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Microplastics in sediments from an interconnected river-estuary region

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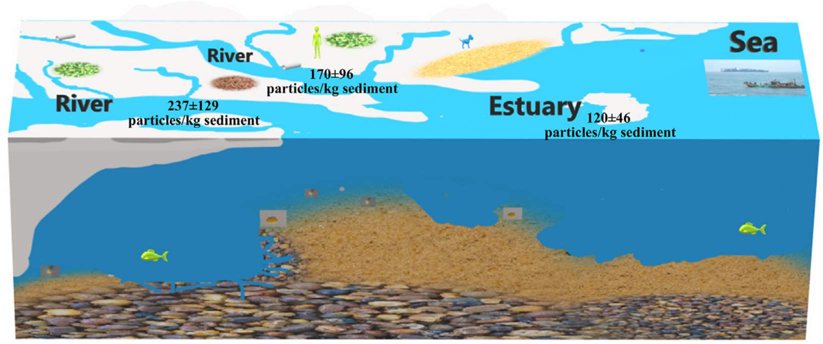
HIGHLIGHTS

- Higher abundance of microplastics was detected in sediments from river than estuary.
- The highest concentration of microplastics was detected at the mouth of river.
- 8 polymers were the most popular microplastics detected in all sediments.
- PE, PE/PP and PET accounted for >50% microplastics detected in sediments.
- Microplastic shapes composition was followed by the orders of film, fragment, fiber, and pellet.

GRAPHICAL ABSTRACT



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ABSTRACT

Microplastics are an emerging pollutant of international concern due to its wide distribution through various pathways. Estuary is an important pathway for land microplastics to enter into the oceans by rivers. In this study, we hypothesized that microplastics would sink into estuary sediment during diffusion and transmission from river before entering into the sea, which results in higher accumulation of microplastics in proximity to river-estuary than in the oceans. In order to demonstrate this hypothesis, sediment samples were collected from an estuary and its two main inputting rivers and the microplastics in these samples were analyzed. In the collected sediment samples, 19 types of polymers, including the three most common polymers (polyethylene, polyethylene terephthalate, and poly(propylene:ethylene)), were identified and confirmed by FT-IR. Eight types of polymers were consistently detected in all samples, while 11 types of polymers were occasionally found in some samples. These microplastics exhibited four shapes and their percentages followed the high-to-low order of film, fragment, fiber and pellet. A relatively lower abundance of microplastics was found in river sediments from Shuangtaizi River with an average of 170 ± 96 particles/kg d.w., compared to that from Daliao River with an average of 237 ± 129 particles/kg d.w., but it was higher than that from Liaohe Estuary with an average of 120 ± 46 particles/kg d.w. Furthermore, the highest concentration of microplastics was found at the mouth of

Abbreviations: ABS, Poly (Acrylonitrile:Butadiene:Styrene); AEM, Ethylene acrylic elastomer; CPA, Chlorinated polyalkene; EPDM, Poly(ethylene:propylene:diene); EPR, Poly (propylene:ethylene); EVA, Ethylene-vinyl acetate copolymer; MF, Melamine-formaldehyde resin; PA, Polyamide (Nylon); PAA-co-PAN, Poly (acrylonitrile:acrylic acid); PAL, Poly(acrylonitrile); PE, Polyethylene; PET, Polyethylene terephthalate; PDMS, Poly (dimethylsiloxane); PP, Polypropylene; PS, Polystyrene; PTFE, Poly tetra fluoroethylene; PVC, Polyvinyl Chloride; PU, Polyurethane; SBS, Poly (styrene-butadiene-styrene).

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rivers, showing high accumulation where the freshwater and saltwater meet. Results from this study, including the abundance, characteristics and spatial distribution of microplastic pollution in sediments from an interconnected river-estuary system, revealed the fate and distribution of microplastics in the river and estuary environment.

1. Introduction

Marine pollution caused by plastic litter and microplastics has become an international concern due to their various adverse impacts. For instance, exposure to microplastics decreased the survival of fish and aquatic invertebrate (Foley et al., 2018), and the potential human health risk rose after ingesting microplastics-polluted seafood and salts (van Cauwenberghe and Janssen, 2014; Yang et al., 2015). Moreover, it is estimated that microplastics pollution costs the aquaculture sector approximately $0.32 million–$503 million in the UK in one year (van der Meulen et al., 2014). Available data from global monitoring programs and field investigations showed that microplastics had already spread from inland to polar region, even to the ocean's deepest area of Mariana trench (Peng et al., 2018; Rochman, 2018). Studies showed that the microplastic accumulation appeared to increase continuously in freshwater and marine environment due to the fast increase of global plastic production and consumption (Bergmann et al., 2015; Cózar et al., 2014; UNEP, 2016). However, the sources and transporting pathways of microplastics and their spatial distribution from rivers to the oceans have not been studied systematically. Therefore, it is difficult to perform a comprehensive assessment of the fate of microplastics in the environment.

Presently, it is estimated that up to 80% of marine plastic litters are from land-based sources due to the mismanagement of solid wastes and wastewater worldwide (Law, 2017), and rivers are believed to be the critical pathway for plastic litters and microplastics to enter into the oceans (Barbuzano, 2019). For instance, a report estimated that 530–1500 tons of plastic wastes were discharged annually into the Black Sea through Danube River, while 20–31 tons were discharged into the North Sea from Rhine River (Van der Wal et al., 2015). Another study using a global river plastic input model showed that 1.15–2.41 million tons of plastic wastes entered the oceans through rivers globally per year, especially more than two thirds of the wastes were transported through the top 20 polluted rivers (Lebreton et al., 2017). Mai et al. (2019) also estimated that about 66 tons of microplastics were discharge into South China Sea through Pearl River Delta. Plastic litters and microplastics would sink from the surface water to water column and then gradually form sediments due to environmental factors and biofouling during the progression from rivers to the oceans (Eo et al., 2019; Kaiser et al., 2017; Kooi et al., 2017). A direct scientific evidence from a field investigation had indicated that the microplastic abundance in deep-sea sediments was significantly higher than that in the surface waters (Woodall et al., 2014). Similarly, the microplastics floating on the river surface could also fall into sediment continuously (Di and Wang, 2018), and the upper layer of sediment was the first depositing location for these microplastics (Willis et al., 2017). Furthermore, studies showed that the microplastics in sediments did not show consistent pattern with that in surface waters. For example, compared with low-density microplastics, such as PP an PE, the high density microplastics, such as PS, showed higher presence in surface sediment at Three Gorges Reservoir (Di and Wang, 2018). Alam et al. (2019) also reported that the amounts of microplastics in sediments showed a significant difference among sampling segments, but no difference was found for the amounts of microplastics in water. Hurley et al. (2018) reported that approximately 30% of the microplastics on river beds remained after a period of severe flooding, suggesting that the microplastics in sediment might be a better indicator to track the historical and current microplastic pollution than those in water.

Estuary is the confluence of freshwater from rivers and saltwater from the oceans, where suspended particles and contaminants from rivers quickly deposit into the sediment under the co-influence of hydrological and sediment transport dynamics (Chapman and Wang, 2001; Yao et al., 2016). Consequently, estuarine sediments are the highly concerned location of microplastic deposition. For example, it was reported that microplastics are accumulated in the estuary sediment with the continuous inflow of river freshwater containing microplastics (Simon-Sánchez et al., 2019), which could play a critical role in the transporting fluxes of microplastics into the oceans. Since each estuary has its own geographical, physicochemical and biological characteristics, including the historic and ongoing sediment contamination, more studies are needed to obtain data for the prediction of microplastic emission from rivers into the oceans through estuary. However, limited researches have been carried out on the microplastics in river sediments and estuary sediments and on their direct comparison (Firdaus et al., 2020; Simon-Sánchez et al., 2019), which are critical to unveil the fates of microplastics from rivers to the oceans.

In this study, we hypothesized that the estuarine sediments are the depositing reservoir of microplastics from the freshwater, and the microplastics are accumulated in estuary sediments when freshwater and saltwater meet. To demonstrate this hypothesis, the abundance and distribution characteristics of microplastics in sediments from an estuary and its two main contributing rivers were investigated. The results showed that the microplastic abundance was lower in estuary sediments than that in river sediments, and the highest concentration of microplastics was detected at the mouth of rivers. Therefore, rivers were not only the transportation highway for microplastics into the oceans but also a temporary repository for microplastic emission before entering into the oceans.

2. Materials and methods

2.1. Sampling sites

Liaohe estuary (LE), which is located at the farthest northern coastline of China, is an important ecological and economic region in northeastern China. It receives the discharge of several rivers, including Daliao River (DR) and Shuangtaizi River (SR) principally, which flow through many cities and rural areas. Liaohe estuary Wetland, which is the LR Delta Wetland and formed from the confluence of DR and SR, is also the largest bulrush wetland in the world and a crucial habitat for many wildlife, especially for some endangered species such as spotted seal (*Phoca largha*) and red-crowned crane (*Grus japonensis*).

In this study, 29 sampling sites from LE and the downstream of DR and SR were selected to investigate the microplastic characteristics and evaluate the transporting profile of microplastics from river to sea. Among these sampling sites, 11 sites were located in LE and distributed around the two river estuaries, 9 sites were along the downstream of DR (DR1 in the mouth), and 9 sites were along the downstream of SR (SR1 in the mouth) (Fig. 1).

2.2. Sampling and treatment

In August, surface sediments from estuary and the center of river were sampled with a steel grab sampler, and they were directly placed into glass containers on site. The samples were transported back to the lab and stored at −20 °C until further analysis. During sampling, the

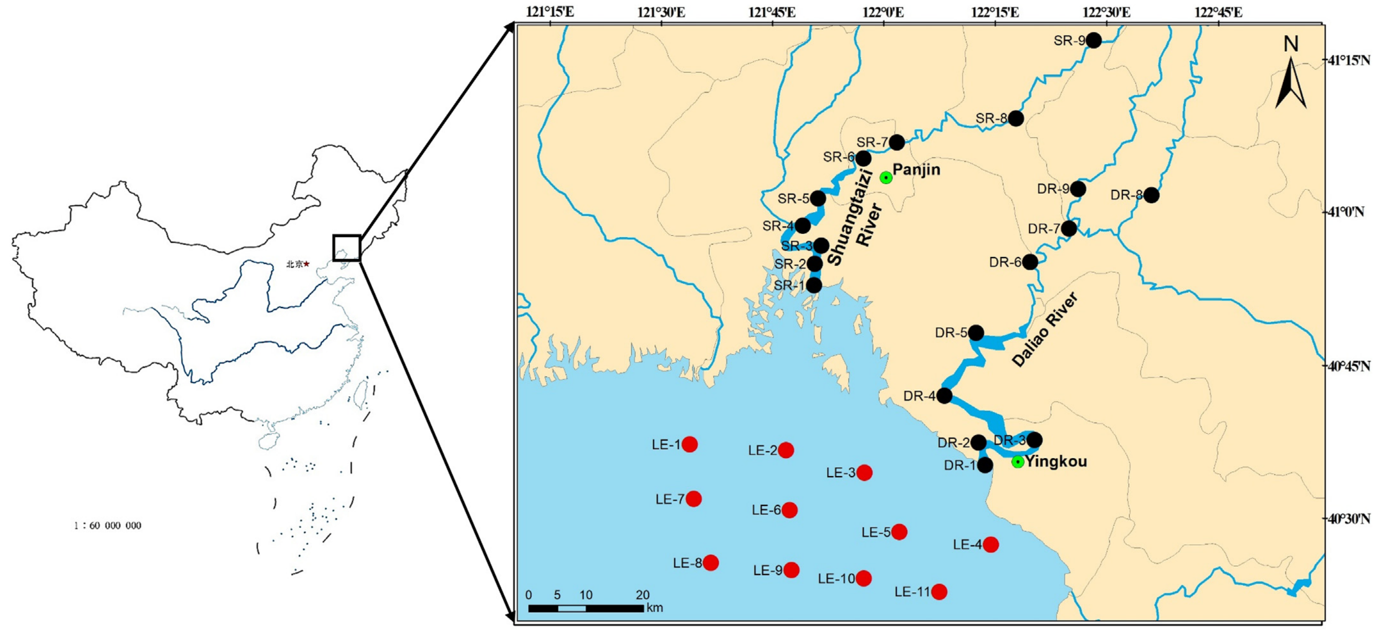

Fig. 1. Sampling sites for microplastic investigation in Shuangtaizi River (SR) and Daliao River (LR), and their confluence of Liaohe Estuary (LE) in northern China. The black dots indicate the sampling sites in rivers, the red dots indicate the sampling sites in estuary, and the green dots indicate the local main cities. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

tools were cleaned with filtered water (pore size = 5 μm), and plastic tools such as rope and net were avoided.

After dried at 50 °C in an incubator and homogenized with a metal spoon, 30–50 g well-mixed sediments were used in the isolation of microplastics following the method described by Xu et al. (2020). In the present study, $ZnCl_2$ solution was used to isolate microplastics in sediments because it allowed the extraction of microplastics with relative high densities, such as PVC (1.14–1.56 g/mL) and PET (1.32–1.41 g/mL). Moreover, it is also cheaper than the other high density solutions such as sodium iodide and sodium polytungstate. Typically, the sediment and $ZnCL_2$ solution (1.5 g/cm^3) were poured into a container and stirred for 30 min, followed by 30 min of sonication. After standing for 6 h without disruption, the surface solution with floating particles was collected by the addition of excessive $ZnCl_2$ solution. Subsequently, the collected microplastics were filtered through a 5 μm stainless steel membrane. This isolation procedure was repeated three times, and all particles were collected on the steel membrane. Afterwards, the membrane along with all particles was immersed in 30% H_2O_2 to remove these natural organic particles. After 72 h of incubation at 50 °C, the residue was filtered through another 5 μm stainless steel membrane before further analysis.

2.3. Microplastic identification

After drying at 50 °C in an incubator, all suspected microparticles on the membrane were picked carefully under an OLYMPUS SZ2 optical stereomicroscope (Shanghai Fulai Optical Technology Co. Ltd., China) with a total magnification of 10–40. These particles were further separated into four groups: pellet, fiber, film and fragment (Please find the Supplementary materials). Particles with sizes of >50 μm were measured individually and imaged with a Leica DM4M digital microscope (Germany).

The Elmer Spotlight 400 Fourier transform infrared spectroscopy (FT-IR) microscopy system (PerkinElmer, USA) equipped with a Specac Golden Gate attenuated total reflectance accessory was used to identify the polymers. All selected microparticles were scanned individually and repeated in triplicate in the spectral range of 4000–650 cm^{-1} with a 4 cm^{-1} resolution and 32 co-added scans. After noise and baseline modification, FT-IR spectrum of each particle was automatically searched against the Bio-Rad KnowItAll® Informatics System 2018 (64-bit)-IR Spectral Library database (Bio-Rad Laboratories, USA) and matched to corresponding polymer with a correlation algorithm (KnowItAll, Bio-Rad). And the particles with mapping over 70% are considered as microplastics. Lastly, the number of microplastics was recalculated after these non-plastic particles were subtracted from the total number of suspected particles.

2.4. Quality control

To eliminate possible contamination, quality control measures had been strictly implemented throughout the sampling in the field and analysis in the lab. For example, all containers such as glass bottles must be washed three times with filtered water, and all samples were freshly prepared and filtered before use. A stainless steel membrane with 5 μm pore size was used instead of organic filter membrane during the analysis process. Three blank samples with purified water were also investigated to monitor the contamination from the analysis process, and no microplastic particles were found in blank samples except for a few fibers which were deducted from the total number of microplastics, suggesting the contamination from background could be ignored.

2.5. Data analysis

The SPSS software 13.0 (SPSS Inc., Chicago, USA) was used in data analysis. To detect the differences among sites along SR, DR and LE, statistical analysis was carried out with One-way ANOVA, followed by the One-sample *t*-test of the independent sample. It is regarded as significantly different when p-value is lower than 0.05.

3. Results and discussion

3.1. Polymer composition of microplastics in sediments from rivers and estuary

Based on FT-IR analysis of particles (Fig. S1), a total of 19 polymers had been identified with >70% matching with the standard, including 8 polymers which were detected in all sites and accounted for nearly 80% of total microplastics, and 11 polymers which were found in some

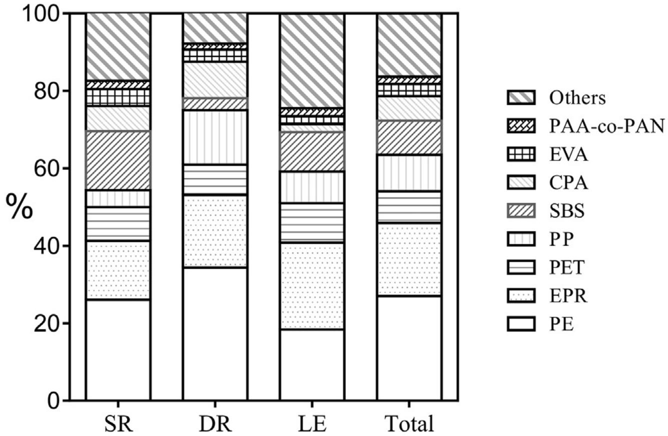

Fig. 2. Percentage of polymers detected in sediment from Shuangtaizi River (SR), Daliao River (DR) and Liaohe Estuary (LE). Others include ABS, AEM, EPDM, MF, PA, PAL, PDMS, PS, PTFE, PU, and PVC.

sites and accounted for about 20% of the total number of microplastics (Fig. 2).

Overall, PE, accounting for about 36% of global non-fiber plastics (Geyer et al., 2017), was the most popular polymer among these identified microplastics. PE was also detected in sediments from Bohai Sea, Yellow Sea, East China Sea (Zhao et al., 2018; Zhang et al., 2019), and even in a heavily urbanized catchment (Tibbetts et al., 2018). Generally, PE was dominant among microplastics detected in surface water for its density is <1.0 g/mL (Koelmans et al., 2019). For example, Zhang et al. (2017) reported that PE microplastics accounted for 51% of the total microplastics on the surface waters of Bohai Sea. However, these floating PE microplastics could sink into sediment after long transmission and diffusion due to attachment with other particles or biomass (Meng et al., 2020). Another study (Zhu et al., 2020) stated that the relative long persistence and enrichment of PE in environment is related with its weaker photodegradability compared with other polymers. The second popular polymer detected in sediments was PE/PP (polyethylene-polypropylene rubber) that was also the most commonly detected polymer in deep-sea sediments from the Western Pacific Ocean (Zhang et al., 2020). PE/PP plastics might come from the weathering and broken of large plastic used in industrial and fishing activities, including the land-based and sea-based sources. Similarly, as one of the most popular polymers in sediments, PET was found in sediments from Bohai Sea, Yellow Sea and other sites globally (Horton et al., 2017; Yao et al., 2019; Zhao et al., 2018). Polyethylene terephthalate is a clear, strong and lightweight polymer, and it is used as a common material for packaging such as bottles and containers, and also widely used for fibers or fabrics. Our investigation found that discarded PET bottles, single-use cups and textile debris were

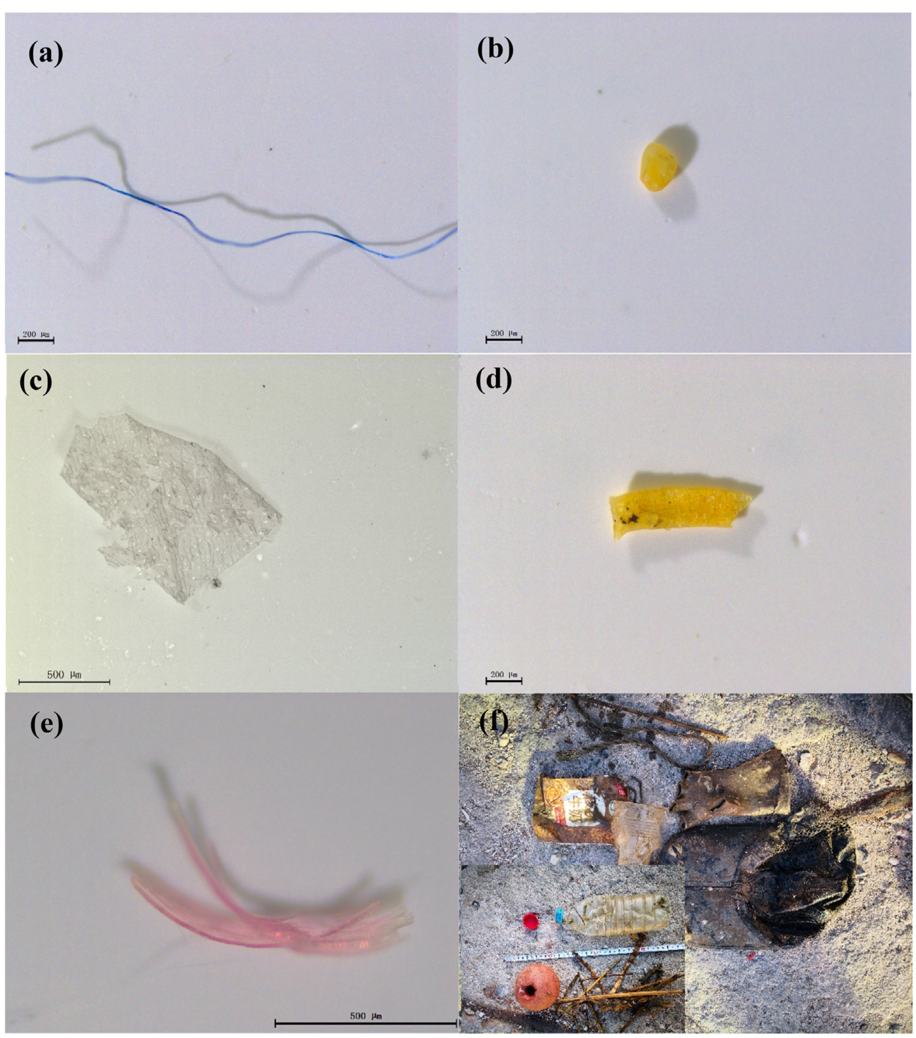

Fig. 3. Representative microplastics detected in sediments from Shuangtaizi River, Daliao River, and Liaohe Estuary. (a): Fiber; (b): Pellet; (c): Film; (d): Fragment; (e) Film; (f). Plastic debris.

widely found in river and estuarine during sampling (Fig. 3(f)). These would be the potential sources of PET microplastics in the environment.

In addition to other polymers detected in sediments, rayon, a type of semi-synthetic fibers used for fashion and industrial applications for its desirable properties of high durability, high resistance, lightweight and low-cost, was detected in all sites with a relative high abundance (22.73%–40.00%). Rayon plastics were also reported widely in sediment (Woodall et al., 2014), water (Lusher et al., 2015) and biota (Li et al., 2018; Lusher et al., 2013) previously. However, rayon is produced from natural wood pulp or cotton and don't belong to synthetic polymer. Therefore, the numbers of rayon were not counted as fossil-based polymer microplastics in this study. The harmful effects caused by microplastics exposure are most dependent on their abundance; thus, the abundance of microplastics should be assessed based on the harmonized definition of microplastics, to avoid the overestimation of their risk in the future.

3.2. Shape of microplastics in sediments from rivers and estuary

As shown in Fig. 3, four different shapes of microplastics were found in all samples, similar with the shapes of microplastics in sediment from Bohai Sea (Zhao et al., 2018). The film shape which was mainly made of PE was found to have the most high abundance in all sediment samples (29/29) from SR, DR and LE, followed by the fragment (27/29) and fiber (26/29), and pellet was only detected at 51.72% of sampling sites (15/29) (Fig. 4(a)–(c)). This distribution pattern was inconsistent with the microplastics in sediment from Bohai and Yellow Sea where fiber is the most common type (Zhao et al., 2018), and also different from that of Lagoon of Venice where only irregular fragments were observed (Vianello et al., 2013) with limited fragments detected in estuaries (Peng et al., 2017, Alves and Figueiredo, 2019). These results indicated that the shape profile of microplastics in sediments from different regions might be related with their sources of microplastics in sediment, that is, the microplastic pollution in LE was depend mainly on the microplastics from DR and SR inputs.

Generally, synthetic textiles from domestic washing are regarded as important sources of microplastics in environment (Hernandez et al., 2017), and the microfibers dominate the microplastic pollution in sediments, such as the highly eutrophic tropical estuary (Alves and Figueiredo, 2019), Tampa Bay (McEachern et al., 2019) and Jagir Estuary (Firdaus et al., 2020). According to the appearance characteristics of microfibers detected in sediments with lengths at 1000 μm in the present study (Fig. 3(b)), it could conclude that some microfibers are probably shed from synthetic textile (Fig. 4(a)), especially in sediments from rivers of DR and SR receiving a large amount of domestic wastewater input. However, some microfibers in sediments, especially from estuary where there are high fishing activities, might come from the breakdown or degradation of plastic rope and fishing net because these microfibers looked like the pieces of rope (Fig. 4(e)). Just like the source of microfibers, fragment, film and pellet microplastics are also likely generated from large pieces of plastics. Based on the appearance of fragment and film microplastics, plastic bags, plastic packaging and farming film collected during the investigation (Fig. 3(f)) might be the microplastic sources, because these plastic products are easily broken down to pieces in environment after natural weathering and physical stress (Teuten et al., 2007; Baldwin et al., 2016; Wang et al., 2016). Similarly, pellet microplastics might be derived from the breakdown of plastic products such as container, cup and fishing tools. In addition, microbeads widely used in personal care products were not detected in sediments from both rivers and estuary in the present investigation, implying that the usage of cosmetic products containing microbeads are limited in China, and the personal care products might not be a critical source of microplastics in environment.

3.3. Abundance of microplastics in sediments from rivers and estuary

According to the FT-IR spectra of particles isolated from sediments, some particles were removed which could not be identified due to be the lack of obviously identifiable peaks, or a spectrum was present but was not recognized in reference database. Overall, 32.65% of the suspected particles collected from all sampling sites were plastic particles verified by μFT-IR, and the number of microplastics was recalculated to express their abundance. The abundance of microplastics in sediments from SR ranged from 67 particles/kg d.w. to 300 particles/kg d.w. with an average of 170 ± 96 particles/kg d.w. A similar level of microplastics was in sediments from DR, ranging from 100 particles/kg d.w. to 467 particles/kg d.w. with an average of 237 ± 129 particles/kg d.w. ($p > 0.05$) (Fig. 5(a), (b)).At the same time, there was a significant difference in the abundance of microplastics per sampling site in SR and DR ($p < 0.05$). Compared with data reported previously, the microplastic abundance in SR and DR was similar to that in Ottawa River (Vermaire et al., 2017), Brisbane River (He et al., 2020) and Beijiang River (Wang et al.,2017b), while it was lower than that in Wei River (Ding et al., 2019), Xiangshan Bay

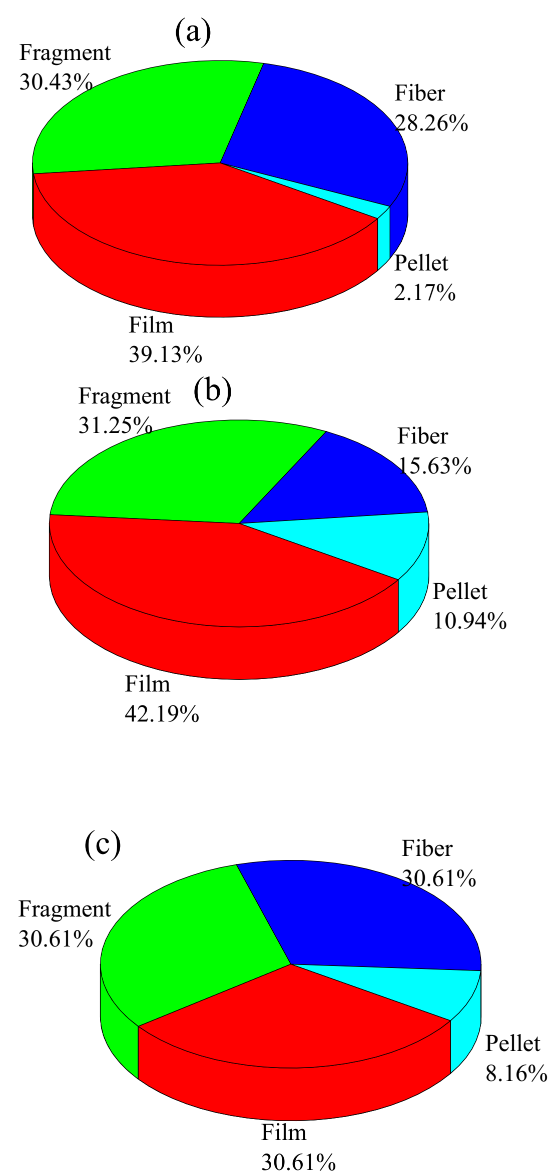

Fig. 4. Percentages of microplastic shapes in sediment from (a) Shuangtaizi River, (b) Daliao River, and (c) Liaohe Estuary.

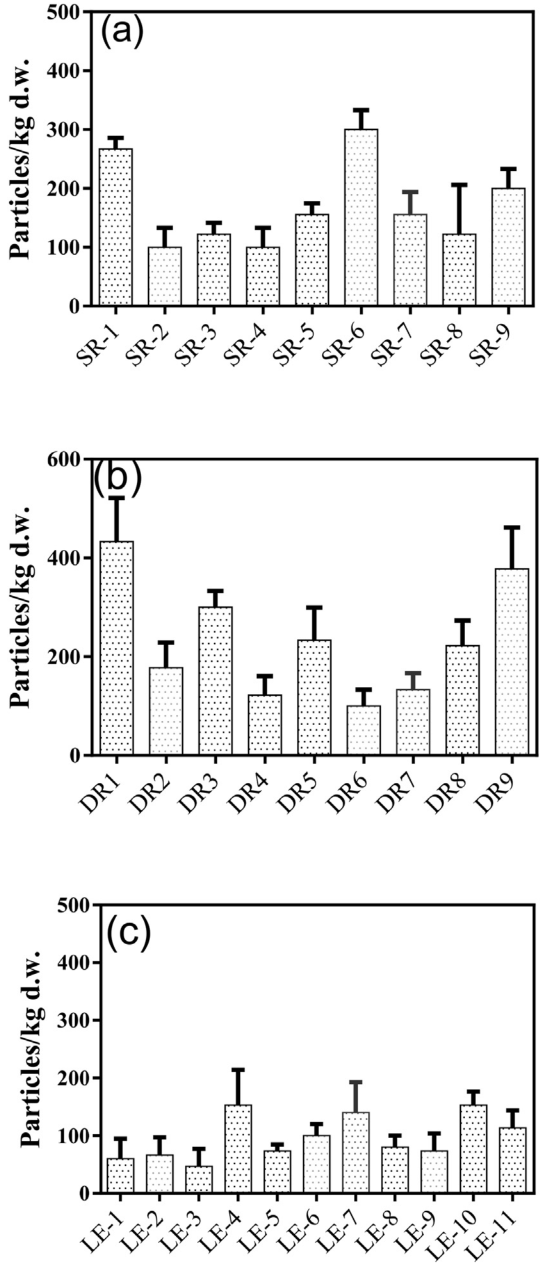

Fig. 5. Abundance of microplastics in sediment from (a). Shuangtaizi River (SR), (b). Daliao River (DR), and (c). Liaohe Estuary (LE) in northern China.

(Chen et al., 2018), Atoyac River basin (Shruti et al., 2018), Rhine and Main (Klein et al., 2015), Rhine and Meuse rivers (Leslie et al., 2017), Pearl River (Lin et al., 2018) and Ebro River (Simon-Sánchez et al., 2019). However, it was higher than that in Yongfeng River (Rao et al., 2019) and Ciwalengke River (Alam et al., 2019) (Table 1). Although there was no consistent increasing or decreasing trend of microplastic abundance along the two rivers (SR and DR), the highest microplastic concentration in each river was found at the mouth of rivers (DR1 and SR1). The same result was also found in Jagir River where the microplastic abundance in sediment was increased toward the mouth (Firdaus et al., 2020). These results indicated that the increased deposition of microplastics occurs at the mouth of rivers because various factors such as turbulence and salinity promote microplastics to sink when freshwater and saltwater meet.

Except for the highest value found at the mouth of rivers, the relatively high microplastic abundance at SR6 and DR3 around the centers of local cities, suggesting that the municipal activities might also affect the abundance of microplastics in environment. Relatively high values were also found in the upstream of rivers (SR9 and DR9) without concentrated industrial activities or sewage treatment, but with many farmlands and some villages, indicating other sources of microplastics, such as the sewage sludge as fertilizer in farmlands, might exist although we did not identify the sources. Furthermore, the abundance of microplastics in river sediment is dependent on some factors, including the microplastic characteristic, river channel and flow dynamics. For example, more microplastics would sink into sediments when water flow becomes slower, resulting in the increase of microplastic concentration in sediments (Kapp and Yeatman, 2018). There are some tributaries to mainstream which might be the important sources of microplastics (Klein et al., 2015), but we did not investigate them in the present study.

In all sampling sites from LE, the average abundance of microplastics was 120 $\pm$ 46 particles/kg d.w., ranging from 80 particles/kg d.w. to 220 particles/kg d.w. (Fig. 5(c)), which was higher than that in the sediments from the central region in Bohai Sea (124.8 $\pm$ 33.2 items/kg d.w.) (Zhao et al., 2018). Compared with that reported in literatures, the microplastic abundance in LE was similar with that in Changjiang Estuary (Peng et al., 2017) and Haihe Estuary (Wu et al., 2019), while lower than that in Jagir Estuary (Firdaus et al., 2020), Maowei Bay (Li et al., 2019), Guanabara Bay (Alves and Figueiredo, 2019), North-East Atlantic (Maes et al., 2017) and Derwent Estuary (Willis et al., 2017), but higher than that in Yondingxinhe Estuary (Wu et al., 2019) and Warnow Estuary (Enders et al., 2019) (Table 1). If the 11 sampling sites were divided into three sections based on the distance to the shoreline, as LE1-LE4, LE5-LE7 and LE8-LE11, the microplastics in the three regions showed similar abundance with no significant difference ($p > 0.05$). The lack of difference in distribution also indicated that these microplastics spread with tidal current and then accumulated in sediments widely, which might be related with the regional geographical feature with a gentle beach slope and width of 3–4 km.

Additionally, the available data are limited to compare the present results with the literature reports, although many investigations on microplastic pollution had been performed in global freshwater and marine environment. This is mainly due to the differences in the concentration units of microplastics in sediments, which were expressed by mass of mg/kg (He et al., 2020) and particles/kg (Wu et al., 2019; Zhao et al., 2018), volume of particles/m^3 (Di and Wang, 2018), area of particles/m^2 (Wessel et al., 2016; Young and Elliot, 2016) and items/cm^2 (van Cauwenberghe et al., 2013). In order to have a global overview of microplastic pollution, the accessible data should be normalized as a critical step, especially a harmonized methodology including sampling design and data report format should also be recommended to establish a regional and national monitoring strategy.

3.4. Microplastics from river and estuary

Our data analysis indicated that the average abundance of microplastics detected in sediments from LE region was significantly lower than that in DR ($p < 0.05$), while slight lower than that in SR ($p > 0.05$). However, it was also significantly lower than those in SR ($p < 0.05$) if the rayon particles were considered as microplastics. These results agreed with the preliminary conclusion that the microplastic pollution in freshwater is more serious than that in marine environment (Hamid et al., 2018). And it also indicated that the majority microplastics retained in river sediment but not flow into the oceans in rainfall season, and the river sediment is the temporary or permanent reservoir for the accumulation of microplastics although no consistent trend was observed among the sampling sites. Meanwhile, the microplastic profile in sediment, including the shape type and polymer composition, showed a similar pattern among the regions of SR, DR and LE (Fig. 2), implying that the microplastic pollution in estuary was mainly affected by river inputs.

Table 1
Abundances of microplastics in sediments collected from global rivers and estuaries.

Environmental compartment	Abundance (particles/kg d.w.)	Location	Reference
River	100–467 (mean: 237 ± 129)	Daliao River, China	Present study
	133–300 (mean: 170 ± 96)	Shuangtaizi River, China	
	5–72	Yongfeng River, China	Rao et al., 2019
	30.3 ± 15.9	Ciwalengke River, Indonesia	Alam et al., 2019
	220	Ottawa River, Canada	Vermaire et al., 2017
	10–520	Brisbane River, Australia	He et al., 2020
	178 ± 69–544 ± 107	Beijiang River, China	Wang et al., 2017a, 2017b
	660 (1–4 mm)	River Thames Basin, UK	Horton et al., 2017
	802 ± 594	urban rivers, China	Peng et al., 2018
	360–1320	Wei River, China	Ding et al., 2019
	1739 ± 2153	Xiangshan Bay, China	Chen et al., 2018
	833.33 ± 80.79–1633.34 ± 202.56	Atoyac River basin, Mexico	Shruti et al., 2018
	672–2175	Rhine and Main, Germany	Klein et al., 2015
	1400–4900	Rhine and Meuse rivers, Netherlands	Leslie et al., 2017
	80–9597 (mean:1669)	Pearl River, China	Lin et al., 2018
	2052 ± 746	Ebro River	Simon-Sánchez et al., 2019
Estuary	80–220 (mean: 120 ± 46)	Liaohe Estuary, China	Present study
	85.0 ± 40.1	Yondingxinhe Estuary, China	Wu et al., 2019
	46–100	Warnow Estuary, Germany	Enders et al., 2019
	20–340 (mean: 121 ± 9)	Changjiang Estuary, China	Peng et al., 2017
	216.1 ± 92.1	Haihe Estuary, China	Wu et al., 2019
	92–590	Jagir Estuary, Indonesia	Firdaus et al., 2020
	520 ± 8–940 ± 17	Maowei bay, China	Li et al., 2019
	160–1000	Guanabara Bay, Brazil	Alves and Figueiredo, 2019
	54–3146 (mean: 421)	North-East Atlantic, BE, UK, NL, FR	Maes et al., 2017
	2430–4200	Derwent Estuary, Australia	Willis et al., 2017

It also found that the microplastic pollution level was related to the distance to urban areas, and the abundances of microplastics in urban areas were significantly higher than that in rural region (Di and Wang, 2018; Wang et al., 2017a) because the population density is also a critical factor affecting the microplastic distribution (Blumenröder et al., 2017). In the present study, the selected SR flows mainly through the Panjin city where the population is about 1.5 million, while DR flows through the Yingkou city where the population is about 2.4 million. And the Panjin city locates on the upstream of SR, while Yingkou city locates on the downstream of DR. These data indicate the population size and the distance of city location have direct impacts on the abundance of microplastics in river due to the mismanagement wastewater and plastic litter. For example, numbers of fossil-based and natural-based fibers which were mainly made of PET and rayon for textiles were detected in sediments from both rivers and estuary, indicating a large amount of domestic wastewater is discharged into rivers directly or indirectly. Therefore, the solutions to address the issue of microplastics should mainly focus on the prevention of land-based sources. For instance, the domestic wastewater should be collected and then treated in sewage treatment plant before discharging (Napper and Thompson, 2016). The abandoned agricultural plastic products, agricultural farming and aquaculture activities along the two rivers should be controlled. In addition, these fragment microplastics discharged by marine fishing, tourism and other maritime activities should also not be overlooked.

4. Conclusions

The present study detected four types of microplastics in sediments from river and estuary, consisting of 19 polymers with PE dominating the microplastic composition. The abundance of microplastics was higher in sediments from rivers than that in the estuary, and the highest abundance was detected at the mouth of rivers reaching Bohai sea, implying that the hydrological dynamics in estuary promote the deposition of microplastics into sediment. Moreover, the microplastic characteristics in sediments in both rivers and estuary showed a similar profile, including the shape and polymer composition. These results suggested that rivers are not only the pathway of microplastics emission from land to the oceans, but also the reservoir for microplastic accumulation before reaching the oceans. Out findings reveal the transportation and fate of microplastics once entering rivers before reaching the oceans.

CRediT authorship contribution statement

Qiujin Xu: Investigation, Writing - original draft.**Ronglian Xing:**Data curation, Writing - original draft.**Mingdong Sun:**Visualization, Software.**Yiyao Gao:**Investigation, Validation.**Lihui An:**Writing - review & editing.

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

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2020.139025.

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