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Wang, Zhenfeng
Su, Bobo
Xu, Xiaoqun
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Preferential accumulation of small (<300 μm) microplastics in the sediments of a coastal plain river network in eastern China

Zhenfeng Wang [a], Bobo Su [a], Xiaoqun Xu [b], Di Di [a], Hong Huang [a], Kun Mei [a], Randy A. Dahlgren [a, c], Minghua Zhang [a, c], Xu Shang [a, *]

[a] Key Laboratory of Watershed Sciences and Health of Zhejiang Province, School of Public Health and Management, Wenzhou Medical University, Wenzhou 325035, China
[b] Laboratory of Marine Ecosystem and Biogeochemistry, Second Institute of Oceanography SOA, Hangzhou 310012, China
[c] Department of Land, Air and Water Resources, University of California, Davis, CA 95616, USA

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ABSTRACT

Microplastics are a global concern for their threat to marine ecosystems. Recent studies report a lack of smaller microplastics (<300 μm) in oceans attributed to a “loss in ocean”. Several hypotheses have been proposed to explain the absence of smaller microplastics, but their fate and transport remain an enigma. Our study recovered high concentrations of microplastics (32947 ± 15342 items kg^{-1} dry sediment) from sediments of a coastal plain river network in eastern China, with the <300 μm fraction accounting for ~85% of total microplastic particles. Microplastic concentrations were generally higher in sediments from tributary streams and streams surrounded by industrial land use. The high variability of microplastics within the watershed indicates that the distribution of microplastics is regulated by several factors, such as distance to source(s), river flow characteristics, buoyancy behavior, degradation, etc. Fragment and foam forms dominated the small microplastics, while fibers were less prevalent in the <300 μm fraction and more abundant in downstream sites. The dominance of small microplastics in riverine sediments in this study provides a possible mechanism to explain the relative absence of small microplastics in the ocean, and advocates for quantification of the whole size spectrum of microplastics in future studies of riverine microplastic fluxes.

1. Introduction

Since development of modern plastics in the early 1900s, these incredibly versatile materials became an indispensable component of human life. Plastics are inexpensive, strong, lightweight, durable, and chemically resistive, with high thermal and electrical insulation properties. All these advantages make plastics widely used throughout the world with annual plastic production increasing from 1.5 million tonnes in the 1950s to more than 320 million tonnes per year currently (Plastics Europe, 2016). Only a small proportion of plastic waste is recycled resulting in large quantities being disposed of in landfills (Barnes et al., 2009) or ending up in the environment as a pollutant (Cole et al., 2011). It is estimated that 4.8 to 12.7 million tonnes of plastic wastes end up annually in oceans through river inputs, and a steady increase is expected in the future (Jambeck et al., 2015). Micro-scale plastic particles of industrial origin, like those used in cleaning and cosmetic products, are suggested as a primary microplastic (MP) source, while plastic debris undergoing mechanical, chemical and biological degradation can result in fragmentation to form secondary MPs (Andrady, 2011; Zettler et al., 2013). MPs are pervasive across global environments (Wright et al., 2013), but there is particular concern about the threat of MPs to aquatic organisms and ecosystems, and even human health (Eerkes-Medrano et al., 2015; Wright and Kelly, 2017).

Size is an important feature regulating the potential hazards of MPs, with small MPs considered an extreme threat to aquatic organisms due to ease of ingestion and the potential to retain other pollutants on their large surface area (Auta et al., 2017). Although the relative absence of small MPs in the surface ocean was previously reported (Cózar et al., 2014; Eriksen et al., 2014), a study in the South China Sea showed that small particles dominated the MPs in the surface water (Cai et al., 2018). These inconsistencies make the

* Corresponding author.
E-mail address: xshang@wmu.edu.cn (X. Shang).

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fate and transport of small MPs in aquatic systems critical to understand. The size distribution of plastic debris on the ocean surface shows peak abundance around 2 mm and a remarkable absence below 1 mm (Cózar et al., 2014). While few studies have found MPs smaller than 200 μm in the surface ocean, others found small MPs in coastal region sediments (Hidalgo-Ruz et al., 2012) implying a possible sedimentation removal mechanism. Sedimentation of small MPs may be enhanced by biofouling (Fazey and Ryan, 2016; Kooi et al., 2017) or ingestion/fecal pellet deposition by marine organisms (Andrady, 2011; Vianello et al., 2013). For example, the MPs "lost" from the surface ocean match the size of common prey for zooplanktivorous fish (Foekema et al., 2013). Further, small MPs can be ingested by zooplankton and deposited with fecal pellets (Cole et al., 2013, 2016). Other factors, such as wind mixing, might also contribute to the suspension/sedimentation dynamics of small MPs within the ocean (Reisser et al., 2015).

Sedimentation mechanisms can partly explain the missing small MP fraction in the surface ocean and this mechanism may also be active in freshwater systems, which are the source of most marine MPs (Hidalgo-Ruz et al., 2012). MPs should preferentially settle in freshwater systems compared to marine systems due to the lower buoyancy of freshwaters and therefore may contribute to the low abundance of small MPs in oceans. Rivers act as the primary vector for plastics entering the ocean (Lechner et al., 2014; Klein et al., 2015). Unlike large continental rivers which carry massive particulate loads from land to ocean, small low-gradient river systems in coastal plains display much less capacity and competence, which likely facilitates sedimentation of MP particles to riverine sediments. Given the high population of urban areas and their prominent use of plastics, these large coastal urban centers may act as the main source of MPs, especially in developing countries where lagging infrastructure often leads to a lack of sewage and trash collection/treatment (Eerkes-Medrano et al., 2015). However, MP sedimentation in these coastal plain river networks may provide an effective retention mechanism that attenuates small MP transport to oceans. While MPs have been found in freshwaters of Europe, North America, and Asia, the environmental behavior (fate and transport) of MPs in freshwater systems has received little research compared to marine systems (Eerkes-Medrano et al., 2015; Wagner et al., 2014).

In this study, we examine MP abundance and size-class distribution in a highly polluted coastal plain river network associated with a large urban population (~9.2 million). We hypothesize that the lack of stream capacity and competence in coastal plain river systems favors MP sedimentation attenuating the transport of small MPs (<300 μm) to the ocean. Retention of small MPs in river sediments might provide a mechanism to explain the missing small MP size fraction in oceans.

2. Materials and methods

2.1. Study area

The study was conducted in the Wen-Rui Tang River watershed, a low-gradient urban river located on the coastal plain in Wenzhou, Zhejiang Province, southeast China (Fig. 1). This area is one of the most rapidly developing regions in China. Rapid development and industrialization have attributed to severe water pollution caused by discharge of large volumes of untreated domestic and industrial wastewater directly into the river system (Yang et al., 2013; Mei et al., 2014).

Waters of the Wen-Rui Tang River are nearly quiescence for several periods of the year as the outlet connections to the much larger Ou River are closed to maintain sufficient water for river transportation and prevent salt water intrusion. Gates to the Ou River are seldom open except during heavy rainfall events, such as during the monsoon season, resulting in diminished hydraulic power to transport the particulate load. We chose 12 sampling sites within the Wen-Rui Tang River watershed to represent the common land use/land cover areas as the river flows though residential,

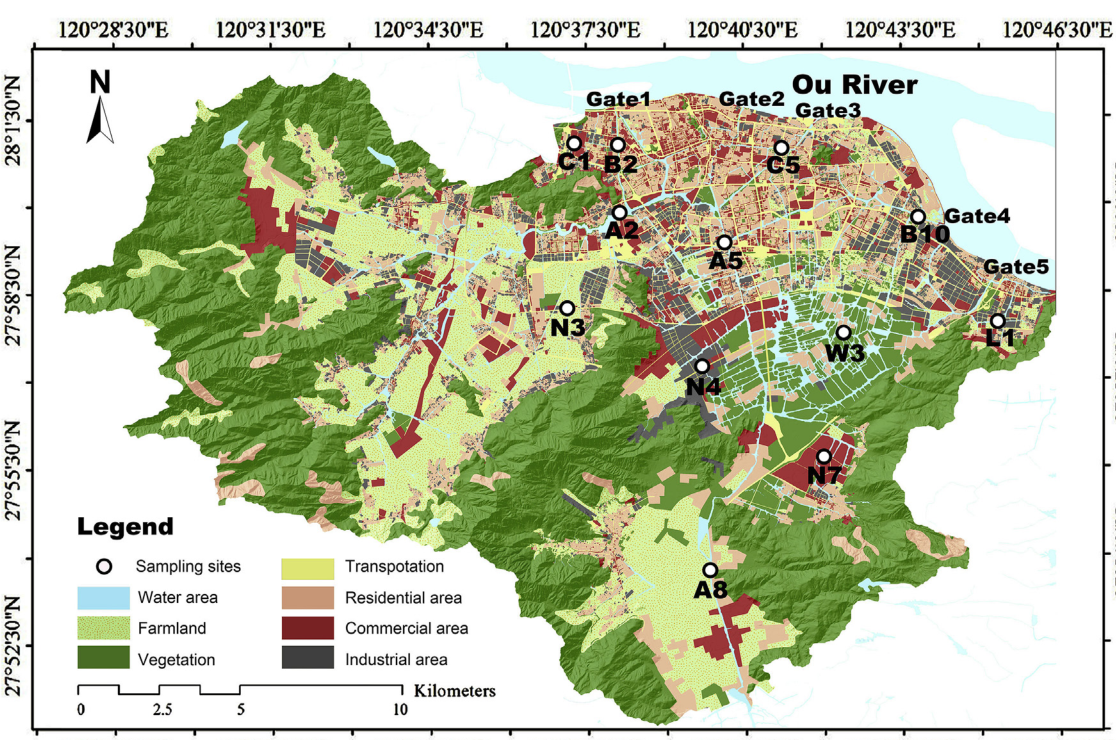

Fig. 1. Location of sampling sites and major land-use units in the Wen-Rui Tang River watershed.

industrial, transportation corridors, farmland, and wetland areas (Fig. 1). Among these sites, 3 sites (A2, A5, A8) are located on the main stream, 4 sites (B2, B10, N3, N4) on major tributaries, and 5 sites (C1, C5, N7, W3, L1) on secondary tributaries. Site A2, A5, B2, B10, C1, C5 and N7 are located in densely populated urban areas, N3 is located near a major highway corridor, N4 and L1 are located in two different industrial districts, W3 is located in a suburban wetland, and A8 is surrounded by farmland. The flow velocity at all sites was <0.05 m s^{-1} during our surveys, but the maximum flow velocity can exceed 2 m s^{-1} during typhoon events, especially at the downstream sites (B10, C5) near the gates to the Ou River. The sediment thickness at the sampling sites ranged from ~0.5 m to more than 1 m.

2.2. Field and laboratory methods

Sampling was carried out in August 2016 before the onset of the typhoon season. At each of the 12 sampling sites, triplicate surface sediment samples were collected using a Peterson grab sampler (32 cm length × 20 cm width × 15 cm depth). A sediment core subsampled from the grab sample (representative of 0–15 cm depth) was placed in pre-washed 500 ml glass bottles for transport to the laboratory. The sampler was rinsed free of sediments between samples to assure no cross contamination.

After thorough mixing, 500 g wet sediment from each sample was dried at 60 °C for 48 h, and the triplicate samples from each site were combined giving a total of 12 composite samples. Extraction of MPs from sediments followed the NOAA protocol, but omitted wet sieving through a 0.3 mm sieve so that the <0.3 mm fraction was included in our study. A 100 g well-mixed, dried sediment sample from each site was sieved through a 5-mm stainless-steel mesh to remove large debris and isolate the <5 mm particle-size fraction for further analysis. The <5-mm fraction was treated by wet peroxide oxidation at 70 °C with 30% hydrogen peroxide to digest natural organic matter present in the sediments (Masura et al., 2015). Following organic matter removal, a salt-water density separation using a $ZnCl_2$ solution (density = 1.7 g ml^{-1}) was utilized to separate MPs through floatation. The density separation step was repeated and the supernatants combined. The supernatant with isolated MPs was filtered through a 0.4 μm polycarbonate membrane filter (HTTP04700, Millipore). To avoid contamination from airborne MPs, the extraction processes were performed in a laminar-flow hood, and all glassware was thoroughly cleaned before use. Laboratory blanks of distilled water were used as "negative control" during extraction and identification processes and no MPs were found on the filters of the duplicate blanks, confirming that background contamination was negligible.

A set (50 items) of randomly selected suspected MPs covering all visual categories of items on each filter were examined to verify polymer composition using a Micro-Fourier Transform Infrared Spectrometer (μ-FT-IR; VERTEX 70 plus HYPERION, 2000; Bruker) to establish a general rule for excluding non-plastic items in the visual examination using a microscope. After that MPs on the filter were stained carefully with 3 drops of 5 μg mL^{-1} Nile Red (NR) for 30 min (Shim et al., 2016), then photographed using a fluorescence stereo microscope (M165FC, Leica) at up to 120 × magnification (Maes et al., 2017). Both fluorescent and non-fluorescent items were enumerated and measured in the images using Image J (https://imagej.nih.gov/ij/). The detailed protocol for identifying, measuring and quantifying MPs based on NR staining is available in Supporting Information.

MP content was converted to a whole sample dry-weight equivalent. MPs were identified based on morphological features of MPs derived from literature data (Norén, 2008; Peng et al., 2017), and classified according to their morphology as fragment (fragment of large plastic waste), fiber (thin or fibrous plastic), pellet (industrial plastic pellet), and foam (lightweight, sponge-like plastic). Because MPs smaller than 20 μm were difficult to identify via μ-FT-IR, MP quantification by NR staining method considered only those particles in the 20–5000 μm range and we report the results in four size classes (μm-based on longest particle dimension): 20–100, 100–300, 300–1000, and 1000–5000. MPs larger than 300 μm are generally considered as "normal" microplastics (NMPs) consistent with MPs reported in most other studies. MPs between 100 and 300 μm are on the threshold of the "missing" MP fraction in oceans; we consider the MP fraction <300 μm the "small" microplastic fraction (SMPs).

3. Results

3.1. Abundance, size composition and distribution of microplastics

Microplastics were found in all sediments collected from the 12 sites in the Wen-Rui Tang River watershed (Table S1). MPs accounted for about 10–50% of the total items collected on the filters after staining with Nile Red (Table S1, Fig. S1), and the result were validated by μ-FT-IR analysis (Fig. S2). Mean concentration of total MPs (20–5000 μm) was 32947 ± 15342 (mean ± std dev) items kg^{-1} sediment, while the average concentration of "normal" microplastics (NMPs 300–5000 μm) was 7-fold less at 4635 ± 2107 items kg^{-1} sediment. Mean concentration of "small" microplastics (SMPs 20–300 μm) that are generally not reported in most studies was 28312 ± 14996 items kg^{-1} sediment. Overall, SMPs accounted for 84.6% of total MPs, while NMPs accounted for the remaining 15.4% (Fig. 2A). Polyethylene (PE), polypropylene (PP), polystyrene (PS), and polyester (PES) were the dominant polymer types identified at most sites based on μ-FT-IR analysis (Table S2). Other common polymer materials included nylon (PA), polyurethane (PU), polyvinyl chloride (PVC), and polyethylene terephthalate (PET).

MP abundance in the river network was highly variable among sites ranging from 18690 items kg^{-1} sediment at A2 to 74800 items kg^{-1} sediment at L1. MP concentrations were slightly lower in the highly populated urban region (28947 items kg^{-1} sediment) compared to the average concentration (32947 ± 15342 items kg^{-1} sediment) for all sites. While MP concentrations in the L1 highly industrialized area was high, the N4 industrial area had average levels. Similarly, MP concentrations in sediment at the A8 suburban agricultural area had lower concentrations compared to the W3

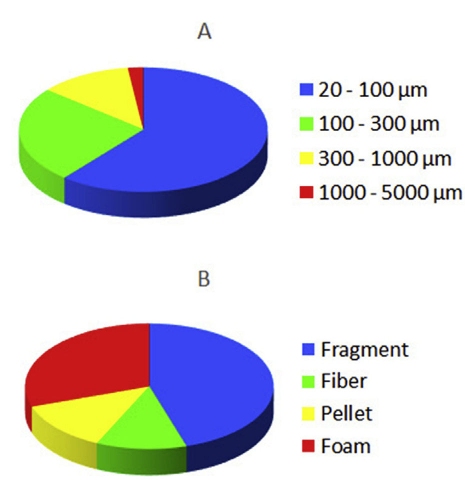

Fig. 2. Average microplastic abundance in river sediments categorized by (A) size and (B) morphological type.

suburban site (Fig. 3). The spatial distribution pattern of MP abundance in river sediments of different class channels followed (items kg^{-1} sediment): main stream (22683 ± 4481) < main tributary (29090 ± 4589) < secondary tributary (42190 ± 20512) (Fig. 4).

The “small” MP (<300 μm) fraction constituted 68.3–94.8% of total MPs. Sites B10 and C5 located at downstream sites near the main outlet gates to the Ou River had the lowest percentages of SMPs largely due to high NMPs at B10 and low SMP concentrations at C5. The low abundance of NMPs at N3 and high abundance of SMPs at L1 resulted in the percentage of SMPs being much higher (>90%) at these sites, although SMPs at N3 and NMPs concentrations at L1 were similar to the average for all sites (Fig. 4).

3.2. Morphological type composition and distribution of microplastics

All four morphological types of MPs were found in sediments at all sites (Fig. S3). Fragments (6290–28070, average 15129 ± 6866 items kg^{-1} sediment) were the most common at all sites and contributed 45.9% of total MPs. Foams (4850–31360, average 9717 ± 7113 items kg^{-1} sediment), pellets (490–11650, average 4232 ± 2706 items kg^{-1} sediment), and fibers (1390–7710, average 3869 ± 1632 items kg^{-1} sediment) were also widespread and contributed 29.5%, 12.8%, and 11.7% of total MPs, respectively (Table S3; Fig. 2B). The percentage contribution of fragments, fibers, pellets, and foams to total MPs varied across sites: fragments 33.7% (A2) to 60.6% (C1), fibers 4.9% (N3) to 23.0% (B10), pellets 2.5% (C5) to 17.7% (N3), and foams 17.4% (C1) to 41.9% (L1) (Fig. 5).

The percentage of fragments was highest in the small microplastic fraction (51.8% in 20–100 μm and 34.8% in 100–300 μm), but less important in the normal microplastic fractions (15.8% in 300–1000 μm and 6.9% in 1000–5000 μm). On the contrary, fibers constituted the majority of NMPs (46.4% in 300–1000 μm and 55.9% in 1000–5000 μm), but were a minor component of the SMP fraction (4.8% in 20–100 μm and 13.1% in 100–300 μm) (Fig. 6A).

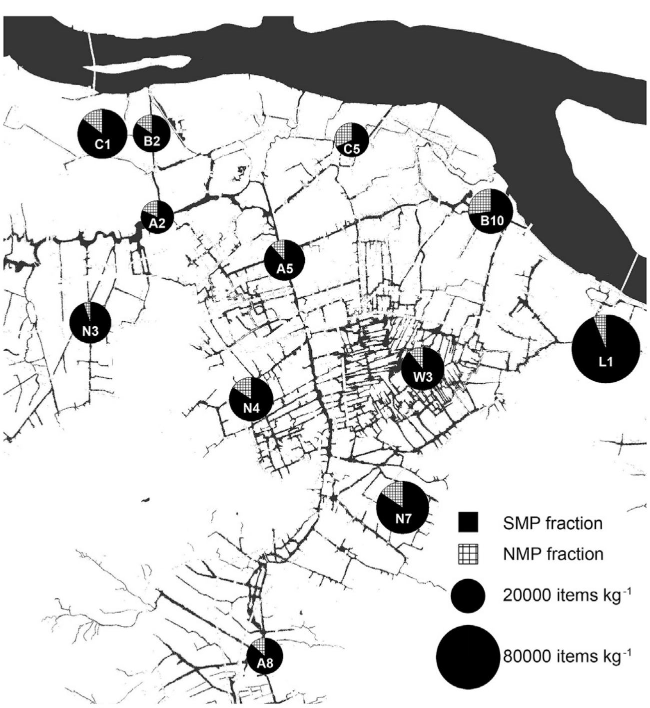

Fig. 3. Microplastic concentrations (items kg^{-1} dry sediment) and percentages of small (SMP, 20–300 μm) and normal (NMP, 300–5000 μm) MP size classes in river sediments.

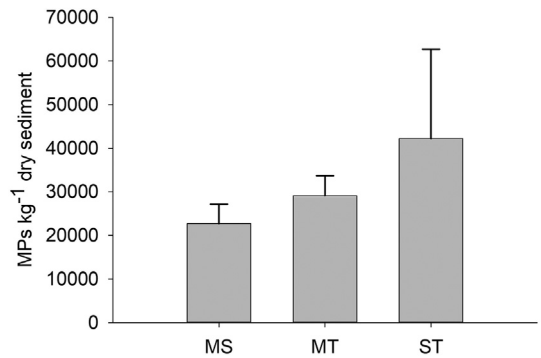

Fig. 4. Microplastic abundance (mean ± std dev) in sediments of different class river channels. MS: Main stream; MT: Main tributary; ST: Secondary tributary.

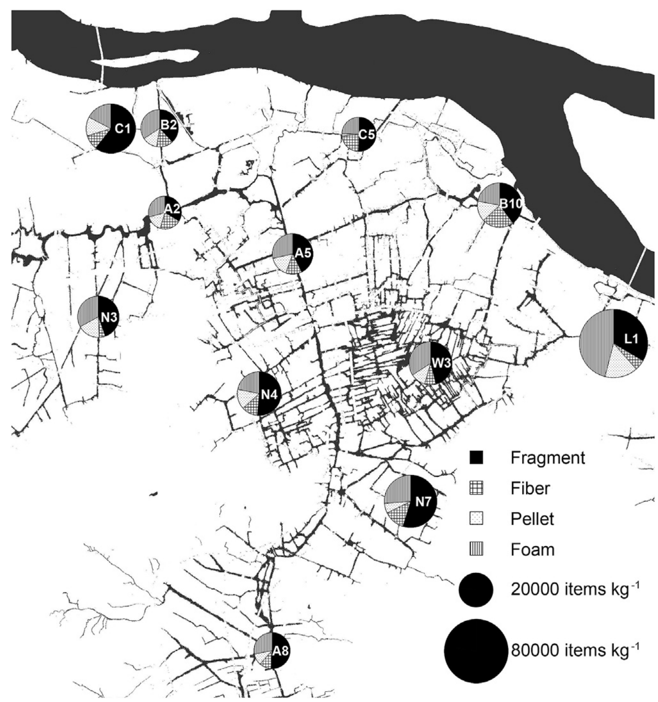

Fig. 5. Microplastic concentrations (items kg^{-1} dry sediment) and the percentage of each morphological type in river sediments.

A total of 68.5% of fragments were <100 μm, while 64.5% of pellets and 63.1% of foams were found in the <100 μm size fraction. Over 90% of MPs in the pellet, foam and fragment morphological classes were SMPs. The size spectrum of fibers was very different from the other morphological classes. About half of fibers were larger than 300 μm, and the fiber contents of the 100–300 μm and 20–100 μm fractions accounted for 28.5% and 22.2% of total fibers, respectively (Fig. 6B).

4. Discussion

4.1. Distribution of microplastics in sediments of the Wen-Rui Tang River network

Microplastics are ubiquitous in sediments from sea floor and coastal beaches in marine environments (Browne et al., 2011; Woodall et al., 2014; Van Cauwenberghe et al., 2015), and rivers and lakes in freshwater environments (Imhof et al., 2013; Eerkes-

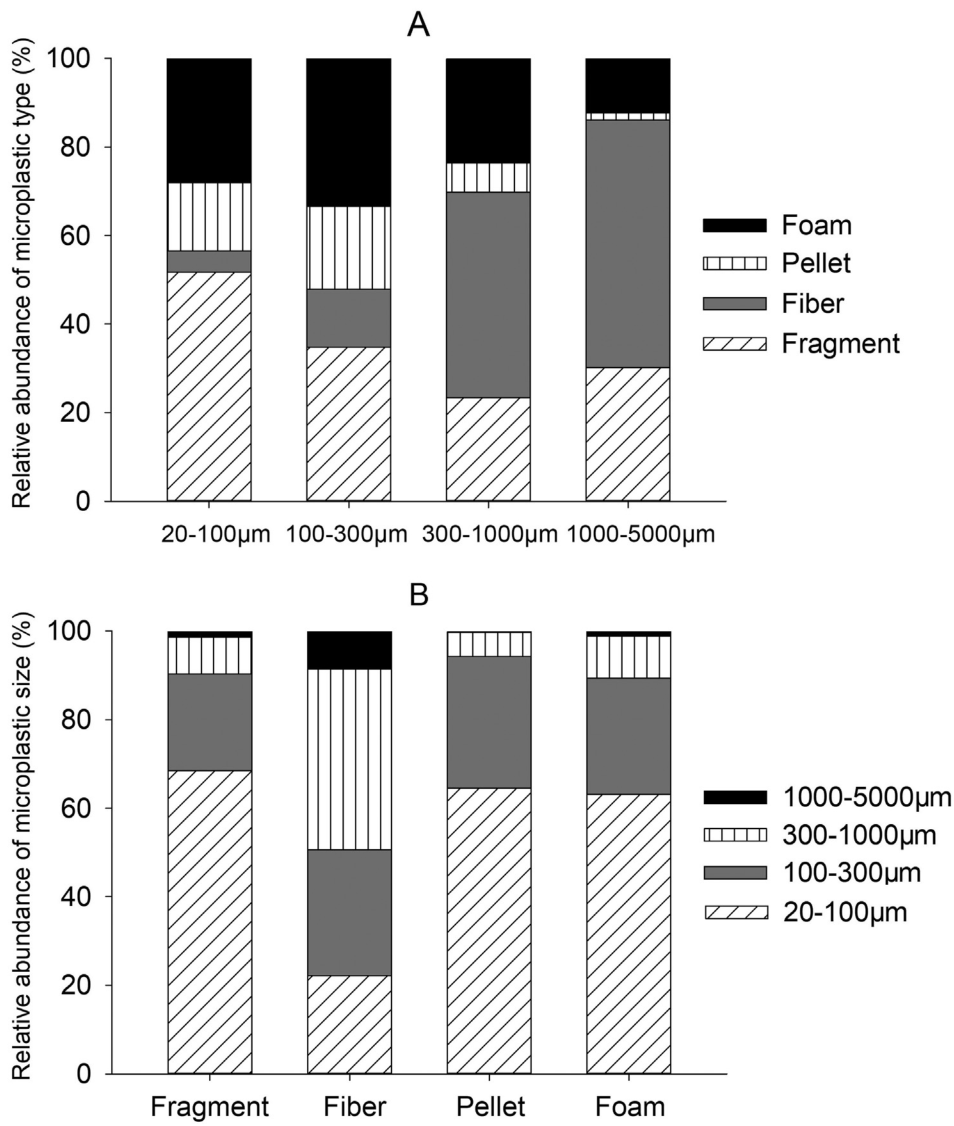

Fig. 6. Composition of microplastics by morphological type (A) and size fraction (B).

Medrano et al., 2015; Klein et al., 2015). However, there are few studies on MP distribution in sediments of semi-closed, coastal plain river systems in densely populated urban areas like the Wen-Rui Tang River. Similar to the accumulation of MPs in low-energy coastal areas (Claessens et al., 2011; Vianello et al., 2013), the unique hydrological dynamics in low energy coastal plain rivers enhance the potential for suspended particle retention in riverine sediments (Noe and Hupp, 2009). In this study, the abundance of MPs in sediments of the Wen-Rui Tang River were up to ~70000 items kg^{-1} sediment, revealing heavy MP pollution in this typical coastal plain river system of eastern China. Although the MPs in this study included the “small” MP fraction (20–300 μm), which is rarely quantified in other studies, the abundance of “normal” size (>300 μm) MPs in Wen-Rui Tang River sediments is still among the highest MP densities reported in freshwater sediments worldwide (Table S4). Among the items recovered from sediment, MPs only account for average 23.6% of total suspected microplastics (Table S1). While the $ZnCl_2$ flotation solution (1.7 g mL^{-1}) used for density separation can isolate heavy plastic materials like PVC, it also suspends more non-plastic items from sediment than lower density flotation solutions. μ-FTIR identification shows these suspected items typically composed of carbon black, pitch, natural fibers, plant detritus, tire wear particles, etc. As μ-FTIR was too slow and difficult for identifying the thousands of items on each filter, the Nile Red staining method couple with fluorescent image processing was developed as an effective and efficient method for identification, measurement and quantification of the small MP fraction.

Several factors are suggested to influence the distribution of MPs in freshwater environments, including population density, distance to urban center or industrial activities, amount of sewage/wastewater overflow, etc. (Eriksen et al., 2013; Free et al., 2014; Eerkes-Medrano et al., 2015). Our study showed MP abundance in sediments from urban sites (i.e., densely populated areas) was not higher than the average values for the entire watershed. Although site in highly industrialized region (L1) had high MP concentrations, the other highly industrialized area (N4) had much lower MP concentrations. The abundance of MPs in river sediments associated with farmlands was low, but sediments from the suburban wetland had average MP values. In general, MP abundance in sediments of different river-order classes showed a tendency of secondary tributaries higher than main tributaries higher than main stream. The complex MP distribution in sediments of the Wen-Rui Tang River network implies that not only the sources, but also other physical, chemical and biological environmental factors are important determinants of MP distribution (fate and transport) in this river network. Water residence time, wind turbulence, and biofouling/defouling are considered important factors affecting the distribution of MPs (Andrady, 2011; Imhof et al., 2013; Fazey and Ryan, 2016). After release into the water column, the buoyancy of MPs allows them to disperse and be driven by wind, rainfall or gate operations. Properties of the particles themselves (e.g. density, size, and shape), flow velocity, biofouling, and ingestion by aquatic animals likely all play important roles in the fate and transport of MPs at the watershed scale (Eerkes-Medrano et al., 2015).

4.2. Size distribution of microplastics in sediments of the Wen-Rui Tang River

There are no standard methods for the isolation and quantification of MPs from sediments (Rocha-Santos and Duarte, 2015; Horton et al., 2017), whereas the common procedures tend to combine size and density separation (Hidalgo-Ruz et al., 2012; Van Cauwenberghe et al., 2015). Given the wide range of MP definitions, from 1 μm to 5 mm, typical sieve apertures used in aquatic ecological studies (53–333 μm) result in the loss of information concerning the entire size spectrum. As a result, the reported size spectrum of MPs is difficult to compare among studies (Hidalgo-Ruz et al., 2012), especially concerning the small-sized MP fraction (<300 μm). The NOAA protocol uses a 300 μm sieve to separate MPs from sediment (Masura et al., 2015), however, sieves of similar or larger size are also commonly used (Rocha-Santos and Duarte, 2015). Therefore, most studies report MPs based on the 300–5000 μm operationally-defined size range while ignoring the "small" MPs in the <300 μm size range. Some recent studies found that the "small" microplastic fraction occurs in much higher abundance than the "large" microplastics. For example, a 63 μm sieve was used in a study of MPs in the sediments of the Rhine-Main Area demonstrating that most MPs occurred in the smallest size fraction (63–200 μm) (Klein et al., 2015). Similarly, a study of sediments from two lakes in central Italy showed a dominance of small MPs (<300 μm) based on a minimum filter pore diameter of 5–13 μm (Fischer et al., 2016). Likewise, most MPs recovered from sediments in an Italian subalpine lake were smaller than 300 μm based on the use of a quartz fiber filter having a retention efficiency of 98% at 2.2 μm (Imhof et al., 2016). In this study, about 85 percent of MPs in the Wen-Rui Tang River sediments were smaller than 300 μm providing additional support for the dominance of SMPs in sediments of freshwater systems. Although the numerical abundance of SMPs is much higher than larger MPs, the contribution of SMPs to the mass or volume of total MPs is limited (Klein et al., 2015). However, the reactivity and ecological risks associated with the SMP fraction maybe disproportionate to their absolute mass/volume as they represent a highly reactive fraction, as smaller plastic particles (nano-size) are reported to have much higher potential to sorb hazardous chemicals resulting in a higher toxicity to aquatic organisms and humans (Velzeboer et al., 2014; Bouwmeester et al., 2015).

Hypotheses for the removal of plastic debris from surface waters include ingestion by marine organisms (Boerger et al., 2010; Foekema et al., 2013), sinking following biofouling (Zettler et al., 2013; Long et al., 2015; Fazey and Ryan, 2016), vertical transport by wind mixing (Reisser et al., 2015), rapid degradation to nano-size particles (Cózar et al., 2014), and most likely a combination of these mechanisms. The effect of biofouling on sedimentation of SMPs has received considerable attention in recent years (Fazey and Ryan, 2016; Kooi et al., 2017). Biofouling leads to the accumulation of organisms and detrital materials on the plastic surface (e.g., biofilms), which in turn affects its hydrophobicity and buoyancy (Ye and Andrady, 1991; Long et al., 2015). The size and shape of floating plastic debris are modified by biofouling as a function of distance from its source with small fragments sink faster than large ones as a result of their higher surface area to volume ratios (Ryan, 2015; Fazey and Ryan, 2016). The eutrophic and low energy water environment in the Wen-Rui Tang River network may enhance sedimentation of SMPs as these two factors both facilitate the development of fouling communities on the surface of MPs. Therefore, the SMPs with a high degree of biofouling would be expected to settle from the water column more quickly than larger materials (Fazey and Ryan, 2016) resulting in their preferential accumulation in sediments. Ingestion by aquatic animals and deposition in fecal pellets may also enhance sedimentation of SMPs, especially given the tremendous zooplankton biomass associated with eutrophic waters (Cole et al., 2016). Compared to large MPs, SMPs are closer to the size of diatoms (mainly 20–200 μm), the main prey of zooplankton facilitating preferential capture and ingestion of the SMP size fraction.

4.3. Morphological characteristics of microplastics in sediments of the Wen-Rui Tang River

Microplastics comprise either manufactured plastics of microscopic size typically used in personal care products or industrial pellets that serve as precursors for manufactured plastic products (primary sources) (Cole et al., 2011; Auta et al., 2017), or fragments or fibers of plastics derived from breakdown of larger plastic products (secondary sources) (Cole et al., 2011; Hidalgo-Ruz et al., 2012; Auta et al., 2017). Plastic fragments were the most abundant among the four types of MPs classified in this study and were especially prominent in the <300-μm fraction. Micro-size fragments are often the product of fragmentation of all kinds of plastic wastes. The percentage of fragments among the various size classes was more evenly distributed than for other MP types implying that the formation/degradation model for fragments is different from foams and pellets (Fig. 7A) (Lambert and Wagner, 2016).

Foams were also abundant in the sediments of Wen-Rui Tang River. The foam to total MPs ratio was positively related to the SMPs to total MPs ratio (Fig. 7D). Foams are one of the most widely used plastics and are typically easy to decompose/degrade under exposure to environmental factors such as UV radiation and mechanical abrasion (Song et al., 2017). Although the density of foams (0.05–0.4 g cm^{-3}) is much lower than water, the buoyancy of micro-size foam can significantly decrease after biofouling and adsorption of sediments, especially in eutrophic and low turbulent waters as found in the Wen-Rui Tang River network. The abundant small foams in the river maybe attributed to runoff of fragmented foam waste degraded on land, or degradation of large foam debris as it is transported downstream in the water column. Because SMPs are more susceptible to sedimentation, the abundance of small foams found in the sediment most likely originates from the biofouling/sediment adsorption mechanisms. The lack of large foam debris in the river sediments implies high buoyancy and a small increase in weight by biofouling (due to low external surface area) that makes deposition more difficult. Thus, the larger foam debris tends to settle on river banks or be transported to the downstream estuary.

The density of plastics comprising micro-size pellets (polyethylene, polyester, and acrylic, typically >0.9 g cm^{-3}) are typically much higher than foams resulting in a higher sedimentation potential. Moreover, small plastic pellets are used as abrasives in industry or as exfoliating agents in personal care products, so they are often more resistant to environmental forces and difficult to

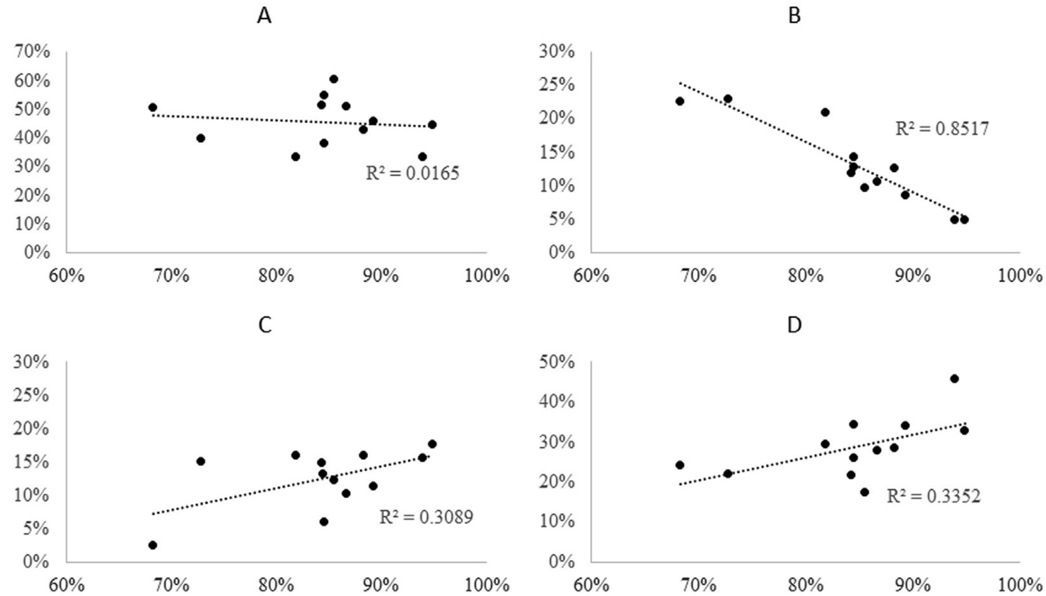

Fig. 7. Relationship between the ratio of microplastic morphological types and small microplastics to total microplastics in river sediments. A: Fragment; B: Fiber; C: Pellet; D: Foam.

degrade. Although the proportion of pellets to total MPs was positively related to the SMPs to total MPs ratio (Fig. 7C), similar to foams, the reason for the size distribution of pellets may differ from foams and depend primarily on their source(s).

In contrast to the three MP types dominated by SMPs (<300 μm), the fibers isolated from river sediments were dominated by fibers >300 μm (Fig. 7B). These findings are consistent with those from Lake Chiusi sediments that showed a much higher abundance of fibers >300 μm compared to those <300 μm (Fischer et al., 2016). Similar to marine phytoplankton, cylindrical phytoplankton with a higher length/diameter ratio are advantageous as the long filament species resist sinking facilitating their dominance in the upper ocean (Padisák et al., 2003). Microplastic fibers may benefit from the same mechanism allowing them to float in the water column longer thereby facilitating their downstream transport. The higher percentage of fibers relative to total MPs at downstream sites (C5 and B10) supports more efficient downstream transport of fibers relative to the foam, pellet and fragment forms that decrease in abundance with downstream transport. Thus, in addition to their widespread domestic source (Salvador et al., 2017), the efficient transport behavior of plastic fibers in riverine systems may explain the high recovery of fiber MPs in sediments from estuarine and coastal areas (Qiu et al., 2015; Ling et al., 2017; Peng et al., 2017).

4.4. *Implications of contrasting MP distribution patterns between freshwater and marine systems*

The negative impacts of MPs on marine ecosystems have received extensive attention (Barnes et al., 2009; Cole et al., 2011; Ivar do Sul and Costa, 2014). In recent years, research on the behavior of MPs in freshwater environments has slowly informed the source of MPs from terrestrial systems (Wagner et al., 2014; Eerkes-Medrano et al., 2015; Horton et al., 2017). However, little is known about the details of MP transport from land to oceans, despite river systems being implicated as important transport pathways (Jambeck et al., 2015; Besseling et al., 2017). Although the ocean floor is considered to be the ultimate fate of marine MPs (Woodall et al., 2014), terrestrial environments such as river and lake sediments might also be a terminal or transient sink for MPs (Imhof et al., 2013; Free et al., 2014). The environmental behavior of different types and sizes of MPs changes during their transport from land to ocean leading to longitudinal variations in the spatial distribution of MPs. The phenomenon of the "missing small microplastic fraction" in the surface ocean has received extensive attention, and some explanation include size-specific sedimentation due to the ballasting effect of biofouling (Fazey and Ryan, 2016; Kooi et al., 2017), zooplankton ingestion (Cole et al., 2013, 2016), wind mixing (Reisser et al., 2015), and rapid decomposition (Cózar et al., 2014) may contribute to the downward transport of SMPs from the surface ocean. Although a recent study in the South China Sea showed a dominant of SMPs in the surface ocean (Cai et al., 2018), the fate of SMPs across the freshwater-seawater interface is still unclear. The abundance of SMP in various size fractions was directly related to their size (i.e., 500 μm > 100 μm>20 μm) in sediments from coastal regions, such as in Changjiang Estuary (Peng et al., 2017) and southeastern Australia (Ling et al., 2017). The lack of SMPs in ocean sediments, coupled with the suspected missing SMPs in ocean surface creates an enigma concerning the absence of SMPs in marine environments.

Our study demonstrated the ubiquitous nature of SMPs in sediments of a coastal plain river network. The MPs recovered from the sediments included both primary and secondary MPs. The primary MPs consisted largely of plastic particles manufactured for industrial and domestic applications and were microscopic in size. The secondary MPs were comprised of fragmentation products from MP debris. Long hydraulic residence time and rapid biofouling caused by severe eutrophication appear to facilitate the retention of MPs in this coastal river network, especially for the SMP fraction (<300 μm). A modeling study demonstrated the potential for river hydrodynamics to affect MP size distributions with profound implications for MP transport to marine systems and retention of some MP size fractions in river systems (Besseling et al., 2017). Although turbulence generated by heavy rainfall during periodic storm events may resuspend some MPs from the sediments, the widespread occurrence of deep sediment layers (50 to >100 cm depth) indicates the potential for persistent storage capacity of MPs in the Wen-Rui Tang River network. However, removal of deposited MPs during channel dredging activities may result in transport to the ocean, as ocean disposal is a common fate for the dredge materials. Additionally, SMPs may be generated from larger MPs due to in situ degradation of retained MPs in riverine sediments. Preferential degradation of larger MPs to SMPs would further enhance the dominance of SMPs relative to total MPs. Thus, the dominance of SMPs in sediments of Wen-Rui Tang River network may not only reflect external inputs, but also result from in situ degradation of MPs within the watershed. The accumulation of large SMP concentrations in coastal plain river systems and other freshwater environments may explain, in part, the "missing" SMP fraction in the surface ocean and marine sediments. Additional research is warranted to determine the fate and transport of MPs from terrestrial to marine systems and to test the hypothesis that SMP sedimentation in freshwater sediments limits their transport to oceans (Besseling et al., 2017).

5. Conclusions

The abundance of MPs found in sediments from a typical coastal plain river network demonstrates an important sink for MPs in riverine sediments. The high proportion of SMPs in freshwater sediments provides a possible mechanism to explain the missing SMP fraction in the surface ocean and marine sediments. The watershed-scale spatial distribution of different MP sizes and types results from the interplay among pollutant sources, plastic chemical composition, shapes, densities, environmental degradation, biological fouling, migration behaviors, etc. China is the largest source of MPs to the ocean (Jambeck et al., 2015). The role of terrestrial water bodies in highly populated and developed areas of southeast China in retaining MPs as a terminal or transient sink for these persistent pollutants, like Lake Taihu (Su et al., 2016) and the Wen-Rui Tang River network, may have a profound effect on SMP transport to oceans. Thus, a more detailed mass balance of riverine MP loads and their fate during transport to the ocean is strongly warranted. The ubiquitous existence of SMPs in freshwater systems and their relative absence in marine systems suggest that future studies in both freshwater and marine systems should consider the whole size spectrum of MPs, especially given the high chemical reactivity and ecological risks associated with the SMP fraction. Moreover, standard methods for sampling and separating MPs from sediments should be revised to include quantification of the small microplastic fraction (<300 μm), with the Nile Red staining method showing excellent potential to identify and track this easily lost MP fraction.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.watres.2018.07.050.

References

Andrady, A.L., 2011. Microplastics in the marine environment. Mar. Pollut. Bull. 62 (8), 1596–1605. https://doi.org/10.1016/j.marpolbul.2011.05.030.

Auta, H.S., Emenike, C.U., Fauziah, S.H., 2017. Distribution and importance of microplastics in the marine environment: a review of the sources, fate, effects, and potential solutions. Environ. Int. 2017 (102), 165–176. https://doi.org/10.1016/j.envint.2017.02.013.

Barnes, D.K., Galgani, F., Thompson, R.C., Barlaz, M., 2009. Accumulation and fragmentation of plastic debris in global environments. Philos. Trans. R. Soc., B 364 (1526), 1985–1998. https://doi.org/10.1098/rstb.2008.0205.

Besseling, E., Quik, J.T.K., Sun, M., Koelmans, A.A., 2017. Fate of nano- and microplastic in freshwater systems: a modeling study. Environ. Pollut. 220, 540–548. https://doi.org/10.1016/j.envpol.2016.10.001.

Boerger, C.M., Lattin, G.L., Moore, S.L., Moore, C.J., 2010. Plastic ingestion by planktivorous fishes in the north pacific central gyre. Mar. Pollut. Bull. 60, 2275–2278. https://doi.org/10.1016/j.marpolbul.2010.08.007.

Bouwmeester, H., Hollman, P.C.H., Peters, R.J.B., 2015. Potential health impact of environmentally released micro- and nanoplastics in the human food production chain: experiences from nanotoxicology. Environ. Sci. Technol. 49 (15), 8932–8947. https://doi.org/10.1021/acs.est.5b01090.

Browne, M.A., Crump, P., Niven, S.J., Teuten, E., Tonkin, A., Galloway, T., Thompson, R., 2011. Accumulation of microplastic on shorelines worldwide: sources and sinks. Environ. Sci. Technol. 45 (21), 9175–9179. https://doi.org/10.1021/es201811s.

Cai, M.G., He, H.X., Liu, M.Y., Li, S.W., Tang, G.W., Wang, W.M., Huang, P., Wei, G., Lin, Y., Chen, B., Hu, J.H., Cen, Z.N., 2018. Lost but can't neglected: Huge quantities of small microplastics hide in the South China Sea. Sci. Total Environ. 633, 1206–1216. https://doi.org/10.1016/j.scitotenv.2018.03.197.

Claessens, M., de Meester, S., Van Landuyt, L., De Clerck, K.D., Janssen, C.R., 2011. Occurrence and distribution of microplastics in marine sediments along the Belgian coast. Mar. Pollut. Bull. 62 (10), 2199–2204. https://doi.org/10.1016/j.marpolbul.2011.06.030.

Cole, M., Lindeque, P., Fileman, E., Halsband, C., Goodhead, R., Moger, J., Galloway, T.S., 2013. Microplastic ingestion by zooplankton. Environ. Sci. Technol. 47, 6646–6655. https://doi.org/10.1021/es400663f.

Cole, M., Lindeque, P., Halsband, C., Galloway, T.S., 2011. Microplastics as contaminants in the marine environment: a review. Mar. Pollut. Bull. 62 (12), 2588–2597. https://doi.org/10.1016/j.marpolbul.2011.09.025.

Cole, M., Lindeque, P.K., Fileman, E., Clark, J., Lewis, C., Halsband, C., Galloway, T.S., 2016. Microplastics alter the properties and sinking rates of zooplankton faecal pellets. Environ. Sci. Technol. 50, 3239–3246. https://doi.org/10.1021/acs.est.5b05905.

Cózar, A., Echevarría, F., González-Gordillo, J.I., Irigoien, X., Úbeda, B., Hernández-León, S., Palma, A.T., Navarro, S., Garcia-de-Lomas, J., Ruiz, A., Fernandez-de-Pelles, M.L., Duarte, C.M., 2014. Plastic debris in the open ocean. Proc. Natl. Acad. Sci. Unit. States Am. 111 (28), 10239–10244. https://doi.org/10.1073/pnas.1314705111.

Eerkes-Medrano, D., Thompson, R.C., Aldridge, D.C., 2015. Microplastics in freshwater systems: a review of the emerging threats, identification of knowledge gaps and prioritisation of research needs. Water Res. 75 (3), 63–82. https://doi.org/10.1016/j.watres.2015.02.012.

Eriksen, M., Lebreton, L.C.M., Carson, H.S., Thiel, M., Moore, C.J., Borerro, J.C., Galgani, F., Ryan, P.J., Reisser, J., 2014. Plastic pollution in the World's oceans: more than 5 trillion plastic pieces weighing over 250,000 tons afloat at sea. PLoS One 9 (12), e111913. https://doi.org/10.1371/journal.pone.0111913.

Eriksen, M., Mason, S., Wilson, S., Box, C., Zellers, A., Edwards, W., Farley, H., Amato, S., 2013. Microplastic pollution in the surface waters of the laurentian great lakes. Mar. Pollut. Bull. 77, 177–182. https://doi.org/10.1016/j.marpolbul.2013.10.007.

Fazey, F.M., Ryan, P.G., 2016. Biofouling on buoyant marine plastics: an experimental study into the effect of size on surface longevity. Environ. Pollut. 210, 354–360. https://doi.org/10.1016/j.envpol.2016.01.026.

Fischer, E.K., Paglialonga, L., Czech, E., Tamminga, M., 2016. Microplastic pollution in lakes and lake shoreline sediments - a case study on Lake Bolsena and Lake Chiusi (central Italy). Environ. Pollut. 213, 648–657. https://doi.org/10.1016/j.envpol.2016.03.012.

Foekema, E.M., De, G.C., Mergia, M.T., van Franeker, J.A., Murk, A.J., Koelmans, A.A., 2013. Plastic in north sea fish. Environ. Sci. Technol. 47 (15), 8818–8824. https://doi.org/10.1021/es400931b.

Free, C.M., Jensen, O.P., Mason, S.A., Eriksen, M., Williamson, N.J., Boldgiv, B., 2014. High-levels of microplastic pollution in a large, remote, mountain lake. Mar. Pollut. Bull. 85 (1), 156–163. https://doi.org/10.1016/j.marpolbul.2014.06.001.

Hidalgo-Ruz, V., Gutow, L., Thompson, R.C., Thiel, M., 2012. Microplastics in the marine environment: a review of the methods used for identification and quantification. Environ. Sci. Technol. 46 (6), 3060–3075. https://doi.org/10.1021/es2031505.

Horton, A.A., Walton, A., Spurgeon, D.J., Lahive, E., Svendsen, C., 2017. Microplastics in freshwater and terrestrial environments: evaluating the current understanding to identify the knowledge gaps and future research priorities. Sci. Total Environ. 586, 127–141. https://doi.org/10.1016/j.scitotenv.2017.01.190.

Imhof, H.K., Ivleva, N.P., Schmid, J., Niessner, R., Laforsch, C., 2013. Contamination of beach sediments of a subalpine lake with microplastic particles. Curr. Biol. 23 (19), 867–868. https://doi.org/10.1016/j.cub.2013.09.001.

Imhof, H.K., Laforsch, C., Wiesheu, A.C., Schmid, J., Anger, P.M., Niessner, R., Ivleva, N.P., 2016. Pigments and plastic in limnetic ecosystems: a qualitative and quantitative study on microparticles of different size classes. Water Res. 98, 64–74. https://doi.org/10.1016/j.watres.2016.03.015.

Ivar do Sul, J.A., Costa, M.F., 2014. The present and future of microplastic pollution in the marine environment. Environ. Pollut. 185, 352–364. https://doi.org/10.1016/

j.envpol.2013.10.036.
Jambeck, J.R., Geyer, R., Wilcox, C., Siegler, T.R., Perryman, M., Andrady, A., Narayan, R., Law, K.L., 2015. Plastic waste inputs from land into the ocean. Science 347 (6223), 768–771. https://doi.org/10.1126/science.1260352.
Klein, S., Worch, E., Knepper, T.P., 2015. Occurrence and spatial distribution of microplastics in river shore sediments of the Rhine-main area in Germany. Environ. Sci. Technol. 49 (10), 6070–6076. https://doi.org/10.1021/acs.est.5b00492.
Kooi, M., Van Nes, E.H., Scheffer, M., Koelmans, A.A., 2017. Ups and downs in the ocean: effects of biofouling on the vertical transport of microplastics. Environ. Sci. Technol. 51 (14), 7963–7971. https://doi.org/10.1021/acs.est.6b04702.
Lambert, S., Wagner, M., 2016. Formation of microscopic particles during the degradation of different polymers. Chemosphere 161, 510–517. https://doi.org/10.1016/j.chemosphere.2016.07.042.
Lechner, A., Keckeis, H., Lamesberger-Loisl, F., Zens, B., Krusch, R., Tritthart, M., Glas, M., Schludermann, E., 2014. The Danube so colourful: a potpourri of plastic litter outnumbers fish larvae in Europe's second largest river. Environ. Pollut. 188, 177–181. https://doi.org/10.1016/j.envpol.2014.02.006.
Ling, S.D., Sinclair, M., Levi, C.J., Reeves, S.E., Edgar, G.J., 2017. Ubiquity of microplastics in coastal seafloor sediments. Mar. Pollut. Bull. 121 (1–2), 104–110. https://doi.org/10.1016/j.marpolbul.2017.05.038.
Long, M., Moriceau, B., Gallinari, M., Lambert, C., Huvet, A., Raffray, J., Soudant, P., 2015. Interactions between microplastics and phytoplankton aggregates: impact on their respective fates. Mar. Chem. 175, 39–46. https://doi.org/10.1016/j.marchem.2015.04.003.
Maes, T., Jessop, R., Wellner, N., Haupt, K., Mayes, A.G., 2017. A rapid-screening approach to detect and quantify microplastics based on fluorescent tagging with Nile Red. Sci. Rep. 7 (44501). https://doi.org/10.1038/srep44501.
Masura, J., Baker, J., Foster, G., Arthur, C., 2015. Laboratory Methods for the Analysis of Microplastics in the Marine Environment: Recommendations for Quantifying Synthetic Particles in Waters and Sediments. NOAA Technical Memorandum NOS-OR&R-48.
Mei, K., Liao, L., Zhu, Y., Lu, P., Wang, Z., Dahlgren, R.A., Zhang, M., 2014. Evaluation of spatial-temporal variations and trends in surface water quality across a rural-suburban-urban interface. Environ. Sci. Pollut. Res. 21 (13), 8036–8051. https://doi.org/10.1007/s11356-014-2716-z.
Noe, G.B., Hupp, C.R., 2009. Retention of riverine sediment and nutrient loads by coastal plain floodplain. Ecosystems 12 (5), 728–746. https://doi.org/10.1007/s10021-009-9253-5.
Norén, F., 2008. Small Plastic Particles in Coastal Swedish Waters. N-research Report, Commissioned by KIMO Sweden.
Padisák, J., Soróczki-Pintér, É., Rezner, Z., 2003. Sinking properties of some phytoplankton shapes and the relation of form resistance to morphological diversity of plankton - an experimental study. Hydrobiologia 500, 243–257. https://doi.org/10.1023/A:1024613001147.
Peng, G., Zhu, B., Yang, D., Lei, S., Shi, H., Li, D., 2017. Microplastics in sediments of the Changjiang estuary, China. Environ. Pollut. 225, 283–290. https://doi.org/10.1016/j.envpol.2016.12.064.
Plastics Europe, 2016. Plastics - the Facts. An Analysis of European Plastics Production, Demand and Waste Data. http://www.plasticseurope.org/plastics-industry/market-and-economics.aspx.
Qiu, Q., Peng, J., Yu, X., Chen, F., Wang, J., Dong, F., 2015. Occurrence of microplastics in the coastal marine environment: first observation on sediment of China. Mar. Pollut. Bull. 98 (1–2), 274–280. https://doi.org/10.1016/j.marpolbul.2015.07.028.
Reisser, J., Slat, B., Noble, K., Plessis, K.D., Epp, M., Proietti, M., de Sonneville, J., Becker, T., Pattiaratchi, C., 2015. The vertical distribution of buoyant plastics at sea: an observational study in the north Atlantic gyre. Biogeosciences 12 (4), 1249–1256. https://doi.org/10.5194/bgd-11-16207-2014.
Rocha-Santos, T., Duarte, A.C., 2015. A critical overview of the analytical approaches to the occurrence, the fate and the behavior of microplastics in the environment. Trac. Trends Anal. Chem. 65 (3), 47–53. https://doi.org/10.1016/j.trac.2014.10.011.
Ryan, P.G., 2015. Does size and buoyancy affect the long-distance transport of floating debris? Environ. Res. Lett. 10 (084019). https://doi.org/:10.1088/1748-9326/10/8/084019.
Salvador, C.F., Turra, A., Baruque-Ramos, J., 2017. Synthetic fibers as microplastics in the marine environment: a review from textile perspective with a focus on domestic washings. Sci. Total Environ. 598, 1116–1129. https://doi.org/10.1016/j.scitotenv.2017.04.172.
Shim, W.J., Song, Y.K., Hong, S.H., Jang, M., 2016. Identification and quantification of microplastics using Nile Red staining. Mar. Pollut. Bull. 113, 469–476. https://doi.org/10.1016/j.marpolbul.2016.10.049.
Song, Y.K., Hong, S.H., Jang, M., Han, G.M., Jung, S.W., Shim, W.J., 2017. Combined effects of UV exposure duration and mechanical abrasion on microplastic fragmentation by polymer type. Environ. Sci. Technol. 51 (8), 4368–4376. https://doi.org/10.1021/acs.est.6b06155.
Su, L., Xue, Y., Li, L., Yang, D., Kolandhasamy, P., Li, D., Shi, H., 2016. Microplastics in Taihu lake, China. Environ. Pollut. 216, 711–719. https://doi.org/10.1016/j.envpol.2016.06.036.
Van Cauwenberghe, L., Devriese, L., Galgani, F., Robbens, J., Janssen, C.R., 2015. Microplastics in sediments: a review of techniques, occurrence and effects. Mar. Environ. Res. 111, 5–17. https://doi.org/10.1016/j.marenvres.2015.06.007.
Velzeboer, I., Kwadijk, C.J.A.F., Koelmans, A.A., 2014. Strong sorption of PCBs to nanoplastics, microplastics, carbon nanotubes, and fullerenes. Environ. Sci. Technol. 48 (9), 4869–4876. https://doi.org/10.1021/es405721v.
Vianello, A., Boldrin, A., Guerriero, P., Moschino, V., Rella, R., Sturaro, A., Da Ros, L., 2013. Microplastic particles in sediments of Lagoon of Venice, Italy: first observations on occurrence, spatial patterns and identification. Estuar. Coast Shelf Sci. 130, 54–61. https://doi.org/10.1016/j.ecss.2013.03.022.
Wagner, M., Scherer, C., Alvarez-Muñoz, D., Brennholt, N., Bourrain, X., Buchinger, S., Fries, E., Grosbois, C., Klasmeier, J., Marti, T., Rodriguez-Mozaz, S., Urbatzka, R., Vethaak, A.D., Winther-Nielsen, M., Reifferscheid, G., 2014. Microplastics in freshwater ecosystems: what we know and what we need to know. Environ. Sci. Eur. 26 (1), 12. https://doi.org/10.1186/s12302-014-0012-7.
Woodall, L.C., Sanchezvidal, A., Canals, M., Paterson, G.L.J., Coppock, R., Sleight, V., Calafat, A., Rogers, A.D., Narayanaswamy, B.E., Thompson, R.C., 2014. The deep sea is a major sink for microplastic debris. R. Soc. Open Sci. 1 (4), 140317. https://doi.org/10.1098/rsos.140317.
Wright, S.L., Kelly, F.J., 2017. Plastic and human health: a micro issue? Environ. Sci. Technol. 51 (12), 6634–6647. https://doi.org/10.1021/acs.est.7b00423.
Wright, S.L., Rowe, D., Thompson, R.C., Galloway, T.S., 2013. Microplastic ingestion decreases energy reserves in marine worms. Curr. Biol. 23 (23), R1031–R1033. https://doi.org/10.1016/j.cub.2013.10.068.
Yang, L., Mei, K., Liu, X., Wu, L., Zhang, M., Xu, J., Wang, F., 2013. Spatial distribution and source apportionment of water pollution in different administrative zones of Wen-Rui-Tang (WRT) river watershed, China. Environ. Sci. Pollut. Res. 20 (8), 5341–5352. https://doi.org/10.1007/s11356-013-1536-x.
Ye, S., Andrady, A.L., 1991. Fouling of floating plastic debris under Biscayne Bay exposure conditions. Mar. Pollut. Bull. 22 (12), 608–613. https://doi.org/10.1016/0025-326X(91)90249-R.
Zettler, E.R., Mincer, T.J., Amaral-Zettler, L.A., 2013. Life in the “Plastisphere”: microbial communities on plastic marine debris. Environ. Sci. Technol. 47 (13), 7137–7146. https://doi.org/10.1021/es401288x.