Occurrence, fate and fluxes of plastics and microplastics in terrestrial and freshwater ecosystems

Theresa Schell[1], Andreu Rico[1], Marco Vighi[1]

[1]IMDEA Water Institute, Science and Technology Campus of the University of Alcalá, Avenida Punto Com 2, 28805, Alcalá de Henares, Madrid, Spain

Corresponding author:
Theresa Schell
IMDEA Water Institute, Avda Punto Com 2, 28805, Alcalá de Henares, Madrid, Spain
Email: theresa.schell@imdea.org
Phone:+34 918305962

E-mail: andreu.rico@imdea.org, marco.vighi@imdea.org

Abstract

Plastics and microplastics are nowadays ubiquitously found in the environment. This has raised concerns on possible adverse effects for human health and the environment. To date, extensive information exists on their occurrence in the marine environment. However, information on their different sources and their transport within and across different freshwater and terrestrial ecosystems is still limited. Therefore, we assessed the current knowledge regarding the industrial sources of plastics and microplastics, their environmental pathways and load rates, and their occurrence and fate in different environmental compartments; thereby highlighting important data gaps which are needed to better describe their global environmental cycle and exposure. This study shows that the quantitative assessment of the contribution of the different major sources of plastics, microplastics and nanoplastics to aquatic and terrestrial ecosystems is challenged by some data limitations. While the presence of MPs in wastewater and freshwater is relatively well studied, data on sediments and especially soil ecosystems are too limited. Moreover, the overall occurrence of large-size plastics, the patterns of microplastic and nanoplastic formation from them, the presence and deposition of plastic particles from the atmosphere, and the fluxes of all kinds of plastics from soils towards aquatic environments (e.g. by surface water runoff, soil infiltration) are still poorly understood. Finally, this study discusses several research areas that need urgent development in order to better understand the potential ecological risks of plastic pollution, and provide some recommendations to better manage and control plastic and microplastic inputs into the environment.

Keywords: plastics, microplastics, nanoplastics, occurrence, environmental exposure, risk management, remediation, surface waters, wastewater, sludge, sediment, soil, agriculture, air, environmental fate, emissions, pollution, water quality, ecotoxicology, impacts, sampling methods, anthropogenic activity, plastic emission, persistence, synthetic fibres

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1. Introduction

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

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

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

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

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

Despite physical effects, some MaPs and MPs have been reported to induce endocrine disrupting effects (Rochman et al. 2014) due to the release of plastic additives such as phthalates, chlorinated paraffins and bisphenols (Stenmarck et al. 2017). Hydrophobic pollutants (e.g. some pesticides, PCBs, PAHs) can also be adsorbed to plastics and may be released into the body of the organisms after ingestion, leading to the so-called Troyan Horse effect (Teuten et al. 2009; Koelmans et al. 2016; Crawford and Quinn 2017; Bouhroum et al. 2019). → add something on plastic being sink for chemicals upon ingestion Furthermore, MPs could not only act as carriers for chemicals, but also can transport bacteria or pathogens attached to them (Keswani et al. 2016; Kirstein et al. 2016) across different environmental compartments and regions.

The continuous emission patterns and the breakdown of plastic litter into smaller fractions in the environment may contribute to future concentrations that are orders of magnitude higher than the ones currently monitored (Everaert et al. 2018), thus contributing to a yet uncertain risk scenario. Policies dedicated to control emissions and manage risks of MaPs, MPs and NPs in the environment require a proper understanding of the main emission routes, the current exposure levels and the fluxes among environmental compartments. The available literature describing the exposure and impacts of plastics in the environment has so far mainly focused on specific emission routes and local monitoring campaigns, and do not provide a comparative assessment of the whole occurrence, transport and fate of plastics in different compartments, which is key to identify suitable management actions.

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

2. Environmental sources of plastics

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

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

Plastics with short-term use expectancy

Single-use items are mainly formed by packaging material, which is the biggest plastic sector worldwide (almost 36% in 2015; Fig. 1) and accounts for almost 50% of the generated plastic waste (Geyer et al. 2017). The vast majority of packaging plastics are PE, PP and polyethylene terephthalate (PET) (Geyer et al. 2017). Except for refillable PET bottles used in some countries, packaging is single-use with a life span of less than six months. Most foods are wrapped in plastic and single-use plastic bags have been widely used all over the world due to their convenience, availability and low price. Plastic bags are known to cause severe environmental and health problems, especially in countries without proper waste management (Adane and Muleta 2009). Thus, many countries have put bans or levies in force to reduce their use or to encourage voluntary reductions (Xanthos and Walker 2017). Many African countries, for instance, have banned single-use plastic bags, while the EU Directive 2015/720 encourages member states to reduce the number of

‘’lightweight’’ carrier bags by 2025. Those bans and restrictions have already reduced the plastic bag use drastically in some countries (e.g. Ireland, England, Italy). Moreover, other single-use items like cutlery, plates, cups and straws are planned to be banned in Europe by 2021 (EC 2019).

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

MPs added to consumer products (e.g. as a component of personal care, cosmetic and cleaning products) are especially manufactured to be used once and then washed down the drain. They are often referred to as microbeads, even though they are mostly irregular in shape in order to obtain an abrasive effect (Fendall and Sewell 2009; Napper et al. 2015; Kalčíková et al. 2017). The majority of microbeads in facial and body scrubs are made of PE, with average concentrations of 4.82 g/100 mL body scrub and 0.74g/100 mL facial scrub (Kalicova et al. 2017, Gouin et al. 2015). Other plastic polymers used in cosmetic products include polylactic acid, PET, polyethylene isoterephthalate, nylon-12, nylon-6, PMMA, polytetrafluoroethylene, and PUR (Leslie 2014; Rochman et al. 2015). Additionally, microbeads are used in industry as abrasives/scrubbers and sand-blasting media as well as in anti-slip, anti-blocking applications and for medical applications. It has been calculated that more than 4000 tons of PE microbeads were used in cosmetic products all over the EU (including Norway and Switzerland) in 2012 (Gouin et al. 2015), and the US is emitting 263 tons of PE microbeads per year (2.4 mg per person per day; Gouin et al. 2011). A ban of microplastics intentionally added to products (i.e., microbeads) has been proposed in the EU, while the US Microbead free waters act of 2015 (US Congress 2015) prohibits the manufacturing, packaging, and distribution of rinse-off cosmetics containing plastic microbeads already. This only applies to rinse-off products, while MPs are still permitted as a component in ‘leave on’ products (e.g. lotions, sunscreens, make-ups and deodorants).

Plastics with mid-term use expectancy

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

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

Plastics with long-term use expectancy

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

3. Pathways of plastic to the environment

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

3.1. Collected solid waste

Collected plastic waste is either landfilled, incinerated or recycled. In Europe 27.3% are landfilled, 30.1% are recycled and 41.6% are incinerated for energy recovery (PlasticsEurope 2018). The percentage of collected plastic waste varies strongly between different countries, depending on the applied waste management plans and policies. While worldwide the plastic recycling rate is still low, it has increased by almost 79% within the last 10 years in the EU, including Norway and Switzerland (PlasticsEurope 2018).

Large scale industrial plastic production began in the 50s, but plastic recycling was not established until the 80s. It is estimated that only 9% of the total produced plastic waste up to 2015 has been recycled (Geyer et al. 2017). From this again only a small portion is submitted to primary recycling in which the recycled plastic is used to replace all or a least a proportion of the virgin polymer resins (Hopewell et al. 2009). While high-income countries have sorting and processing facilities, in low income countries plastic recycling is not well established. Moreover, certain types of plastic are difficult to recycle. For example, thermoset plastics, including textiles, are usually not recycled.

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

3.2. Wastewater

Both MPs as well as MaP enter wastewater either directly if products containing plastic are flushed down the drain (e.g. fibres detached during laundry of textiles, microbeads in consumer products, cotton buds or sanitary products), or in combined sewer systems from street dust and litter. MaP escape wastewater treatment only on rare occasions and mainly enter the environment with untreated wastewater due to combined sewer overflows e.g. after heavy rainfall events or snowmelts (Williams and Simmons 1999), or if untreated wastewater enters the environment because WWTPs are not in place. Although high-income countries treat on average 70% of the wastewater, yet globally only 20% of the generated wastewater is treated (Sato et al. 2013). For MPs, the situation is different, due to their small size, they can escape the treatment and are also released with treated effluents (Ziajahromi et al. 2016). This pathway for MPs has been increasingly investigated. To date, 21 studies have measured MPs in wastewater (Tab. S1), from which two do not exclusively assessed MPs but included other litter items in the micro range (microliter; Michielssen et al. 2016; Talvitie et al. 2017b). Such studies were mainly carried out in (northern and western) Europe (13 studies), followed by north-America (5 studies).

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

WWTPs have in general a large retention potential for MPs, often higher than 95% (Tab. S1). However, in treated wastewater the number of MPs varies greatly too, from less than 1 MP/L (Browne et al. 2011; Carr et al. 2016; Murphy et al. 2016; Ziajahromi et al. 2017) to several hundred (Simon et al. 2018), and up to several thousand MP/L (Vollertsen and Hansen 2017; Fig. 2). Larger MPs are usually better retained during the treatment, so the most frequently observed MPs in treated wastewater are smaller than 300 µm (Dris et al. 2015; Mintenig et al. 2017; Gündoğdu et al. 2018; Magni et al. 2019; Talvitie et al. 2017a; Lee and Kim 2018; Wolff et al. 2018; Liu et al. 2019). For example, Magni et al. (2019) found that 94% of the MPs between 5–1 mm were retained by an Italian WWTP, while only 65% of the MPs between 0.1–0.01 mm were retained (Magni et al. 2019). Moreover, the number of MPs seems to be increasing with decreasing particle size. Wolff et al. (2019) reported the results of small-size MPs measured in treated wastewater and indicated that the 44% of measured MPs are between 10 and 30 µm, while 51% are between 30 and 100 µm. Furthermore, current research indicates that the amount of MPs retained by WWTPs is not only influenced by the size, but also by the particle shape. Usually, fibres are better retained in WWTPs as compared to microbeads or other irregular particles (Magnusson and Norén 2014; Talvitie et al. 2017b; Gündoğdu et al. 2018). Fibres and fragments are the most frequently occurring MP types in WWTP effluents (Tab. S1). Regarding polymer composition, PE particles or PES fibres are the most common plastic types (Tab. S1). Although a huge amount of tyre debris is suspected to enter WWTPs (Kole et al. 2017), they have not been frequently reported in treated effluents (Tab. S1). Only Dyachenko et al. (2017) and Lee and Kim (2018) have reported the presence of black particles possibly being tyre fragments.

Interestingly, concentrations of MPs in wastewaters show some seasonal and diurnal variations related to water consumption rates and human activity (Mintenig et al. 2017; Talvitie et al. 2017b; Lares et al. 2018). For instance, Talvitie et al. (2017b) reported that night time concentrations were slightly lower (average concentrations 476.7 and 0.8 mircolitter/L in influent and effluent respectively) compared to day time concentrations (584 and 1.7 mircolitter /L in influent and effluent, respectively). Therefore, MPs occurrence seems to be highly variable and depending on a variety of different environmental (weather, season) and behavioural variables but also methodological procedures (i.e. sampling method, including mesh sizes and sample volume), extraction method, and determination method. Despite the high retention of MPs by WWTPs, considering the large volumes treated daily, it is considered that more than one million particles can enter the aquatic environment via this pathway per WWTP (Ziajahromi et al. 2017; Gündoğdu et al. 2018), which constitutes one of the main sources of MPs into the environment.

3.3. Sludge and other agricultural amendments

The majority of MPs is already retained by WWTPs during pre- and primary treatment (mechanical treatment and sludge settling processes) and therefore concentrated in the grease or sludge phase (Murphy et al. 2016; Leslie et al. 2017; Talvitie et al. 2017b). While solids intercepted by grids and grease removal steps are disposed on landfills, sludge is often reused as fertilizers in agriculture. The amount trapped in the sludge roughly constitutes 50-90% of the MPs present in raw wastewater (Tab. S2; Magnusson and Norén 2014; Carr et al. 2016; Lee and Kim 2018). MP concentrations measured in sludge range between 650 MPs/Kg dw to more than 240,300 MPs/Kg dw (Fig. 3, Tab. S2). Murphy et al. (2016) found significant bigger sized MPs in the sludge phase compared to MPs in treated wastewater, confirming the differential retention potential of WWTPs regarding MPs size. Furthermore, the sludge treatment process (thickening, digestion, drying, stabilization, dewatering) may have an effect on the MP size (Mahon et al. 2017). Similar to wastewater, sludge samples usually show high numbers of fibres, followed by fragments (Tab. S2), and the main detected polymer is usually PES (particularly when there are many fibres present), followed by PE and PP.

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

4. Occurrence and fluxes of plastics in environmental compartments

4.1. Air

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

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

4.2. Soil

It has been suggested that agricultural soils could constitute larger MP sinks than marine ecosystems (Hurley and Nizzetto 2018). However, research on the quantification of plastics in soils (for both MaPs and MPs) is still very limited and mostly contracted to the last four years. We identified twelve studies reporting plastics in soil, from which three considered only a limited number of plastic types (Tab. S3). The available studies provide first indications of the scale of the pollution and suggest the ubiquitous presence of MPs in terrestrial ecosystems, also beyond agricultural areas. Most studies report plastic quantities in terms of particles, while some others provide concentrations based on mass measurements, which hampers to some extent direct comparisons among them. The highest MP concentration based on mass has been measured in soils from an industrial area in Australia, which was historically used to produce chlorinated plastic, containing 6700 mg

MP/kg dw (Fuller and Gautam 2016). The highest concentration based on the number of MP particles was provided by Vollertsen and Hansen (2017), who described Danish agricultural soils containing about 145 000 MPs/kg, in the size range of 20 to 500 µm which was based on weight however only 12 mg/kg. Also Chinese farmland soils were found to contain a high MP content, ranging between 70 and 18,760 MPs/kg dw (Fig. 4; Liu et al. 2018; Zhang and Liu 2018; Zhang et al. 2018). In contrast farmlands in Germany showed a much lower MP occurrence (0.34 MPs/ kg dw; Piehl et al. 2018). This might be partly related to differences in the considered MP sizes during the study and due to differences in agricultural practices. While Piehl et al. (2018) assessed MPs of a size between 1 and 5 mm, the study by Vollertsen and Hansen (2017) considered MPs between 20 μm and 500 μm. However, the different ranges in concentrations seem mostly attributed to the presence of different input sources.

The application of sewage sludge as agricultural fertilizer (biosolids) is considered to be a major source of MPs to soils. Nizzetto et al. (2016) estimated that between 63,000-430,000 and 44,000-300,000 tons of MPs could be yearly added to agricultural land in Europe and north America, respectively. Corradini et al. (2019a) found that increasing number of sludge applications were positively correlated to increasing MP concentrations in soils. Zubris and Richards (2005), report up to 1,210 fibres/kg in soils five years after sewage sludge application and detected fibres still 15 years after application, which is another indication for MPs accumulation in soil due to sludge application. On the other hand, almost twice the concentration of MPs was found in Danish fields not treated with sludge compared to treated fields (Vollertsen and Hansen 2017). Additional studies investigating the presence of MPs in soil after application of wastewater sludge are fundamental to better estimate the importance of this pathway.

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

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

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

The fate of MPs within the soil is not completely clear yet. MPs in soils may be transported along with water runoff and soil erosion into adjacent streams and rivers. So far, there is no knowledge on the importance of this pathway as it has not been experimentally proven. Translocation into deeper soil layers can occur through soil cultivation (Hurley and Nizzetto 2018) or transport by soil organisms. Earthworms and collembola have been shown to ingest and transport MPs from the soil surface into deeper soil layers (Huerta Lwanga et al. 2017a; Maaß et al. 2017; Rillig et al. 2017). Also other animals e.g. birds or domestic animals, which have been shown to take up MPs (Zhao et al. 2016; Huerta Lwanga et al. 2017b) can transport MPs over longer distances. To date, it is yet unclear whether low sized MPs can be transported through soil pores into ground water, but low concentrations of MPs (0 to 7 MPs/m^3) have been reported in raw drinking waters from groundwater wells (Mintenig et al. 2019). Uptake of plastics by plants is another potential source of mobilization of plastics from soil ecosystems, particularly for NPs, however no studies have investigated this using whole plants (Ng et al. 2018). The only study available in this respect is the one provided by Bandmann et al. (2012), who demonstrated uptake of 20 and 40 nm PS beads by tobacco BY-2 cells in cell culture via endocytosis, while 100 nm beads were excluded.

4.3 Surface waters

Plastic pollution along rivers has been already observed and assessed in the 1990s (Williams and Simmons 1996, 1999). Nevertheless, few studies have reported plastic pollution in freshwaters until the whole environmental movement was initiated few years ago. Some studies assessing litter in rivers have not exclusively focused on plastic, but also included other litter items like glass, paper and wood. Those studies show that about 80% of the litter items are plastics, but do not provide concentrations or mass estimates (Crosti et al. 2018; González-Fernández et al. 2018; Castro-Jiménez et al. 2019).

Studies focusing on providing concentrations of MaPs in the environment are very limited (Tab S4). MaPs concentrations have been reported for example for the Los Angeles river, in California (819 MaPs/m^3; Moore et al. 2011), the Yangtze river in China (8.74x10^3 MaPs/km^2; Xiong et al. 2019), and in Lakes (1,800 MaPs/km^2) and Rivers (0.012 MaPs/m^3) in Switzerland (Faure et al. 2015). It has been estimated that in the river Seine in France, 28,000 kg of floating plastic are trapped annually by floating debris retention booms (Gasperi et al. 2014) and floating MaP in the Saigon river in Vietnam were estimated to range between 7,500 – 13,700 tons per year (van Emmerik et al. 2018). As only buoyant plastics were considered in those studies, the total loads may be underestimated as plastic is also transported by sub-surface transport (Morritt et al. 2014). The most common MaPs reported in freshwater environments are plastic bottles, food packaging items, plastic bags and sewage-related plastic like handles from buds of cotton wool and sanitary towels (Tab. S4). Regarding polymer composition, PP and PE are the plastic types that were omnipresent, and to a lesser extent PS and PET have been reported (Table S4).

MPs in water have been reported in different units (i.e. particles per water volume, or particles per area). To be able to compare the results of the different studies, we choose 37 studies which either reported the number of MPs per water volume or gave sufficient information to transform the reported unit. Like in other environmental compartments the concentrations varied greatly among studies (Fig. 5, Tab S5). Most studies in Europe found average concentrations of less than 1 to less than 100 MP/m^3, while the highest average concentration of 100,000 MPs/ m^3 (with a maximum concentration of 187,000 MPs/ m^3) was measured in the Amsterdam Canals (Leslie et al. 2017). Furthermore, Lui et al. (2019a) reported up to 22,849 MPs/m^3 (average: 1,409 MPs/m^3) in storm water ponds receiving urban runoff in Denmark. The highest peak concentration from all studies was found in the Snake River in North America and was as high as 5,405,000 MPs/m^3 (average: 91 MPs/m^2) (Kapp and Yeatman 2018). The second highest peak concentration was reported by Lahens et al. (2018), and corresponds to 519,223 MPs/m^3 (minimum 17,210 MPs/m^3) monitored in the Saigon River (Vietnam). Overall, reported concentrations of MPs appear to be higher in Asia, as compared to Europe and North America (Fig. 5). However, most of the studies carried out in Asia were performed in China and focused on assessing lower size classes that those studied in Europe. The only two studies conducted in Europe that considered a very low size (MPs below 20 µm), were the ones by Leslie et al. (2017a) and Lui et al. (2019), who observed by far the highest concentrations. Current research shows that smaller particles (<0.5 mm) are usually the most frequent ones (e.g. Leslie et al. 2017; Yan et al. 2019). Therefore, the higher concentrations found in Asia may be not exclusively related to a higher pollution but also to the sampling methods used.

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

Fig. 2 Overview on most common sampling methods used for freshwater MPs sampling

Although MPs have been found in remote locations and rural areas, there is evidence that MPs concentration increases with proximity to cities (Wang et al. 2017b; Di and Wang 2018; Tibbetts et al. 2018). A modelling study identified the Yangtze River catchment as the catchment transporting the highest plastic loads into the ocean (Schmidt et al. 2017a). The four case studies looking at MPs concentrations in the Yangtze river found highly variable concentrations, but were also amongst the highest observed (Zhang et al. 2015; Wang et al. 2017b; Di and Wang 2018; Xiong et al. 2019). However, concentrations in the same order of magnitude were also monitored in other rivers in China such as the Pearl river, which was also ranked under the top ten catchments transporting plastic into the ocean (Schmidt et al. 2017a).

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

Fragments and fibres formed by PE and PP are the most frequently observed particles across all studies evaluating MP pollution in freshwater ecosystems; whereas pellets or beads are only rarely reported as the main occurring plastic types (Tab. S5). The latter are mainly found in studies along the rivers Rhine and Danube, in the proximity to plastic processing plants and are thus assumed to be pre-production pellets (Lechner et al. 2014; Lechner and Ramler 2015; Mani et al. 2016). The prevalence of secondary MPs (fragments and fibres) suggests wastewater and runoff as sources for plastic pollution in freshwater ecosystems (Tab.S5). Several studies confirmed that by demonstrating that MP concentrations are higher downstream of WWTP as compared to sampling sites in upstream areas (McCormick et al. 2014; Estahbanati and Fahrenfeld 2016; Vermaire et al. 2017; Kay et al. 2018). For example, in the Ottawa River (Canada), 0.71 particles/m^3 were found upstream of a WWTPs compared to 1.99 MPs/m^3 downstream. In the Raritan River and the North Shore Channel (USA) 24 MPs/m^3 and 1.94 MPs/m^3 were found upstream the WWTP, and 71.7 particles and 17.93 MPs/m^3 were detected downstream, respectively (McCormick et al. 2014; Estahbanati and Fahrenfeld 2016; Vermaire et al. 2017). As mentioned above, the majority of MPs in wastewater is smaller than 300 µm, thus it may be presumed that larger MPs enter via different pathway like surface runoff, or steam from the breakdown of MaPs directly in the aquatic environment. However, with untreated wastewater, for instance during sewage overflows, MaPs can enter river ecosystems. For example, Morritt et al. (2014) identified polllution hotspots in the vicinity of WWTPs that were mainly constituted of sanitary products. MPs hotspots were also detected in areas with low population density but high agricultural use, pointing also to agricultural runoff as an important source (Kapp and Yeatman 2018)._Finally, poor waste management likely increases plastic input into aquatic ecosystems (Lahens et al. 2018), where they can break down into smaller particles. Xiong et al. (2019), for example, found that the abundance of microplastic is positively related to the presence of MaPs.

4.4. Sediments

Similar to MaP in surface waters also MaPs in sediments are only rarely assessed and the way MaP occurrence is reported is highly variable and difficult to compare (Tab. S6). MaPs along river banks have been observed while assessing buoyant litter in general (Williams and Simmons 1999; Rech et al. 2014), and river beach sediments in Switzerland contained on average 90 MaPs/m^2 (Faure et al. 2015). Across different lake shores, MaPs concentrations have been shown to vary notably (Imhof et al. 2013; Fischer et al. 2016). While high MaPs concentrations have been observed at the south shore of Lake Garda (Italy; with an average concentration of 483 MaP/m^2), the occurrence at the north shore was significantly lower (i.e. 0-8.3 MaP/m^2; Imhof et al. 2013). Food packaging is among the most frequently observed MaPs but also bottles, bags and ropes are described by several studies. Regarding the polymer composition, PE and PP as well as Styrofoam (PS) are reported (Tab S6).

As for MaPs and the other compartments, the concentration of MPs in freshwater sediments has not been reported in consistent units across all studies. Therefore, we focused on studies that have reported the concentration in MPs/kg sediment. However, studies reporting MPs per sediment area, which gave sufficient information to estimate the concentration in MPs/kg, were also included. Therefore, from the 33 studies that were found during the literature search, 30 were chosen for comparisons (Fig.6, Tab. S7). The highest sediment concentration of 2,071 MPs/kg dw has been found in the urban canals of Amsterdam, where also the highest water concentrations were observed (Leslie et al. 2017). MP concentrations in river bed sediments seem, in general, higher than in river beach and shore sediments (Fig. 6; Tab S7). Most studies on MPs in river bed sediments report concentrations between 100 MP/kg and a few thousands. Studies from Asia were exclusively carried out in China, and reported similar concentration ranges as those described in Europe. Interestingly, the study on the Yangtze River (China), which has been estimated to be the highest contributor of plastic to the sea (Schmidt et al. 2017a) and amongst the highest MPs concentrations reported in water (Fig. 5, Tab. S5), had a comparably low sediment concentration 7-66 MP/kg. The only study carried out in Africa assessing the concentration of MPs in river sediment reports notable differences between concentrations in summer (1-14.61 MP/kg dw) and winter (13.3 - 563.8 MP/kg dw; Nel et al. 2018), which were related to a reduced flow condition in winter. Subsequently, the hydrological variation shown by many rivers seems to be one of the main factors contributing to MPs deposition and re-mobilization from river beds. This was also demonstrated by Hurley et al. (2018a), who report that about 70% of the MPs in the sediments of the upper Mersey and Irwell catchments (UK) were exported after a flooding event. Several studies show that, after transportation with the river flows, MPs tend to (re-)deposit in low energy environments, such as meanders, deltas, dams, harbours and coastal lagoons (Claessens et al. 2011; Vianello et al. 2013; Shruti et al. 2019). The deposition of low-density polymers in sediment environments is also related to a density increase by biofouling (e.g. Ye and Andrady 1991; Andrady 2011; Zettler et al. 2013; McCormick et al. 2014).

For lakes, mainly beach and shore sediment concentrations have been reported. In Europe average concentrations for beach and shore sediments ranged between 0.94 and 44 MP/kg, while beach and shore sediments from Lake Ontario (Canada) contained much higher concentrations (20-27,830 MPs/kg; Fig. 6, Tab. S7). Several studies have noted that plastic concentrations differ strongly between different areas of the same lake (Zbyszewski and Corcoran 2011; Imhof et al. 2013; Zbyszewski et al. 2014; Zhang et al. 2016), suggesting that accumulation is patchy and form contamination hotspots influenced by winds, waves and/or beach morphology (Imhof et al. 2016, 2018). Similar observations were made at Lake Huron (Canada), in which 94% of all monitored pellets were found to accumulate in one single beach (Zbyszewski and Corcoran 2011). In the Taihu Lake (China), MPs concentrations ranged from 11 to 235 MP/kg in different bed areas, and the average MPs abundance in sediments in the northwest area was approximately six times higher than that of the southeast area (Su et al. 2016).

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

4.5. Marine

Rivers are estimated to be the main pathways for plastics entering the oceans. Estimations on the amount of plastic waste entering the ocean through this pathway range between 0.41 and 4x10^6 tons per year (Lebreton et al. 2017; Schmidt et al. 2017b). From the top ten river catchment that transport 88-95% of the global plastic load into the oceans, eight are located in Asia (Schmidt et al. 2017b). Oceans have been assumed to be the final sink for MaPs and MPs. As this review is focused on terrestrial and freshwater ecosystems, this compartment will not be discussed in detail. A number of articles and reviews have been published on the topic within the last few years which describe plastic occurrence in the oceans and its effects on marine life (see Barboza and Gimenez 2015; Jambeck et al. 2015; Auta et al. 2017).

5. Discussion

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

5.1. The need for standardization of sampling and analysis

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

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

Besides this, existing methods for the identification and counting of MPs are quite variable. Until recently, it was common practice to solely rely on visual detection (using a microscope), which may lead to false positive or false negatives. In more recent studies, visual examination is usually combined with FTIR (Fourier Transform Infrared) or Raman Spectroscopy, which allows polymer Identification. This is, however, time-consuming and thus frequently only a sub-sample is subjected to spectroscopic methods. Other studies use different methods like SEM (Scanning Electron Microscopy), XRF (X-Ray Fluorescence), Pyr-GC/MS (Pyrolisis interfaced with gas chromatography/mass spectrometry). It has been observed that MP abundance often varies with the methods used (Song et al. 2015; Mai et al. 2018; Picò and Barcelo 2019), so analytical results may be difficult to compare across studies.

There is an urgent need for a harmonisation of methods for sampling in different environmental compartments, sample processing, MP extraction, identification, and counting, as well as for the units to be used for reporting data. A recent report from GESAMP (Group of Experts on the Scientific Aspects of Marine Protection) describes and compares methods for sampling and analysing MaPs and MPs, with particular focus on the marine environment (GESAMP 2019). Although many problems remain unsolved (e.g. the need for sampling small size MPs and NPs), the report may represent a valuable starting point for the development of protocols for large scale monitoring of plastic litter in the environment.

5.2. Small size micro-nanoplastics: the largest unknowns

Most procedures commonly applied to date allow sampling, processing and measuring particles down to a minimum size of 20 μm. Only very few studies measured smaller particles, down to 10 μm (e.g. (Leslie et al. 2017; Simon et al. 2018). In theory, very small particles and, especially NPs, should be more abundant in the environment, and their concentrations are expected to increase. Moreover, from a toxicological point of view, NPs are particularly interesting because it is possible that below a given size (still unknown) they cross cellular membranes and enter into the cells, with possible interactions in the cellular content and structure. This represents a substantial difference in comparison to MaPs or MPs. Indeed MPs cannot be accumulated in biological organs and tissues and may produce mainly physical stress on living organisms, although the consequences of that may result in physiological and metabolic alterations. The development of methods for the evaluation and quantification of small-size MPs and NPs is one of the major research needs to assess the potential risks for human and environmental health. In particular, detection technologies to identify nano-sized plastic particles are still lacking (Mai et al. 2018). A promising approach, at least to quantify the mass and the composition (if not the number of particles), could be the use of Pyr-GC/MS (Hendrickson et al. 2018, Mintenig et al. 2018) coupled with methods of small size particle separation based on ultrafiltration membrane technologies (Mulder 1998; Judd and Jefferson 2003).

5.3. Towards a microplastic mass balance and suitable evaluation of environmental fluxes

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

From the data reported in Fig. 2 and Table S1, it can be concluded that the range of particles in effluents from WWTPs that include secondary and tertiary treatments spans from 1 to 5,800 MPs/L, with a geometric mean around 29 MPs/L. In non-treated wastewaters the concentrations range from few particles/L up to more than 100,000, with a geometric mean of about 242 MPs/L. These data are in reasonable agreement with the percentage of retention by WWTPs reported by several authors, which ranges from 80% to 99% of the inflowing particles number (see Section 4.3).

The approximated per capita consumption of water in Europe is 140 L per day (EUROSTAT, 2015). Although with some regional differences, it may be estimated that about 85% of the EU population (525 millions in the EU plus Norway and Switzerland) is connected to WWTP with secondary or tertiary treatment, while the rest (15%) is connected to a WWTP with only primary treatment or not connected at all (Table 1).

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

- from treated wastewater: an average value of 1,800E+9 particles per day (possible range from 9E+9 to 130E+12 particles/day)
- from untreated wastewater: an average value of 2,700E+9 particles per day (possible range from 27E+9 to 1,400E+12 particles/day).

Transforming these data on a weight basis is not easy because, in general, only numbers of MPs are reported, while size/weight conversion factors are not readily available. Combined data on numbers and weight are reported in a Danish report (Vollertsen and

Hansen 2017) assessing MPs occurrence in ten different WWTP, and in the study by Simon et al. (2018). However, both studies took only MPs between 10 or 20 and 500 μm into account. Therefore, estimating the load on a weight basis from the particle numbers is not possible.

Despite their wide range of variability, these estimates give a first approximation of the load of MPs in surface waters from urban wastewater and allow the following observations. First, the load that may be attributed to the relatively small percentage of European untreated wastewaters is much higher than the load deriving from treated wastewater, which points towards a definite need of implementing secondary and tertiary WWTPs in areas that are still not connected to reduce total MPs emission. Taking into account that untreated wastewater is concentrated in south-eastern Europe, it may be hypothesized that some watersheds (e.g. lower Danube) are subject to higher contamination than those located in other European regions (Lechner et al. 2014). Unfortunately, data on MP concentrations in surface waters of south-eastern Europe are not available. Due to the scarcity of data of water consumption and WWTP implementation, a comparable evaluation cannot be done for other continents. However, it may be hypothesized that the percentage of treated wastewater in Asia and Africa is much lower than in Europe or North America.

The problem is also complicated by the fact that only a relatively small part of the population in connected to sewerage systems. Data from the WHO/UNICEF Joint Monitoring Programme (JMP), referred to 2015, indicate that in Eastern, South-eastern and Central Asia, with a population of more than four billion inhabitants, only 25% of the population is connected with sewerage systems; and in Sub-Saharan Africa the percentage is lower than 6% (WHO/UNICEF 2019). The high concentrations of MPs in surface waters of Asia (mostly in China), as compared to those measured in Europe (Fig. 5), supports the hypothesis regarding the large influence of WWTP on surface water emissions.

The total values calculated in this study seem relatively low, particularly for treated waters, if one considers that they represent emissions at the continental level. Nevertheless, the The low concentrations of MPs in surface waters of Europe, as compared to Asia for example, seem to be directly related to ther ow values estimated from wastewater concentrations may justify the relatively low values measured in surface waters in Europe (Fig 5 and Tab. 5), all referred to north, central and south European water bodies. On the other hand, the high values measured in Asia (mostly in China), supports the hypothesis regarding the large influence of WWTP on surface water emissions.

The lowest average weight of MPs in effluents compared to influents (almost 50%) indicates that the removal efficiency is higher on bigger particles. The dominant shape in WWTP effluents were fibres, followed by fragments. Only in one case a minor amount (<10%) of pellets that may be assimilated to primary microbeads was observed.

Obviously, wastewater represents only one of the possible pathways of MPs into surface waters, and as discussed in this study, there is no doubt that surface runoff from agricultural and urban soils may also represent a major source. Unfortunately, a comparable estimate of MPs emissions from soils due to water runoff is not possible due to field data limitations. On the other hand, this review shows that MP concentrations in WWTP sludge (mainly from Europe) range between 10E+3 and 10E+5 particles/kg dw. Nizzetto et al. (2016) estimated that the total yearly input of MPs from sewage sludge to farmland is about 63,000-430,000 tons in Europe, and 44,000-300,000 tons in North America. Data on MP concentrations in soil are scarce and scattered (Fig. 4 and Tab S3). The majority of data on agricultural soils refer to China and indicate a reduced range of variability (from about 60 to 200 particles/kg dw), except for a couple of higher values (more than 10,000 particles/kg dw) from soils sampled in a greenhouse. Overall this study shows that soil could be considered as a sink as well as a source of MPs to surface water. Therefore, further research is urgently required to assess fluxes of MPs from soils into surface water ecosystems and to assess the fate of MPs in the soil ecosystems, investigating its retention potential and the capacity of MPs to reach groundwater ecosystems. An additional source of MPs to soil and surface water may be atmospheric fall-out (Dris et al. 2016). However, the information available to date does not yet allow a quantitative estimate (Wetherbee et al. 2019).

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

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

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

5.4. Microplastics in environmental compartments: what does it mean in terms of exposure for living organisms?

As discussed above, information on the presence of MPs in environmental compartments is often biased by the inconsistency of units (e.g. n/L, n/m^2, mg/L, n/kg, mg/kg), by the variability in size classes sampled and measured, and by the complexity in shape and composition that are often not clearly reported. These inconsistencies make the assessment of their possible impact on living organisms rather complex, so the actual environmental risks of different plastics and their associated chemicals remain largely unknown (Koelmans et al. 2017). It is important to highlight that the effects of MPs on living organisms cannot be quantified by a simple concentration-response relationship of the whole mass of MPs of certain type found in environmental samples, as for most chemical contaminants. Their impacts on aquatic organisms depend on a number of factors such as:

- the shape: the physical effect determined by long and thin fibres may be completely different from those determined by microspheres or by irregular fragments (Au et al. 2015; Lambert et al. 2017);

- the size range: the definition of MPs in term of size is extremely wide (from 5 mm to 1 μm) and the living organisms that may be affected by MPs are also extremely variable in size. For example, in the aquatic environment, from fish to zooplankton; for any type and size of organism, different MP size classes may be ingested and thus effective, including small sizes (below 20 μm) and NPs, that are practically never measured;
- the composition: for most MP polymers, being the effects mainly physical, it may be hypothesised that the response is not related to the polymer composition; however, for some particular MP particles, such as for tyre debris, the composition is much more complex and the effects may also be determined by the leaching of non-polymeric chemicals.

It follows that the available information on the presence of MPs in the environmental compartments does not allow, to perform an ecological risk assessment based on a comparison between an environmental exposure (e.g. a PEC: predicted environmental concentration) and an effect level (e.g. a PNEC: predicted no effect concentration). So far only An ecological risk assessment of MPs would require much more detailed information on MP exposure with a precise assessment of number (or weight) of particles per size classes, shape and composition. Considering that current methods for the analysis of MPs are complex, expensive and time consuming, this level of detail is, to date, difficult to be achieved. Moreover, ecotoxicological tests have been frequently carried out using PE microspheres, while other polymers and especially other shapes like fragments and fibres are expected to be more abundant in the environment (de Sá et al. 2018). Further research must be devoted to both areas, to refine exposure assessments and to perform effect assessments taking into account ecologically relevant combinations of organisms and MPs sizes, shapes and types. It is most likely that future risk assessments need to necessarily consider MP particle mixtures taking into account different polymer type, shape and size, and that exposure and risk indicators are derived taking all these variables into account.

Regarding the effect assessment, the major unknown issues are related to small and very small particles (Koelmans 2019). As mentioned above, the size threshold below which these particles may enter in the cells is still unknown. Moreover, once they enter in the cells, the possible interactions of these, theoretically chemically inert polymeric molecules, with cell structure and functioning are also unknown. Recent studies on NPs performed with reference materials painted with fluorescent dye demonstrate their capacity to be taken up, enter tissues, and accumulate in small organisms (Cui et al. 2017; Lee et al. 2019). However, some authors discuss that this can be an artefact created by the leaching of those dye paints, which can be taken up into cells or due to the autofluorescence of the evaluated biological tissues (Catarino et al. 2019; Schür et al. 2019).

5.5 How can MP inputs in the environment be controlled?

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

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

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

6. Conclusions

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

There is enough experimental evidence demonstrating that the presence of MaPs in aquatic ecosystems represent an environmental risk, particularly for large animals. Regarding MPs, a risk for human and environmental health has not been demonstrated (EC SAM 2019; GESAMP 2019). All available toxicological evidence indicates that some effects on aquatic and terrestrial organisms, vertebrates and invertebrates, have been observed only at concentrations that are orders of magnitude higher than the maximum levels measured in the environment (Lenz et al. 2016; Redondo Hasselerharm et al. 2018). Other possible effects, such as a potential increase in the bioaccumulation of chemicals due to their transport into the organisms adsorbed on MPs (the “Trojan horse effect”) seems to be context dependent, and negligible in comparison to direct accumulation from the surrounding environment (e.g. from water) or from food (Koelmans et al. 2013, 2014; Lohmann 2017; Mohamed Nor and Koelmans 2019). However, research is still needed to demonstrate this experimentally.

Current knowledge gaps regarding environmental fluxes and breakdown of MPs and NPs are still large in order to assess future risks for man and for the environment. Furthermore, the bias on sampling and analysis makes a precise quantification challenging. This is particularly difficult for small MPs and NPs, which are probably the more concerning particles from a toxicological point of view. Moreover, although present exposure seems to be far away from levels of concern, it is difficult to predict future emission patterns since they will be closely related to plastic use and management policies. This review shows that the construction of waste-water treatment facilities and the proper management of sludge applications in agriculture are efficient means to reduce MPs emissions. Moreover, the ban of single-use plastics, the substitution of some plastic polymers with biodegradable compounds, and the reduction of MPs emission at a source are key to control plastic pollution. From now onwards, we expect technological solutions to be developed and implemented in this direction. There is no doubt that plastics changed our life in the middle of last century, and the control of plastics will again change our life in the near future.

7. List of abbreviations

ATR	Attenuated Total Reflectance
ECHA	European Chemical Agency
EEA	European Environmental Agency
ERA	Ecological Risk Assessment
EVA	Ethylene-vinyl-acetate
FTIR	Fourier Transform Infrared
MaP	Macroplastic
MP	Microplastic
NP	Nanoplastic
PAH	Policyclic aromatic hydrocarbons
PC	Polycarbonate
PCB	Polichlorinated biphenyl
PE	Polyethylene
PEC	Predicted Environmental Concentration
PES	Polyester
PET	Polyethylene-terephtalate
PMMA	Poly-methyl-metacrylate
PNEC	Predicted No Effect Concentration
PP	Polypropylene
PS	Polystyrene
PUR	Polyurethane
PVC	Polyvinylchloride
Pyr-GC/MS	Pyrolisis-Gas Chromatography/Mass Spectrometry
SAPEA	Science Advice for Policy by European Academies
SEM	Scanning Electron Microscopy
WWTP	Waste Water Treatment Plant
XRF	X-Ray Fluorescence

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

	No treatment or no connection	**Primary**	**Secondary**	**Tertiary**

	with sewerage			
Northern	15.1	5.6	2.3	77
Central	3.4	0	16.5	80.1
Southern	23	2.2	21.3	53.4
Eastern	26	0.2	13.6	60.6
South-Eastern	40	16.7	22.8	20.6
Weighted average respect to population	13	2	18	67

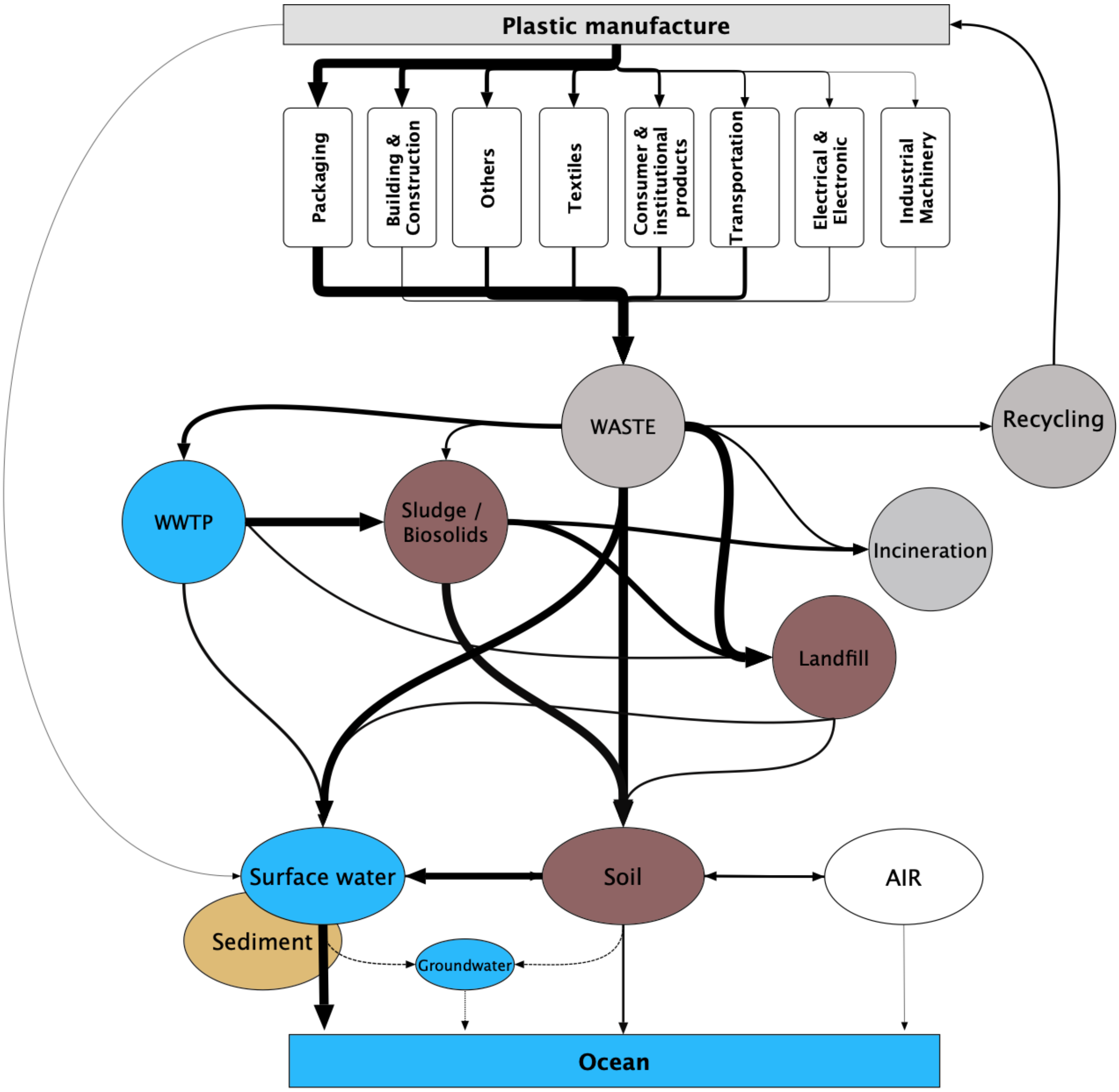

Fig. 1. Production and pathways of plastics into the different environmental compartments. Thickness of the different arrows are related to the relevance of the different mass flows. The relevance of the different plastic

source mass flows is based on Geyer et al. (2017), while the relevance of the environmental flows is based on the reviewed literature or assumptions. Dashed lines indicate yet completely unexplored pathways with unknown relevance.

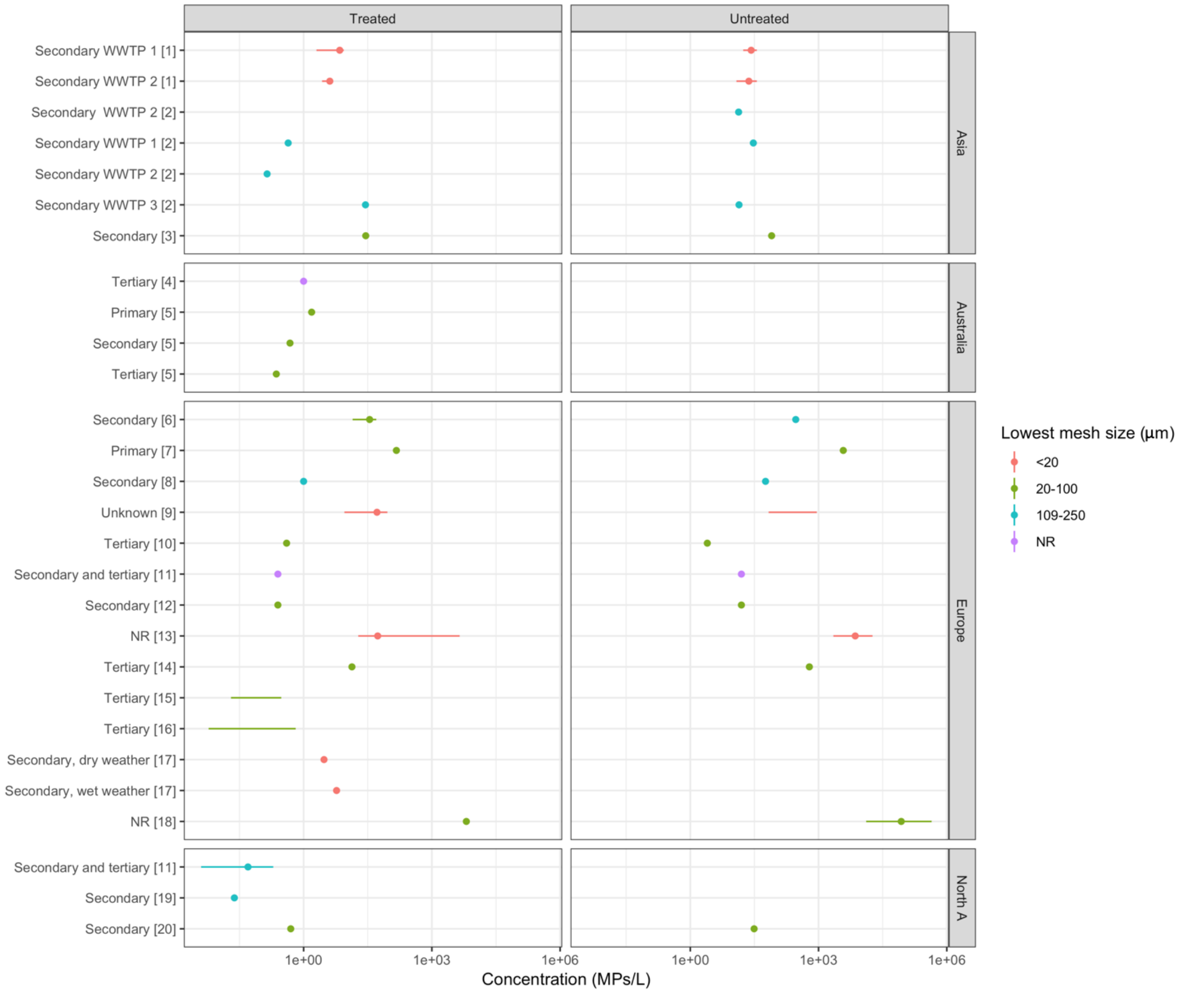

Fig. 2. MP concentrations in untreated and treated wastewaters (MPs/L) from WWTPs with different treatment types. NR= not reported. [1] Gündoğdu et al. (2018) [2] Lee and Kim 2018 (2018) [3] Liu et al. (2019b) [4] Browne et al. (2011) [5] Ziajahromi et al. (2017) [6] Dris et al. (2015) [7] Helcom (2014) [8] Lares et al. (2018) [9] Leslie et al. (2017) [10] Magni et al. (2019) [11] Mason et al. (2016) [12] Murphy et al. (2016) [13] Simon et al. (2018) [14] Talvitie et al. (2015) [15] Talvitie et al. (2017a) [16] Talvitie et al. (2017b) [17] Wolff et al. (2018) [18] Vollertsen and Hansen (2017) [19] Dyachenko et al, 2017 (2017) [20] Gies et al. (2018).

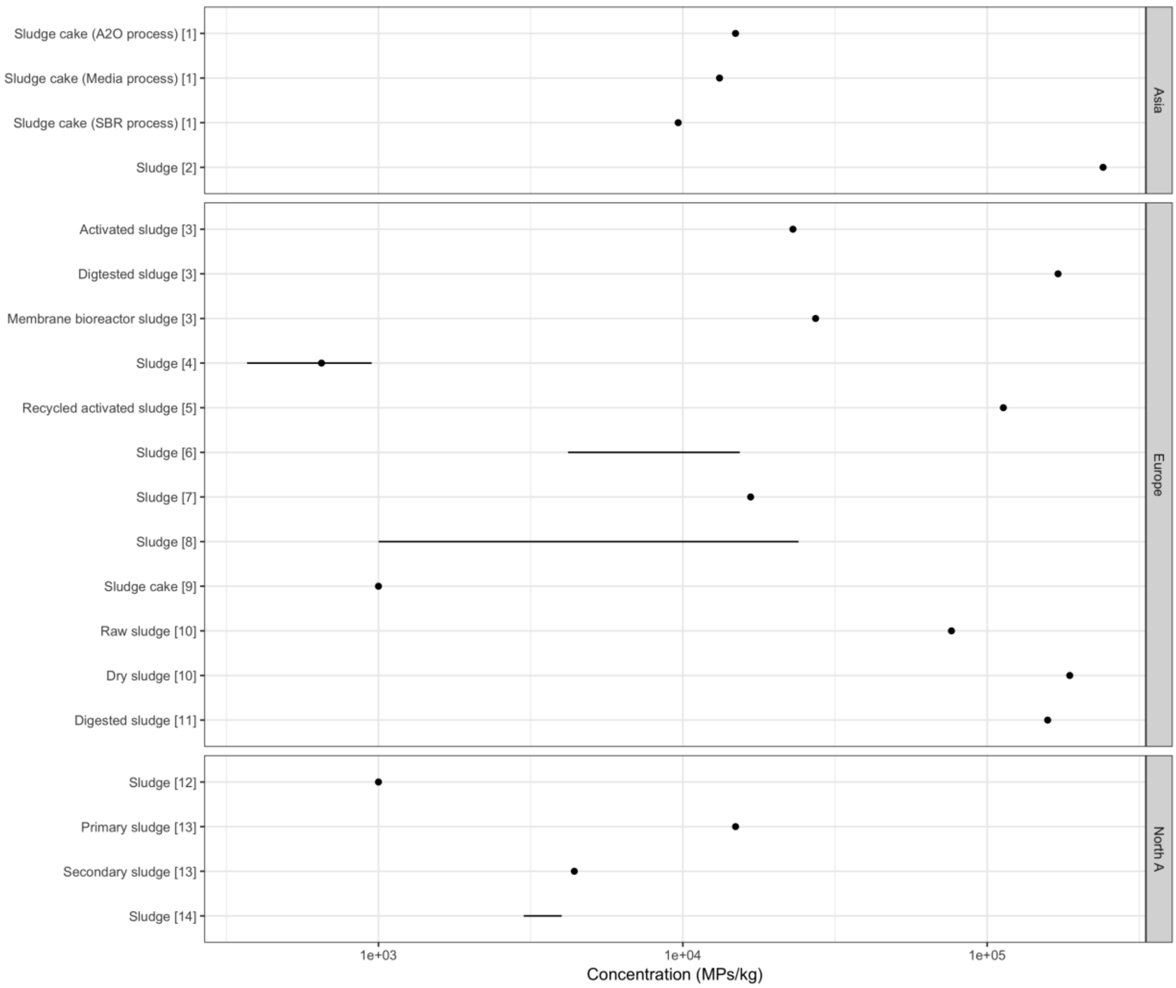

Fig. 3. MP concentrations in sludge samples (MPs/kg dw). North A = North America; A2O = anaerobic-anoxic-aerobic; SBR= sequence batch reactor [1] Lee and Kim (2018) *[2] Liu et al.* (2019b) *[3] Lares et al. 2018* (Lares et al. 2018) *[4] Leslie et al.* (2017) *[5] Magni et al.* (2019) *[6] Mahon et al.* (2017) *[7]Magnusson and Norén* (2014) *[8] Mintenig et al.* (2017) *[9] Murphy et al.* (2016) *[10] Talvitie et al.* (2017b) *[11] Vollertsen and Hansen* (2017) *[12] Carr et al.* (2016) *[13]Gies et al.* (2018) *[14] Zubris and Richards* (2005)

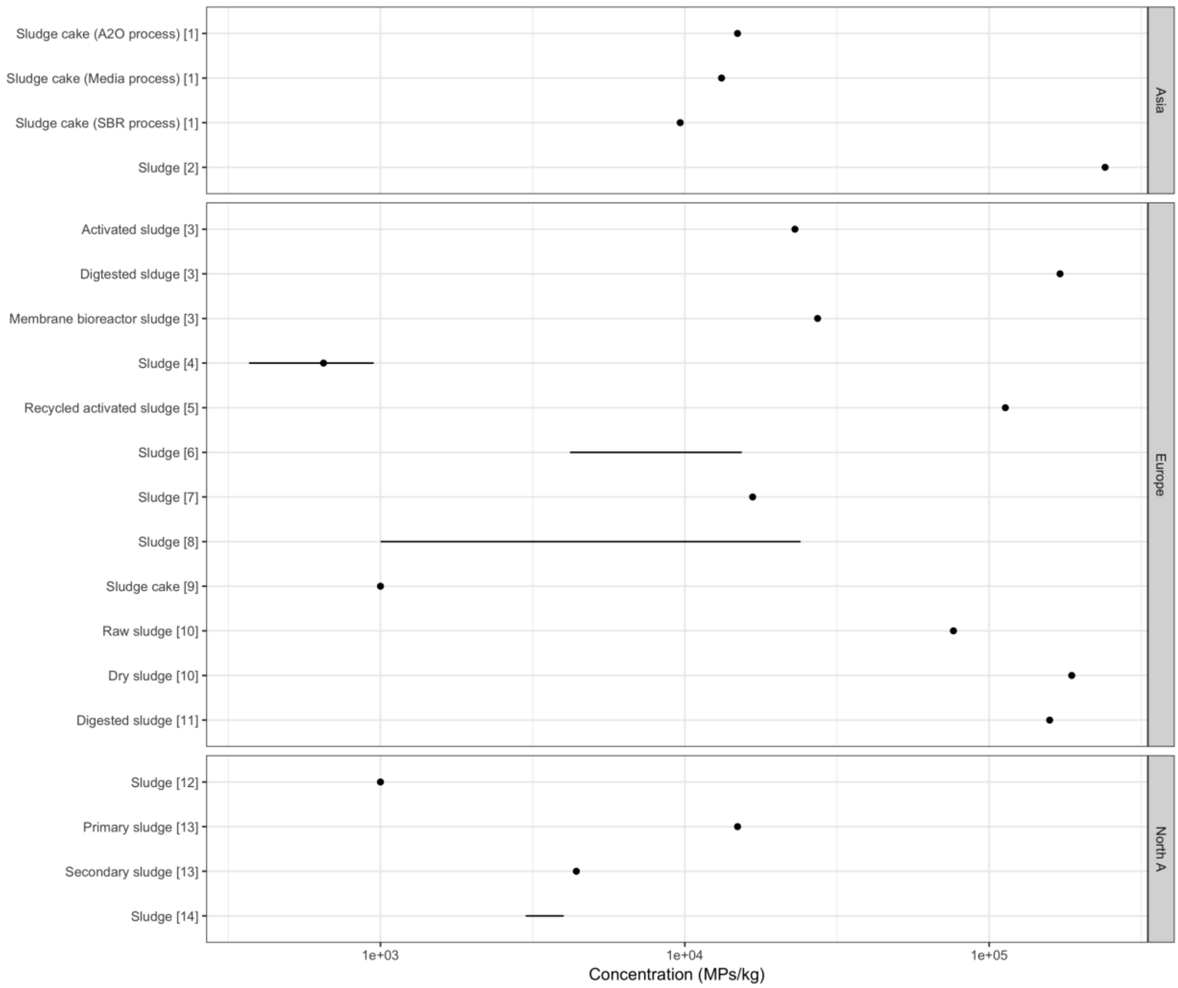

Fig. 4. MP concentrations in different soil samples (MPs/kg dw). AUS = Australia; C.A. = Central America; N.A = North America. [1] Fuller and Gautam (2016) [2] Zhang et al. (2018) [3] Zhang and Liu (2018) [4] Liu et al. (2018) [5] Huerta Lwanga et al. (2017b) [6] Corradini et al. (2019b) [7] Vollertsen and Hansen (2017) [8] Piehl et al. (2018) [9] Scheurer and Bigalke (2018) [10] Zubris and Richards (2005).

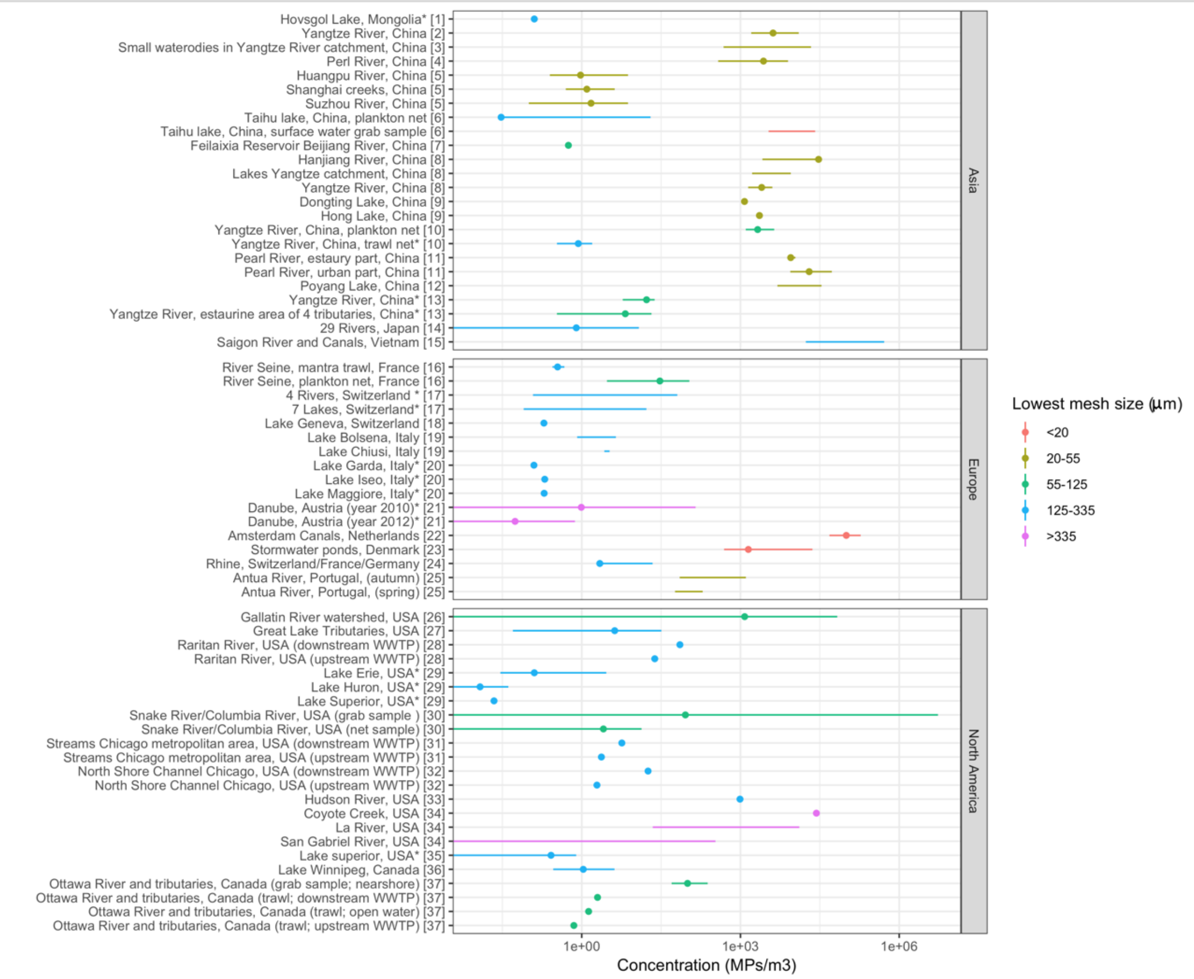

*Fig. 5. MP concentrations in surface water samples (MPs/m^3). * Concentration in MPs/ m^3 was estimated by dividing the reported concentration in particles per area by the height of the net used for sampling. [1] Free et al. (2014) [2] Di and Wang (2018) [3] Hu et al. (2018) [4] Lin et al. (2018) [5] Luo et al. (2018) [6] Su et al. (2016) [7] Tan et al. (2019) [8] Wang et al. (2017a) [9] Wang et al. (2018) [10] Xiong et al. (2019) [11] Yan et al. (2019) [12] Yuan et al. (2019) [13] Zhang et al. (2015) [14] Kataoka et al. (2019) [15] Lahens et al. (2018) [16] Dris et al. (2015) [17] Faure et al. (2015) [18] Faure et al. (2012) [19] Fischer et al. (2016) [20] Sighicelli et al. (2018) [21] Lechner et al. (2014) [22] Leslie et al. (2017) [23] (Liu et al. 2019a) [24] Mani et al. (2016) [25] Rodrigues et al. (2018) [26] Barrows et al. (2018) [27] (Baldwin et al. 2016) [28] Estahbanati and Fahrenfeld (2016) [29] Eriksen et al. (2013) [30] Kapp and Yeatman (2018) [31] McCormick et al. (2016) [32] McCormick et al. (2014) [33] Miller et al. (2017) [34] Moore et al. (2011) [35] Hendrickson et al. (2018) [36] Anderson et al. (2017) [37] Vermaire et al. (2017).*

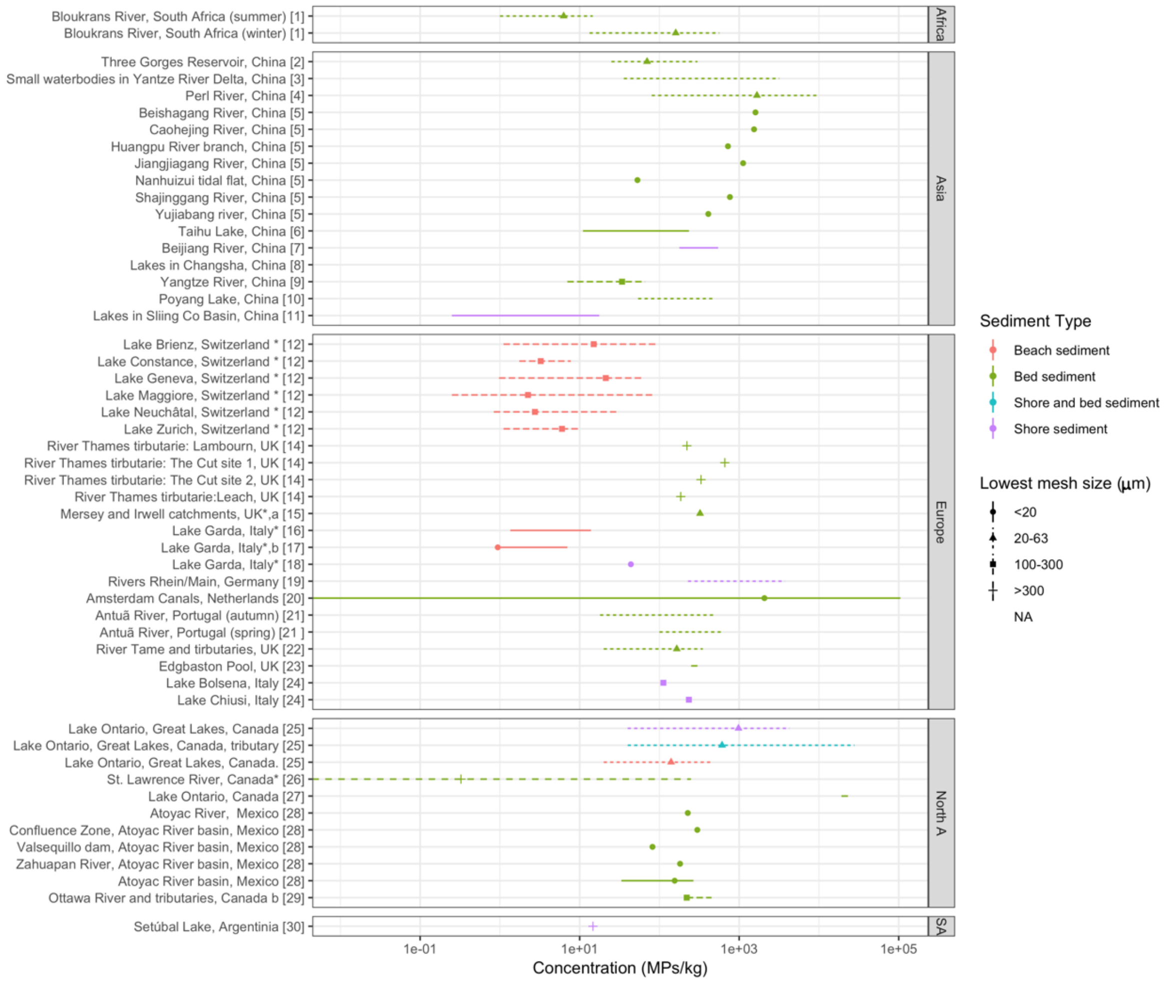

Fig. 6. MP concentrations in different type of sediment samples (MPs/kg). Notes: Concentration in MPs/kg was estimated by using the sample depth and assuming a density of 1.6 g/cm^3 for the sediment [a] maximum value is shown; [b] no lower value reported. North A. = North America; SA = South America.[1] Nel et al. (2018) [2] Di and Wang (2018) [3] Hu et al. (2018) [4] Lin et al. (2018) [5] Peng et al. (2018) [6] Su et al. (2016) [7] Wang et al. (2017a) [8] Wen et al. (2018) [9] Xiong et al. (2019) [10] Yuan et al. (2019) [11] Zhang et al. (2016) [12] Faure et al. (2015) [14] Horton et al. (2017) [15] Hurley et al. (2018a) [16] Imhof et al. (2013) [17] Imhof et al. (2016) [18] Imhof et al. (2018) [19] Klein et al. (2015) [20] Leslie et al. (2017) [21] Rodrigues et al. (2018) [22] Tibbetts et al. (2018) [23] Vaughan et al. (2017) [24] Fischer et al. (2016) [25] Ballent et al. (2016) [26] Castañeda et al. (2014) [27] Corcoran et al. (2015) [28] Shruti et al. (2019) [29] Vermaire et al. (2017) [30] (Blettler et al. 2017*

Supplemental material

Table S1. Concentration and removal of MPs in municipal WWTPs for different treatment types; **bold** numbers represent the median concentration instead of the mean concentration; NR = not reported.

Treatment type (no. of WWTP)	Location	Mean concentration in MP/L[a] ± SD or (minimum - maximum)		Removal rate (%)	Lowest mesh sizes or lowest size limit (µm)	Identification method	Dominant shapes in effluent	Dominant polymer composition in effluent	Reference
		Influent	Effluent						
Tertiary (1)	Australia	NR	1	NR	NR	Visual and FTIR	Fibres	Polyester, Acrylic	[1]
Primary (1)	Australia	NR	1.54	NR	25	Visual and FTIR	Fibres	PET	[2]
Secondary (1)		NR	0.48	NR					
Tertiary (1)		NR	0.28	NR					
Secondary (3)	Korea	29.8	0.435	98-99	109	Visual and FTIR	Fibres, black particles[c]	NR	[3]
		13.5	0.14						
		13.8	0.28						
Secondary (2)	China	79.9 ± 9.3	28.4 ± 7	64.4	47	Visual and Raman	Fragments	Nylon	[4]
Secondary (2)	Turkey	26.6 (17.3 - 36)	7 (2 - 8.7)	73	5	Visual and µ-Raman	Fibres	PET	[5]
		23.4 (12 - 36)	4.1 (2.7 - 4.7)	79					
Secondary (1)	France	293 (260 - 320)	35 (14 - 50)	NR	100	Visual	Fibres	NR	[6]
Primary (1)	Russia	3787[b]	148[b]	96	20	Visual	Black particles		[7]
Secondary (1)	Finland	57.6 ± 12.4	**1.0 ± 0.4**	98.3	250	Visual and FTIR/Raman	Fibre	PES	[8]
NR (7)	The Netherlands	68 - 910[d]	51 - 81[d]	72	10	Visual and FTIR	Fibres		[9]
Secondary (1)	Sweden	15.1 ± 0.89	0.00825 ± 0.00085	99.9	300	Visual and FTIR	Fibres		[10]
Tertiary (1)	Italy	2.5 ± 0.3	0.4 ± 0.1	84	63	Visual and FTIR	Lines and Films	PES	[11]

Secondary and tertiary (12)	Germany	NR	> 500 µm: 0-0.05; <500 µm: 0.01-9	NR	20	Visual and FTIR	Fibres	PES	[12]
Secondary (1)	Scotland	15.7 ± 5.23	0.25 ± 0.04	98.4	65	Visual and FTIR	Flakes	PES, Polyamine	[13]
Secondary (10)	Denmark	7,771 ± 4,283 [k] (2,223 - 18,285), **7216;** 341 ± 323.9 µg/L [k] (61 - 1189 µg/L), **250 µg/L**	114.3 ± 133.5 [k] (min-max: 19 - 447) **54**; 4.3 ± 4.25 µg/L [k] (0.5-11.0 µg/L), **3.7 µg/L**	99.3; 98.3[e]	10[f]	FTIR and infrared map	Particles	PE and Polyester	[14]
Tertiary (1)	Finland	180 fibres; 430 particles	4.9 ± 1.4 fibres; 8.6 ± 2.5 particles	NR	20	Visual	Particles	NR	[15]
Tertiary (4)[g]	Finland	NR	0.02-0.3	NR	20	Visual and FTIR	NR	PES	[16]
Tertiary (1)	Finland	NR	0.006-0.651[b]	> 99	20	Visual and FTIR	Fragments	Cotton, Polyester[h]	[17]
Secondary (1)	Germany	NR	5.9 (wet weather); 3 (dry weather)		10	Raman	Fragments	PET	[18]
NR (10)	Denmark	127,000; **86,000;** 8,000 µg/L; **5900 µg/L**	5,800**, 6,400;** 34 µg/L, **16 µg/L**	99.7%	20	Visual and FTIR	NR	Nylon	[19]
Tertiary (1)	USA	1	0.0009	99.9	150	Visual and FTIR	NR	NR	[20]
Secondary (1)	USA	NR	0.024 (24h sample); 0.17 (2h peak flow event)	NR	125	Visual and FTIR	Fragments	NR	[21]
Secondary and tertiary (17)	USA	NA	0.05 ± 0.024 (0.004-0.195)	NR	125	Visual	Fibres	NR	[22]
Secondary (1)	USA	NA	5.9[b]	95.6	20	Visual	Fibres	NR	[23]
Tertiary (1)		NA	2.6[b]	97.2					
Tertiary (1)[j]		NA	0.5[b]	99.4					
Secondary (1)	Canada	31.1 ± 6.7	0.5 ± 0.2	97.1-99.1	63	Visual and FTIR	Fibres	Polyester	[24]

Notes: [a] If not indicated otherwise [b] Anthropogenic litter in the micro range in general and not only microplastic considered, [c] Suspected tyre particles; [d] Range of mean concentrations between different WWTP; [e] Retention based on particle mass; [f] Upper size limit was 500 µm; [g] Four different advanced treatment methods were tested; [h] Only fibres considered; [j] Pilot scale anaerobic membrane bioreactor; [k] numbers were calculated based on data in the publication. [1] Browne et al. (2011) [2] Ziajahromi et al. (2017) [3] Lee and Kim (2018) [4] Liu et al. (2019b) [5] Gündoğdu et al. (2018) [6] Dris et al. (2015) [7] HELCOM (2014) [8] Lares et al. (2018) [9] Leslie et al. (2017) [10] Magnusson et al. (2016) [11] Magni et al. (2019) [12] Mintenig et al. (2017) [13] Murphy et al. (2016) [14] Simon et al. (2018)

[15] Talvitie et al. (2015) [16] Talvitie et al. (2017a) [17] Talvitie et al. (2017b) [18] Wolff et al. (2018) [19] Vollertsen and Hansen (2017) [20] Carr et al. (2016) [21] Dyachenko et al. (2017) [22] Mason et al. (2016) [23] Michielssen et al. (2016) [24] Gies et al. (2018)

Table S2. Concentration of MPs in sludge from municipal WWTPs. **Bold** numbers represent the median concentration instead of the mean concentration; NR = not reported.

Sludge type (no of WWTPs)	**Location**	**Mean Concentration ± SD in in MP/kg dw[a] or minimum-maximum**	**Retained in sludge (%)**	**Lowest mesh sizes or assessed size (µm)**	**Identification method**	**Dominant shapes in effluent**	**Dominant polymer composition in effluent**	**Reference**
Secondary sludge after thickening and dehydration	Korea,	14,895	49.3	106	Visual and FTIR	Fragments (mainly black)	NR	[1]
Secondary sludge after thickening and dehydration	Korea	9,655	44.7	106	Visual and FTIR	Fragments (slightly more than fibres (only fibres and fragments reported)	NR	[1]
Mix of primary and secondary sludge after thickening and dehydration	Korea	13,200	49.0	106	Visual and FTIR	Fragments (slightly more than fibres (only fibres and fragments reported)	NR	[1]

Mix of primary and secondary sludge	China	240,300	NR	20	Visual and Raman	Fragments	Nylon	[2]
Sludge	Spain	NR	NR	20 / 200	visual and FTIR, differential scanning calorimeter	NR	NR	[3]
Sludge (3)	The Netherlands	650 (370-950)[b]	72	10	visual and FTIR	Fibres	NR	[4]
Recycled activated sludge (1)	Italy	113,000 ± 57,000[b]	NR		visual and FTIR	Fibres	PES	[5]
Sludge (7)	Ireland	4,196 - 15,386	NR	45	visual and FTIR/Raman	Fibres	PE, PES acrylic, PET, PP, polyamide	[6]
Activated sludge (1)	Finnland	23,000 ± 4,200	NR	20	Visual and FTIR/Raman	Fibres	Polyester	[7]
Digested sludge (1)	Finnland	170,900 ± 28,700	NR	20				
Membrane bioreactor sludge (1)	Finnland	27,300 ± 4,700	NR	20				
Sludge (1)	Sweden	16,700 ± 1,960; 720 ±112[b]	NR	300	Visual	Fibres	NR	[8]
Primary Sludge (6)	Germany	1x 10^3 - 2.4 x 10^4	NR	20	Visual and FTIR	Fibres	PE	[9]
Sludge (1) (24 h duplicate)	Scotland	800[b,c]	NR	65	Visual and FTIR	NR	PES, acrylic, PP, alkyd, PS	[10]

Raw sludge	Finnland, Europe	76,300		20	Visual and FTIR		NR	[11]
Dry sludge		186700	99.9					
Digested sludge (5)	Denkmark, Europe	4.5 mg/g; 169 000 MPs/g **158,000** MPs/g; **6.5** mg/g			Visual and FTIR	NR	PE	[12]
Sludge as biosolid (1)	USA, North America	1,000	99.9 (in grid and biosolids)	20		NR	NR	[13]
Primary sludge (1)	Canada, North America	14,900[b]	NR	1	visual	Fibres	NR	[14]
Secondary sludge (1)		4,400[b]	NR	1				
Sludge as biosolid	North America	3,000 - 4,000	NR	NR	visual	Fibres	NR	[15]

Notes: [a] If not indicated otherwise; [b] concentration in wet weight, [c] concentration estimated from figure

[1] (Lee et al. 2019) [2] (Liu et al. 2019b) [3] (Bayo et al. 2016) [4] (Leslie et al. 2017) [5] (Magni et al. 2019) [6] (Mahon et al. 2017) [7] (Lares et al. 2018) [8] (Magnusson and Norén 2014) [9] (Mintenig et al. 2017) [10] (Murphy et al. 2016) [11] (Talvitie et al. 2017a) [12] (Vollertsen and Hansen 2017) [13] (Carr et al. 2016) [14] (Gies et al. 2018) [15] (Zubris and Richards 2005)

Table S3. Concentration of MPs and MAPs in different soil types. **Bold** numbers represent the median concentration; ** Most common shape or polymer type observed. NR = not reported. Dw = dry weight; ww = wet weight.

Soil type (number of fields)	Location	Plastic type (size in mm)	Mean concentration MPs/kg dw[a] (minimum-maximum)	Mean concentration of MeP or MaP/kg	Identification method	Reported shapes	Reported polymer composition	Reference
Industrial	Australia	MPs (< 1)	300 - 67,500 mg/kg	NR	FTIR	NR	**PVC,** PE, PS,	[1]
Agricultural (vegetable fields)	China	MPs (0.02- 5) / MePs (4- 20)	0-3 cm sample depth: 78.00 ± 12.91; 3-6 sample	0-3 cm sample depth: 6.75 ± 1.51; 3-6 cm	Visually, FTIR	**Fibres, Fragments,** films,	**PP, PE,** PES	[2]

			depth cm: 62.50 ± 12.97	sample depth: 3.25 ± 1.04		pellets		
Agricultural (Greenhouse) (4)	China	MPs /MePs (0.05-10)	18,760 (7,100 - 42,960)[d]	NR	Visually	**Fibres,** fragments, films	NR	[3]
Buffer (former crop land)			14,360 (8,180 - 18,100)[d]	NR				
Agricultural	China		0-10 cm sample depth: 40 ± 126; 0.008 ± 0.025mg/kg; 10-30 cm sample depth: 100 ± 141; 0.368 ± 0.740 mg/kg	NR	Visually	PE, PP		[4]
Fruit field			0-10 cm sample depth: 320 ± 329; 0.540 ± 0.603 mg/kg; 10-30 cm sample depth: 120 ± 169; 0.460 ± 0.735 mg/kg	NR				
Agricultural (Greenhouse)			0-10 cm sample depth: 100 ± 254; 0.130 ± 0.307 mg/kg; 10-30 cm sample depth: 80 ± 193; 0.024 ± 0.051 mg/kg	**NR**				
Agricultural	Germany	MPs (1-5) and MaPs (>5)	0.34 ± 0.36	206 MaPs/ha or 0.066 kg MaPs/ha;	Visuall and FTIR	**Fragments, Films,**	MaPs: **PE**; PS ,PP, PVC, PET,	[5]

						Fibres	PMMA MPs: **PE**, PP, PS	
Floodplain	Switzerland	MPs (0.125-5) and MePs (5-25)	5 mg/kg dw (0-55mg/kg dw)	**NR**	FTIR	**PE,** PA, natural latex, PS, PVC, SBR, PP		[6]
Agricultural	Denmark	MPs (0.02-0.5)	145,000[f] ;12 mg/kg[f]	NR	Visual and FTIR	PE, Nylon and PP		[7]
Agricultural (sewage sludge applied)			71,000[f] ; 5.8mg/kg[f]					
Agricultural	USA	Synthetic fibres	580 (± 403) – 1210 (± 250)[g]	NR	Polarized microscopy	Fibres		[8]
Agricultural (6) 0-25 cm	Chile	MPs[i]	**1.37 - 4.38[h]** (0.73 -12.9) mg/kg dw	**NR**	Visual	**Fibres,** **F**ilms, Fragments, Pellets		[9]
Rural home gardens (agro-foresty land-use system)	Mexico	MPs (<5[i])/ MaPs (> 5)	870 ± 1,900	744,000 ± 204 000 PE bottles /ha 74 000 ± 65 000 MaPs/m^2	Visual	NR	NR	[10]
Horticulture	Argentina	PE MaP (> 20)	NR	3 ± 1.9 g/m^2	Visually	Films[k]	PE[k]	[11]
Dessert	USA	MaP	NR	0.056 – 0.344 bags /ha; 0.392-0.627 balloon clusteres/ha	Visually	Bags, balloons[l]	PE, latex	[12]

Notes: [a] If not indicated otherwise; [b] Wet weight; [c] Calculated mean concentration from all dept fraction; [d] Not distinguished into concentration of MPs and MePs but 95% of the observed plastic particles are in the MPs size (0,05 -1mm) ; [e] Extraction method was only suitable for low density plastics; [f] Not reported if dry or wet weight; [g] Only synthetic fibres concentration assessed; [h]Range of medians is shown, which was increasing with increasing number of sludge applications; [i] Lower detection limit not reported; [k] Only PE film concentration assessed; [l] Only bags and balloons assessed. [1] Fuller and Gautam (2016) [2] Liu et al. (2018) [3] Zhang and Liu (2018) [4] Zhang et al. (2018) [5] Piehl et al. (2018) [6] Scheurer and Bigalke (2018) [7] Vollertsen and Hansen (2017) [8] Zubris and Richards (2005) [9] Corradini et al. (2019) [10] Huerta Lwanga et al. (2017) [11] Ramos et al. (2015) [12] Zylstra (2013)

Table S4. MaPs reported in surface waters.

Waterbody (number of waterbodies)	**Location**	**Sample type (sampling type)**	**Reported Concentration range**	**Lowest assessed size (mm)**	**Observed plastic types**	**Reference**
Saigon River	Vietnam	Water (floated plastic intercepted by nets)	Median estimated amount entering the river: 4.43 **(**0.96 - 19.9) g/inhabitant/day	20	Bags, bottles, drinking recipients, plastic cutlery PE (79%), PP (15%), PET (4%)	[1]
Saigon River	Vietnam	Water (floated plastic intercepted by nets)	0.2 - 0.3 tons per day emitted to the ocean	50	PS food container fragments), PS foam polyolefin bags and food wrappings, caps, lids, polyolefin hard plastic, fragments, cups, bottles, straws, others	[2]
Yangtze River	China	Water (trawl net)	Mean: 8.74 x 10^3 items/km^2 (1.94 x 10^3-2.78 x 10^4 items/km^2)	5[b]		[3]
Rhone River	France	Water (visual observation from elevated points)	NR	70	Bags, sheets, bottles, covers/packaging, others	[4]
Tiber River	Italy	Water (visual observation from elevated points)	1,270 litter items/km^2; 190 litter items > 20 cm/km^2[c]	25	Plastic pieces, bottles, covers, polystyrene pieces, cover/packaging, foam	[5]
Lakes (7)	Switzerland	Water (Manta trawl)	Mean: 1,800 ± 3,100 items/km^2, 44,000 ± 80,000 mg/km^2; median: 860 items/km^2, 12,000 mg/km^2	5	PE (mainly packaging films), PP (mainly fragments), PS (mainly foams)	[6]

Rivers (5)	Switzerland	Water	Mean: 0.012±0.034 items/m^3, 0.43 ± 12 mg/m^3; median: 0	5		[6]
River Seine	France	Water (plastic trapped by floating debris retention booms)	27,000 tons floating plastic intercepted annually		PP, PE, PET	[7]
Upper Thames estuary	UK	Water (plastic intercepted by nets close to riverbed)	Total number of items: 8480[c]	NR	Bags, cups, plates, forks, food wrappers, Tabaco packaging, sanitary components, others	[8]
Los Angeles River	USA	Water (plastic intercepted by nets)	819 items/m^3	4.75	NR	[9]
San Garbiel River	USA	Water (plastic intercepted by nets)	125 items/m^3	4.75	NR	[9]

Notes: [b] only mesoplastics and not MaPs assessed but the upper size limit is unknown; [c] Floating litter in general assessed and not only plastic. [1] Lahens et al. (2018) [2] van Emmerik et al. (2018) [3] Xiong et al. (2019) [4] Castro-Jiménez et al. (2019) [5] Crosti et al. (2018) [6] Faure et al. (2015) [7] Gasperi et al. (2014) [8] Morritt et al. (2014) [9] Moore et al. (2011)

Table S5. Concentration of MPs in different waterbodies with sample type, mesh size limit, identification methods, reported shapes and polymer compositions. **Bold** numbers represent the median concentration; * Concentration in MPs/ m^3 was estimated by dividing the reported concentration in particles per area by the height of the net used for sampling; ** Most common shape or polymer type observed. NR = not reported. Dw = dry weight; ww = wet weight.

Waterbody (number of waterbodies)	**Location**	**Mean concentration in MP/m$^{3\ a}$ ± SD or (minimum - maximum)**	**Sample type**	**Lowest mesh sizes or lowest size limit (μm)**	**Identification method**	**Reported shapes**	**Reported polymer composition**	**Reference**
Hovsgol Lake (1)	Mongolia	0.127 (0.01 - 0.28)*	Manta trawl	333	Visual	Fragments films, lines/fibres	NR	[1]

Yangtze River (1)	China	4,700 ± 2,800; **4130** (1600 - 1.26 x10^4)	Water pumped through stainless steel sieve	48	Visual and Raman	Fibres**, Fragments, (Pellets)	PS**, PP, PE, PC, PVC, Vinyl chloride, others	[2]
Small Waterbodies in Yangtze River catchment (25)	China	480 - 21,520	Grab water	20	Visual and FTIR	Fibres	PES	[3]
Perl River (1)	China	2,724 (379 - 7,924)	Water sieved	20	Visual and FTIR	Fibres	PE, PP	[4]
Shanghai creeks (1)	China	1.25 (0.5 -4 .2)[b]	Pump or metal pail	20	Visual and FTIR	Fibres**, Fragments, Films/Pellets	PES**, Rayon, PP	[5]
Suzhou River (1)		1.5 (0.1 - 7.5)[b]						
Huangpu River (1)		0.95 (0.25 - 7.5)[b]						
Taihu lake (1)	China	0.03 (20[d])	Plankton net	333	Visual and FTIR	Fibres**, Fragments, Films/Pellets	Cellophane**, PET, PES, Terephtalic acid, PP	[6]
		3,400 - 25,800	Surface water grab sample	5				
Feilaixia Reservoir Beijiang River (1)	China	0.56	Plankton net	112	Visual and FTIR	Foams**, Fragments, Films, Fibres	PP**, PE, EPS, PS, PET, PVC	[7]
Yangtze River (1)	China	2,516 (1,400 - 4,000)	Water pumped through stainless steel sieve	50	Visual and FTIR	Fibres**, Granules, Films, (Pellets)	PET**, PP, PE, Nylon, PS	[8]
Hanjiang River (1)		29,933 (2,600 - 3,200)						
Surface waters of Wuhan: Lakes (20), Yangtze River (1) and Hanjiang River (1)		1,660 ± 6,391 – 8,925 ±1,591						
Dongting Lake (1)	China	1,191.7	Water pumped through stainless steel sieve	50	Visual and Raman	Fibres**, Granules, Films	PE**, PP**, PS, PVC	[9]
Hong Lake (1)		2,282.5						
Yangtze River (1)	China	0.86 (0.34 - 1.58)*	Trawl net	333	Visual and Raman	Sheets**, Fragments**, Foams, Lines	PP**, PE, PS, others	[10]
Yangtze River (1)		2,113 (1,260 - 4,340)	Water filtered through plankton net	64				
Pearl River, urban	China	19,860 (8,750 - 53,250)	Water filtered	50	Visual and Raman	Films**,	Polyamide**,	[11]

part (1)			through stainless steel sieve			Granules, Fibres	Cellophane, PP, PE	
Pearl River, estuary part (1)		8,902 (7,850 - 1.1x10^4)				Granules**, Films**,Fibres		
Poyang Lake (1)	China	5,000 – 34,000	Water filtered through stainless steel sieve	50	Visual and Raman	Fibres**, Films, Fragments, Pellets	PP, PE, Nylon, PVC	[12]
Yangtze River (1)	China	16.8 (5.96 - 23.83)*	Trawl net	112	Visual and FTIR	Sheets**, Fragments, Lines, Foam	PE**, PP**, PS	[13]
Yangtze River Tributaries (4)		6.663 (0.34 - 20.81)*						
Rivers (29)	Japan	**7.9;** 1.6 ± 2.3 (0 - 12)	Plankton net	335	FTIR	Fragments (only mentioned)	PE**, PP, PS, others	[14]
Saigon River and Canals (1)	Vietnam	10 - 223	Net sample			Fragments	PE**, PP, PE-PP, PS, others	[15]
		17,200 - 519,000	Grab water sample	300	Visual and FTIR	Fibres**	PES**, PET PE, PP, others	
River Seine (1)	France	30 (3 - 108)	Plankton net	80	Visual	Fibres	NR	[16]
		0.35 (0.28 - 0.47)	Mantra trawl	330		Fibres, Fragments, Spheres		
Lakes (7)	Switzerland	0.51 ± 0.67; **0.27***	Manta trawl	300	Visual and FTIR	Fragments**, Foams, Films, Fibres, Others	PE**, PP, PS	[17]
Rivers (4)		7 ± 0.2; **0.36**						
Lake Geneva (1)	Switzerland	0.193*	Manta trawl	300	Visual	NR	NR	[18]
Lake Bolsena (1)	Italy	0.82 - 4.42	Manta trawl	300	Visual (UV microscope) and SEM	Fragments**, Fibres**	NR	[19]
Lake Chiusi (1)		2.68 - 3.36				Fibres**, Fragments		

Lake Iseo (1)	Italy	0.2*	Manta trawl	300	Visual and FTIR	Fragments**, Filaments, sheets, others	PE**, EPS; PP, others	[20]
Lake Maggiore (1)		0.195*						
Lake Garda (1)		0.125*						
Danube (1,Year 2010)	Austria	0.983 (0 - 141.66)*	Conical drift nets	500	Visual	Spherules**, Flakes, Pellets	NR	[21]
Danube (1,Year 2012)		0.055 (0 - 0.75)*				Pellets**, Flakes, Spherules		
Amsterdam Canals (1)	Netherlands	100,000 ± 49,000 (48,000 - 187,000)	Grab sample	10	Visual and FTIR	Fibres**, Spheres, Foils	NR	[22]
Storm water ponds (1)	Denmark	1,409 (490 - 22,894)	Filtered through stainless steel mesh	10	Visual and FTIR	NR	PP**, PVC ,PES, PE, PS, Others	[23]
Rhine (1)	Switzerland, France, Germany	4.96; **2.196;** 2, 684^{d,*}	Manta trawl	300	Visual and FTIR	Spherules**, Fragments, Fibres	PS**, PP, Others	[24]
Antua River, Portugal, March (1)	Portugal	58 - 193	Surface and bottom water filtered	55	Visual and FTIR	Fragments**, Fibres, Foams, Films, Pellets	PE**, PP, Others	[25]
Antua River, Portugal, October (1)		71 - 1,265						
Gallatin River watershed (1)	USA	1,200 (0 - 67,500)	Grab sample	100	Visual and FTIR	Fibres**, Fragments, (Beads)	Semi-synthetic cellulose**, PET, PES, PVA, Neoprene	[26]
Great Lake Tributaries (29)	USA	1.9; **4.2** (0.05 - 32)	Neuston net	333	Visual	Fibres**, Fragments, Foams, Films	NR	[27]
Raritan River	USA	24.0	Plankton net	125	Visual	NR	NR	[28]

upstream WWTP (1)								
Raritan River downstream WWTP (1)		71.7						
Lake Superior (1)	USA	**0.022** (0.01 - 0.08)*	Manta trawl	333	Visual and SEM/EDS	Pellets**, Fragments, Foams, Films, Lines	NR	[29]
Lake Huron (1)		**0.012** (0 - 0.04)*						
Lake Erie (1)		**0.127** (0.03 - 2.91)*						
Snake River/Columbia River (1)	USA	91 ± 1,140 (0 - 5 405,000)	Grab sample	100	Raman	Fibres**, Fragments, Beads, Films	PP, PE, PET, PES	[30]
Snake River/Columbia River (1)		2.57 ± 2.95 (0 - 13.5)	Net sample	100				
Lakes in Lubbock Texas (1)	USA	53 - 105 µm: 0.79 - 1.56 mg/L; 106 - 179 µm: 0.31 - 1.25 mg/L	Grab sample	53-179	Visual	Beads, Filamentous MPs, Irregular MPs	NR	[31]
Wetlands in Texas (1)		53 -1 05 µm: 0.64 - 5.51 mg/L; 106 - 179 µm: 0 - 1.79 mg/L						
Streams Chicago metropolitan area, upstream WWTP (1)	USA	2.355 ± 0.375	Neuston nets	333	Visual and Py-GCMS	Pellets**, Fibres**, Fragments**, others	PP**, PE**, PS**	[32]
Streams Chicago metropolitan area, downstream WWTP (1)		5.733 ± 0.850						

North Shore Channel Chicago, upstream WWTP (1)	USA	1.94 ± 0.81	Neuston nets	333	SEM	Fibres**, Fragments	NR	[33]
North Shore Channel Chicago, downstream WWTP (1)		17.93 ± 11.05				Fibres**, Fragments, Pellets, Styrofoam		
Hudson River (1)	USA	980[c]	Grab sample	330	FTIR	Fibres	NR	[34]
La River (1)	USA	22 - 12,932	Manta trawl, Hand nets, Rectangular nets, streambed sampler	1000	Visual	Foamed plastic**, Pellets, Fragments, Films, Lines, whole items	NR	[35]
San Gabriel River (1)		0 - 337				Foamed plastic**, Fragments, Films, whole items, Lines, Pellets		
Coyote Creek (1)		27,211				Fragments**, Foams, Lines, Pellets, whole items		
Estuarine Rivers Chesapeake Bay (1)	USA	0 - 0.036g/m^{3}*	Surface trawl	330	Visual and Raman	Fragments**, Sheets**, Fibres, EPS, others	PE	[36]

Lake Superior (1)	USA	0.263 ± 0.193 (0-0.786)* 0.086 (0 - 0.0253 mg/$m^{3)}$	Manta trawl	333- 4 mm?	Visual and yr-GC/MS and ATR-FTIR	Fibres**, Fragments, Films, (Beads, Foams, Others)	PE, PVC, PP, PET, others	[37]
Lake Winnipeg (1)	Canada	1.07 (0.29 - 4.16)	Manta trawl	333	Visual and SEM-XDS	Fibres**, Fragments, Films, Foams	NR	[38]
Ottawa river and tributaries, nearshore (1)	Canada	100 (50 - 240)	Grab sample filtered	100	Visual	Fibres	NR	[39]
Ottawa river and tributaries, open water (1)		1.35	Manta trawl			Fibres**, Fragments, Beads		
Ottawa river upstream WWTP (1)		0.71				NR		
Ottawa river downstream WWTP (1)		1.99				NR		

Notes: [a] If not indicated otherwise; [b] Numbers for different sample sites were estimated from graph and mean numbers were calculated; [c]Only fibres included; [d] Maximum observed concentration. [1] Free et al. (2014) [2] Di and Wang (2018) [3] Hu et al. (2018) [4] Lin et al. (2018) [5] Luo et al. (2018) [6] Su et al. (2016) [7] Tan et al. (2019) [8] Wang et al. (2017b) [9] Wang et al. (2018) [10] Xiong et al. (2019) [11] Yan et al. (2019) [12] Yuan et al. (2019) [13] Zhang et al. (2015) [14] Kataoka et al. (2019) [15] Lahens et al. (2018) [16] Dris et al. (2015) [17] Faure et al. (2015) [18] Faure et al. (2012) [19] Fischer et al. (2016) [20] Sighicelli et al. (2018) [21] Lechner et al. (2014) [22] Leslie et al. (2017) [23] Liu et al. (2019a) [24] Mani et al. (2016) [25] Rodrigues et al. (2018) [26] Barrows et al. (2018) [27] Baldwin et al. (2016) [28] Estahbanati and Fahrenfeld (2016) [29] Eriksen et al. (2013) [30] Kapp and Yeatman (2018) [31] Lasee et al. (2017) [32] McCormick et al. (2016) [33] McCormick et al. (2014) [34] Miller et al. (2017) [35] Moore et al. (2011) [36] Yonkos et al. (2014) [37] Hendrickson et al. (2018) [38] Anderson et al. (2017) [39] Vermaire et al. (2017)

Table S6. Concentration of MaPs in sediments. **Bold** numbers represent the median concentration

Waterbody (no of waterbodies)	**Location**	**Sample type**	**Reported mean (min-max) concentration[a]**	**Lowest assessed size (mm)**	**Observed plastic types**	**Reference**
Rivers (6)	Switzerland	Beach sediment	90 ± 250 items/m^2; 14,000 ± 33,000 mg	5	PE (mainly packaging films), PP (mainly fragments), PS (mainly	[1]

			/m; **11** items /m^2, **480** mg/m^2		foams)	
Lake Bolsena	Italy	Beach sediment	North shore: 2.57 items/m^2, 2.6 g/m^2; South shore: 0.28 items/m^2, 1.1 g/m^2	5	Industrial packaging (PE, PP), food packaging (PE), net/rope/string/cord (polyamide, PVC, polyacrylonitrile), others	[2]
Lake Chiusi	Italy	Beach sediment	East shore: 5 items/m^2, 4.5 g/m^2; west shore: 0.22 items/m^2, 0.2 g/m^2	5	Cigarette butts (cellulose acetate), net/rope/string/cord (polyamide, PVC, polyacrylonitrile), others	[2]
Albegna River	Italy	Sediment	Winter: 7-12 items/kg; Summer: 16 -43 items/kg	5.1 – 25	Filaments, fragments, others	[3]
Osa River			Winter: 7-12 items/kg; Summer: 16 -43 items/kg			
Ombro River			Winter: 0 items/kg; Summer 0-14 items/kg			
Lake Garda	Italy	Beach sediment	North shore: 483 ± 236 items/m^2; South shore: 0 - 8.3 items/m^2	5	NR	[4]
Lake Garda	Italy	Beach sediment	5 ± 9 items/m^2	5	NR	[5]
Edgbaston Pool,	UK	Sediment	2- 20 items per sampling site	NR	Food wrappers, bottle caps, plastic bags, Styrofoam, bottles, films, fragments, ropes, straws, syringe, cosmetic tubes, fibrous clothing	[6]
River Taff	UK	River bank	584 plastic items/0.1 km river bank [c]	NR	Plastic, packaging, others	[7]
Lake Ontario	Canada	Beach sediment	366 items/kg[b]	5	Fragments	[8]
Lake shoreline	Argentina	Shoreline sediment	meso: 25 items/m^2, 19 g/m^2; MaP: 1.15	meso: 5 -25 MaP > 25	Food wrappers (PP &PS), bags (PE), bottles (PET), Styrofoam food	[9]

			items/ m^2; 4.9 g m^2;		containers (expanded PS)	
Rivers (4)	Chile	River bank	up to **3.4** items/m$^{2, c}$	1.5	NR	[10]

Notes: [a] If not indicated otherwise; [b] Mean has been calculated across different sampling dates and includes only fragments; [c] Persistent buoyant litter in general assessed and not only plastic. [1] Faure et al. (2015) [2] Fischer et al. (2016) [3] Guerranti et al. (2017) [4] Imhof et al. (2013) [5] Imhof et al. (2018) [6] Vaughan et al. (2017) [7] Williams and Simmons (1999) [8] Corcoran et al. (2015) [9] Blettler et al. (2017) [10] Rech et al. (2014)

Table S7. Concentration of MPs sediments of different waterbodies with sample type, mesh size limit, Identification methods, reported shapes and polymer compositions.
Bold numbers represent the median concentration; * Concentration in MPs/kg was estimated by using the sample depth and assuming a density of 1.6 g/cm^3 for the sediment; ** Most common shape or polymer type observed. NR = not reported.

Waterbody (number of waterbodies)	**location**	**Sample type**	**Average concentration in MP/kg dry weight ±SD or (minimum-maximum)[a]**	**Type of analysis**	**Lowest measured size (µm)**	**Reported shapes**	**Reported polymer compositions**	**Reference**
Bloukrans River (summer)	South Africa	Bed sediment	6.3 ± 4.3 (1 - 14.61)	Visual	63	Fibres	NR	[1]
Bloukrans River (winter)			160.1 ± 139.5 (13.3 - 563.8)					
Three Gorges Reservoir	China	Bed sediment	82 ± 60 (25 - 300)[b]	Visual and Raman	48	Fibres**, Fragments, Pellets, Film, Styrofoam	PS**, PP, PE	[2]
Small waterbodies in Yangtze River Delta	China	Bed sediment	35.8 - 3185[c]	Visual and FTIR	20	Fibres**, Fragment, Granule	PP, PE, PES	[3]
Pearl River	China	Bed sediment	1,669 (80 - 9,597)[c]	Visual and FTIR	20	Fibres, Fragments, Films	PE**, PP,	[4]
Rivers Shanghai (6)	China	Bed sediment	802 ± 594	Visual and FTIR	1	Spheres**, Fibres, Fragments	PP**, PES, Rayon, Others.	[5]
Vembanad Lake	India	Bed sediment	252.8 ± 25.76 (96 - 496)	Visual and Raman		Films**, Foams**, Fragments, Fibres	High density PE, Low density PE, PS; PP	[6]

			MPs/m^{2}[g]					
Taihu Lake	China	Bed sediment	11 - 234.6	Visual and Raman	1.2	Fibres, Fragments, Films, Pellets	Cellophane**, PET, PES, terephthalic acid, PP	[7]
Beijiang River	China	Shore sediment	178 ± 69 - 544 ± 107[d]	Visual and FTIR	1	NR	PE**, PP, Copolymer	[8]
Lakes in Changsha (12)	China	Bed sediment	270.17 ± 48.23 - 866.54 ± 37.96[d]	Visual and Raman	<500	Fragments**, Fibres, Films, Foams	PS**, PE, PET, PP, Polyamide, PVC	[9]
Yangtze River	China	Bed sediment	34 (7 - 66)[c]	Visual and Raman	333	NR	NR	[10]
Poyang Lake	China	Bed sediment	54 - 506	Visual and Raman	50	Fibres**, Fragments, Films, Pellets	PP**, PE PVC, Nylon	[11]
Lakes Siling Co Basin (4)	China	Shore sediment	<0.125 – 17.63 ± 35.3	Visual and Raman	1	Sheets, Lines, Fragments Foams	PP**, PE**, PVC, PET, PS	[12]
Lakes (6)	Switzerland	Beach sediment	16.25 ± 25 (10 - 86.25)*	Visual and FTIR	300	Foams, Fragments, Fibres, Films, Pellets. Lines, Beads	PE, PP, PS, PVC	[13]
Lakes Bolsena	Italy	shore sediment	40.04 ± 13.8*	UV microscope and Scanning Electronic Microscope	300	Fragments**, Fibres		[14]
Lake Chiusi			44.10 ± 14.48*			Fibres**, Fragments		
Thames tributaries								[15]
Leach	UK	Bed sediment	185 ± 42	Visual and Raman / x-XRF	1000[e]	Fibres**, Fragments, Films	PES**, PET**, PP polyacrylsulphane, PE, PS PVC and others	
Lambourn			221 ± 95					
Cut Site 1			665 ± 77					
Cut Site 2			332 ± 161*			Fragments**, Fibres, Films		
Upper Mersey and Irwell catchments	UK	Bed sediment	323.13*[f]	Visual and FTIR	50	Beads**, Fragments, Fibres	NR	[16]

Lake Garda	Italy	Beach sediment	43.85 ± 110.69*	Visual and Raman	1	NR	NR	[17]
Lake Garda	Italy	Beach sediment	0.94 ± 1.68	Visual and Raman	1	Plastic and paint particles	Polyamide, PE, PS, PP and others	[18]
Lake Garda	Italy	Beach sediment	1.35 ± 0.69 - 13.85 ± 12.29*	Visual and Raman	9	Fragments**, Fibres	PS, PE	[19]
Rivers Rhine and Main	Germany	Shore sediment	228 - 3 763	Visual and FTIR	63	Fragments**, Fibres**, Spherules	PS**, PE, PP, Polyamide, Others	[20]
Amsterdam canals	Netherlands	Bed sediments	2,071 ± 4,246 (0 - 10,500)	Visual and FTIR	10	Spheres, Fibres, Foils	NR	[21]
Antuã River (Spring)	Portugal	Bed sediment	100 - 624	Visual and FTIR	55	Fragments**, Fibres, Foams, Pellets	PE**,PP**, PS, PET	[22]
Antuã River (Autumn)			18 - 514					
River Tames and tributaries	UK	Bed sediment	165 (20 - 350)	Visual and FTIR	63	Fragments**, Fibres, Spheres, Foames, Films, irregular Spheres, commercial Fragments	PE**, PVC, Polyamide,	[23]
Edgbaston Pool	UK	Bed sediment	250 - 300	Visual	500	Fibres, films, foams, Fragments	NR	[24]
Lake Ontario	Canada	Shore and beach sediment	760 (20 - 27,830)	Visual and Raman/x-XRF (X-ray fluorescence spectroscopy)	63	Fragments**, Fibres, Beads	PE**, PS, Polyurethane, PP, PVC, PSS, Others	[25]
St Lawrence River	Canada	Bed Sediment	85.99 ± 86.53*; **0.325*** (0 -248.75*)	Visual	500	NR	NR	[26]
Lake Ontario	Great Lakes, Canada	Beach sediment	19,175.2 – 27,774.79[h]	Visual and Raman, FTIR	<1	Pellets**, Fragments and polystyrene	PE**, PP, NC	[27]
Atoyac River	Mexico	Bed sediment	155.17 (33.33 - 266.67)[d]	Visual	1.2	Films**, Fragments, Fibres, pellets		[28]
Ottawa River and tributaries	Canada	Bed sediment	220	Visual	100	Fibres**, beads fragments		[29]
Lake Huron	Great	Beach	408 MPs/m^{2,}	Visual and	NR[i]	Pellets**,	PE**, PP, PET	[30]

	Lakes, Canada	sediment	[f,g]	FTIR/ SEM		fragments, Styrofoam		
Lakes Erie	Canada	Beach sediment	0.36 - 3.7 MPs/m^{2}[g]	Visual and FTIR	NR[i]	Fragments**, Pellets, Styrofoam	PE, PP	[31]
Lake St. Clair			0.18 - 8.38 MPs/m^{2}[g]			Fragments**, Styrofoam, Pellets,		
Lake Huron			0.98 - 34 MPs/m^{2}[g]			Pellets**, Fragments, Styrofoam		
Setúbal Lake	Argentina	Shore sediment	14.67*	Visual and FTIR	350	Hard plastic, Fibres		[32]
River Albegna	Italy	Bed sediment	Winter: 305 - 477; Summer: 202-253	Visual	10	Filaments, Fragments, (Films)	NR	[33]
River Osa			Winter: 312; Summer 259					
River Ombrone			Winter: 75 - 188; Summer: 137 - 168					

Notes: [a] If not indicated otherwise; [b]Wet weigth; [c]Not mentioned if wet or dry, [d] Range of means between different sampling sites, [e]Upper size limit 4000 µm; [f]Maximum observed concentration; [g]Not sufficient information available to estimate the number in MPs/kg, [h] Calculated from data in the paper.
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