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Microplastic abundance, distribution and composition in the Pearl River along Guangzhou city and Pearl River estuary, China

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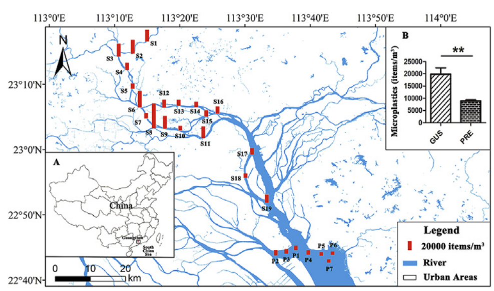
HIGHLIGHTS

- First comparison of microplastics between estuary and urban section of the Pearl River.
- Films were the main shape in the Pearl River.
- PA and cellophane were the dominant polymer types in the Pearl River.
- Waste water effluents from urban city might be a main source of microplastics.

GRAPHICAL ABSTRACT



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ABSTRACT

Like many urban rivers, the Pearl River in China is contaminated with microplastics. Compared with marine environments, microplastic pollution in freshwater is less understood, especially in urban rivers. In the present study, the abundance and distribution of microplastics in water from the Pearl River was investigated, including the estuary and the urban section along Guangzhou. The average abundance of microplastics was 19,860 items/m^3 and 8902 items/m^3 in the urban section and estuary, respectively. Wastewater effluents from cities might be a main source of microplastics in the Pearl River, and the urban tributaries might act as retention systems for microplastics. Among these microplastics, over 80% of them were less than 0.5 mm. The main shapes of microplastics were film, fragment, and fiber, mostly blue or transparent. Moreover, the most common polymer types of these microplastics were polyamide (26.2%) and cellophane (23.1%). This study reveals the contamination and characteristics of microplastics in the Pearl River, and provides important data for further research on microplastics in freshwater ecosystems.

1. Introduction

Plastics are organic synthetic polymers, which have been used widely around the world mainly due to their affordability, bio-inertia, and high strength-to-weight ratio. It was estimated that over 250,000 tons of discarded plastics were deposited from land into the ocean in 2014 (Eriksen et al., 2014). After weathering and

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ultraviolet radiation, larger plastic debris degrade into smaller pieces, and those measuring less than 5 mm are named microplastics (Thompson et al., 2004). Apart from this source, microbeads from daily beauty and health products such as toothpastes and cleansers also contribute to microplastics pollution. Most of these tiny microbeads are made of polyethylene, which can easily flow past water filtration systems and enter lakes and oceans (Auta et al., 2017). Tiny fibers produced during washing are also microplastics. Due to the various sources, microplastics in the environment occur in different shapes, such as fragments, spheres, and fibers. As a potential threat to humans, microplastics pollution has become a growing concern in the world.

Microplastics in the marine environment are easily introduced into the food chain of the ocean because of their more available small size for ingestion. A wide range of marine organisms have been reported to take up microplastics, such as zooplankton, bivalves, shrimp, fish, and whales (Cole et al., 2013; Lusher et al., 2015a; Ferreira et al., 2016). The intake of these tiny particles can cause great harm to organisms, including reduced growth rate, pathological stress, oxidative stress, and reproductive complications. Moreover, toxic chemicals attached to the particles also pose a great risk to marine organisms because of the bigger specific surface area and stronger adsorption ability of microplastics (Reisser et al., 2014). Previous studies have reported that microplastics often accumulate in the tissues of animals, and are difficult to remove and ingest (Auta et al., 2017). Acting as vehicles to transport pathogens and toxic pollutants, accumulated microplastics in animals can eventually be transferred to humans through the food chain, causing serious health problems (Wang et al., 2016).

Microplastics have been detected in growing numbers in waters and sediments throughout the world, at especially high levels in lakes and rivers. In recent years, these small plastics have even been observed in deep-sea sediments and Arctic polar water (Lusher et al., 2015b; Van Cauwenberghe et al., 2015). Studies also demonstrate that microplastics are widely detected in waters in Spain (Iniguez et al., 2017), England (Martin et al., 2017), Australia (Reisser et al., 2013), and the USA (Gray et al., 2018). Microplastics found in waters are mainly composed of polypropylene (PP), polyethylene (PE), polystyrene (PS), polyethylene terephthalate (PET) and polyvinyl chloride (PVC). Microplastics composed of different materials have different environmental behaviors. Those mainly composed of PET and PVC are more likely to sink, while PP, PE, and PS more easily float. Polyamide (PA) and polyvinyl alcohol are common components of microplastics as well (Carr et al., 2016). As these materials are difficult to degrade by microorganisms, microplastics often persist in our environment in all forms, including table salt, beer, sugar, dust in our homes, and even bottled water samples (Kosuth et al., 2018). The concern about the impact of microplastics is increasing, along with a significant increase in the amount of microplastics in the environment.

China is the largest plastics producer in the world (Jambeck et al., 2015). Guangzhou, one of the most important megacities in China, is a representative city with massive plastic production and waste due to the huge population and intensive anthropogenic activities. In 2016, nearly 10 million tons of plastics were produced in Guangdong (Li et al., 2018a). The extensive use and discarding of plastics in Guangzhou greatly increase the microplastics burden in the Pearl River (Lin et al., 2018). Eventually, microplastics will enter the South China Sea through the Pearl River Estuary (PRE). As transitional zones between rivers and oceans, estuaries play a vital role in microplastics transportation. To date, coastal areas along the PRE have become hotspots of microplastics pollution (Fok et al., 2017). Several studies have reported that microplastics pollution was observed in the Pearl River, Hong Kong and Guangdong coastal areas, and the PRE (Fok and Cheung, 2015; Li et al., 2018a; Lin et al., 2018). However, the difference and relationship between microplastics pollution in the PRE and the Pearl River, especially in urban sections, is still unknown.

In this study, we focus on microplastic contamination in the estuary of the Pearl River and its urban section along Guangzhou, in particular, to figure out whether urban tributaries act as retention systems with higher microplastic densities than estuaries. The abundance, size, color, and shape of microplastics in surface water from 26 sites were investigated. We also explored the polymer types of microplastics by Raman spectra. This study will provide basic data for monitoring microplastics in the water resources of southern China.

2. Materials and methods

2.1. Sampling

Water samples were collected from 26 sites in the Pearl River in December 2017, as shown in Supplementary Table S1. Before sampling, all tools were cleaned using distilled water. Samples of 20 L of surface water were collected using a 5 L water sampler and then passed through a 50 μm stainless steel sieve. The residue on the sieve was washed with pure water and removed into 50 mL glass bottle (Lin et al., 2018). Two water samples were taken in the same way at each location. Before the experimental analysis, samples were preserved at 4 °C.

2.2. Microplastic extraction

In the laboratory, to dissolve the natural organics in the water sample, samples were treated with 30% H_2O_2 for 24 h at room temperature in the dark (Nuelle et al., 2014). Then the samples were filtered through 0.45 μm filter paper under a vacuum pump, and the filter papers were placed in a dish and air-dried at room temperature.

2.3. Microscope inspection

The particles were observed on the filter paper with a stereomicroscope (Optec SZ680) and measured with an eyepiece micrometer. Based on previous studies (Cole et al., 2011; Hidalgo-Ruz et al., 2012), microplastics can be divided into three types according to their morphology: fiber, fragment, or film. They were also divided into five categories according to color: green, blue, transparent, red, and other. According to the size, microplastics are divided into six classes: class 1, <0.5 mm; class 2, 0.5–1 mm; class 3, 1–2 mm; class 4, 2–3 mm; class 5, 3–4 mm; and class 6, 4–5 mm. The quantity, type, color, and size of the microplastics in each sample were recorded.

2.4. Microplastics identification

Microplastics cannot be completely accurately identified by visual observation alone (Silva et al., 2018). Raman spectroscopy can be used to analyze the composition of sample particles, as previously reported (Araujo et al., 2018). In this study, 130 samples were randomly selected and analyzed by micro-Raman spectroscopy (Thermo Fisher Scientific DXR2, 532 nm laser, Raman shift 50-3500 cm^{-1}). The obtained spectra were compared with the spectral libraries on the instrument. In addition, several particles were selected for analysis by scanning electron microscope (SEM; Hitachi S-4800, Japan). Particles were placed on double-sided tape and coated with evaporated gold before SEM observation. The number of microplastics for each sample was recalculated after removing the non-microplastics, as previously reported (Di and Wang, 2018).

2.5. Quality assurance and control

To prevent external pollution from affecting the research results, the experimenter needs to wear a cotton test suit and cannot wear plastic gloves. The sampler and stainless steel sieve used for sampling need to be rinsed with pure water in advance. Sampler and sieve need to be washed with pure water after sampling at each location. The water-filled glass container needs to be rinsed 3 times with pure water and baked at 120 °C for 4 h.

2.6. Statistical analysis

Numerical data are presented as the mean ± standard error (SE). Data analysis was performed by SPSS. The means of 2 groups were compared by independent-samples *t*-test. The differences were regarded as significant at $^{*}p < 0.05$ and extremely significant at $^{**}p < 0.01$ in all cases.

3. Results

3.1. Abundance and distribution

Microplastics were widely detected in all water samples collected from 31 sites in the Pearl River, with significant spatial variations in their distribution. The abundance of microplastics in the Guangzhou urban section (GUS) of the river and PRE varied from 8725 to 53,250 and 7850 to 10,950 items/m^3 of water, respectively (Fig. 1). A high density of microplastics of over 20,000 items/m^3 was detected in S1, S2, S3, S6, S8, S9, and S11, all of which were located near industrial parks or logistics parks. The lowest abundance (<8000 items/m^3) was found in P5 and P7, which were at the PRE and far from the city center. The microplastics levels showed less variance in samples from PRE (Fig. 1B). In addition, the average microplastics abundance in GUS (mean 19,860 items/m^3) was over two times higher than that in PRE (mean 8902 items/m^3) ($p < 0.01$). Therefore, microplastics pollution in the urban section of the Pearl River was more serious than in PRE.

3.2. Microplastic characteristics

The size of microplastics surface water from GUS was similar to that from PRE, ranging from 0.05 mm to 5 mm. Over 80% of the microplastics were less than 0.5 mm in all detected samples, while only a small amount of 4–5 mm was observed (Fig. 2A). A higher proportion of microplastics of 0.5–2 mm was found in waters from PRE, though the difference was not significant due to the limited samples. The amount of microplastics decreased as the length increased. Microplastics of 2–3 mm were significantly reduced in waters from PRE than from GUS ($p < 0.05$), and microplastics of 4–5 mm were not detected in waters from PRE (Fig. 2B). The color of microplastics was also recorded. In this study, blue and transparent items were prevalent in all water samples, constituting 38% and 37% of microplastics, respectively. Smaller proportions of green and red plastic items were also found in these samples (Fig. 3).

Typical microplastics are shown in the photographs in Fig. 4A–D. Microplastics in these samples were classified into three shapes: film, granule, or fiber. Briefly, film is a thin piece of plastic debris, granule is a spherical or cylindrical piece or fragment, and fiber is a thin and long item. When an item could not be defined as fiber or film, it was classified as granule. Different proportions of the three shapes were observed at different sampling sites (Fig. 4E). Film was the most dominant component, with a proportion of 52% in samples from GUS, followed by granule and fiber, constituting 41% and 7%, respectively. In waters from PRE, the abundant components were granule and film, with proportions of 48% and 43%, respectively. Similarly, the amount of fiber was the least, accounting for only 9% of microplastics. No significant difference was observed in the shapes of microplastics from these two areas (Fig. 4F). Typical plastic-like particles were further analyzed by SEM. Generally, the surface of polymers was unregulated or smooth (Fig. 5A–F). Moreover, a large amount of transparent pellets (Fig. 4C and D, red arrow) observed in this study were determined as diatoms according to the regular holes on their surface (Fig. 5G and H).

3.3. Composition of the microplastics

To identify the composition of the microplastics, a total of 130 items were randomly selected and observed by micro-Raman

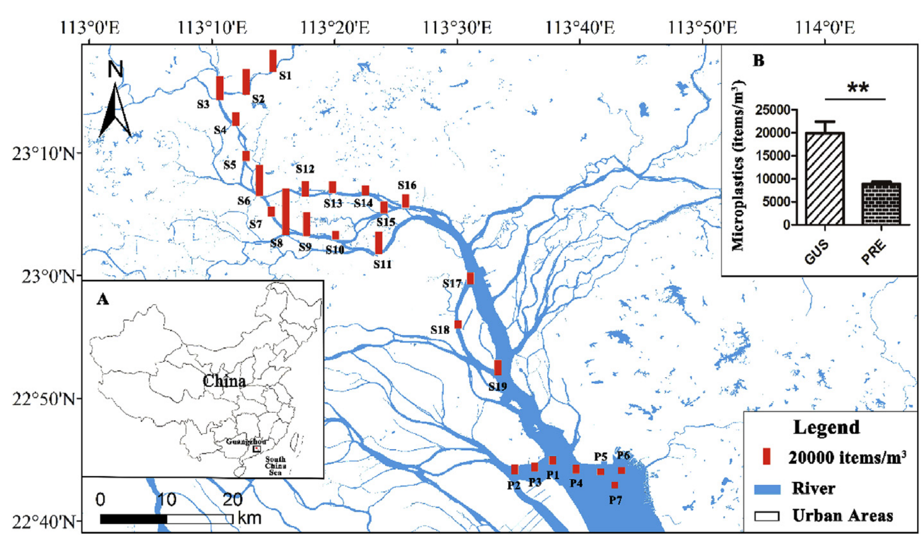

Fig. 1. The abundance of microplastics in the Pearl River. Red column represents the abundance of microplastics in the surface water. Inset A shows the positions of sampling sites in Guangdong, China. Inset B shows a comparison of microplastic abundance between the estuary and the urban section along Guangzhou ($^{**}p < 0.01$). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

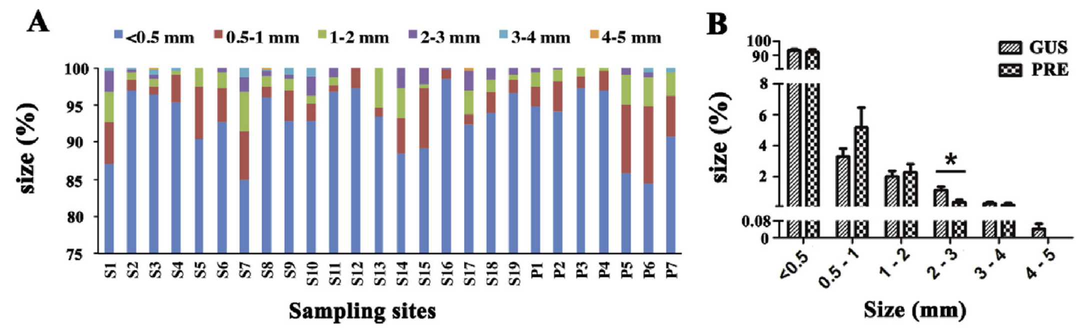

Fig. 2. Sizes of microplastics in the Pearl River. (**A**) Percentages of different-size microplastics at 26 sampling sites. (**B**) Comparison of different-size microplastics between the estuary and the urban section along Guangzhou (*$p < 0.05$).

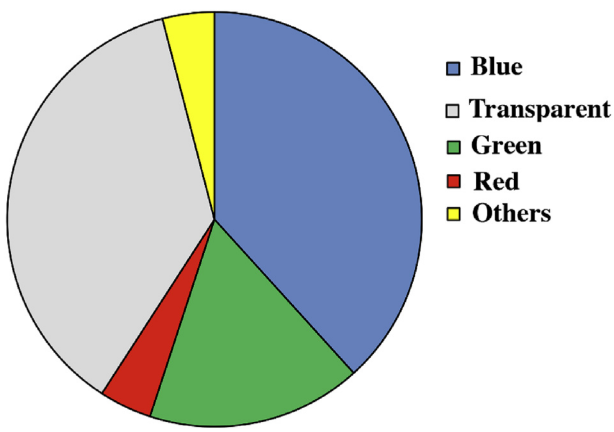

Fig. 3. Colors of microplastics in the Pearl River. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

spectroscopy. The results showed that 112 of them were microplastics. For all samples, polyamide was the most common polymer type (26.2%), followed by cellophane (23.1%), polypropylene (13.1%), and polyethylene (10.0%). A few items were identified as vinyl acetate copolymers (VACs) and polyvinylchloride (Fig. 6A). The Raman spectra of typical microplastics are shown in Fig. 6B. The other 18 items were determined as non-microplastics as their spectra were matched below 70% when compared with the spectra database.

4. Discussion

In the present study, the abundance of microplastics in samples from GUS was significantly higher than from PRE, indicating that wastewater effluents from urban cities might be a main source of microplastics in the Pearl River and the urban tributaries might act as retention systems for microplastics. Seven of the 26 sampling sites near industrial parks or logistics parks showed a value of over 20,000 items/m^3 in our study, which was not surprising, because microplastic inputs are expected to be much higher in industrialized watersheds (Anderson et al., 2016). Besides, insulated boxes are widely used in logistics parks for transporting, which can easily enter water drainage systems when they are improperly disposed,

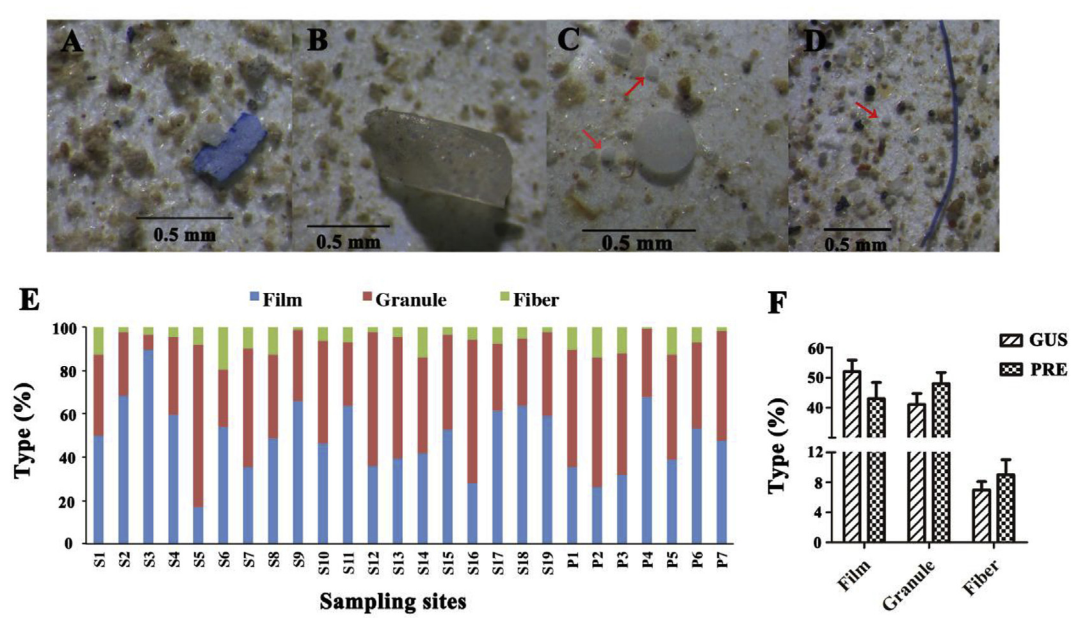

Fig. 4. Types of microplastics in the Pearl River: (**A**) film, (**B,C**) granule, (**D**) fiber. (**E**) Distribution of microplastics at 26 sampling sites by type. (**F**) Comparison of different types of microplastics between the estuary and the urban section along Guangzhou; no significance was observed. Red arrows indicate diatoms. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

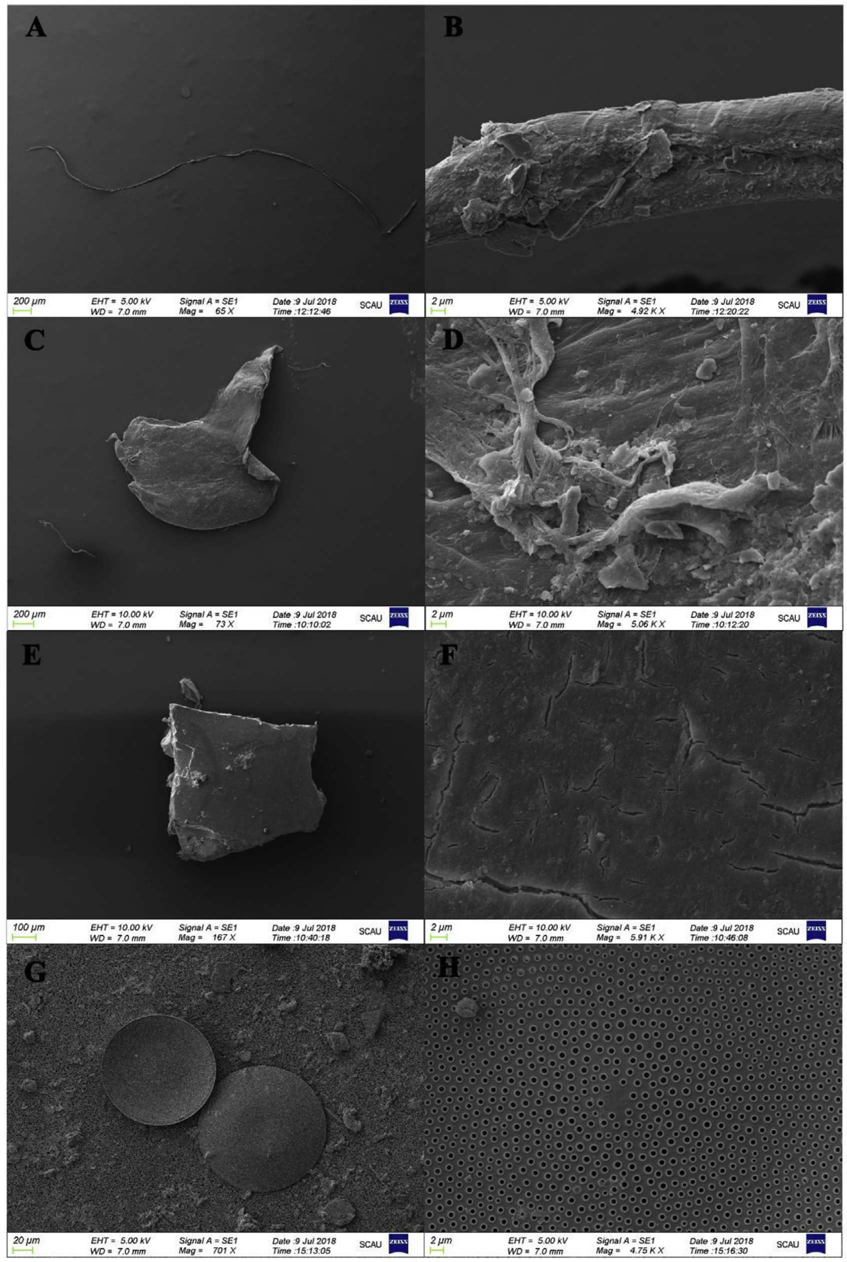

Fig. 5. SEM images of microplastics in the Pearl River: (**A,B**) fiber, (**C,D**) film, (**E,F**) granule, (**G,H**) diatoms.

thus contributing to the microplastics pollution nearby (Fok and Cheung, 2015). Although S10 is next to Xiyu Industrial Park, it is also located near the famous Guangzhou Haizhu National Wetland Park. Lower population density and human activities resulted in a lower concentration of microplastics.

As the monitoring method is still not unified, it was difficult to compare our results with other studies. However, we can compare the microplastic abundance with that reported in other research using similar methods and expressive units. In comparison with worldwide microplastic pollution (Supplementary Table S2), the abundance of microplastics in the Pearl River was much lower than that of the Saigon River in Vietnam (Lahens et al., 2018). Levels of microplastics in GUS were almost two times those of Taihu Lake and Hong Kong beaches in China (Fok and Cheung, 2015; Su et al., 2016), similar to that in the Seine River of France (Dris et al., 2015). The microplastic abundance in PRE was similar to that in the Three Gorges Reservoir (Di and Wang, 2018), but still much higher than the levels monitored in the Antuã River of Portugal and the Great Lakes tributaries of the USA (Baldwin et al., 2016; Rodrigues et al., 2018). A previous study reported that microplastics were widely distributed in the Pearl River along Guangzhou, ranging from 379 to 7924 items/m^3 in waters sampled in July 2017, which were much lower levels than those in our study (Lin et al., 2018). As reported previously, the distribution of microplastics in water can be affected by various factors, such as weather, the ambient environment, and nearby human activities (Thiel et al., 2003; Browne et al., 2011; Kukulka et al., 2012). Seasonally, the microplastic abundance in water would be expected to be lower in July, since the rain events in summer are more intense than in winter in Guangdong Province. Microplastics were also detected in oysters along the PRE, which ranged from 1.4 to 7.0 items per individual and were positively related to those in surrounding water (Li et al., 2018a). These results

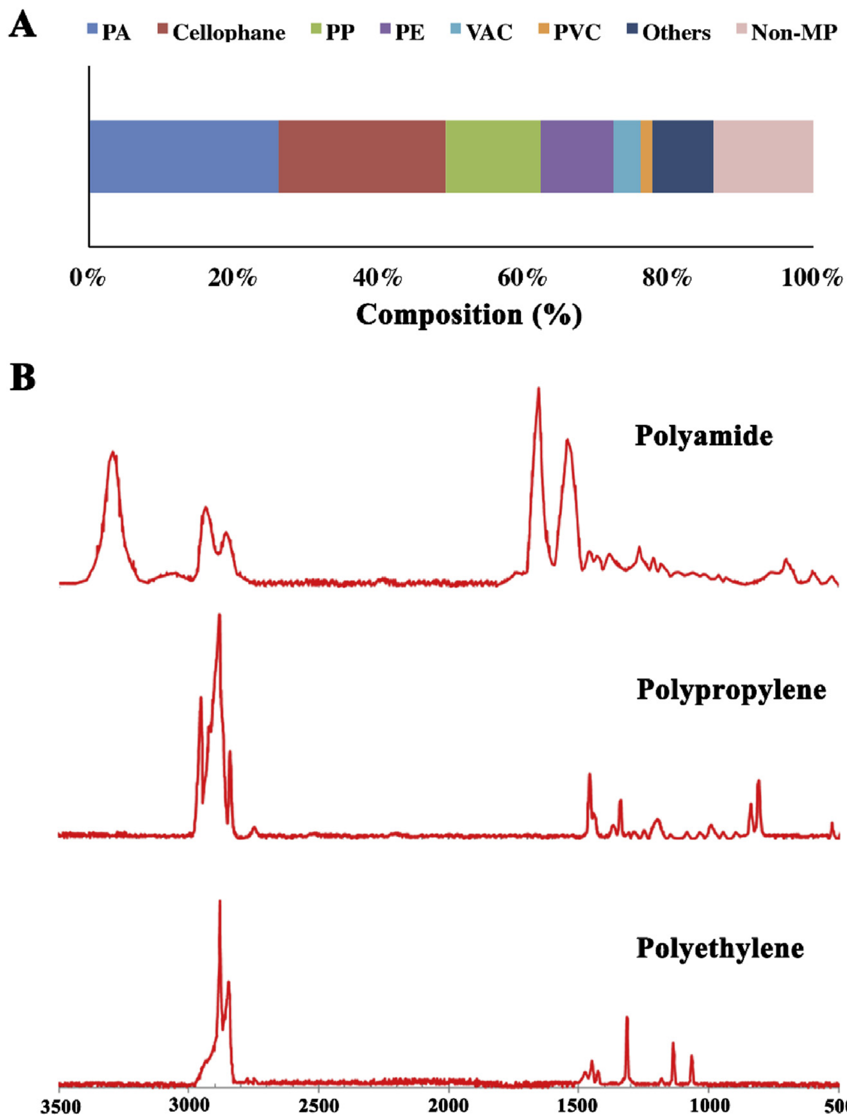

Fig. 6. Composition of selected items identified by micro-Raman spectroscopy. (**A**) Percentage of plastic types in the selected items; (**B**) Raman spectra of typical microplastics.

indicate that microplastic may transfer in the food chain and pose potential threats to aquatic organisms and human.

The high proportion of microplastics smaller than 500 μm was not surprising, and correlated with some previous reports. Similarly, the most common size of microplastics observed in the Yellow Sea and Bohai Sea was in the range of 50–500 μm (Zhao et al., 2018). In Taihu Lake, sizes ranging from 100 to 1000 μm were more frequent in the observation of microplastics (Su et al., 2016). Small microplastics (<250 μm) were predominant and large size classes (>1000 μm) were barely represented in the Saigon River (Lahens et al., 2018). The enrichment of smaller-size microplastics may be because microplastics from wastewater treatment plants are mainly less than 0.5 mm in size (Mason et al., 2016). Another explanation may be that large pieces of plastic could gradually be split into small particles (Zhang et al., 2015). In addition, a small amount of large-size microplastics (4–5 mm) was observed only in the GUS and not in the PRE. Small particles can more easily be carried away by runoff, thus larger ones remained in the tributaries (Hurley and Nizzetto, 2018).

Moreover, we found that most of the microplastics here were classified as film and granule. High proportions of fragment and film were also determined in water from Lake Hovsgol in Mongolia and Tamar Estuary in the UK (Free et al., 2014; Sadri and Thompson, 2014). However, these results were quite different from some previous studies, in which fiber was the most dominant shape (Lusher et al., 2014; Lin et al., 2018). This indicates that microplastic characteristics may be related to the sampling area and the source of plastic in the water. Granules are widely used as material for cosmetic scrubbers or plastic production. Cosmetic products such as facial cleanser and toothpaste contain numerous plastic granules (Napper et al., 2015). Due to the high density of human activities along the Pearl River, these microplastics in the shape of film or granule are likely delivered from urban wastewater or caused by degradation and fragmentation of plastic debris, such as bottles, bags, and wrappers (Free et al., 2014). Currently, the specific mechanism of how plastic degrades and fragments is still unknown, and the effect on determining microplastic density in water needs further investigation.

Determining the origins of microplastics in water is not easy, but their polymer types may provide a potential indication. In this study, PA, cellophane, PP, and PE were observed most frequently in the Pearl River. These polymers are widely used in the packaging industry, which indicates that urban pollution might be an important source of these microplastics. Polypropylene and PE are the most frequently reported polymer types in microplastics from coastal environment (Hidalgo-Ruz et al., 2012), which accounted for a relatively minor proportion here. A previous study, which was focused on sewage sludge from wastewater treatment plants in China, revealed that microplastics in the shape of films were mainly composed of PA (Li et al., 2018b). Polyamide is widely used in the food packaging industry and as monofilament in fishing line, which indicates an urban origin of these particles (Naji et al., 2017). Since cellophane was defined as a kind of microplastic, it has been reported to be prevalent in water systems worldwide (Woodall et al., 2014; Yang et al., 2015; Castillo et al., 2016). As an organic cellulose-based polymer, cellophane is commonly used in cigarette and food wrappers, and acts as a release agent for the manufacture of rubber and fiberglass products as well (Yang et al., 2015). Some particles were determined as non-microplastics here. However, most of them were identified as additives or compositions of plastics. For example, cyclopentanone is a common characteristic compound added to PA (Dekiff et al., 2014). Thus, component analysis is quite important for the identification of microplastics.

5. Conclusion

This study reveals microplastics pollution in waters from the Pearl River. The abundance of microplastics ranged from 8725 to 53,250 items/m^3 in the GUS and 7850 to 10,950 items/m^3 in the PRE. The highest density of microplastics was detected in sampling sites near industrial parks or logistics parks. The concentration of microplastics in GUS was much higher than in PRE, indicating that intensive human activities might be an important cause of microplastics pollution in the Pearl River. Most of the microplastics were less than 500 μm in size. The main shapes of observed microplastics were films, fragments, and fibers, mostly colored blue or transparent. Moreover, polyamide and cellophane were the most common polymer types among these microplastics. Overall, these results highlight the microplastics contamination in the Pearl River and provide important data for further research on microplastics in freshwater ecosystems.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2018.11.093.

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