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Comparison of microplastic pollution in different water bodies from urban creeks to coastal waters☆

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Although freshwater and estuary systems are recognized as origins and transport pathways of plastics to the oceans, there is a lack of comparison of microplastics in different water bodies or river networks. In the present study, the spatial distribution of microplastics was compared across different water bodies, including city creeks (Shanghai), rivers (Suzhou River and Huangpu River), an estuary (Yangtze Estuary) and coastal waters (East China Sea) in the Yangtze Delta area. Significant spatial differences of microplastic abundances were revealed across the sampling areas. The results showed that the abundance of microplastics was higher (1.8–2.4 items/L) in freshwater bodies than that in estuarine and coastal water (0.9 items/L). In the Suzhou River and the Huangpu River, microplastics showed trends of increasing abundance downstream, where the peak of microplastic pollution is closer to the city center and the estuary. In respect of abundance, microplastics are likely to be transported from pollution sources to sink areas via river networks. The proportion of fibers was the highest in city creeks (88%), followed by the Suzhou River (85%), the Huangpu River (81%), the Yangtze Estuary (66%) and the East China Sea (37%). Similarly, polyesters dominated in city creeks and rivers. The results suggest that both the abundance and properties of microplastic pollution varies across different water bodies. Microplastic pollution in small freshwater bodies is more serious than in estuarine and coastal waters. Therefore, we support prioritization of water monitoring for microplastics within entire river networks, instead of single water body surveys.

1. Introduction

Microplastics are considered potential environmental hazards due to their ubiquitous presence. The ecological risk associated with environmentally relevant microplastic pollution is unclear; however, some field observations and laboratory studies have shown that microplastics are likely to threaten the life and development of biota via direct and indirect pathways, including ingestion, adherence and transfer throughout food chains (Desforges et al., 2015; Farrell and Nelson, 2013; Long et al., 2015). Although more efforts are required to assess the potential for negative impacts on organisms, there exists a growing body of evidence that suggests that microplastics are becoming more commonplace in aquatic eco-systems (Law, 2017). The current quantities of microplastics in these systems will inevitably increase due to degradation of larger plastic items, ultimately breaking down into smaller, even nanosized, plastic pieces (Cózar et al., 2014; Corcoran et al., 2015; Eriksen et al., 2014).

Microplastic pollution was initially addressed for marine environments and, by extension, coastal shorelines. World-wide research on marine microplastic pollution from polar regions to shorelines has been a research topic since the 1970s (Carpenter and Smith, 1972; Barboza and Gimenez, 2015; Cole et al., 2011). In recent years, research regarding plastic pollution has focused particularly on the source, transportation, and fate of microplastics in natural habitat (Cole et al., 2011; Law, 2017; Zhang, 2017). Large-scale gyre investigations revealed the pelagic plastic pollution accumulation in open oceans and the existence of the "Great Pacific

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garbage patch" (Andrady, 2017). The levels of microplastic pollution in the open ocean have also been determined by large-scale and long-term voyage investigations using trawls. Microplastics have been detected at depths ranging from deep sea floors to microlayers on sea surfaces (Song et al., 2014; Van Cauwenberghe et al., 2013). The spatial distribution of microplastic pollution, specifically its high abundance in near-shore sea water, demonstrates a close relationship between pollution sources from land and marine microplastic abundance.

While microplastic pollution in marine waters has been widely documented, similar studies in estuarine and fresh water are comparatively scarce (Li et al., 2017). In light of pollutant control, it is important to trace the source and behavior of microplastics from terrestrial ecosystems. Freshwater systems can directly receive microplastics from multiple primary sources, such as manufacturing processes and landfill operations (Browne et al., 2011; Eerkes-Medrano et al., 2015). The occurrence of microplastics has been reported in freshwater from lakes, rivers and wastewater treatment plants (Eriksen et al., 2013; Estahbanati and Fahrenfeld, 2016; Yonkos et al., 2014). It has been shown that pollution sources, anthropogenic impacts and hydrodynamics have the potential to influence the rates at which microplastics accumulate and are transported (Browne et al., 2011; Eerkes-Medrano et al., 2015; Horton et al., 2017). Though the proportion of wastewater-derived plastics in freshwater is largely unknown, effluent from industrial and domestic sources makes an important contribution to microplastic pollution. The efficacy of microplastic removal strategies is varied among different waste water treatment plants in urban areas (Carr et al., 2016; Estahbanati and Fahrenfeld, 2016; Mintenig et al., 2017). However, the current case studies, which have aimed to quantify microplastic pollution in aquatic environments, have seldom included freshwater tributaries within urban and peri-urban river systems (Zhang et al., 2018).

Light plastic material introduced to the marine environment is buoyant, while biological and physicochemical processes can change the density of heavier plastic materials, thus potentially conferring buoyant properties. Before settling down in the sediment, water bodies are major pathways to transport land-sourced microplastics, as well as providing a temporary reservoir in the short term (Rocha-Santos and Duarte, 2015; Siegfried et al., 2017). Freshwater systems, especially rivers, are likely to transport microplastics from land-based sources to estuaries and the open ocean. The marine environment is a primary sink when considering the life-span of microplastics. Ultimately, microplastics introduced to this environment will either accumulate at the shoreline or sink to the sediment from surface seawater (Siegfried et al., 2017; Woodall et al., 2014). While the hydrodynamic mechanisms involved in microplastic transportation within freshwater and estuarine systems are still unclear, the high abundance of microplastics at coastal water mouths and estuaries around the globe is clear (Browne et al., 2010; Fok and Cheung, 2015; Yonkos et al., 2014). The higher abundance of microplastics observed in rivers and other small water bodies is believed to have comparatively more significant impacts on these ecosystems and their inherent biota (Horton et al., 2017; Rillig, 2012).

Water monitoring is a simple and straightforward way to track down the fate of pollutants. However, specific sampling and monitoring of microplastics has been inadequately frequent, so the degree to which urban river systems are polluted by microplastics is largely unknown. Urban water bodies and river systems represent important sinks for pollutants discharged from adjacent pollution sources, including domestic and industrial land uses. The major rivers across coastal cities are primary pathways between source of pollution and the open ocean. We hypothesized that these rivers represent different reservoirs, transport pathways and deposition sections for microplastics. This study was designed to investigate spatial trends within and between sample locations in the Shanghai river systems and to establish the link between microplastics pollution and mass watercourse systems. Investigating the role of inland water bodies in the distribution of microplastics is key to understanding microplastic transport pathways from land sources to the marine environment.

2. Materials and methods

2.1. Investigation area

Shanghai is one of the most populated cities in China, with a large industrial focus. It is located on the Yangtze River Estuary, in which the river plume is an important hydrological process that affects the distribution and transportation of water-borne particles such as microplastics (Zhang, 2017). While previous research on sediments, fish and mussels has revealed the high level of microplastics in this area, information based on river nets is still lacking (Li et al., 2015; Su et al., 2018). The Suzhou River and Huangpu River are two main rivers that connect thousands of branching rivers within Shanghai (Fig. 1). The fluvial processes of this system direct most water to the Yangtze River Estuary and the East China Sea. In this investigation, the People Squire of Shanghai was considered as the city center, where anthropogenic activities are the most intensive within the urban areas. Meanwhile, creeks samples from the southern and northern parts of the Suzhou River were treated as two different categories because the northern part is a traditional industrial zone in Shanghai (Fig. 1).

2.2. Sample collection

Surface water samples from 43 sites were collected from April to September 2017. The research area covers urban, suburban, and peri-urban lands and includes fresh to estuarine sections of the Yangtze River and the East China Sea (Fig. 1) (Table S1). Based on the size of the watershed, these sites were clustered into five types. Sampling sites located at the small water bodies (S1–S14), the Suzhou River (S15–S23) and the Huangpu River (S24–S31) within Shanghai belong to urban creeks and rivers types. These small water bodies are believed to be influenced by intense anthropogenic activity inside the city, and transport water containing microplastic contaminants to estuarine and marine waters. Sampling sites located on the Yangtze River (S32–S38) and the East China Sea (S39–S43) span estuarine and coastal sections, representing primary watercourses that receive material from upstream in Shanghai. Five liters of surface water were sampled by using a metal pail. At the sampling sites from the estuary and coastal waters, 5 L surface water was collected by an air lift pump from a boat. The surface water was collected three times at each site.

2.3. Isolation of microplastics

Surface water was filtered through nylon filters of 20 μm pore size based on our previously established methods (Su et al., 2018). The substances collected on the filters were immediately washed into glass bottles by using KOH solution (10% w/v). Approximately 250–300 mL KOH solution was added to each bottle to dissolve the organic matter of the surface water in each bottle. The glass bottles were covered and placed in an oscillating incubator at 65 °C and 80 rpm for approximately 24–48 h (depending upon the dissolution level). After the dissolution process, samples were filtered again with the same size filter, and the filters were stored in dry Petri dishes for further observation.

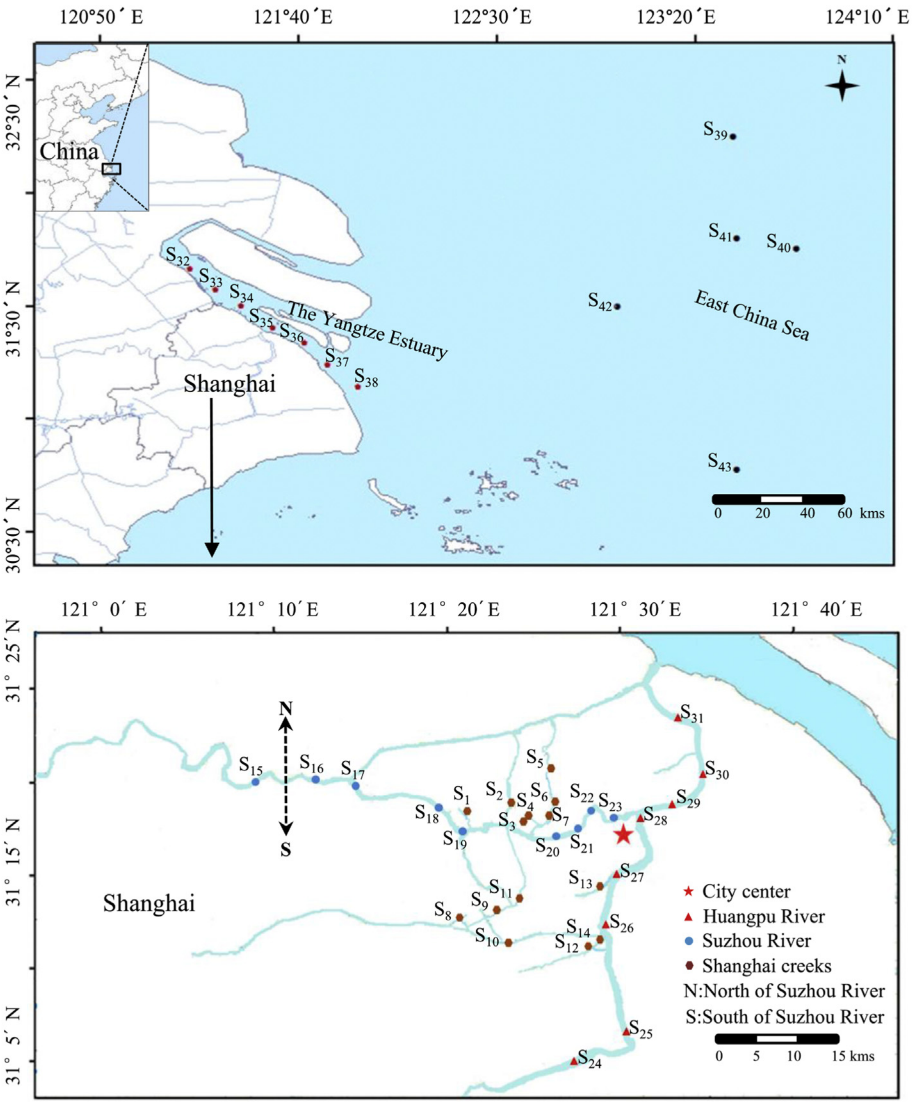

Fig. 1. Locations of sampling sites within the East China Sea, the Yangtze River (A) and the Huangpu River, the Suzhou River and city creeks (B).

2.4. Observation and validation of microplastics

Whole filters were visually inspected under a Carl Zeiss Discovery V8 Stereo microscope (Micro Imaging GmbH, Gottingen, Germany), and images were taken with an AxioCam digital camera. A visual assessment was applied to identify the types of microplastics according to the physical characteristics of the particles. The number, size, shape, and color of microplastics for each sample was recorded. The microplastics were classified into the following four morphotypes: fiber, pellet, film and fragment (Li et al., 2016).

Visually-identified particles were randomly selected for validation. They represented the most common types of the visually identified particles. A total number of 887 items were recovered from samples and 285 (32.1%) items were verified via FTIR. The polymer composition was measured under the attenuated total reflection mode of a μ-FT-IR (Bruker, LUMOS). All spectra were compared with a database from Bruker for verification. The spectra matching with a quality index more than 60% were accepted. The final number of microplastics was recalculated by removing verified nonplastics.

A series of blank controls without water samples were done in current paper. There were 12 items recovered from 69 blanks, representing 0.045 ± 0.1 items/L. Blanks accounted for only 2.6% of the average microplastic abundance in water samples, which can be ignored in results.

2.5. Data analysis

The difference between the quantities of microplastics for more than two groups was determined by one-way analysis of variance (ANOVA) followed by Tukey's HSD test (homogeneous variances) or the Tamhane-Dunnett test (heterogeneous variances), along with multiple comparisons. A significance level of 0.05 was chosen, and the difference between two groups was analyzed using Student's t-test. The geographic information used for regression analysis was acquired via satellite imaging and ArcGis 10.0.

3. Results

3.1. Validation and composition of microplastics

Of the 285 randomly selected items, 206 items were confirmed as plastics (success rates of visual identification ranged from 70% to 74% within the five water body types) (Table S2). Overall, ten polymer types were identified. The dominant polymer was fibrous polyester

(PES) (27.7%) (Fig. 2A), followed by rayon (14.4%) (Fig. 2E) and polypropylene (PP) (8.7%) (Fig. 2B) (Table S2). Nonplastic compounds such as cotton, pigment and paper were also confirmed in our samples. Samples from the East China Sea contained the lowest proportion of PES but the highest proportion of PP. The major river, the Huangpu River, contained the highest amounts of PES, which were roughly double the amount of that detected in the East China Sea and Estuary.

3.2. Abundances of microplastics in different water bodies

Microplastics were found in all water samples and ranged from 0.08 items/L to 7.4 items/L (Fig. 3). The abundance differed significantly among the 43 sampling sites, and the measured concentrations ranged over 10 orders of magnitude ($p < 0.05$) (Fig. 3). In terms of different water bodies, microplastic concentration ranged from 0.9 items/L to 2.4 items/L, and the differences were significant from creek to coastal water samples ($p < 0.05$). The highest abundance of microplastics (7.4 items/L) was found at S22 from the Suzhou River. The lowest abundance (0.08 items/L) was also found within the same river, but further upstream (S16) (Fig. 3).

For those small water bodies, sampling sites located in the north of the Suzhou River showed higher abundance than those located in the south ($p < 0.05$) (Fig. 4B). In terms of spatial distribution, the abundance of microplastics significantly increased when sampling sites were closest to the city center ($p < 0.05$) (Fig. 4C). Again, the abundance of microplastics significantly increased when sampling sites were closer to estuaries (Fig. 4D).

3.3. Types, sizes and colors of microplastics in different water bodies

The characteristics of microplastics were similar across different sampling areas. Fiber was the most dominant component, with a proportion of 37–88% ($p < 0.05$) (Fig. 5). The proportion of fibers was the highest (88%) in Shanghai creeks, followed by those in the Suzhou River (85%), the Huangpu River (81%), the Yangtze Estuary (66%) and the East China Sea (37%).

Blue and red items were prevalent in all samples, accounting for 46–76% of the overall microplastics ($p < 0.05$) (Fig. 6A). However, in the Huangpu River, the proportion of gray items was orders of magnitude higher than the average levels from other sites (Fig. 6A). Within the size range from 20 to 5000 μm, microplastics between 100 and 1000 μm were more frequently observed than other size fractions ($p < 0.05$), accounting for 57–80% of all microplastics detected (Fig. 6B).

4. Discussion

4.1. Comparison of microplastic levels in different water bodies

The current work is the first study to monitor microplastics in surface water from different types, namely, the creeks, rivers and coastal water in urban, peri-urban and estuarine sections of the natural waterways of a large city. Microplastics quantification and distribution data for comparison in city inland waters is very

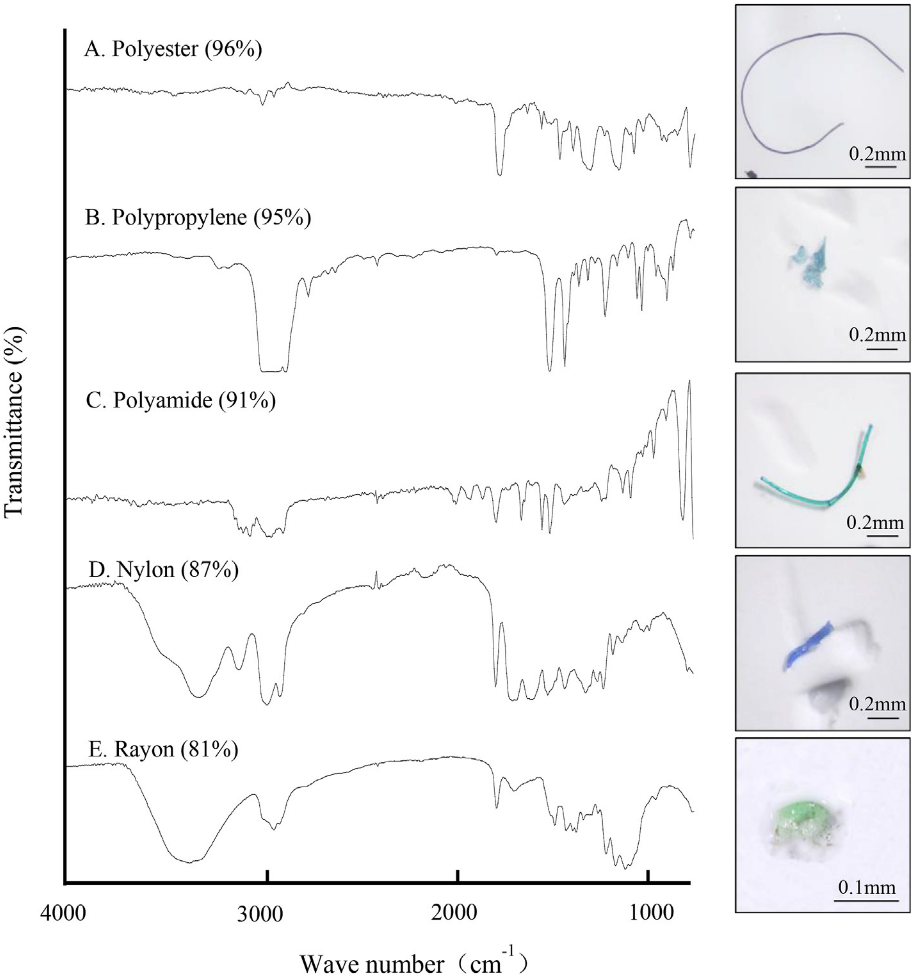

Fig. 2. Selected items for identification and their composition.

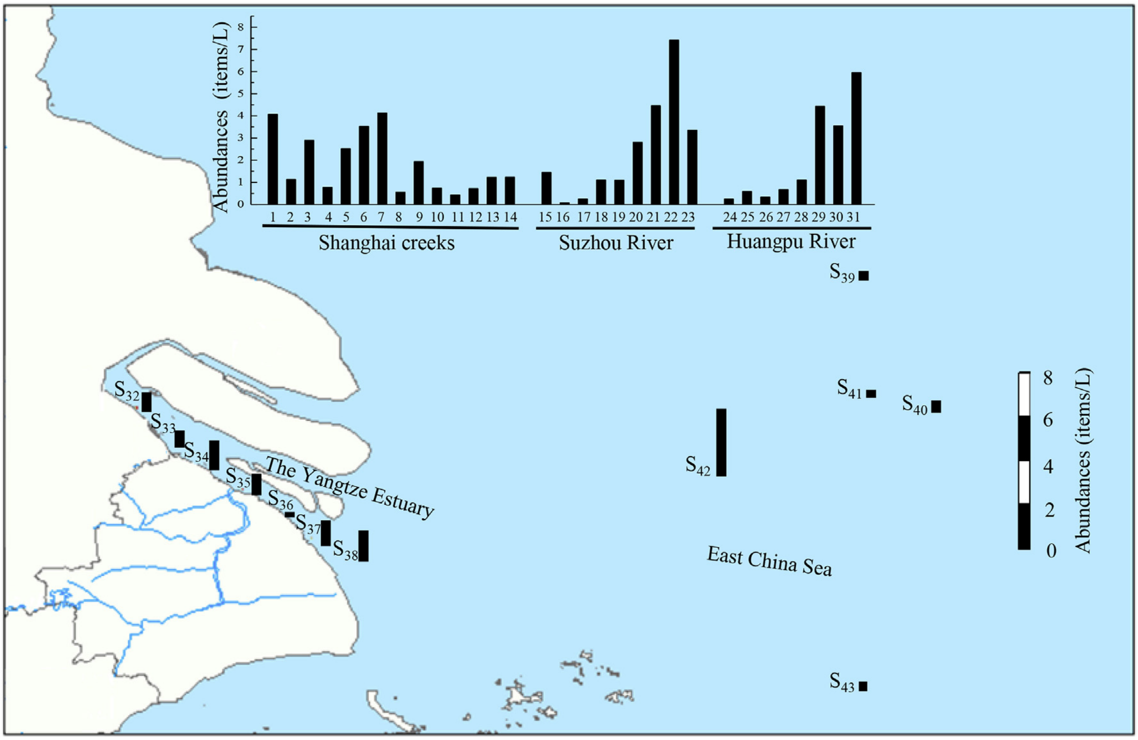

Fig. 3. Comparison of microplastic levels across different water ways.

limited. Here, we have compared our results with previous reports from rivers, estuaries and shoreline water. It should be noted that interstudy comparisons must be made with caution as significant inconsistencies in study design result from differences in reporting protocols.

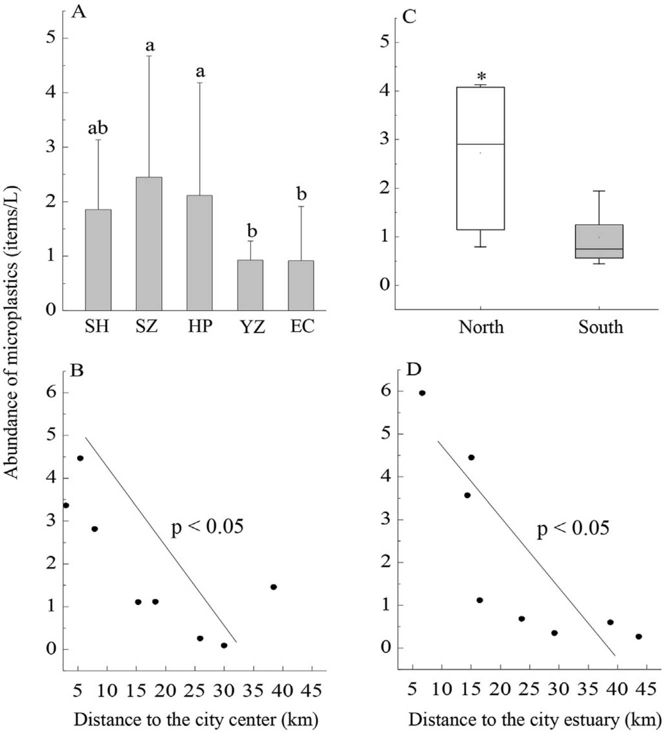

Fig. 4. Associations between watershed characteristics and microplastic concentrations in water bodies: (A) water ways; (B) locations of sampling sites within city creeks; (C) distance to the city center from rivers; (D) distance to estuary from streams. Abbreviations: SH, Shanghai creeks; SZ, Suzhou River; HP, Huangpu River; YZ, Yangtze Estuary; EC, East China Sea.

Overall, the microplastic levels in our report are within the ranges detected from previous research on microplastic pollution in the freshwater and estuarine systems of China (<0.1 items/L-10.9 items/L, Zhang et al., 2018). However, the highest microplastic concentration, documented at S22 from the Suzhou River, was close to the record from urban rivers, the Wuhan River (8.9 items/L) and the Yangtze River Estuary (10.9 items/L), which were considered hotspots for freshwater and estuarine microplastic pollution in China (Zhao et al., 2014). In a global sense, the peak of our results exceeds the microplastics abundance detected downstream of wastewater treatment plants (0.1 items/L); however, abundance of microplastics was less than that recorded in heavily polluted sea water in Germany and Australia (>50 items/L) (Salvador Cesa et al., 2017).

In addition to baseline monitoring, our study provided insight into the distribution of microplastics in water from small water bodies towards the open sea. First, while a strong correlation between the distance downriver and microplastic concentration was not observed, the level of microplastics from creek and river water averaged twice as much as those detected from estuary and coastal waters. This indicates that dilution during transport could account for the decrease in concentration of microplastics downstream (Lattin et al., 2004; Nizzetto et al., 2016). Furthermore, urban rivers were suggested as potential sources of microplastics, and their concentrations were generally higher than what found in the open ocean (Law, 2017; Mason et al., 2016).

Second, the spatial distribution indicated a clear tendency of microplastic abundance to increase in urban and estuary waterways (Fig. 3). Of the sites within the Suzhou River, microplastic abundance was significantly higher near the city and southern areas, where industrial pollution is expected to be the cause. Studies in a variety of urban areas generally support a positive correlation between microplastic quantities and proximity to

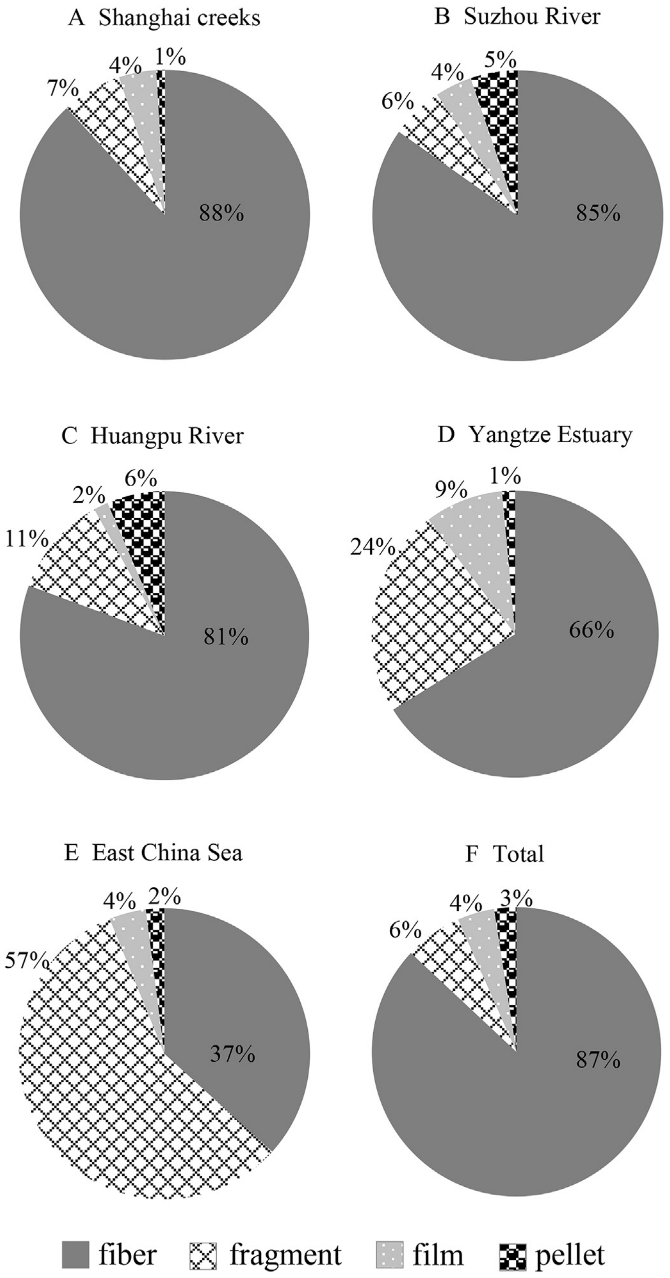

Fig. 5. Comparison of microplastic shapes from different water bodies.

densely populated or industrial areas, which is not surprising given the anthropogenic origin of plastic materials (Lares et al., 2018). However, interestingly, peaks in microplastic concentrations were observed from estuarine sections of the Huangpu River, which are far from the city or any obvious pollution source. We posit that mainstream hydrological processes might significantly contribute to the accumulation of microplastics, in addition to pollution source and input. Large rivers discharge significant amounts of particulate materials and a large fraction of the river-borne particulate organic matter is initially deposited near the mouth (Boldrin et al., 2005; Dagg et al., 2004). The behavior of microplastics is quite similar to suspended solids and particulate materials, so microplastics may also be accumulated by the same process involved in the Yangtze River plume (Zhang, 2017).

4.2. Comparison of microplastics properties in different water bodies

Fibers are among the predominant forms of microplastics found in water bodies ranging from sea beds to remote inland freshwater lakes (Salvador Cesa et al., 2017). Likewise, our study revealed a high proportion of fibers across different sampling sites. What is more interesting is the decreasing tendency of fiber abundance from small urban water bodies to the sea. Generally, the urban effluent, especially domestic pollution, was considered as a primary factor contributing to microfiber abundance. Experiments sampling wastewater from domestic washing machines demonstrated that a single garment can produce >1900 fibers per wash (Browne et al., 2011). Studies on treated wastewater (TWW) in Germany found that synthetic fibers dominated in more than 80% of the samples (Mintenig et al., 2017). The presence of fiber in TWW could account for their presence and abundance in urban waterways via riverine transports.

In our case, more than 80% of microplastics in small urban rivers and main rivers are fibers, while the Yangtze River and coastal water samples contained less than 50% fiber. In regard to polymers, which are commonly used as garment materials, urban rivers and creeks contained more polyester than coastal water. Our results suggest that microfibers are more likely found closer to shorelines where effluents are discharged. Such a spatial pattern was also reported in seawater investigations (Sherman and van Sebille, 2016; Yonkos et al., 2014). Clearly, in addition to discharged effluents, urban water bodies can directly receive airborne fibers and those carried by storm water. In the short term, these processes have the potential to significantly increase the concentrations of fiber in small water bodies. Upon entering larger water bodies, however, they tend to diffuse and are covered by other long-term sources; for example, the fragmentation of floating debris (Browne et al., 2011). On the other hand, we found more microplastics in small freshwater river systems than in estuarine and marine water bodies with larger water volumes. The dominant dense fiber in rivers may reflect a drop in fiber proportion from the estuary to the ocean. Hydrologic processes in river systems are considered more intense than those in the open sea with larger water bodies. In rivers, dense plastic fibers are more likely to be resuspended instead of settling down in sediment (Nizzetto et al., 2016). Microplastic morphology could be considered an important part of pollution finger printing while more specific field investigations are required in future.

4.3. The fate of microplastics in different water bodies from source to sink

The concentration of microplastics differed from water bodies with a decreasing rate from urban river networks to coastal waters, whereas an increasing tendency was observed from major rivers to estuaries. When combined, we can hypothesize that the following are two critical processes involved in microplastic transportation: source discharge and transport to sink. Microplastics were discharged into small water bodies through point and nonpoint pollution sources. This can occur when water runs over or through land, accumulates pollutants, and deposits them into nearby waterways (Ouyang et al., 2018). The composition of small water bodies may impact the health of local biota. Evidence from our previous study confirmed an adverse impact on tadpoles after ingesting microplastics (Hu et al., 2018). Our results clearly reveal a serious concentration of microplastics in small rivers and other water bodies in the urban districts of Shanghai. The microplastic concentrations are significantly higher in these districts than in

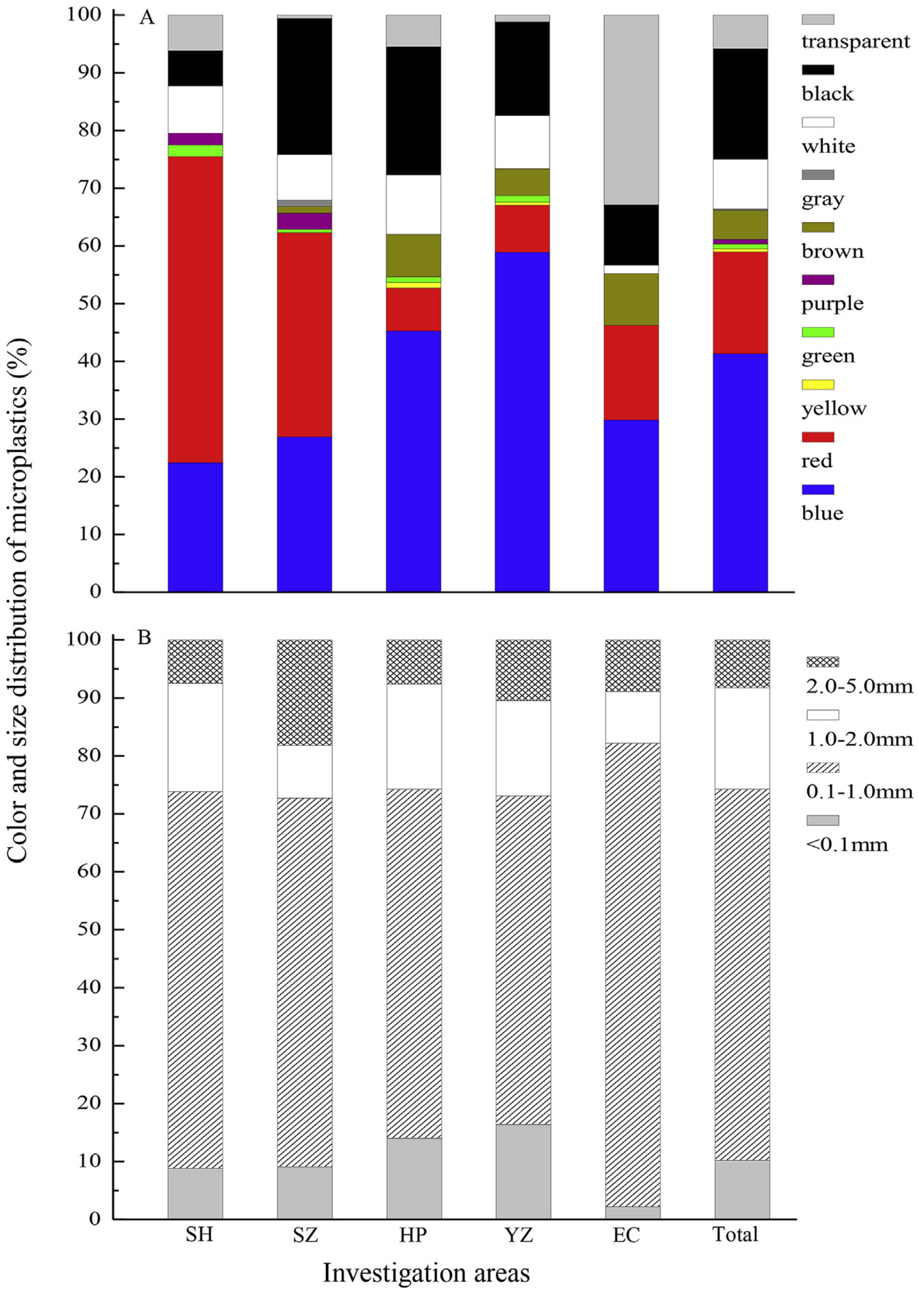

Fig. 6. Color (A) and size (B) distribution of microplastics from different water bodies. Abbreviations: SH, Shanghai creeks; SZ, Suzhou River; HP, Huangpu River; YZ, Yangtze Estuary; EC, East China Sea. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

open oceans with larger water volumes. Nevertheless, these water bodies remain the least investigated of all aquatic environments and are largely excluded from microplastic monitoring (Sutton et al., 2016; Yonkos et al., 2014).

After transportation by the initial depositor of microplastics, rivers with large watersheds will receive them from various branches as introduced by runoff. The high rates of particulates and water discharge cause these microplastics to be ultimately transported to the river mouth area. River plume processes are affected by a suite of complex factors that are not fully understood (Dagg et al., 2004). It is relatively clear, however, that estuarine water bodies are hot spots of microplastic abundance (Browne et al., 2011; Kim et al., 2015; Wang et al., 2016; Yonkos et al., 2014). Transport of microplastic particles within these zones will generally be affected by the same factors that influence sediment transport. The surveys conducted at the Peal River Estuary, the Yangtze River Estuary and the estuarine rivers of the Chesapeake Bay all documented high levels of microplastics in comparison with nearby sea water (Fok and Cheung, 2015; Yonkos et al., 2014; Zhao et al., 2014). In addition to hydrological conditions, meteorological processes such as wind and rain are also believed to contribute to microplastic accumulation in water (Barboza and Gimenez, 2015; Browne et al., 2011; Driedger et al., 2015). Overall, the riverine transport of microplastics in major rivers is more likely ruled by non-anthropogenic factors than by anthropogenic factors. In addition, as we can see, pollution sources and hydrological factors contribute to the spatial distribution and transportation of microplastics from inland rivers to the open ocean. However, the transportation process is irreversible on a large scale, and sometimes it is not easy to predict transportation pathways. In deeper waters, sediments may become a permanent sink for plastics that either descend out of the water column directly or are transported over and down the continental slope.

5. Conclusion

Our studies indicate that the spatial distribution of microplastics in water bodies varies across different types of water bodies. Smaller water bodies are more likely to be affected by pollution sources, while the transportation of microplastics within main rivers is likely due to hydrological processes. Our results suggest that both the abundance and properties of microplastics showed significant variations in different water bodies. Microplastic pollution in small water bodies is more serious than in estuary and coastal waters. Therefore, we support prioritization of water monitoring for microplastics within entire river networks, instead of single water body surveys. Measurements of microplastics present in different types of water bodies is essential to understand their source and sink.

Notes

The authors declare no competing financial interest.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2018.11.081.

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