent: 0.7 (± 0.6)–3.5 (± 1.3) Composite (24-h) samples: Influent: 390–900 Effluent = 1.4–2.8 Blank: 0.4–0.8 Sludge and reject water: Excess + raw sludge: 76.3 (± 4.3) par/g; 6,3611 (± 3544) par/L Dry sludge: 188 (± 26) par/g Reject water: 12.9 (± 0.3) par/g; 12,867 (± 275) par/L > 99% retention after secondary treatment 66% of fibers were natural Clear PE fragments in effluent were similar to those in scrubs, but most clear particles were hard crystals that could not be identified	Average discharge: 270,000 m^3/day Average MP releases: 1.7×10^6 to 1.4×10^8 MPs/day to Gulf of Finland, Baltic Sea Most (> 97%) microliter removed by pre-treatment; further removal in sludge Produced 60,000 tonnes dried sludge annually. Huge amounts of microlitter released in sludge Identified only 18% of MPs/micro-litter in wastewater. Weak spectral matches due to interfering peaks (1000–2500 cm^{-1}) of biofilms SEM–EDS could assist analysis (e.g., confirm inorganics)

Table 4 continued

Study	Sample and plant/study details	Sample collection	Sample[a] preparation	Analysis[b]	Main findings[c]	Comments
Talvitie et al. (2017b) Helsinki, Finland 4 municipal WWTPs with advanced final treatments PEq = 5×10^4–8.0×10^5 CAS + BAF + DF* (Efflux = 100) CAS + disinfection or MBR* (Efflux = 5) CAS + DAF (Efflux = 7) CAS + RSF (Efflux = 32) Efflux: million m^3/year *pilot	Triplicate samples before and after treatments Primary, secondary, tertiary (BAF, DF, MBR, DAF, RSF) MBR treated primary effluent. RSF, DAF, and DF treated secondary Sampled after CAS unit at 1 plant to compare with MBR Sampled April 2014–August 2015	RSF and DAF wastewater pumped into custom filtering device (300, 100 and 20-µm) Collected DF and MBR samples using taps designed for sampling into device Volume varied with water quality and filter (0.5–1000 L) Also collected automated 24-h composite samples (4.0–27.4 L)	No further processing	Visual (microscope) Imaging FTIR (FTIRi) of representative particles (> 20 µm) Commercial polymer and additives spectral library, and user-made textile fiber library	Influent and effluent (MP/L), respectively, and removal (%): Grab samples (means, $n = 3$) MBR = 6.9 and 0.005 (99.9%) RSF = 0.7 and 0.02 (97.1%) DAF = 2.0 and 0.1 (95.0%) DF10 = 0.5 and 0.3 (10-µm filter) (40%), DF20 = 2.0 and 0.03 (20-µm filter) (98.5%) Composite (24-h) samples (smaller volumes) RSF = 0.4 and < 0.04 (> 90%) DAF = 2.3 and 1.2 (48%) MBR = 3.2 and 0.2 (94%) CAS = 3.2 and 0.5 (84%) PES dominate (mean = 60%), then PE (14%), polyacrylates (7%), PVC (5%), PS (4%), and PP (3%). Most PES MPs were textile fibers, and PE MPs were mainly microbeads	Discharge = 1.75×10^6–8.22×10^7 MPs/day Efflux = 8.03×10^2–8.82×10^7 m^3/y (Sun et al. 2019) Advanced final-stage treatment can substantially reduce MPs; needed to remove small MPs (< 100 µm). Small fraction should be included to assess releases 13 polymers identified: PES, PE, PP, PS, PU, PVC, PA, acrylamide, polyacrylate, alkyd resin, polyphenylene oxides, ethylene vinyl acetates

Table 4 continued

Study	Sample and plant/ study details	Sample collection	Sample[a] preparation	Analysis[b]	Main findings[c]	Comments
Vollertsen and Hansen (2017) Denmark 10 WWTPs	Influent and effluent (20 samples for each of 3 sampling events) Anaerobic digested sludge from 5 plants 10 farmland soils: 5 treated with sludge and 5 untreated (See Table 3)	Collected 1-L raw influent as flow proportional 24-h samples using autosamplers at plant Effluent collected in bottles. Filtered on-site with 3 steel filters (10-µm) until filters clogged (4.1–81.5 L) Collected about 1 kg sludge (at dewatering unit after sludge digestion)	Pre-filtered 1 L with 500 µm sieve. 1 mL SDS (150 g/L) added before wet sieving. Treated 200 mL of filtrate (by hydrolysis: 48 h, 40 C, cellulase, and oxidation: 50% H_2O_2 + catalyst), then concentrated MPs in 5 mL ethanol Effluent filters treated similarly to influent, and MPs concentrated 5 mL ethanol 0.1 g sludge (dry matter) suspended in 50 mL water and processed similarly to influent. MPs concentrated in 5 mL ethanol Analyzed particle 20–500 µm	µ-FTIR-FPA imaging. 0.02–0.3 mL of ethanol suspension applied to slide (Kevley MirrIR) for reflectance mode and to ZnSe transparent slide for transmission (700 × 200 µm area) Reflectance on particles ≥ 80 µm, and transmission on smaller particles Fast scan to reject non-MPs (MPs absorb in 2820–2970 cm^{-1} range). Unrejected particles analyzed	Large variability between plants: Influent: mean = 127,000 MP/L (8.0 mg/L), range = 13,000–442,000 MP/L (0.22–29.6 mg/L), median = 86,000 MP/L (5.9 mg/L) Effluent: mean = 5800 MP/L (0.034 mg/L), median = 6400 MP/L (0.016 mg/L) 92.6% retention by number, 99.7% by mass Median size in effluent (42.5 µm) 20% smaller than in influent (50 µm) Particle Types (% by number): Influents (181 particles) mainly polyamide/nylon (76.8%) and PE (13.3%). 6.6% Zn stearate, 2.2% PP, and 0.6% each PVC and PP-PE copolymer Effluents (150 particles) similar: nylon (77.3%) and PE (16.7%), 4% Zn stearate, 2% PP Digested wastewater sludge: MPs about 2% of total dry matter (25–30%) Mean = 169,000 MP/g (4.5 mg/g) Median = 158,000 MP/g (6.5 mg/g) Identified MPs (29): 65.5% PE, 31,0% nylon, and 3.4% PP by number (79.2% PE, 20.3% nylon, and 0.5% PP by mass)	Cannot identify MPs with Zn stearate coating (used on household plastics and others), but coating indicates MP Authors concluded MP mass is better for assessing removal because it is conserved, but number important in assessing impact Lassen et al. (2015) estimated total Danish MP emissions (1 µm–5 mm) to aquatic environment of 600 to 3100 tons/year. Relative to authors' estimate of MPs in effluents (11 tons/year in 20–500 µm range) they concluded that wastewater from Danish WWTPs is a minor fraction of total MP emissions Mass concentrations based on MP size (estimated volume), polymer density, and scaling to total sample

Table 4 continued

Study	Sample and plant/study details	Sample collection	Sample[a] preparation	Analysis[b]	Main findings[c]	Comments
Ziajahromi et al. (2017) Australia Sydney 3 major WWTPs: 1 primary, 1 secondary, and 1 tertiary with reverse osmosis (RO) treatment PEq = 0.15–1.2 × 10^6	Wastewater at different treatment stages Population and plant capacity (ML/day): Primary = 1.2 million and 308; Secondary = 67,130 and 0.48; Tertiary = 150,870 and 0.28 Sampled over 3 days (10/2015)	Filtered 3–200 L with stacked-sieves (500, 190, 100, and 25-µm), depending on water type. Removed 25-µm screen if blocked (e.g., clogged after 8 L of primary effluent). Tertiary and RO samples were 200 L. Sampled 1-h at max. of 10 L/min. Stored screens in foil-wrapped petri dishes	Samples concentrated to 100 mL in 90 °C oven. Digested (30% H_2O_2) and treated by density separation in NaI (1.49 g/mL) Centrifuged (15-mL tubes, 5 min, 3500xg). Filtered (25 µm) buoyant particles and rinsed Stained with Rose Bengal to reduce false positives	Visual (microscope) ATR-FTIR on particle subset	Mean effluent concentrations (MPs/L): Primary = 1.54, Secondary = 0.48, Tertiary = 0.28, RO = 0.21 > 90% removal of MPs in primary effluent 100% removal of particles > 190 µm PET fibers and irregular PE particles most common. Attributed to clothing and personal care products. No MPs in controls Main (75% of suspect) MPs in 1° effluent of tertiary plant: PE (42%), PET (36%), PS (15%), and PP (8%). Most were white and blue irregular particles, followed by black fibers (100–190 µm) PET fibers 65% and 88% of MPs in tertiary and RO samples, respectively. 75% of suspect MPs in tertiary and 60% in RO samples confirmed	Treated 13–308 × 10^6 L/day Discharge = 3.6–460 × 10^6 MP/day Some differences in plastic composition along treatment train may relate to different sample volumes Despite rigorous protocol, 22–90% of suspect MPs were non-plastic (e.g., castor oil, stearic acid/stearate. Authors stressed need for careful analyses and long-term monitoring Recoveries of PS MPs (250–500 µm) in spikes were 92% (25-µm screen) and 99% (500-µm screen)

A^2/O anaerobic, anoxic, oxic (aerobic), *AS* activated sludge, *BAF* biologically active filter, *CAS* conventional activated sludge, *DAF* dissolved air flotation, *DF* disc filter, *FPA* Focal plane array (detector), *HCl* hydrochloric acid, *H_2O_2* hydrogen peroxide, *MBR* membrane bioreactor, *MCT* mercury–cadmium–telluride, *µ-ATR-FTIR* Fourier transform infrared spectroscopy coupled with an attenuated total reflectance microscope objective (µ-ATR), *NaCl* Sodium chloride, *par* particles, *PEq* population equivalent, *PTFE* polytetrafluoroethylene, *RSF* rapid sand filtration (filters), *SDS* sodium dodecyl sulfate, *SEM* scanning electron microscopy, *WPO* wet peroxide oxidation, *WWTP* wastewater treatment plant

largest plant, which employed primary clarification, had the highest efficiency (97.6 ± 1.2%.), while the two smaller facilities averaged 85.2 ± 6.0% and 85.5 ± 9.1%, suggesting that retrofitting secondary plants with primary clarifiers could improve removal. Source modeling indicated that MP fiber loads in influent were consistent with laundering textiles. The estimated total discharge from all three plants was 500 million to 1 billion MPs/day, equivalent to an annual discharge of 0.34–0.68 g MP per capita in treated wastewater. This release may account for < 0.1% of the annual plastic input to this metropolitan area's surface waters on a mass basis, considering mismanaged waste. However, as discussed, N&MPs in wastewater may pose unique toxicological risks because of their potential for sorption of toxins and biological uptake.

Yang et al. (2019) reported mean concentrations in influent and final effluent of 12.03 ± 1.29 and 0.59 ± 0.22 MPs/L, respectively, for samples collected at an advanced sewage treatment plant (STP) in Beijing. The plant treats mainly domestic effluents using: an aerated grit chamber, a primary sedimentation tank, A^2/O (anaerobic, anoxic, and aerobic) treatment followed by secondary sedimentation, and a final series of advanced processes (denitrification, ultra-filtration, ozonation and UV. Primary aerated grit treatment removed 58.84 ± 8.05% of the MPs. Subsequently, 54.47 ± 14.73% and 71.67 ± 11.58% of the MPs were removed after the A^2/O and final treatments, respectively. An overall reduction > 95% was found. The authors indicated further improvement would be difficult as advanced technology was being used. For example, post-filtration units can lower fiber concentrations (Mintenig et al. 2017), and this technology (ultra-filtration) was used after secondary sedimentation. Microfibers were dominant (85.92%) in influents and effluents, and > 60% were removed by the aerated grit chamber. The authors suggested that an aerated grit chamber treatment might be an effective alternative for reducing microfiber loads. Of the 18 polymers identified, PET, PES and PP accounted for > 70% of the detected MPs. The PET and PES MPs were mainly fibers, thought to originate from domestic washing. In contrast to some studies, PE accounted for just 1.64% of the total MPs and was found in only one of three sample sets. The small PE fraction relative to studies in Europe (average = 14%) (Talvitie et al. 2017a) and the US (> 90%) (Carr et al. 2016) may relate to differences in MP use between China and Europe/US. Some (about 2%) non-fiber MPs contained both PE and PP, and MPs (1.30%) identified as synthetic rubber were found in effluents, possibly from tire wear. Most MPs were black (37%), transparent (34%), or blue (12%), indicating different sources. Particles < 50 μm could not be isolated because high levels of suspended particulate matter quickly clogged the filters. Also, for confirmatory analysis by μ-FTIR, collected MPs were isolated by hand, under a microscope, and MPs < 50 μm could not be transferred for analysis. The authors noted that a previous study (Erni-Cassola et al. 2017) found higher MP concentrations when filters with a smaller particle capture size were used and recommended future work on these smaller MPs.

As mentioned, direct comparisons of studies may not be valid due to procedural differences, but all studies of WWTPs have found significant MP reductions. Primary treatment alone removed an average of 65% (50–78%) of MPs in influents. Secondary

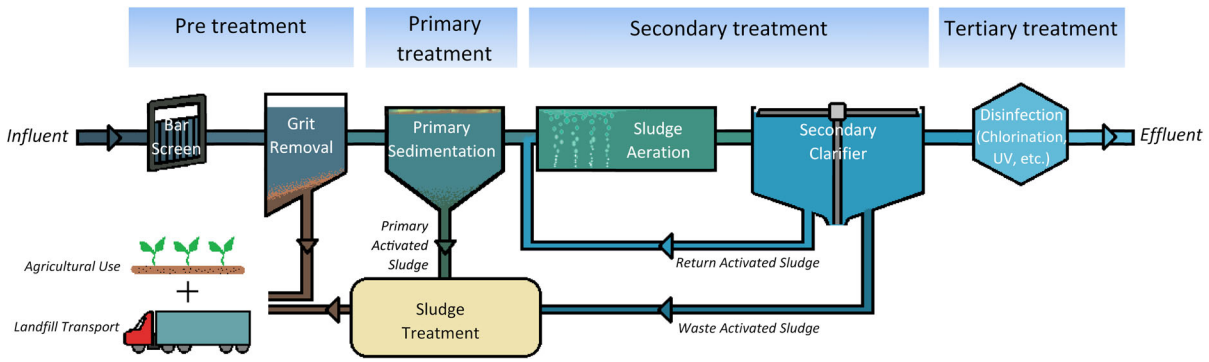

Fig. 2 Illustration of serial treatment stages of a tertiary wastewater treatment plant

treatment increased removal to 97.7% (96–99%), while a final tertiary treatment averaged 94% removal (range = 90–99.9%) (Burns and Boxall 2018). Overall, typical removal efficiencies range from 96 to 99%. For example: 99% removal after mechanical, chemical and biological treatments (15,000 particles/L in influent and 8 in effluent) (Magnusson and Norén 2014); 96% after secondary treatment (467 textile particles/L in influent and 16/L in effluent) (Talvitie and Heinonen 2014); and 98% after tertiary treatment (430 particles/L and 180 textile fibers/L in influent, and 4.9 and 8.6, respectively, in effluent), with estimated daily discharges of 2.3×10^9 particles and 1.3×10^9 fibers, based on a flow rate of 270 million L/day (Talvitie et al. 2015). Carr et al. (2016) reported 99.9% removal for tertiary U.S. WWTPs, and a total daily discharge of 9.3×10^5 MPs. Murphy et al. (2016) reported 98.4% removal for a secondary plant in Scotland. vollertsen and Hansen (2017) reported releases of 0.3% of influent MPs (i.e., 99.7% removal) in Danish WWTPs, with effluent MPs being mostly fibers and fragments.

Though treatment technologies were not designed for MP removal, retentions of 96%–99.9% represent high capture efficiency. However, even small levels in effluents can result in substantial MP releases due to the high discharge volumes (e.g., Conley et al. 2019; Magni et al. 2019; Ziajahromi et al. 2017). Nevertheless, their relative contribution may be small (Burns and Boxall 2018; Conley et al. 2019). Vollertsen and Hansen (2017) estimated that effluents contribute only 3% of the total MP load to the environment. Based on modeling, recipient rivers were expected to retain the most common MP sizes (Besseling et al. 2017). If so, freshwater sediments are the most relevant compartment to consider with respect to discharges to rivers (Burns and Boxall 2018; Nizzetto et al. 2016a). However, if most (> 95%) MPs are trapped in sludge (Bayo et al. 2020; Magni et al. 2019; Magnusson and Norén 2014; Michielssen et al. 2016; Talvitie et al. 2015, 2017a; Yang et al. 2019), sludge may be a more relevant route of environmental release (Alimi et al. 2018; Magni et al. 2019; Nizzetto et al. 2016b; Rillig et al. 2017a; Talvitie et al. 2017a; Zubris and Richards 2005). Estimates of the MP contents of sludge from 28 WWTPs across 11 provinces in China ranged from 1.60 to 56.4×10^3 MPs/kg (dry wt.), with an average ($n = 79$) of $22.7 \pm 12.1 \times 10^3$ MPs//kg (Li, XW et al. 2018d). For China, the average environmental release of sludge-based MPs was estimated at 1.56×10^{14} particles per year (Li et al. 2018d). According to an estimate for Europe and North America, the annual amount of MPs transferred from WWTPs to agricultural soils as biosolids exceeded the total burden in oceans (Nizzetto et al. 2016b). Organic fertilizers from biowaste composting and digestion also may be an important source of environmental MPs: 20–24 and 14–895 MPs/kg material were found in biowaste composts and digests, respectively (Weithmann et al. 2018).

4 Current status and research needs

The environmental impacts and human health risks of N&MPs have attracted increasing scientific, public, and regulatory interest over the past decade. Microplastics have been found in surface waters, sediments, and WWTPs globally, with fibers and fragments being dominant. Growing evidence of environmental N&MPs and their uptake by a wide range of organisms (e.g., Curren and Leong 2019; Eerkes-Medrano et al. 2015; Lusher 2015) has drawn attention to these emerging, persistent contaminants. Concerns about ecotoxicity and other adverse effects have prompted numerous studies, especially of aquatic systems. As discussed, findings of concern include: environmental releases by WWTPs, adverse effects on aquatic organisms, bioaccumulation and amplification in the food chain, sorption/release of toxic organic compounds and metals, biofilms with toxic bacteria (Curren and Leong 2019; McCormick et al. 2016), ingestion and translocation, and the presence of MPs in human stool. Efforts to ban products such as microbeads were a positive step, but major releases of plastic items continue, including microbeads in unregulated products. Moreover, although microbeads have been a focus of attention, synthetic fibers may be a much larger issue (Boucher and Friot 2017; Henry et al. 2019; Ziajahromi et al. 2017).

The relative contributions of N&MPs from terrestrial and freshwater sources are not well understood, slowing advancement of source management. Fahrenfeld et al. (2019) reviewed the evidence and methods for possible source apportionment of MPs in freshwater systems, based on MP characteristics and mass balance techniques. The authors concluded that the data indicated potential for differentiating some

sources within studies, based on differences in polymer types and shapes, but that clear cross-study patterns were lacking (Fahrenfeld et al. 2019). Major obstacles to source apportionment, and to N&MP studies generally, include challenges in identifying polymer types and surface contaminants; different classifications of particle shapes; lack of data for terrestrial sources; and poor understanding of fate, transport, and weathering processes. Accurate data on particle composition and size are key to effective pollution prevention and mitigation. A better understanding of the actual risks of N&MPs is needed to establish effective waste management policies and a sound regulatory framework for plastic pollutants. Priority knowledge gaps are discussed below.

4.1 Potential risks

Further study is needed to assess their ecotoxicological and human health risks, especially of NPls (Andrady 2011; Curren and Leong 2019; de Sa et al. 2018; Galgani et al. 2015; Hidalgo-Ruz et al. 2012; Lehner et al. 2019; Peng et al. 2017; Smith et al. 2018; Triebskorn et al. 2018; Vethaak and Leslie 2016; Wang et al. 2018, 2019). Studies of a wide range of organisms, polymers, particles sizes, and exposure times were recommended to better assess potential effects (Browne et al. 2008; Wright and Kelly 2017). Several major knowledge gaps must be addressed to assess the potential environmental and human health risks of N&MPs. There is a critical need to understand the mechanism(s) of action and ecotoxicological effects of *environmentally relevant* N&MPs (de Sa et al. 2018). Though N&MPs are a negligible fraction of plastic waste by mass, their potential for efficient sorption of toxins and biological uptake due to small size may present unique toxicological risks, especially for discharges from WWTPs (Conley et al. 2019). In particular, NPls have much higher surface area for chemical sorption and their uptake by organisms, tissues, and cells is more likely. However, high concentrations of natural particles with similar size ranges have been reported. Triebskorn et al. (2018) found that < 10 in 10^6 particles in river surface water were plastics. These particles also burden ecosystems and their consideration may be relevant to understanding the relative ecotoxicological risks of N&MPs. However, as recognized by others, the authors acknowledged that the relevance of toxicity assays is suspect due a mismatch between the concentrations and types of materials tested and those in the environment (e.g., differences in particle types, sizes, surface contaminants, and concentrations).

The possible impacts of N&MPs on aquatic organisms and humans depend on their environmental fates. In the case of nanomaterials, aggregation and deposition greatly affect availability in aquatic environments. Because different nanomaterials can have widely different surface chemistry (e.g., adsorbed species, extent of ionization) and colloidal stability, and their properties depend on environmental conditions (e.g., pH, ionic strength), it is not possible to generalize the behavior of these materials (e.g., Wiesner et al. 2009). Similarly, N&MPs undergo transformations in the environment, making their fate and impacts difficult to predict. However, trophic transfer of N&MPs through the food web has been observed (e.g., Farrell and Nelson 2013; Setala et al. 2014), and inhalation/ingestion are human exposure routes (Cox et al. 2019; Toussaint et al. 2019; Wright and Kelly 2017). Contamination of drinking water and food and beverage items has raised concerns about the possible human health effects of N&MPs, but uncertainty in the existing data has impeded reliable risk assessments. Based on 50 studies, Koelmans et al. (2019a) concluded that MPs are present in water (and some foods) but found no evidence of harmful effects. However, the authors reported inadequate overall data quality. Limitations mainly relate to a lack of standardized methods and quality assurance measures. In the field of toxicology, the validity of studies is commonly assessed by consensus criteria (e.g., Klimisch score, CRED [Criteria for Reporting and Ecotoxicity Data]) (Hermsen et al. 2018). Such criteria have not yet been established for studying the impacts of plastic debris (Koelmans et al. 2017).

4.1.1 Dietary intake in humans

Relatively little is currently known about the daily intake, biodistribution, and physiological persistence of N&MPs. The extent of contamination in the food supply is uncertain, but MPs and plastic-related contaminants have been found in seafood (Li et al. 2015, 2016; Van Cauwenberghe and Janssen 2014), honey and sugar (Liebezeit and Liebezeit 2013), table salts (Karami et al. 2017a; Yang et al. 2015), beer (Liebezeit and Liebezeit 2014), bottled water

(Mintenig et al. 2014; Ossmann et al. 2018; Schymanski et al. 2018), and drinking water and its freshwater (Koelmans et al. 2019a) and groundwater sources (Mintenig et al. 2019). Textiles and packaging can be sources of N&MPs/additives in food and beverages. Given the widespread contamination, it is not surprising that MPs (PET) were found in all human stool samples collected in a preliminary study of eight international participants. Particles were attributed to bottled water consumed as part of the study (Parker 2018), but airborne fibers may have contributed. For example, Catarino et al. (2018) reported that the risk of plastic consumption was greater from exposure to airborne fibers than seafood (mussels).

High-quality data on N&MP abundances in food, drinking water, and other beverages are needed. Available evidence on N&MPs in a normal diet is scarce and unreliable. Toussaint et al. (2019) used an indirect approach to access exposure. They reviewed peer-reviewed publications (since 2010) that document the presence of N&MPs in food products, and in animals (> 200 species) in the human food chain. Methods and quality criteria applied in the studies were considered. Overall, the authors concluded that precise data to accurately assess dietary exposure to N&MPs cannot be produced until standardized methods are available. Meanwhile, based on an assessment of human consumption, Cox et al. (2019) reported that avoiding bottled water might significantly reduce MP exposure. They further recommended studies of MP contamination in food groups representing major sources of nutrition globally (grains, vegetables, beef, poultry). Where existing data are suspect, due to use of older (unreliable) methods, further studies were recommended. The new data generated will provide better estimates of human consumption of plastic particles, ultimately permitting improved assessments of the potential ingestion risks (Cox et al. 2019).

4.1.2 Nanoplastics

Nanoscale particles can have markedly different properties than larger forms of the same material. The cytotoxic effects (e.g., AshaRani et al. 2009; Lewinski et al. 2008) and potential environmental impacts (e.g., Klaine et al. 2008; Lead et al. 2018) of nanomaterials are well documented. Similarly, NPls may pose greater risks than their MP counterparts. Small size and hydrophobicity may result in cell entry and cytotoxicity (Bothun 2008; Hoet et al. 2004; Lehner et al. 2019; Oberdorster et al. 2005; Rasch et al. 2010; Yousefi and Tufenkji 2016; Yousefi et al. 2016). Based on the definition of 'nanoparticle,' manufactured nanoscale (≤ 100 nm diameter) plastics have been used in laboratory studies, polystyrene latex (PSL) beads in particular (Bergami et al. 2017; Lehner et al. 2019; Lu et al. 2016; Nolte et al. 2017; Rist et al. 2017). These studies may be relevant to PSL beads but their relevance to environmental NPls is unclear (e.g., Lehner et al. 2019; Phuong et al. 2016) as nanomaterials are not representative of environmental NPls. The former have specific properties and sizes by design (Brar et al. 2010; Gigault et al. 2016, 2018; Ju-Nam and Lead 2008; Weinberg et al. 2011), while environmental NPls have highly polydisperse sizes and compositions (Gigault et al. 2016, 2018; Lambert and Wagner 2016; Ter Halle et al. 2017). Also, NPls typically occur as colloids with heterogeneous aggregates of natural/anthropogenic materials (Hotze et al. 2010; Huffer et al. 2017), with structures and surfaces that depend on environmental conditions (e.g., pH, salinity, organic matter (Li et al. 2018a). As mentioned previously, their presence in polluted waters may pose special risks due to sorption of a variety of toxins.

4.2 Monitoring needs

As emphasized in this review and numerous publications, a lack of standardized methods remains a major impediment to risk assessment and N&MP studies generally. Targeted particle sizes and analytical procedures have varied widely and been error prone, limiting available data on environmental concentrations and size distributions, especially for NPls and very small MPs. In some cases, concentrations may be significantly underestimated. Conkle et al. (2018) reviewed 50 aquatic surveys and reported that about 80% accounted only for MPs ≥ 300 µm. They also analyzed 770 personal care products containing microbeads and found 1649–31,266 MPs/g of product (1.9–71.9 mg/g), > 95% of which were < 300 µm. Studies employing screens with a size cut > 125 µm also can underestimate N&MPs (Conley et al. 2019). Further, common analytical techniques have inadequate size resolution (e.g., about 10–20 µm for µ-FTIR). Size distributions in several recent studies appeared to be dominated by relatively small particles (e.g., 81–92% 1–10 µm [(Pivokonsky et al. 2018)],

96% 4–20 μm [(Triebskorn et al. 2018)], 61% 10–300 μm [(Leslie et al. 2017)], and 73% 50–500 μm [(Yuan et al. 2019)]. However, other investigators (Di et al. 2019; Luo et al. 2019; Wang et al. 2018) reported a majority of MPs in larger size classes (Novotna et al. 2019). Improved monitoring methods are needed to resolve apparent discrepancies and generate accurate data, especially for NPls and small MPs as these small particles may pose greater risks and concentrations could be much higher than currently realized. Combined with suitable protocols to collect and isolate N&MPs from environmental samples, emerging analytical techniques are expected to advance environmental studies of N&MPs. Research gaps include dietary intake, sources, urban watersheds, environmental transformations (e.g., weathering, and interactions with contaminants, natural materials, and animals) and their impact on particle transport and toxicity, source apportionment, degradation of 'biodegradable' polymers (and others), NPls (all matrices), and WWTP technologies.

4.3 WWTPs

4.3.1 Long-term studies

Studies of urban watersheds, especially those with high populations, highlight WWTPs as potential sources of N&MPs. Most studies have relied solely on visual identification of MPs, with counts based on specific physical characteristics (e.g., Conley et al. 2019). Confirmation by μ-FTIR and μ-Raman is becoming more common, though only a sub-sample of MPs is usually examined. Long term studies are needed to capture temporal variations, using improved methods and sampling strategies (Conley et al. 2019; Lares et al. 2018; Leslie et al. 2017). Seasonal and weather-related variations occur, and diurnal variations in influents may exist. Automated composite sampling could provide more representative samples, but low sampling volumes (10–20 L) may cause false negatives, and care must be taken to avoid contamination by textile fibers, which may be significant (30%).

Additional research on treatment processes is needed to better understand factors that affect N&MP retention. A comparison of three unit processes indicated that granular sand filtration and MBRs are more efficient in removing small anthropogenic litter (SAL or 'microlitter'), though the final effluent had higher fiber content than effluent treated by activated sludge (Michielssen et al. 2016). Studies of WWTPs that treat water from combined sewers may be especially relevant as MPs in stormwater may differ from those in municipal wastewater, and because high MP levels were reported for combined systems in 17 US municipalities (Mason et al. 2016). In addition to contamination concerns, N&MP loads in WWTPs could adversely impact their operational stability (Enfrin et al. 2019a). They enter different treatment stages to varying extents, depending on size and composition. Shear forces during processing may generate smaller, undetectable particles. Degradation reportedly increased the number of NPls/MPs in water by an order of magnitude (Enfrin et al. 2019b). Their impact on different stages is unclear, but membrane filtration processes could be affected (Enfrin et al. 2019a). Considering degradation and removal, the total N&MPs discharged can be double the number in influent, with significantly increased particle surface area and porosity for sorption organic pollutants, metals, and pathogens (Enfrin et al. 2019a; Li et al. 2019). Systematic studies of WWTPs using standardized methods could have important implications for source management. New water treatment processes and approaches to limit N&MPs in water and wastewater are needed, to maintain water quality and reduce risks to ecosystems (Enfrin et al. 2019a). Methods to trap microfibers at their sources (e.g., laundry) could greatly reduce levels in influents and accumulation in sludge. New fabrics with reduced shedding also could provide significant reductions.

4.3.2 Sludge and fibers

The potential ecological risks of MP-contaminated sludge should be assessed prior to agricultural or other applications. For such assessments, improved methods are needed for processing sewage sludge and wastewater with high levels of suspended solids. Even after rigorous oxidation, naturally occurring organic debris may remain in the samples (Dyachenko et al. 2017; Lares et al. 2018). Incomplete digestion of natural fibers has caused potential bias, especially in studies based solely on visual identification of MPs. An acid digestion step post oxidation (H_2O_2) was reported to reduce interferences of cotton and semi-synthetic textiles, but it also can reduce any acid-sensitive

MPs (Conley et al. 2019), and any residual debris may confound results. In some studies, no digestion or separation steps were used to remove non-plastic SAL, rather this microlitter was designated as a category of interest (Michielssen et al. 2016; Talvitie et al. 2017a). Conley et al. (2019) reported thin fibers with white/translucent appearances as suspect MPs. Identification by μ-FTIR-ATR was not possible due to problems with ATR crystal contact, especially with particles < 500 μm (Mintenig et al. 2017). Similarly, Leslie et al. (2017) could not obtain spectra for 75% of the thin transparent fibers in some samples. Staining procedures have been applied to assist fiber identification but improved spectral matching through use of a custom library could better expedite MP identification. As discussed, Talvitie et al. (2017a) obtained poor spectral matches for natural textile fibers with commercial libraries. After generating a custom library, using model fibers, all fibers were successfully identified. Improved FTIR, Raman, and/or other techniques to identify a wide variety of fibers, including plastic, natural, and semi-synthetic materials, especially for smaller particles, is a priority research need (e.g., Lares et al. 2018). Semi-synthetic fibers, cellulosic materials, and other microliter also may have risks (Remy et al. 2015). Selective measurement could help identify and mitigate sources, if appropriate, and allow assessment of the risks of different materials.

5 Conclusions

The potential impacts of N&MPs have been a major focus of plastic pollution. They have been found in air, water, soil, food and beverages, biota, and human stool. Plastic debris can be transported long distances by air and water. During transport, it can absorb toxins and exchange between environmental compartments, with potential for harm to many species. Most N&MPs in marine environments originate from land-based activities. Some major MP types in marine and freshwater systems have been identified but the sources are uncertain. Studies of urban watersheds are especially relevant as most plastic pollution originates in urban areas and relatively few have been reported. Further research is needed to identify the types, sources, and abundances of N&MPs in urban watersheds, and to better understand their transport, transformations, fate, size distributions, associated toxins, and potential toxicity. In future studies, particular attention should be paid to transformations that affect transportation. Studies of NPls are especially needed. Their small size and high surface area may pose unique risks and little is known about this fraction, mainly due to the monitoring challenges for nano and sub-μm scale particles. Methods applied to other fields (e.g., biology, nanomaterials) also may be useful for NPl measurement.

Abundances of MPs in freshwaters are highly variable (e.g., 10^{-5}–10^{5}/L), but MP pollution is widespread and tends to be much higher in urbanized areas. Wastewater treatment plants generally have high overall removal efficiencies for MPs (e.g., $\geq 98\%$), but particle releases can be substantial due to the high discharge volumes, and through sludge applications. Atmospheric fallout of N&MP pollution in urban areas, especially fibers, also contaminates freshwaters (and land) and has potential for long-range transport.

Studies of N&MPs and plastic-related contaminants in food, beverages, and other consumer products are needed. Long-term, systematic studies of WWTPs also are needed to better characterize N&MPs, evaluate treatment technologies and the impact of particle capture, and assess the risks of contaminated sludge. A lack of standard methods and reference materials have slowed progress in determining the environmental prevalence and impacts of N&MPs. To properly conduct these and other studies, robust, standardized protocols to collect, process, and analyze N&MPs in complex matrices (soil, sediment, sludge, wastewater, biological, etc.) are a critical research priority. Representative reference materials are essential to method validation, QA/QC, interlaboratory comparisons, and environmentally relevant toxicity studies. Automated, accurate methods that maximize N&MP recovery and identification will permit comparisons across studies and advance our understanding of their potential risks. User-built spectral libraries can assist particle identification. Accurate measurement and source identification in urban watersheds are necessary to reduce N&MPs in all environments.

Ultimately, the most effective way to reduce N&MP pollution is to reduce plastic production, use, and improper disposal. Concurrent, sustained efforts, including improved waste management and development of new materials and technologies (e.g.,

recycling, waste-to-energy) will be required, evolving towards a circular economy to preserve resources and minimize waste (Koelmans et al. 2019b). This will be aided by future studies involving mass balance between production, recycling, and release to the environment of all plastics. Regardless of whether current levels pose ecological and human health risks, plastic pollution is an enormous, escalating problem. Left unchecked, environmental levels of N&MPs are expected to increase due to increasing plastic production and use, projected emissions, and continuous fragmentation of long-lived plastic reservoirs.

6 Supporting information

Additional data table for WWTP studies prior to 2017.

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