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Microplastics in mangrove sediments of the Pearl River Estuary, South China: Correlation with halogenated flame retardants' levels

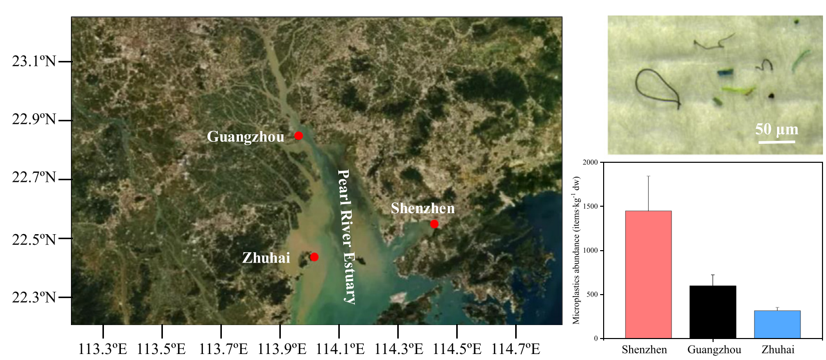
Linzi Zuo [a], Yuxin Sun [a,b,*], Hengxiang Li [a,b], Yongxia Hu [a,e], Lang Lin [a,d], Jinping Peng [c], Xiangrong Xu [a,b,*]

[a] *CAS Key Laboratory of Tropical Marine Bio-resources and Ecology, Guangdong Provincial Key Laboratory of Applied Marine Biology, South China Sea Institute of Oceanology, Chinese Academy of Sciences, Guangzhou 510301, China*
[b] *Innovation Academy of South China Sea Ecology and Environmental Engineering, Chinese Academy of Sciences, Guangzhou 510301, China*
[c] *Faculty of Chemical Engineering and Light Industry, Guangdong University of Technology, Guangzhou 510006, China*
[d] *University of Chinese Academy of Sciences, Beijing 100049, China*
[e] *School of Environment and Guangdong Key Laboratory of Environmental Pollution and Health, Jinan University, Guangzhou 510632, China*

HIGHLIGHTS

- Microplastics were reported in mangrove sediments from the PRE.
- Mangrove sediments had a relatively higher abundance of microplastics.
- Microplastic abundance had potential positive correlations with population density and GDP.
- PBDEs, DBDPE, BTBPE and HBCDD may have same pollution sources with microplastics.

GRAPHICAL ABSTRACT



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ABSTRACT

Marine microplastic pollution of intertidal mangrove ecosystem is a matter of concern. However, the relationship between microplastic distribution and other pollutants such as halogenated flame retardants (HFRs) is unknown. In this study, forty-eight sediment samples were collected from three mangrove wetlands of the Pearl River Estuary (PRE), South China to investigate the distribution of microplastic and discuss the possible relationship between HFRs and microplastic abundance in mangrove sediments. The abundance of microplastic in mangrove sediments from the PRE ranged from 100 to 7900 items$\cdot$kg^{-1} dry weight (dw), with an average of 851 $\pm$ 177 items$\cdot$kg^{-1} dw, which was at a relatively higher level compared to other regions worldwide. The highest abundance of microplastic was observed in Shenzhen mangrove sediments. The abundance of microplastic was significantly and positively correlated with population density and gross domestic product of the PRE. The microplastics with size <500 μm were predominant in mangrove sediments, accounting for a proportion of 69.4% in all microplastic samples. Polypropylene-polyethylene copolymer, green/black, and fibers/fragments were the dominant type, color and shape in all microplastic samples, respectively. The correlation between HFRs and microplastic abundance demonstrated that polybrominated diphenyl ethers, decabromodiphenyl ethane, 1,2-bis(2,4,6-tribromophenoxy)ethane and hexabromocyclododecane may have the same pollution source as microplastics.

* Corresponding authors at: CAS Key Laboratory of Tropical Marine Bio-resources and Ecology, Guangdong Provincial Key Laboratory of Applied Marine Biology, South China Sea Institute of Oceanology, Chinese Academy of Sciences, Guangzhou 510301, China.
E-mail addresses: sunyx@scsio.ac.cn (Y. Sun), xuxr@scsio.ac.cn (X. Xu).

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1. Introduction

Microplastics are small plastic particles <5 mm in size (Thompson et al., 2004). The global production of plastics in 2018 was 359 million tonnes, of which approximately 30% of plastics was produced in China (Statista, 2018). Microplastics have been widely found in water, sediment and biota samples (Ivar do Sul and Costa, 2014; Van Cauwenberghe et al., 2015; Auta et al., 2017; Rezania et al., 2018). Microplastics are always associated with various organic pollutants, including plastic additives in the environment (Schrank et al., 2019; Fred-Ahmadu et al., 2020). For example, halogenated flame retardants (HFRs) are used as additives to endow flame resistance to plastics. These HFRs can be released into the environment because most of them are directly doped into plastics without chemical bonds (Choi et al., 2009; Khaled et al., 2018; Sun et al., 2019). When microplastics are mistakenly ingested as food by organisms, HFRs can accumulate in the biota, even transfer into the food chains and bring potential risks to organisms or humans (Wang et al., 2018).

Mangroves occur in the intertidal zones of tropical and subtropical coastlines and are the most productive forest ecosystems worldwide, providing food and habitat to marine and terrestrial organisms. Known for their plentiful detritus and abundant organic carbon, mangrove wetlands can act as important sinks for varieties of contaminants (Zhu et al., 2014; Wu et al., 2016). Nor and Obbard (2014) firstly reported microplastics in Singapore mangrove wetlands with an abundance of 63 items kg^{-1}. In China, a comprehensive study in mangrove wetlands along the south-eastern coastal zones revealed that microplastic pollution had a strong spatial heterogeneity in abundance and characterization, mainly caused by difference in pollution sources. In the Beibu Gulf of China, abundant microplastics existed in mangrove wetlands of the Qinzhou Bay, and fragments of white polystyrene from local mariculture were the predominant microplastics (Li et al., 2018b; Li et al., 2019). Mangrove wetlands have well-developed root systems and abundant organic matter, which result in large numbers of microplastic being retained in mangrove ecosystems (Li et al., 2019; Zhou et al., 2019; Li et al., 2020). In our previous studies, various pollutants including HFRs have been found in the mangrove wetlands (Zhang et al., 2015; Hu et al., 2019; Zhang et al., 2019). However, the relationship between microplastic distribution and these pollutants remains unknown.

The Pearl River Estuary (PRE), one of China's largest subtropical estuaries, is situated in the Pearl River Delta (PRD), South China. It is a highly urbanized and economically dynamic area. Rapid urbanization and industrialization of the PRD region resulted in large numbers of contaminants including microplastics to accumulate in the PRE. It was estimated that about 1.09–2.31 × 10^4 t of plastic wastes have been discharged through the PRE to the South China Sea each year (Lebreton et al., 2017). Hong Kong, located on the east of the PRE, is a hotspot of marine plastic debris (Cheung et al., 2016). Mangrove wetlands distributed in the PRE, acting as buffers for estuaries and barriers against land-based contaminants, are believed to suffer severe microplastic pollution. However, information on microplastic pollution, as well as their relationship with the presence of HFRs in mangrove wetlands in the PRE, is extremely sparse.

In this study, microplastic pollution in mangrove sediments was investigated from three mangrove wetlands in the PRE. The objectives of this study were to (1) investigate the abundance and spatial distribution of microplastics in the PRE; (2) explore the characterization of microplastics in mangrove sediments; (3) discuss the possible relationship between microplastic pollution and HFRs, whose distribution in these three mangrove sediments has been reported in our previous study (Hu et al., 2019).

2. Materials and methods

2.1. Sampling area and sediments collection

Forty-eight sediment samples were collected in November 2015 in the PRE of Guangdong Province, South China. The three sampling sites included Futian Mangrove Nature Reserve in Futian district of Shenzhen City ($n = 21$), Tantou Mangrove Nature Reserve in Nansha district of Guangzhou City ($n = 13$), and Qi'ao Island Mangrove Nature Reserve in Xiangzhou district of Zhuhai City ($n = 14$) (Fig. 1). The details of these three sampling sites are given in Table S1. A stainless steel shovel sampler was used to take out the top 5 cm sediments and 1 kg of wet sediment samples was stored in an aluminum container, sent back to the laboratory and kept in cold storage (−20 °C) until further treatment.

2.2. Microplastics extraction

Sediment samples were dried at 40 °C in glass petri dishes for 72 h. To prevent airborne contamination, the petri dishes were covered with aluminum foil throughout the whole experiment process. Branches, leaves and rocks in the sediment samples were removed before drying. Organic carbon in sediments was removed by wet peroxide

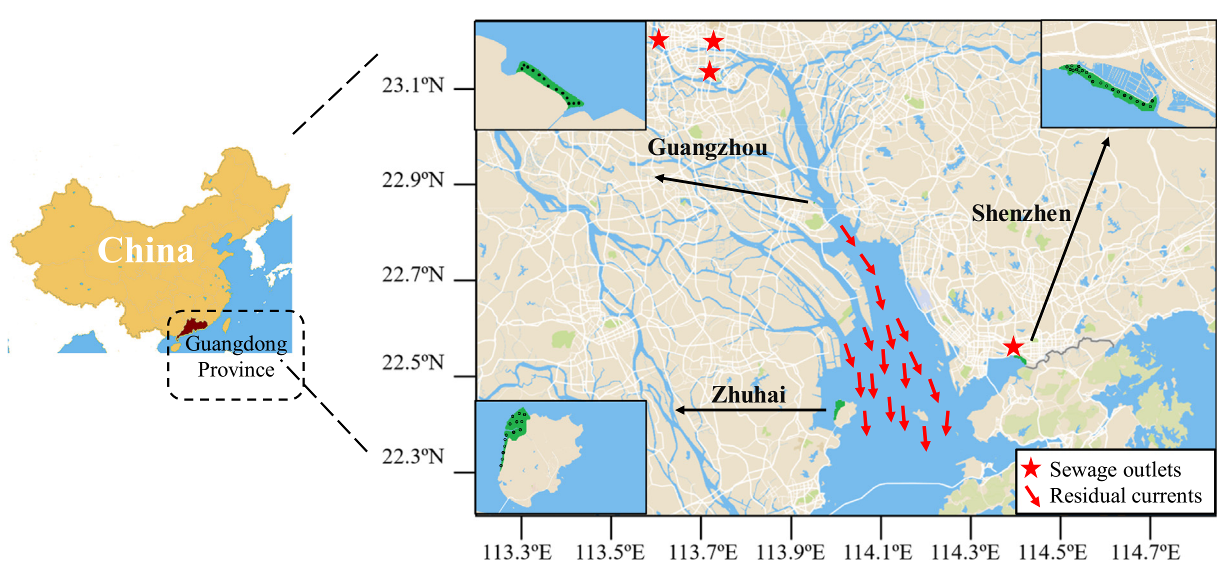

Fig. 1. Map of sampling sites.

oxidation before the extraction of microplastics. Briefly, 80 mL 0.05 M $FeSO_4$ and 20 mL of 30% H_2O_2 were added to a 250 mL Erlenmeyer flask with 10 g oven-dried sediments. The sample was heated in a water bath to 70 °C for 30 min. The mixture was then transferred into a 1 L beaker and 500 mL saturated NaCl (1.2 g·L^{-1}) solution was added. Samples were then stirred continuously with a glass rod and allowed to settle overnight for density separation. Vacuum filtration was applied to the supernatant with a 1.2 μm pore size using a 47 mm diameter glass microfiber filter (GF/C, Whatman, UK). The filtered microplastics were transferred to a clean petri dish with a clean glass microfiber filter and covered with aluminum foil prior to further analysis.

2.3. Morphology and identification of microplastics

Microplastics were identified using a stereo microscope (Olympus SZX10, Tokyo, Japan). Subsequently, a digital camera (Olympus DP80, Tokyo, Japan) was used to take photos of microplastics on the filter membrane. Microplastics were photographically assessed by their outer morphological and physical characteristics including count, size, shape and color as described in detail in Nor and Obbard (2014). In this study, the abundance of microplastics was expressed as the number of particles per kilogram of dried sediments (items·kg^{-1} dw). The software of Image J (1.46r, National Institutes of Health, USA) was adapted to measure the size of microplastics in photographic images. Microplastics were classified into four types based on their shape: fiber, fragment, pellet and sheet and divided into six categories based on color: transparent, black, blue, green, brown and pink.

Microplastic particles were identified by a Micro Fourier Transform Infrared Spectrometer (μ-FTIR) (Thermo Scientific Nicolet iN10, U.S.A). The mercury-cadmium-telluride detector of μ-FTIR spectrometer was cooled with liquid nitrogen and analysis was conducted in transmittance mode. Microplastics were transferred to a microscope slide with a needle prior to analyses. For each particle analyzed, the infrared spectrum between 4000 cm^{-1} and 400 cm^{-1} was recorded by three times, with a spectral resolution setting at 8 cm^{-1}. Because the spatial resolution was 5 μm, particles having a diameter exceeding 5 μm could be identified in this study. The difference between recorded spectra and the standard FTIR spectrum databases were compared using OMNIC software (Thermo Fisher, USA). The polymer types of microplastics were verified when the similarity between microplastics and standard exceeded 80%.

2.4. Quality assurance and quality control (QA/QC)

To prevent microplastic contamination in the experiment process, all reagents, including distilled water, saturated NaCl, H_2O_2 and $FeSO_4$ solution, were filtered through a 0.7 μm glass fiber filter (GF/F, Whatman) before use. Aluminum foil was used to cover the containers before and after use. For Shenzhen, Guangzhou and Zhuhai samples, three procedural blanks were performed simultaneously in the experimental process to measure background contamination. Microplastics were not detected in the procedural blank samples. Microplastics were extracted five times based on a preliminary experiment and the presence of microplastics in solution was rare after five extractions.

2.5. Statistical analysis

Statistical analysis was performed with SPSS 17.0 (SPSS Inc., Illinois, USA) and p value below 0.05 was taken to be statistically significant. Differences between microplastic abundance and size among the three mangrove wetlands were discriminated by one-way analyses of variance (ANOVA). Pearson correlation analysis was conducted to explore the relationship between microplastic abundance and population density and gross domestic product (GDP), and the relationship between HFRs and microplastic pollution of mangrove sediments.

3. Results and discussion

3.1. Abundance of microplastics in mangrove sediments of the PRE

The abundance of microplastics in mangrove sediments of the PRE is presented in Table 1. The microplastic abundance varied between 100 and 7900 items·kg^{-1} dw, with an average value of 851 ± 177 items·kg^{-1} dw. The microplastic abundance in this study was one or two orders of magnitude higher than those in mangrove sediments from Singapore (12–63 items·kg^{-1} dw) (Nor and Obbard, 2014), and slightly greater than those in mangrove sediments from Qinzhou Bay, South China (15–2310 items·kg^{-1} dw) (Li et al., 2018a, 2018b; Li et al., 2019). Zhou et al. (2019) reported that the mean microplastic abundance of Shenzhen and Zhuhai mangrove sediments in the PRE were 157 and 141 items·kg^{-1} dw, respectively, which were much lower than the present study. This could be attributed to the use of different microplastic extraction methods and different microfiber filters.

The comparison of microplastic abundance in different sediments is listed in Table 1. Microplastic pollution levels in mangrove sediments of the PRE were more severe than those in coastal areas from Europe and America, including Baltiysk Strait of Russia (34 items·kg^{-1} dw), Biscay Bay of France (67 items·kg^{-1} dw), national parks of USA (43–443 items·kg^{-1} dw) and the Isle of Rügen in Germany (40–140 items·kg^{-1} dw) (Zobkov and Esiukova, 2017; Hengstmann et al., 2018; Phuong et al., 2018; Yu et al., 2018) (Table 1). Microplastic abundance in this study was in the same range as those in the western (100–900 items·kg^{-1} dw) and eastern (2433 ± 2000 items·kg^{-1} dw)

Table 1
Comparison of microplastics abundance in sediments with other studies.

Location	Microplastic abundance (items·kg^{-1} dw)			Reference
	Range	Median	Mean	
Mangroves of PRE in China	100–7900	567	851	This study
Shenzhen and Zhuhai Mangroves of PRE in China			149	Zhou et al. (2019)
Mangroves in Qinzhou Bay, China	15–2310			Li et al., 2018b; Li et al. (2019)
Mangrove in Singapore	12–63			Nor and Obbard (2014)
Sanggou Bay, China			2178	Wang et al. (2019)
Xiangshan Bay, China			1739	Chen et al. (2018)
Guangzhou City, China	80–9597			Lin et al. (2018)
Changjiang Estuary, China	20–340		121	Peng et al. (2017)
North Yellow Sea, China			37	Zhu et al. (2018)
Baltiysk Strait, Russia			34	Zobkov and Esiukova (2017)
Bay of Biscay, France			67	Phuong et al. (2018)
National Parks in the USA	43–443			Yu et al. (2018)
The Isle of Rügen, Germany	40–140			Hengstmann et al. (2018)
Western Mediterranean, Spain	100–900			Alomar et al. (2016)
Eastern Mediterranean Basin, Lebanese			2433 ± 2000	Kazour et al. (2019)

Mediterranean, respectively, which is the largest semi-enclosed sea in the world (Alomar et al., 2016; Kazour et al., 2019). Compared with the sediments of coastal areas of China, microplastic abundance in this study was much higher than Changjiang Estuary (20–340 items·kg^{-1} dw) and North Yellow Sea (37 items·kg^{-1} dw) (Peng et al., 2017; Zhu et al., 2018). Only Sanggou Bay (mean, 2178 items·kg^{-1} dw) and Xiangshan Bay (mean, 1739 items·kg^{-1} dw) had higher microplastic abundance than this study (Chen et al., 2018; Wang et al., 2019). Higher microplastic abundance (80–9597 items·kg^{-1} dw) was recorded in the sediments from the Pearl River along Guangzhou City, which is located on the upstream of the PRE (Lin et al., 2018). Extremely high microplastic abundance was also found in water from the Pearl River along Guangzhou City (mean, 19,860 item·m^{-3}) and the PRE (mean, 8902 item·m^{-3}), South China (Yan et al., 2019). A combination of these results with our study indicates that the PRE has become a hotspot for microplastic pollution. The PRD region had a high population (76.6 million people in 2015) and a prosperous economy (gross domestic product with 6.8 trillion RMB in 2015) (Guangdong Statistics Bureau, 2016). Large numbers of microplastics may be released into the PRE because of its large population and rapid economic development of the PRD. Therefore, a high abundance of microplastics was found in mangrove sediments of the PRE.

Significant differences for microplastic abundance were found in sediments among the three mangrove wetlands ($F = 4.70$, $p = 0.014$) in the PRE (Fig. 2). Microplastic abundance from Shenzhen mangrove sediments was significantly higher than those in Guangzhou ($p = 0.039$) and Zhuhai ($p = 0.006$). One reason for this observation was that Shenzhen has a higher population density than those of Guangzhou and Zhuhai (Supplementary Material, Table S2). Shenzhen also has a higher GDP than Guangzhou and Zhuhai (Table S2) and the Futian mangrove wetland in Shenzhen is the only natural reserve region located in an urban area in China. Therefore, a high abundance of microplastics was expected in Shenzhen mangrove sediments. The median and mean values of microplastic abundance in mangrove sediments of Guangzhou (500 and 597 ± 124 items·kg^{-1} dw) were higher than those of Zhuhai (249 and 316 ± 38 items·kg^{-1} dw), but no significant difference was found between these two mangrove wetlands ($p = 0.509$). This may be because mangrove wetlands in Guangzhou and Zhuhai are both located in a suburban district, and they have similar population density and GDP (Table S2). To further verify the relationship between microplastic pollution and anthropogenic activities, we calculated the correlation between microplastic abundance and social-economic indices based on Li et al. (2020) and the present study. The microplastic abundance in mangrove sediments was positively correlated with population density ($r = 0.961$, $p = 0.003$) and GDP ($r = 0.908$, $p = 0.033$), suggesting that microplastic pollution in the PRE was related to the anthropogenic activities. Similar results were also found in estuarine rivers of Chesapeake Bay, USA and estuaries in southeast China (Yonkos et al., 2014; Tang et al., 2018).

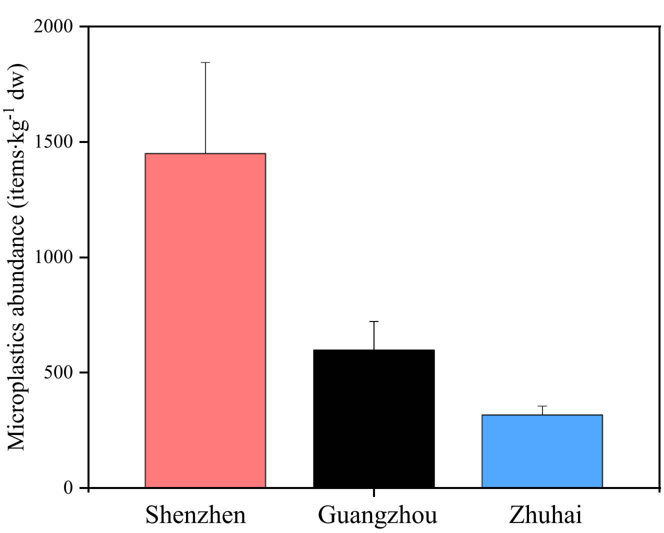

Fig. 2. Microplastic abundance in mangrove sediments of the PRE, South China.

3.2. Characterization of microplastics in mangrove sediments of the PRE

The features of microplastics in mangrove sediments of the PRE were presented in Fig. 3. The average sizes of microplastics in Shenzhen, Guangzhou and Zhuhai mangrove sediments were 452 ± 33, 811 ± 95 and 777 ± 84 μm, respectively. For Shenzhen mangrove sediments, the range of sizes of microplastics were dominated by 50–100 μm (30.0%) and 100–200 μm (29.9%), followed by 200–500 μm (18.5%) and 500–2000 μm (18.5%), with smaller contributions from <50 μm (8.0%) and >2000 μm (4.2%) (Fig. 3a). Microplastics in Guangzhou and Zhuhai mangrove sediments had similar size distributions. The dominant microplastic size class was 500–2000 μm (38.6% and 39.8%), followed by 100–200 μm (21.6% and 20.5%) and 200–500 μm (23.9% and 19.3%). There were significant differences in microplastic sizes between Shenzhen, Guangzhou and Zhuhai mangrove ($p < 0.001$). Shenzhen mangrove sediments contained more microplastics with size smaller than 500 μm (77.3%) than those of Guangzhou (52.3%) and Zhuhai (50.6%). The Shenzhen mangrove wetland is located in a semi-closed bay. Consequently, smaller particles were more difficult to remove by water flow (Yan et al., 2019). Sediments from all three mangrove wetlands in the PRE had a high proportion of microplastics with size smaller than 500 μm. The high proportion of small microplastics could promote the negative effects on organisms in mangrove wetlands because microplastics between 125 and 500 μm were easily captured, for example, by two benthic bivalve organisms (*Ennucula tenuis* and *Abra nitida*) and caused severe effects on these two species (Bour et al., 2018).

Colors of microplastics in mangrove sediments are shown in Fig. 2b. Black and green were the dominant colors of microplastics in mangrove sediments of the PRE and accounted for 28.3% and 43.1% for Shenzhen, 35.2% and 27.0% for Guangzhou, 47.6% and 21.1% for Zhuhai, respectively. Colored and transparent microplastics in mangrove sediments of the PRE had average proportions of 88.0% and 12.0%, respectively. Compared with transparent microplastics, colored microplastics are more easily captured by marine biota. For example, blue microplastics were preferentially ingested by Amberstripe scads (*Decapterus muroadsi*) (Ory et al., 2017).

Four shapes including fiber, fragment, pellet and sheet in mangrove sediments were identified in the present study (Fig. 3c). The distribution of different shapes across three mangrove wetlands was similar. Fibers were the dominant shapes of microplastics, which contributed >69.7% to all shapes of microplastics, followed by fragments (27.8–28.9%), with smaller contributions of pellets (0–1.2%) and sheets (0–1.2%). Fibers were also the dominant microplastics in mangrove sediments of Singapore, non-mangrove sediments from Changjiang Estuary and Pearl River along Guangzhou City of China (Nor and Obbard, 2014; Peng et al., 2017; Lin et al., 2018). The shape distribution of 48 sampling sites showed pellet and sheet microplastics only appeared in two sites of Shenzhen and two sites of Guangzhou mangrove wetlands (Fig. S1), suggesting an infrequent occurrence of pellet and sheet microplastics in the PRE mangrove wetlands.

The FTIR results showed microplastics in mangrove sediments included polypropylene-polyethylene copolymer (PP&PE), polyethylene terephthalate (PET), polystyrene (PS), low-density polyethylene (LDPE) and cellophane (Fig. 4). PP&PE was the predominant polymer, with contributions of 77.5%, 68.8% and 56.4% for Shenzhen, Guangzhou and Zhuhai mangrove sediments, respectively (Fig. 3d). PE (36%) and PP (21%) are the largest two groups in plastic production globally, and their copolymers are also commonly used in packaging, textiles and fishing gears (Geyer et al., 2017; Cai et al., 2018). Consequently, PP&PE are widely detected in the marine environment.

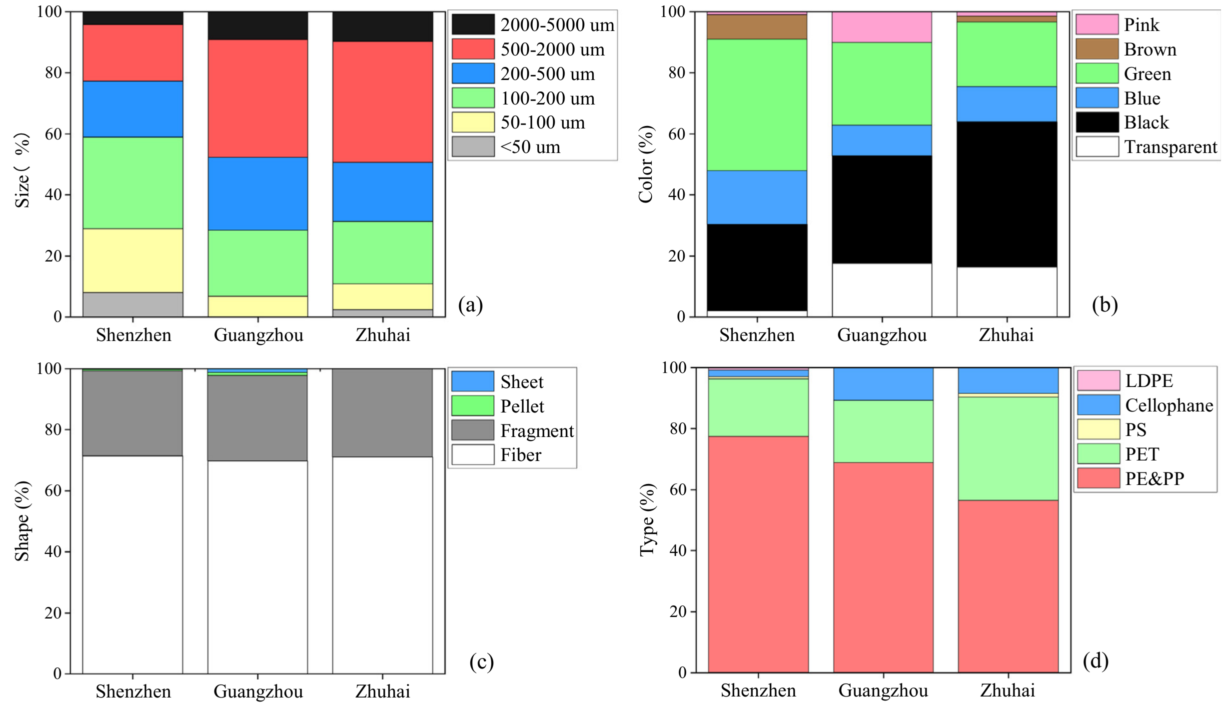

Fig. 3. Characterization of microplastics in mangrove sediments of the PRE.

The characterization of microplastics showed the color, shape and polymer types of microplastics in the three mangrove wetlands were similar. Thus, these microplastics might be from a similar source. Pellet microplastics are mainly generated from industrial sources as raw material. The low frequency detection of pellet microplastics indicated that industrial emission was not the major source. In our previous study, we simultaneously investigated microplastic pollution in the Pearl River along Guangzhou City and in wastewater treatment plants (WWTPs) and found that WWTPs were an important source of microplastic pollution in the Pearl River and fibers were the dominant shape in the Pearl River along Guangzhou City, as influents and effluents of WWTPs (Lin et al., 2018). Thus, wastewater effluents from upstream might be a source of microplastics in this study (Fig. 1). Prevailing weather and currents played significant roles in the transportation of microplastics. We collected the sediment samples in winter and typhoons and rainstorms generally occur in summer in the PRE. However, typhoons and rainstorms would tend to increase the diversity of microplastic types (Lo et al., 2020). Sporadic events seem to have little effect on microplastic transportation in this study. Residual currents are tightly connected to the transportation of suspended matter. Zheng et al. (2019) found that large quantities of microplastics were detected in areas with residual currents. In this study, Guangzhou and Zhuhai mangrove wetlands are located in areas having residual currents (Fig. 1) (Zhou et al., 2012; Zheng et al., 2014). However, the highest microplastic abundance was found in the Shenzhen mangrove wetland. This may be because the Shenzhen mangrove wetland is close to a sewage outlet (Fig. 1). Firstly, the diffusion of water in the bay near Shenzhen mangrove wetland mainly depends on tidal currents. The bay represents a bottle-neck, topographically, and the water of the inner bay exchanges minimally with the open sea. After transportation by currents, the low energy of mangrove wetlands enhanced the potential for microplastic retention in sediments as turbulence and bottom currents subsided (Ballent et al., 2016). Meanwhile, the introduced microplastics could be trapped by mangrove pneumatophores and prop roots, and then deposited (Huang et al., 2020). Finally, biofouling and adsorption to natural substances in mangrove wetlands would jointly promote the deposition of microplastics (Ballent et al., 2016).

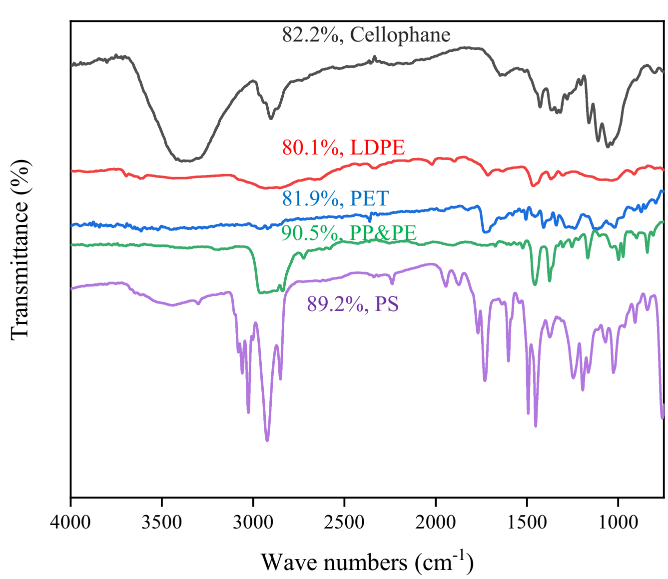

Fig. 4. The FTIR spectrum of selected microplastics and the match degrees with the standard spectrum.

3.3. The correlation between microplastic and HFRs in mangrove sediments of the PRE

The presence of polybrominated diphenyl ethers (PBDEs), decabromodiphenyl ethane (DBDPE), 1,2-bis(2,4,6-tribromophenoxy)ethane (BTBPE), tetrabromobisphenol A (TBBPA), hexabromocyclododecane (HBCDD) and dechlorane plus (DP) in mangrove sediments of the PRE were reported in our previous work (Hu et al., 2019). To calculate the correlation between microplastic and HFRs, microplastic abundance (items·kg^{-1} dw) should be converted into microplastic concentration (μg kg^{-1} dw). The average sizes of microplastics in mangrove sediments from Shenzhen, Guangzhou and Zhuhai were 452, 811 and 777 μm, respectively, and we assumed all microplastics were PP&PE fibers (average diameter, 30 μm). The density of PP&PE was 0.89–0.95 $g \cdot cm^{-3}$, and we assumed the density of microplastics in this study was 0.92 $g \cdot cm^{-3}$. Thus, the average weights of microplastic particles in sediments of Shenzhen, Guangzhou and Zhuhai mangrove were 0.35, 0.62 and 0.60 μg. The value of microplastic concentration ($\mu g \cdot kg^{-1}$ dw) was multiplied by microplastic abundance ($items \cdot kg^{-1}$ dw) and average microplastic weight. The correlation analysis showed that the concentrations of PBDEs ($r = 0.537$, $p = 0.000$), DBDPE ($r = 0.300$, $p = 0.046$), BTBPE ($r = 0.544$, $p = 0.000$) and HBCDD ($r = 0.538$, $p = 0.000$) were positively correlated with microplastic concentration in mangrove sediments. But this was insufficient to prove that microplastics in mangrove wetlands were the source of these four HFRs. We subsequently found the contents of PBDEs, DBDPE, BTBPE and HBCDD in plastics as previously reported in the literature (Table S3). The content of PBDEs in plastic did not exceed 7.9%. Thus, the average theoretical concentrations of PBDEs were 40.1, 29.2 and 15.0 μg/kg dw in Shenzhen, Guangzhou and Zhuhai because we assumed all PBDEs in microplastics were released (Table S4). However, the average environmental concentration of PBDEs in Shenzhen, Guangzhou and Zhuhai were 72.2, 61.2 and 20.3 $\mu g \cdot kg^{-1}$ dw (Table S4). Similarly, it was demonstrated that the environmental concentration of DBDPE and HBCDD were also higher than their theoretical concentrations (Table S4). This revealed that PBDEs, DBDPE and HBCDD in the PRE mangrove sediments were not released from microplastics, but their positive correlation suggested that these compounds may have the same source or exhibit similar environmental behavior to microplastics. For example, PBDEs, DBDPE, HBCDD and microplastics might come from the degradation products of plastic debris. For BTBPE, their environmental concentrations were 0.35, 0.19 and 0.10 $\mu g \cdot kg^{-1}$ dw in Shenzhen, Guangzhou and Zhuhai, and their theoretical concentrations were 3.04, 2.24 and 1.14 $\mu g \cdot kg^{-1}$ dw. Thus, BTBPE in mangrove sediments of the PRE was possibly derived from microplastics. However, more evidence is needed for this hypothesis.

No significant correlations were found between microplastic concentration and levels of DP ($r = 0.286$, $p = 0.057$) and TBBPA ($r = 0.232$, $p = 0.126$) in mangrove sediments. DP was mainly used as additives into electrical hard plastic connectors (Sverko et al., 2011). Therefore, the major sources of DP might be e-waste. The major sources of TBBPA are TBBPA-based product manufacturing, and e-waste recycling. The half-life of TBBPA is short and its concentrations decreased with increasing distance from point sources (Malkoske et al., 2016). Choi et al. (2009) found that the release of TBBPA from plastics pellets reached an equilibrium state within one day. Thus, TBBPA in mangrove wetlands were not from the same pollution source as microplastics.

4. Conclusions

This study investigated the occurrence and distribution of microplastics in mangrove sediments from the PRE, South China. The microplastic abundance in mangrove sediments of the PRE was higher than comparable regions worldwide. Microplastic abundance in mangrove sediments was significantly correlated with population density and GDP. The predominant size, color, type and shape of microplastics in mangrove sediments were smaller than 500 μm, polypropylene-polyethylene copolymer, green/black, and fibers/fragments, respectively. Significant relationships between concentrations of PBDEs, DBDPE, BTBPE, and HBCDD and microplastic in mangrove sediments indicated that they might originate from same source in the environment. The effect of microplastics on mangrove biota should be a great concern in the future.

Author statement

L. Zuo, Y. Sun, Y. Hu, and X. Xu co-designed the experiment. Y. Sun, and Y. Hu sampled the sediments. L. Zuo, Y. Sun, H. Li, L. Lin and J. Peng coanalyzed the microplastics in mangrove sediments. L. Zuo and Y. Sun analyzed the data. All authors contributed to the interpretation of results and wrote the manuscript.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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