Environmental Pollution 251 (2019) 862–870

Contents lists available at ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol

Distribution, sedimentary record, and persistence of microplastics in the Pearl River catchment, China★

Yujuan Fan [a, b], Ke Zheng [a, b], Zewen Zhu [a, b], Guangshi Chen [a, b], Xianzhi Peng [a, *]

[a] State Key Laboratory of Organic Geochemistry, Guangzhou Institute of Geochemistry, Chinese Academy of Sciences, Guangzhou, 510640, China
[b] University of Chinese Academy of Sciences, Beijing, 100049, China

ARTICLE INFO

Article history:
Received 28 January 2019
Received in revised form
12 April 2019
Accepted 11 May 2019
Available online 15 May 2019

Keywords:
Microplastics
River
Sedimentary core
Vertical distribution
Spatial and seasonal patterns

ABSTRACT

Microplastics (MPs) in the environment have become an issue worldwide. However, data about MPs in freshwater systems are still limited so far. This study investigated sources, fate, and seasonal and spatial distribution of MPs in the main stream Pearl River and its tributaries, as well as in the Pearl River Estuary (PRE), China. MPs were widely detected in the river water, river bed sediment, and estuarine sediment, with abundances of 0.57 ± 0.71 items L^{-1}, 685 ± 342 items kg^{-1} dry weight (dw), and 258 ± 133 items kg^{-1} dw, respectively. Sheet, fragmental, and fibrous polyethylene, polypropylene, and ethylene-propylene copolymers were predominant, suggesting that MPs in the Pearl River catchment be mainly derived from fragmentation of discarded plastic wastes. In addition, municipal wastewater was also an important MPs source, especially for polyethylene terephthalate (PET) fibers. Polymers of higher density, such as PET and polyvinyl alcohol were relatively more abundant in the sediment than in the river water, especially in the estuarine sediment. Upward increase of the MP abundance was observed in the sedimentary core, probably indicating increasing release of plastic wastes due to growing production and uses of plastic products. On the other hand, percentage of finer MPs increased with increasing depth. The results revealed persistence and potential downward dispersion of the fine MPs. The MPs abundance was positively related with population density and gross domestic product, demonstrating impacts of human activities and economic development on the MPs contamination. Higher MPs abundance was detected in dry season than in wet season in the river water, suggesting dilution effect of precipitation. It's estimated that 15963 tons of MPs could be released annually into the PRE from the main stream Pearl River and its tributaries.

1. Introduction

Plastics are widely used in manufacture of a variety of industrial and daily life products due to their unique properties, such as buoyancy and strong tear resistance (Laist, 1987). Global production of plastics has been increasing drastically since their commercial development in 1940s and has reached 350 million tons in 2017 (PlasticsEurope, 2018). As a result, large amounts of plastic wastes may end up in the environment (Derraik, 2002; Fendall and Sewell, 2009; Ashton et al., 2010; Browne et al., 2011; Hidalgo-Ruz et al., 2012; Jambeck et al., 2015; Wright and Kelly, 2017), which may pose long-term risks to the ecosystem considering poor decomposability of the plastics.

Once entering the environment, plastic wastes may fragment into smaller and smaller pieces, facilitating their long range transport and dispersion in the environment, even being ingested by various organisms (Baird and Hooker, 2000; Leite et al., 2014). Environmental risks of plastic wastes have been extensively studied. In addition to well-known blockage and abrasion of gastrointestinal and ingestive tracts of organisms by large plastic pieces (>5 mm), microplastics (MPs, <5 mm) may significantly impact assimilation efficiency and energy balance, causing negative repercussions on reproduction, disrupting feeding habits, and increasing mortality (Cole et al., 2013; Lee et al., 2013; Wright et al., 2013; Gardon et al., 2018). In addition, MPs may act as a vector and a source of chemical contaminants and pathogens (Andrady, 2011; Bakir et al., 2012; Besseling et al., 2013; Fisner et al., 2013; McCormick et al., 2014; Setala et al., 2014; Mai et al., 2018a; Paluselli et al., 2019). MPs have therefore been included in

★ This paper has been recommended for acceptance by Maria Cristina Fossi.
* Corresponding author.
E-mail address: pengx@gig.ac.cn (X. Peng).

https://doi.org/10.1016/j.envpol.2019.05.056
0269-7491/

emerging contaminants (Browne et al., 2007; Lambert and Wagner, 2018).

MPs have been found ubiquitous in oceans, from the North Pacific gyre to remote Arctic areas, from sea surface to hadal bottom (Barnes et al., 2009, 2010; Van Cauwenberghe et al., 2013; Obbard et al., 2014; Woodall et al., 2014; Zhang et al., 2016; Cincinelli et al., 2017; Pazos et al., 2018; Peng et al., 2018), in soil and terrestrial system (Fuller and Gautam, 2016; Zhou et al., 2016; Liu et al., 2018; Scheurer and Bigalke, 2018; Zhang and Liu, 2018). In contrast, occurrence of MPs in freshwater systems was much less documented, which is attracting growing attentions (Wanger et al., 2014; Horton et al., 2017; Hu et al., 2018). Previous researches have revealed large amount of plastic wastes being transported into the sea through rivers (McCormick et al., 2014; Lebreton et al., 2017; Schmidt et al., 2017; Hendrickson et al., 2018). Recent studies have demonstrated presence and high abundance of MPs in rivers and lakes worldwide (Zbyszewski and Corcoran, 2011; Lechner et al., 2014; Castaneda et al., 2014; Sadri and Thompson, 2014; Mani et al., 2015; Klein et al., 2015; Fischer et al., 2016; Zhang et al., 2015; Biginagwa et al., 2016; Vaughan et al., 2017; Nel et al., 2018; Wang et al., 2018). For example, plastic particles in Austrian Danube have outnumbered fish larvae (Lechner et al., 2014). Polyethylene (PE) microbeads in sediment of St. Lawrence River, Canada were up to 13832 items m^{-2} (Castaneda et al., 2014). MPs were observed ubiquitous in Taihu Lake and the Yangtze River catchment, China (Su et al., 2016; K. Zhang et al., 2017; Luo et al., 2019; Xiong et al., 2019).

China has the largest production of plastics, accounting for 26% of the global production (PlasticsEurope, 2018). The Pearl River system was ranked the third most polluted river in the world with estimated midpoint mass inputs of plastic wastes of $>1.1 \times 10^5$ tons per year (Lebreton et al., 2017). Occurrence of MPs in the Pearl River and the Pearl River Estuary (PRE) has been primarily reported (Lin et al., 2018; Lo et al., 2018). However, distribution patterns, detailed characterization, and environmental fate of the MPs in the Pearl River catchment are far from well-studied.

This study aimed to (1) characterize in detail the MPs in the water and sediment to discuss sources of the MPs in the Pearl River catchment, (2) investigate spatial and seasonal patterns of the MPs to illustrate factors impacting the MPs contamination, and (3) study sedimentary record to reveal fate and persistence of the MPs in the environment. The results of this research would not only reveal impacts of anthropogenic activities on the plastics contamination but also shed an insight in fate of plastic wastes in the environment.

2. Materials and methods

2.1. Study area and samplings

The main stream Pearl River (PR) and its 3 major tributaries, named Xijiang River (XJ), Beijiang River (BJ), and Dongjiang River (DJ), flow through Guangdong Province, South China and finally merge into the South China Sea via the PRE. A detail description of the study area has been provided previously (X. Peng et al., 2017,). A sketch map of the study area with the sampling sites was shown in Fig. S1 in the Supplementary Information (SI). Eleven sampling sites were set, 1 in XJ (R1), 1 in BJ (R2), 3 in PR (R3-R5), 2 in DJ (R6-R7), and 3 in the PRE (E1-E3). In addition, a sedimentary core was collected in the PRE (C1).

Surface water samples were only collected in the rivers. Samplings were performed seasonally in 2016 and 2017 during March to May (spring), July to August (summer), and November to next January (fall). The suspended particles in the water were collected using surface trawling with a 160 μm mesh size Watertools WT002120 plankton net (Hydro-Bios Apparatebau GmbH, Kiel-Altenholz, Germany). A Hydro-Bios Model 438-115 digital flow meter was attached at the mouth of the trawl frame to in-situ measure water volume flowing through the net. Water velocity was measured using a Nanshui Current Meter (Baowei Instrument Co., Ltd, Nanjing, China). After trawling for about 15 min with filtered water volumes of 18860–138134 L, the nets were lifted and flushed from outside with river water to ensure all particles into the end bucket located at the bottom of the nets. The buckets and nets were then wrapped with clean aluminum foils and kept in covered stainless steel barrels. The trawling times, water volumes running through the trawling net, weather conditions (e.g., air temperature and precipitation), and water parameters (e.g., surface water temperature) were recorded during each sampling event. All the water suspended particles were collected in duplicate at each site.

The river bed sediments were collected using a grasp bucket, wrapped with clean aluminum foils, and sealed in ziploc polyethylene bags. The estuarine surface sediment and sedimentary core were sampled in April 2013, which has been described in detail previously (X. Peng et al., 2017). The sedimentary core (22°16′, 113°38′) was collected using a gravity corer with a depth of 54 cm and was sliced at the site with intervals of 2 cm.

All samples were kept on ice packs during transport to the laboratory. Upon arriving at the laboratory, the suspended materials captured by plankton nets were immediately eluted with deionized water into 5 L glass beakers followed by digestion and filtration within 24 h. The sediment samples were stored at −20 °C prior to further treatment.

2.2. Sample treatment

2.2.1. Suspended particles of river water

The suspended particles collected in the glass beakers were first sifted through 5 mm stainless steel sieves and the large debris (>5 mm) were discarded. The filtrates were added with 30% H_2O_2 solution (30%, V/V) to digest living organisms and organic matter at room temperature in dark for 3 d as described in literature (Nuelle et al., 2014; Tagg et al., 2015). The digested samples were then filtrated through 0.7 μm glass microfiber filters (GF/F, Whatman International Ltd., Maidstone, England) on a vacuum system. The filters containing the particles were placed in clean glass petri dishes and dried at 45 °C.

2.2.2. Sediment

The sediments were freeze-dried and homogenized. Large plastic debris (>5 mm) was visually picked out and put away. An aliquot of 1 kg of each sediment sample was then sieved successively through 1 mm, 0.45 mm and 0.1 mm mesh sieves. Each portion was collected separately. MPs in each portion of the sediment were then extracted using density separation as reported in literature (Thompson et al., 2004; Hidalgo-Ruz et al., 2012; Mai et al., 2018b) with moderate modifications. The sediment was put into a 1 L glass beaker, followed by addition of 750 mL of potassium formate (KF) solution with density of 1.5 g cm^{-3}. The mixture was homogenized, vigorously stirred for 30 min, and was kept stationary overnight before the supernatant was collected. This process was repeated three times. The supernatants were combined and filtered through 8 μm cellulose nitrate membrane filters (Whatman International Ltd., Maidstone, England). The filters were stored in clean glass petri dishes and were dried at 45 °C prior to further observation.

For the sediment core, each 3 consecutive slices (6 cm of depth) were combined and homogenized to make a composite sample due to sample volume. MPs in the composite samples were then treated as described above for the surface sediment samples.

2.3. MPs analysis

The particles retained on the filters were visually examined carefully under a Leica EZ4 stereomicroscope (Leica, Wetzlar, Germany) and the suspected MPs were primarily picked out with stainless tweezers based on their shapes (Fig. S2), surface texture, colors, and luster. The particles were classified according to size (the longest dimension), color, and shape. In each size class, the particles were counted and weighed in order to facilitate quantification of the MP abundances and concentrations. However, in the sedimentary core, the MPs were not weighed due to too low in mass.

Polymer identification was conducted on a Nicolet iN10 integrated infrared microscope interfaced with a Fourier transformation spectroscopy (μ-FTIR) (Thermo Fisher, Atlanta, GA, USA) and equipped with a liquid nitrogen-cooled mercury cadmium telluride (MCT) detector in transmission mode. The spectra were recorded as an average of 128 scans in the spectral range of 675–4000 cm^{-1} at a resolution of 8 cm^{-1} and an aperture of 100 × 100 μm. The infrared spectrum was monitored three times from different points on each particle to obtain better spectra. The obtained spectra of the samples were matched with polymeric material database using OMNIC software (Thermo Fisher, Atlanta, GA, USA). A MP polymer was identified when a matching degree with the standard spectrum was >80%. Some randomly selected particles were further confirmed by a Raman spectroscopy (HORIBA Scientific, Kyoto, Japan).

2.4. Quality assurance and quality control

In order to efficiently extract the MPs from the sediment, sodium chloride (NaCl, density of 1.2 g cm^{-3}), sodium iodide (NaI, density of 1.8 g cm^{-3}), zinc chloride (ZnCl2, density of 1.6–1.7 g cm^{-3}), and potassium formate (KF, density of 1.5 g cm^{-3}) were tested as separation solutions. KF was finally chosen due to relatively better recovery (Fig. S3) and easily visual examinations. Lab coats, cotton clothing and gloves were worn during the whole experiments to reduce potential contamination. All glass vessels were washed with distilled water and covered with aluminum foils prior to use. Sample pretreatment were performed in fume hood to reduce contamination from the ambient air. Visual examination and pickup of the suspected MPs were carried out in the ultra-clean bench. All liquid reagents/solutions were filtered with glass microfiber filters (0.7 μm, Whatman GF/F) before use. Procedural blanks and laboratory blanks were set to monitor laboratory contamination, which has been described in detail in the SI. No plastic debris was found in the blanks. Recovery tests were performed by spiking polyethylene (PE) pellets of 125 μm (Sigma-Aldrich, St, Louis, Missouri, USA) in tap water and clean sediment. The recoveries were 97 ± 2% and 96 ± 3%, respectively. The MP abundance and concentration were reported in items L^{-1} and mg L^{-1} in the water and items kg^{-1} and mg kg^{-1} in the sediment, respectively. Duplicated analyses were performed for each water samples and the results were presented as mean ± standard deviation (SD).

2.5. Statistical analysis

Data processing was conducted using Origin 8.5 (OriginLab Corporation, Northampton, MA, USA), IBM SPSS 24.0 (IBM, Armonk, NY, USA), and Excel 2010 (Microsoft, Seattle, WA, USA). Nonparametric Kruskal–Wallis test was used for multiple comparisons. If Kruskal–Wallis test indicated significant difference, nonparametric Mann–Whitney *U* test for pairwise comparisons was further performed to identify significantly different groups. The significance was set at $p < 0.05$ unless specified. Mapping of the spatial distribution was performed using ArcGIS10.2 (ESRI, Redlands, CA, USA).

3. Results and discussion

3.1. MPs in river surface water

3.1.1. Occurrence

A total of 813 randomly selected particles were analyzed by μ-FTIR, in which 94.4% were identified as the MPs. The MP abundances and concentrations in the water of PR and the tributaries were 0.57 ± 0.71 items L^{-1} and 0.051 ± 0.036 mg L^{-1}, respectively (Table 1, Fig. S1). As illustrated in Table S1, this result was comparable to those reported in the Pearl River along Guangzhou city and Jiaojiang River, China (Lin et al., 2018; Zhao et al., 2015), relatively lower than those in Taihu Lake and Yangtze River tributaries, China (Hu et al., 2018; Su et al., 2016; Wang et al., 2016), Seine River, and Danube River, Europe (Dris et al., 2015; Lechner et al., 2014), but higher than those observed in Great Lake tributaries, USA (Baldwin et al., 2016; McCormick et al., 2014), Lake Bolsena and Lake Chiusi, central Italy (Fischer et al., 2016).

3.1.2. MPs characterization

The MPs in the river water were categorized into 4 size classes: 1–5 mm, 0.45–1 mm, 0.25–0.45 mm, and <0.25 mm considering that the trawling net with mesh of 160 μm possibly could not collected particles <0.1 m. The finest (<0.25 mm) represented 60.6 ± 10.6% of the observed MPs, followed successively by MPs of 0.25–0.45 mm, 0.45–1 mm, and 1–5 mm, showing decrease in number as increase in size (Fig. 1a). Similar size distribution patterns have been documented for MPs in coastal soils adjacent to the Bohai Sea and the Yellow Sea (Zhou et al., 2018). Increasing numbers of fine MPs was ascribed to continuous mechanical and chemical weathering of plastic wastes in the environment (Goldstein et al., 2013; Jayasiri et al., 2013; Free et al., 2014; Nor and Obbard, 2014; Baldwin et al., 2016). On the other hand, the largest portion (1–5 mm) made up 56.4 ± 9.4% of the MPs mass. Contrary to the abundance, the MPs demonstrated decrease in mass with decline in size (Fig. 1a), probably suggesting that the MPs in the river water be mainly sourced from fragmentation of discarded large plastics wastes.

Morphologically, sheet MPs were the predominant in the river water, followed successively by fragments, fibers, foams, and spherules (Fig. 1b). MPs in the environment come from primary sources such as industrial raw material (Mato et al., 2001) and microbeads applied in some detergents and personal care products (Gregory, 1996; Fendall and Sewell, 2009), and secondary sources generated mainly by mechanical and photo-oxidative degradation of various discarded plastic wastes into smaller pieces (Browne et al., 2007; Singh and Sharma, 2008; Cole et al., 2011). Furthermore, laundry wastewater can also be a significant source of MP fibers in the environment, especially in urban areas (Hartline et al., 2016; Hernandez et al., 2017; Kay et al., 2018; Li et al., 2018). The predominance of sheet and fragmental MPs in the river water further suggested that the MPs in the PR and the tributaries be mainly derived from fragmented and degraded plastic wastes, indicating that proper management of discarded plastic products is especially important in reduce MPs contamination in the area. In addition, large quantities of fibrous MPs might imply significant contribution of laundry wastewater to the MP contamination in the rivers. In contrast, industrial raw material (pellets, flakes and spherules) accounted for 79.4% of the plastic debris in Austrian Danube, suggesting predominantly primary source of the MPs in Danube River (Lechner et al., 2014).

In term of colors, white/transparent MPs were predominant,

Table 1
Abundance and concentration (mean ± standard deviation) of the MP in the Pearl River catchment.

	Water						Sediment	
	Abundance (items L^{-1})			Concentration (mg L^{-1})			Abundance (items kg^{-1})	Concentration (mg kg^{-1})
	Spring	Summer	Winter	Spring	Summer	Winter		
Mainstream PR	0.37 ± 0.05	0.35 ± 0.08	1.96 ± 0.90	0.078 ± 0.038	0.046 ± 0.022	0.083 ± 0.044	928 ± 210	216 ± 130
BJ	0.14 ± 0.01	0.14 ± 0.01	0.36 ± 0.01	0.011 ± 0.002	0.013 ± 0.002	0.014 ± 0.008	132	40
DJ	0.25 ± 0.05	0.24 ± 0.08	0.64 ± 0.33	0.055 ± 0.021	0.062 ± 0.024	0.058 ± 0.050	604	247
XJ	0.22 ± 0.02	0.27 ± 0.11	0.43 ± 0.12	0.039 ± 0.010	0.035 ± 0.012	0.018 ± 0.014	586	109
PRE	NA	NA	NA	NA	NA	NA	258 ± 133	14 ± 14

NA: not analyzed.

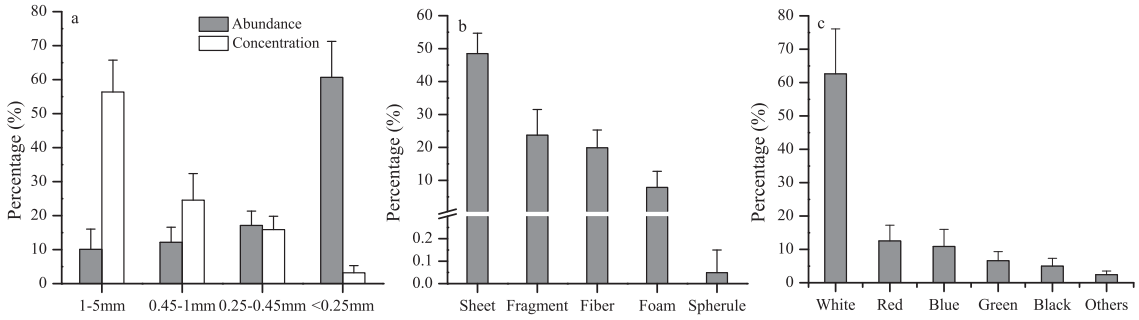

Fig. 1. Distributions of size (a), shape (b), and color (c) of microplastics (mean ± SD) in the water of the Pearl River and the tributaries.

accounting for 62.6 ± 13.5%, followed by red, blue, green, and black. Yellow and purple MPs were only sporadically observed (Fig. 1c). Similar result was reported in the sewage sludge from 11 provinces of China (Li et al., 2018). This might be related to colors of plastic products produced and used in China. In addition, yellowing/weathering effect by photodegradation after being discharged in the environment could also bleach out color of the plastic debris, leading to increasing portion of white/transparent MPs.

3.1.3. Polymer compositions of the MPs in the river water

Eighteen polymers were identified as illustrated in Fig. 2. Overall, polypropylene (PP) and low density polyethylene (LDPE) were the most abundant, accounting for 38.1 ± 10.3% and 31.1 ± 11.6%, respectively. Overall, PP, LDPE, PE-PP copolymers, and high density polyethylene (HDPE) summed up to 64.5–83.7% of the total MPs. Polyethylene terephthalate (PET), polystyrene (PS), polyvinyl chloride (PVC), polyvinyl alcohol (PVA), polyamide/nylon (PA), ethylene vinyl acetate (EVA), and alkyd were also widely distributed. Furthermore, acrylonitrile butadiene styrene (ABS), polycarbonate (PC), polyurethane (PU), acrylic/poly (methyl methacrylate) (PMMA), and polyacrylonitrile (PAN) were observed in PR. This result was basically consistent with those reported for waters worldwide in which PP and LDPE have been the predominant MP polymers (McCormick et al., 2014; Wang et al., 2016; Tsang et al., 2017; W. Zhang et al., 2017; Lahens et al., 2018; Rodrigues et al., 2018).

Specifically by morphology, the sheet and fragmental MPs mainly consisted of PE, PP, and PP-PE copolymer (Fig. 3a), probably deriving from fragmentation and breakup of discarded such plastic wastes as plastic bottles and bags. Fibrous MPs were mainly made up of PET, PP, and PA (Fig. 3b). PP fibers are widely used in products as carpets and thick ropes and may enter the environment via washing and discard of these products (Claessens et al., 2011; Halstead et al., 2018). while PET fibers are widely applied in textiles and may be released into the environment via municipal wastewater discharge (Browne et al., 2011; Halstead et al., 2018; Kay et al., 2018). PA fibers found in BJ and XJ (Figs. S4a and e) might be sourced from lost/discarded fishing gears (Claessens et al., 2011). The spherules were only detected at one site in PR (R3) nearby a large scale sewage treatment plant (Fig. S1) and consisted mainly of PE, PS, and PP (Fig. 3c), probably sourced from microbeads applied in detergents and personal care products released with wastewater. The foams in the river water were all expanded PS.

3.1.4. Spatial and seasonal patterns of the MPs

Spatial distribution pattern of the MPs in the river water was illustrated in Fig. 4a. Generally, the MPs abundance and concentration were the highest in PR (0.89 ± 0.92 items L^{-1}; 0.069 ± 0.038 mg L^{-1}), followed by DJ (0.40 ± 0.28 items L^{-1}; 0.059 ± 0.026 mg L^{-1}), XJ (0.30 ± 0.13 items L^{-1}; 0.031 ± 0.014 mg L^{-1}), and BJ (0.21 ± 0.11 items L^{-1}; 0.013 ± 0.004 mg L^{-1}). Closely positive correlation was found between the MPs abundance and population density as well as the gross domestic product (Fig. 5), implying significant impact of human activities and economic development on the MPs contamination. Urban development and population density have been reported to significantly influence MPs contamination in the environment (Browne et al., 2011; Yonkos et al., 2014; Peters and Bratton, 2016; Luo et al., 2019). Furthermore, municipal wastewater and surface runoff were found to be important sources of MPs in the aquatic environment (Mason et al., 2016; Lasee et al., 2017; Barletta et al., 2019). The highest MPs abundance observed at R3 in PR was probably related with the outfall of a largest scale sewage treatment plant of Guangzhou in the vicinity (Fig. S1).

More specifically, fibrous MPs were relatively richer whereas fragmental MPs were relatively less abundant in BJ than in the other rivers (Fig. S4a). Besides, percentages of the finest MPs (<0.25 mm) were higher in PR and XJ than in DJ and BJ (Fig. S4b). Regarding polymer compositions, PE was more abundant in BJ than in the other tributaries whereas PP was more abundant in DJ and XJ without significance (Fig. S4c). No spatial difference was observed for color distribution of the MPs (Fig. S4d). These results suggested

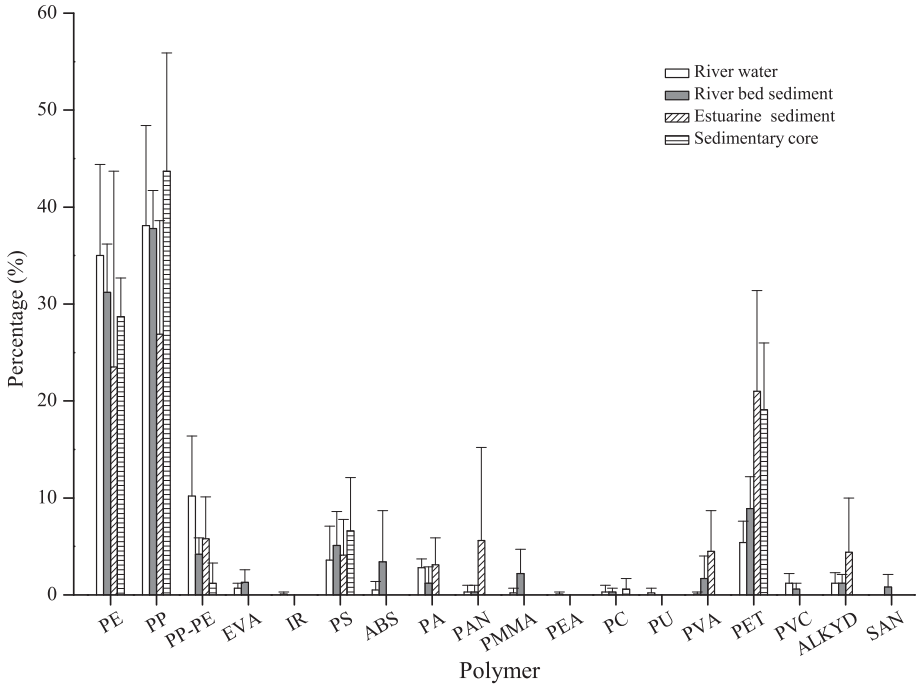

Fig. 2. Polymer composition of the MPs in the Pearl River catchment. PP: polypropylene; PE: polyethylene, including LDPE and HDPE; PET: polyethylene terephthalate; PS: polystyrene; PVC: polyvinyl chloride; EVA: ethylene vinyl acetate; IR: isoprene rubber; ABS: acrylonitrile butadiene styrene; PA: polyamide/nylon; PAN: polyacrylonitrile; PMMA: acrylic/poly (methyl methacrylate); PEA: polyethylacrylate; PC: polycarbonate; PU: polyurethane; PVA: polyvinyl alcohol; ALKYD: alkyd resin; SAN: styrene acrylonitrile.

discharge of similar type of plastic wastes in the PR catchment.

The MPs abundances were significantly higher in fall (1.19 ± 1.01 items L^{-1}) than in spring (0.29 ± 0.10 items L^{-1}) and summer (0.27 ± 0.11 items L^{-1}, Fig. 4b, $p = 0.001$). Annual precipitation in the Pearl River Delta averages >2000 mm, mostly in wet season (spring to summer). Therefore, lower MPs abundances in the river water in spring and summer could be ascribed to dilution effect of the precipitation. In contrast, the MPs concentrations were comparable in fall (0.057 ± 0.045 mg L^{-1}) and spring (0.056 ± 0.037 mg L^{-1}), while slightly lower in summer (0.044 ± 0.027 mg L^{-1}). The MPs of 1–5 mm accounted for the major of the MPs mass but represented the minor of the MPs particles (Fig. 1a), which might explain the difference in seasonal patterns of the MPs in number and in mass. However, significantly higher abundances and weights were reported in wet season than in dry season in the PRE on the west of Hong Kong, which was ascribed to transportation of plastic litter by surface runoff during the wet season (Cheung et al., 2016). Higher MPs abundance observed in rainy season in Goiana Estuary, Brazil was also attributed to high freshwater inflow flushed MPs (Lima et al., 2015, 2016). There were not significantly seasonal differences in distribution of size, color, shape, and polymer compositions of the MPs (Fig. S5), implying possibly similar discharges of plastic waste types in all seasons. In addition, seasonal patterns of the MPs did not show obvious difference among the rivers (Figs. S6 and S7).

3.2. MPs in the river bed sediment

The MPs abundance and concentration in the river bed sediment were 685 ± 342 items kg^{-1} dry weight (dw) and 174 ± 115 mg kg^{-1} dw, respectively (Table 1, Fig. S1). This result was comparable to those obtained in sand from littoral zone of BJ (Wang et al., 2017), the sediments from small water bodies of the Yangtze River Delta, China (Hu et al., 2018), River Thames, UK (Klein et al., 2015; Horton et al., 2017), and rivers in the Mediterranean sea (Martellini et al., 2018). However, the MPs abundance obtained in the sediment of the PR and the tributaries were relatively higher than those in the sediments of Taihu Lake (Su et al., 2016), middle and lower reaches of the Yangtze River, China (Xiong et al., 2019), Lake Bolsena and Lake Chiusi, central Italy (Fischer et al., 2016), and River Kelvin, UK (Blair et al., 2019).

Like in the river water, the MPs abundance in the bed sediment of PR (928 ± 210 items kg^{-1}) was higher than those in DJ (604 items kg^{-1}), XJ (586 items kg^{-1}), and BJ (132 items kg^{-1}) though the differences were not significant (Fig. S8). Similar phenomenon was observed in the sediment of Taihu Lake, with higher MPs in the most heavily contaminated northwest area of the lake (Su et al., 2016).

The MPs in the river bed sediment showed similar distribution patterns to those in the water in terms of size, shape, and color (Fig. S9). The finest fraction (<0.1 mm) accounted for $45.0 \pm 4.3\%$ of the MPs in number whereas the coarsest (>1 mm) portion accounted for $64.5 \pm 7.0\%$ of the MPs in mass (Fig. S9a). Sheets was the predominant, making up to $70.0 \pm 4.2\%$ of the total MPs in the river bed sediment, followed by fibers and fragments. Spherules and foams were occasionally observed (Fig. S9b). Density of expanded PS (0.015–0.035 g cm^{-3}) is much lower than water, which could explain its rare detection in the sediment. On the other hand, percentage of spherule MPs in the sediment (0.9%) was relatively higher than in the water (0.1%), indicating higher likeliness of the spherical MPs to settle down to the sediment than to float in the water, which is reasonable considering the relatively smaller surface areas of the spherical MPs. In addition, percentage of sheet MPs appeared higher in the sediment than in the water, probably due to easier curling of the sheet MPs, leading to reduction of surface areas and consequently sinking. White/transparent MPs were the predominant, accounting for $51 \pm 7\%$, followed by red, green, red, black, yellow, and purple MPs (Fig. S9c).

PP, PE, and PP-PE copolymers were also dominant polymers in the bed sediment (Fig. 2), which were similar to the river water. PE

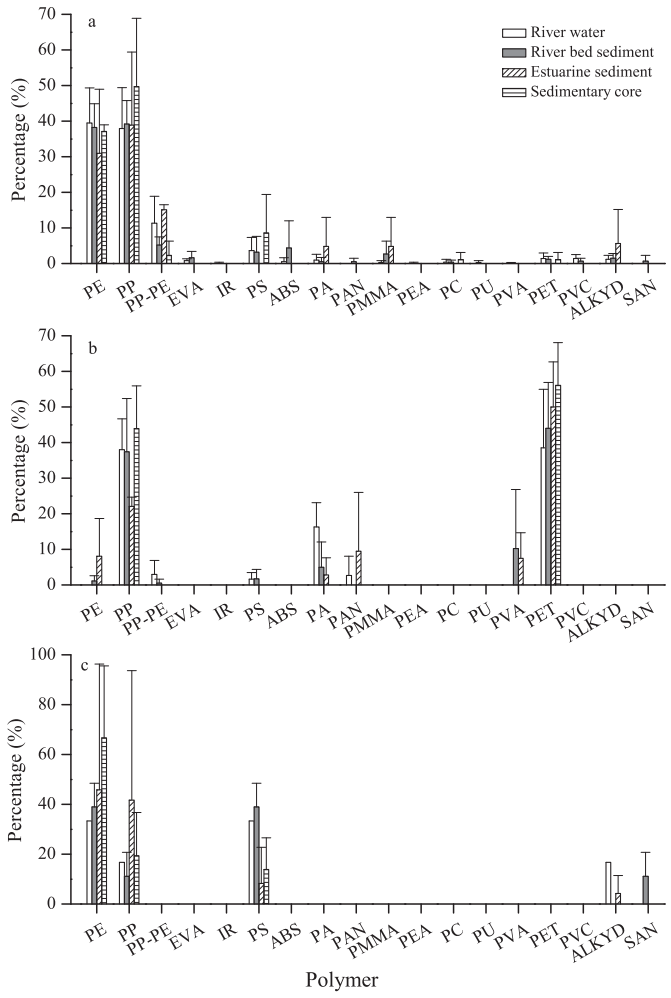

Fig. 3. Polymer composition of the microplastics in different shape. (a) sheet/fragment, (b) fibrous, (c) spherical. Abbreviations of the polymer names see Fig. 2.

and PP have been widely detected as predominant polymers in sediments (Zhang et al., 2016; Sruthy and Ramasamy, 2017; Rodrigues et al., 2018; Zhou et al., 2018). Previous research revealed that buoyancy of plastics can be influenced by biofouling, causing low-density plastics to sink (Lobelle and Cunliffe, 2011), which might be the reason for wide observation of PP and PE in sediments regardless their density less than water. Others polymers, such as PET, PVC, PVA, PA, EVA, ALKYD, ABS, PC, and PU were also detected (Fig. 2). It's worthy to note that high density polymers such as PET, PVA, ABS, PMMA (Table S2), and SAN were more abundant in the sediment than in the water (Fig. 2), indicating their higher inclination to sink into the sediment due to higher density.

3.3. Distribution and vertical pattern of the MPs in the Pearl River Estuary

The MPs abundance and concentration in the estuarine surface sediment of the PRE were 258 ± 133 items kg^{-1} dw and 14 ± 14 mg kg^{-1} dw, respectively, lower than those in the river bed sediment (Table 1, Fig. S8). This result fell in range of those detected in coastal area of Hong Kong (Tsang et al., 2017), Yangtze Estuary (G. Peng et al., 2017), Bohai Sea, and the Yellow Sea (Zhao et al., 2018). The finest particles (<0.1 mm) represented the majority of the MPs in the estuarine sediment (Fig. S10a). White/transparent were the most abundant color, followed by red and blue (Fig. S10b). Morphologically, fibers were the most prevalent, followed by fragments, which was somewhat different from those in the river bed sediment and river water. The prevalence of the fibrous MPs may be sourced from lost/discarded fishing gears considering active fishing activities in the estuary. Spherical and membranous MPs were also observed (Fig. S10c). These results were basically consistent with those in the sediment of Yangtze Estuary, China (G. Peng et al., 2017).

Nine polymers were identified, including PP, PE, PET, PP-PE, PAN, PVA, ALKYD, PS, and PA (Fig. 2), which agreed well with those detected in coastal sediment of Hong Kong (Tsang et al., 2017) and beach sand of the Bohai Sea (Yu et al., 2016). In the sediment of Yangtze Estuary, however, the most abundant MP polymers were rayon, polyester, and acrylic, which were suggested to be derived from laundry wastewater (G. Peng et al., 2017). Specifically by morphology, fibrous MPs in the estuarine sediment were mainly PP, PET and PAN, which was similar to that in the river water and river bed sediment (Fig. 3b). However, the percentages of PET and polymers of higher density (e.g., PAN and PVA) were relatively higher in the estuarine sediment than in the river bed sediment and river water (Fig. 2).

The MPs abundances in the sedimentary core ranged from 140 to 820 items kg^{-1}, showing an upward increase (Fig. 6a). As mentioned above, production and use of plastic products keep increasing globally since their commercial development (PlasticsEurope, 2018). Our result suggested that accumulation of the MPs in the estuarine increase in line with the plastic production and uses. However, it's interesting to note that the percentage of fine MPs (<0.45 mm) showed obviously downward increase, reaching 100% at the depth of 42 cm and there down (Fig. 6b). These results suggested persistence and potential downward dispersion of the MPs in the sediment, which could be of concerns considering detrimental effects of the MPs on benthic and sediment-ingesting organisms.

The MPs in the core were most fragments, fibers, and spherules

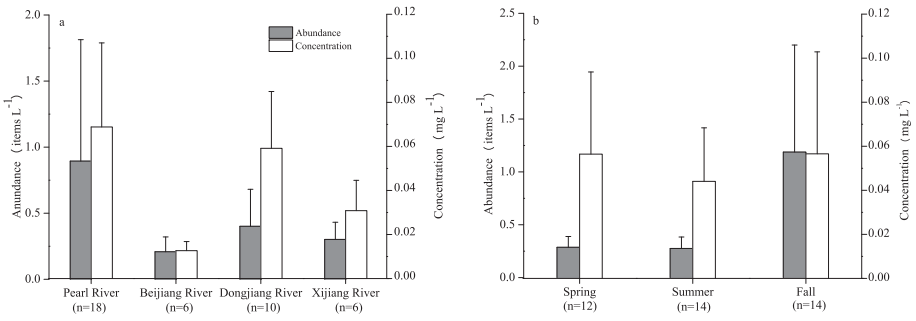

Fig. 4. Spatial (a) and seasonal (b) patterns of the microplastics abundance and concentration in the river water of the Pearl River catchment.

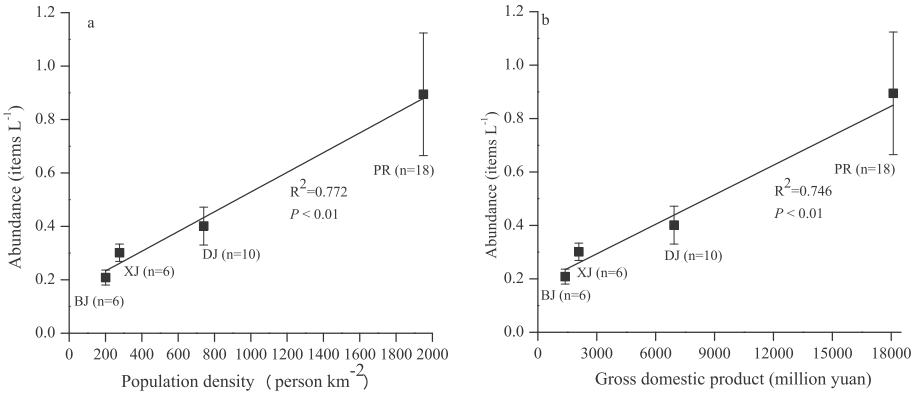

Fig. 5. Correlations between the microplastics abundance in the river water and population density (a) and gross domestic product (b). Data about population density and gross domestic product were obtained via http://www.gdstats.gov.cn/tjzl/tjgb/.

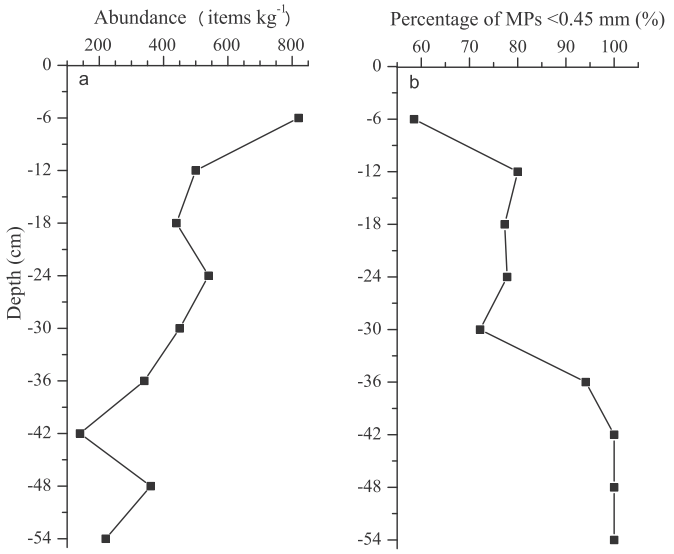

Fig. 6. Vertical distribution of the microplastics in the sedimentary core of the Pearl River Estuary. (a) microplastics abundance, (b) percentage of the microplastics <0.45 mm.

(Fig. S11a). Like in the riverine and estuarine surface sediment, the MPs were predominated white/transparent (Fig. S11b). Polymers of the MPs in the sedimentary core were mainly PP, PE, PP, and PET. PS and PC were also detected (Fig. 2). Specifically, the spherical MPs were comprised of PE, PP, and PS while the fragmental MPs comprised of PP and PE. The fibrous MPs were mainly made up of PET and PP (Fig. 3).

As one of the most important and busiest shipping channels in the world, spills of preproduction pellets during transport are likely happen in the PRE. In addition, treated wastewater from the Pearl River Delta is generally discharged into PR and its tributaries and finally reaches the PRE, which may also cause overtime accumulation of plastic microbeads contained in various detergents and personal care products in the PRE. However, conclusions need to be made with cautiousness considering limited data about the MPs in wastewaters as well as in the sediment.

4. Conclusions

Distribution and characterization of the MPs were revealed in the water and sediment of the Pearl River catchment, China. The MPs abundances and concentrations in the river water were 0.57 ± 0.71 items L^{-1} and 0.051 ± 0.036 mg L^{-1}, respectively. Based on this result, an estimated 15963 ± 11268 tons of MPs would be annually transported to the South China Sea via PR and its tributaries. The MPs were widely present in the riverine and estuarine sediment. In terms of polymer composition and morphology, PP and PE fragments, sheets and fibers were predominant MPs in the PR catchment. Furthermore, polymers of higher density s were relatively more abundant in the sediment than in the river water, especially in the estuarine sediment. The MPs abundances in the river water were obviously lower in wet season than in dry season, probably due to dilution effect of the precipitation. Population density and economic development showed significant impacts on the MPs contamination. The vertical distribution of the MPs in the PRE demonstrated upward increase, indicating growing release of plastic wastes in this area. In addition, percentage of the MPs <0.45 mm increased with increasing depth of the sediment core, suggesting persistence and possible downward dispersion of the MPs.

Acknowledgements

This work was financially supported by National Natural Science Foundation of China (No. 41572330 and No. 41877365) and SKLOG2016-A04. The authors have no conflicts of interest to declare. This is contribution No.IS-2700 from GIGCAS.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2019.05.056.

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