Chemosphere 280 (2021) 130800

Contents lists available at ScienceDirect

Chemosphere

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

Vertical microplastic distribution in sediments of Fuhe River estuary to Baiyangdian Wetland in Northern China

Zeyan Zhou [a], Panyue Zhang [a,*], Guangming Zhang [b], Siqi Wang [a], Yajing Cai [a], Hongjie Wang [c]

[a] College of Environmental Science and Engineering, Beijing Forestry University, Beijing, 100083, China
[b] School of Energy and Environmental Engineering, Hebei University of Technology, Tianjin, 300000, China
[c] Xiong'an Institute of Eco-Environment, Hebei University, Baoding, 071002, China

A R T I C L E I N F O

Handling Editor: Derek Muir

Keywords:
River estuary
Microplastic
Sediment
Vertical distribution

A B S T R A C T

Microplastics exist widely in water environment. The microplastic distribution in sediments can better reflect the long-term microplastic pollution, especially the vertical distribution. However, the vertical microplastic distribution in sediments is diverse and unclear. This paper is the first study on vertical microplastic distribution in estuary sediment of Fuhe River, the main upstream river flowing to Baiyangdian Wetland in the northern China. The typical feature of Fuhe River is that the effluent of municipal wastewater treatment plants is its main water source. Microplastics in 15 sampling sites and different depths (0–50 cm) were examined. Results showed that the microplastic content decreased with the increase of sediment depth, and the highest content was 1049 ± 462 items/kg in the topmost sediment layer (0–5 cm). The particle size of microplastics was smaller in deeper sediment layers. The proportion of colored microplastics in deeper sediment layers was larger than that in shallower layers. Polyethylene (PE) and polypropylene (PP) were the main plastic polymer types in all sediment samples. The spatial distribution characteristics of microplastics in sediments were closely related to human activities, and the microplastic content was higher in the areas with more intense human activities. This study is helpful to understand the detailed distribution characteristics of microplastics in typical rivers in the northern China, and can provide guidance for reducing microplastic pollution.

1. Introduction

With the increasing demand for plastics in everyday life, industrial and agricultural production, the development of plastic industry has been obviously accelerated (Lee et al., 2015). As a result, more than 18% of plastic wastes were send to landfill in 2018, and more and more plastic wastes have also been accumulating in the environment (PlasticsEurope, 2019). It was estimated that by 2050 there would be roughly 12,000 metric tons of plastic wastes entering in landfill or natural environment (Geyer et al., 2017). Lee et al. (2015) pointed out that the plastic pollution has become one of the highly serious global environmental problems like climate change (Shen et al., 2020).

When plastic wastes enter into environment, the microplastics smaller than 5 mm in size may pose a potential risk to health of aquatic organisms and human beings through food chains (Thompson, 2004; Tang et al., 2020). Recently, microplastics have been extensively studied, including microplastic distribution in rivers, oceans and soils, and interaction between microplastics and organic pollutants (Guo and Wang, 2019; Cordova et al., 2021; Díaz-Jaramillo et al., 2021). Microplastics in the sediments of rivers and oceans not only cause serious pollution risk to the environment but also may threaten the safety of aquatic organisms (Gerolin et al., 2020; Huang et al., 2021). Microplastics are easily to be swallowed by aquatic organisms and keep stable in the organisms, which may cause malnutrition and mechanical damage of their digestive systems. Therefore, the predatory activities of organisms may reduce, even the organisms starve to death (Kramm and Volker, 2018; Possatto et al., 2011).

Microplastics in inland freshwater bodies not only are the main source to marine environment, but also may have a negative impact on local ecosystems (Lu et al., 2019). Currently, there is the highest plastic production and consumption in China, but the microplastic pollution in freshwater bodies of China has not been systematically and comprehensively investigated, and the basic data and corresponding understanding about microplastic distribution in many important rivers and

* Corresponding author. College of Environmental Science and Engineering, Beijing Forestry University, Qinghua East Road 35, Haidian District, Beijing, 100083, China.

E-mail address: panyue_zhang@bjfu.edu.cn (P. Zhang).

https://doi.org/10.1016/j.chemosphere.2021.130800
Received 22 February 2021; Received in revised form 23 April 2021; Accepted 30 April 2021
Available online 5 May 2021

lakes is insufficient (Tang, 2014; Zhang et al., 2018), especially in the rivers in northern China where the effluent of municipal wastewater treatment plants (MWTPs) becomes the base flow. The huge amount of microplastics in effluent of MWTPs makes it difficult to prevent and control microplastic pollution. Moreover, the microplastic distribution in sediments better reflects the long-term microplastic pollution in the area, compared with that in river water, especially the vertical distribution. The vertical microplastic distribution involves in microplastic properties, and their transportation and sedimentation in water environment. However, there are few studies on the vertical microplastic distribution in river sediments in northern China.

Baiyangdian Wetland is located in Xiong'an New Area, a new engine of modern economic system of China and is the largest inland freshwater wetland in northern China. Baiyangdian Wetland plays a significant role in promoting local ecological and economic construction by providing natural resources, regulating the local climate, and controlling flood, and so on. Currently there is no relevant research about microplastic pollution in this area. The target river of this study is Fuhe River, an important river flowing to Baiyangdian Wetland. In this paper the microplastic distribution was studied, especially the vertical distribution characteristics, including content, shape, particle size, color and type in estuary sediments of Fuhe River, and the factors influencing the microplastic distribution were discussed. The finding will fill the knowledge gaps of microplastic pollution in Baiyangdian Wetland watershed and provide a valuable reference for mechanism study of microplastic transportation and sedimentation, and policy setting of scientific microplastic control.

2. Materials and methods

2.1. Study area and sample collection

The Fuhe River is located in Baoding City, Hebei Province (38°10′ N-40°00′ N, 113°40′ E–116°20′ E) with a watershed area of 781 km^2. There are few industries in Fuhe River estuary to Baiyangdian Wetland, thus the microplastics come mainly from the effluent of municipal wastewater treatment plants, life and agricultural residues. Nine large villages are located along Fuhe River estuary (Fig. 1) and the information of villages is shown in Table S1. Subaqueous sediment samples were collected along Fuhe River estuary to Baiyangdian Wetland at 15 different sites (F1–F15), and the distance between two sampling sites is about 2 km. Global position system (GPS) was used to record the geographical coordinates of each sampling site (Table S2). The sediment samples with a depth of 50 cm were collected by a columnar sampler (inner diameter = 7 cm, height = 100 cm) in typical sampling sites F1, F4, F6, F7, F8, F12 and F15. Then the samples were cut into 10 layers with a stainless steel knife, and each sediment layer was 5 cm deep according to the method of Wu et al. (2019a). The samples were stored in sampling bottles for further experiments.

2.2. Sample treatment and microplastic extraction

The microplastics were extracted from sediment samples with density separation method (Thompson, 2004; He et al., 2020), and three replicates were carried out for each sample. The sediment samples were treated with 30% H_2O_2 to degrade the organic matters, and then dried at 50 °C until the sample weight did not change. The dried sediment of 50 g was weighted for microplastic extraction. Saturated sodium chloride (NaCl, Sinopharm Chemical Reagent Co., Ltd., China) solution was used as extracting solution because NaCl was cheap and environmentally friendly. Saturated NaCl solution of 1 L and dry sediment of 50 g were mixed in a glass beaker with a magnetic bar for 15 min, and then the beaker was kept in an ultrasonic bath (KQ5200E, Kunshan, China) for 15 min. After settling for 12 h, the supernatant was filtered from the mixture by 1 μm glass fiber filter papers (47 mm diameter, Millipore) with a vacuum filter (SHZ-DIII, Yuhua, China), and this procedure was repeated three times for each sample. The filter papers were dried at 50 °C until the paper weight did not change. The dried filter paper was observed with a stereoscopic microscope with a 40-100 × magnification (XTD-7045A, Coissm, China), and the possible microplastics were collected. Finally, 0.63–5.00 mm particles were analyzed by a Fourier transform infrared spectrometer (FTIR) (Vertex 70, Bruker, Germany) to identify the polymer type, and the scan range was from 4000 to 400 cm^{-1}. The FTIR spectra were compared with the Hummel Polymer and Additives Library.

2.3. Quality assurance and control

The glass vessels were cleaned three times with deionized water before each procedure, and covered with tin foil when the vessels were not in use. Three procedural blanks were conducted to assess potential pollution from the laboratory. For blank experiments, deionized water was filtered through the glass fiber filter papers with a vacuum filter. The filter papers were exposed to the laboratory environment for 24 h,

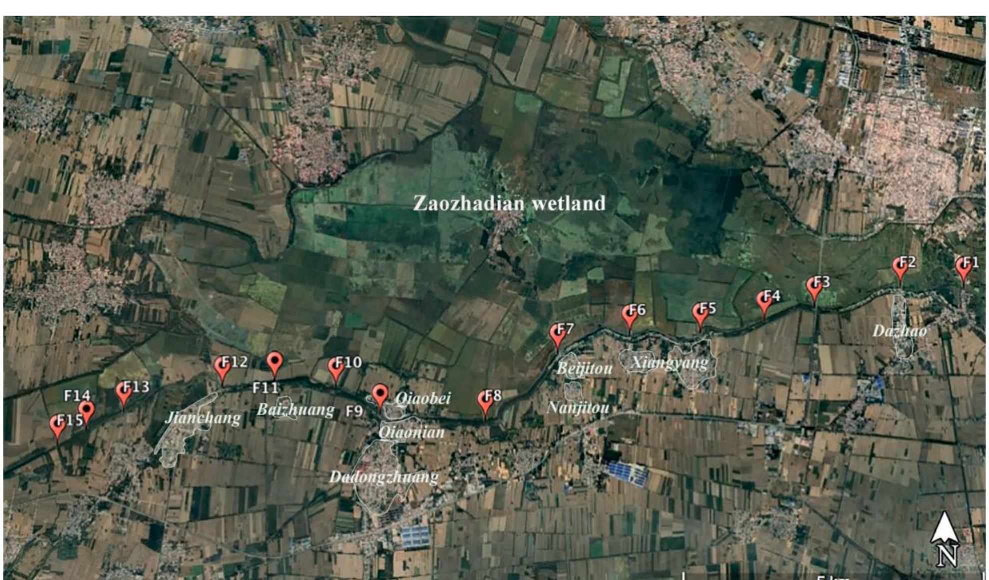

Fig. 1. Sampling sites along Fuhe River estuary to Baiyangdian Wetland.

and then the microplastics on the filter papers were observed with the stereoscopic microscope. No microplastic was observed on these filter papers, confirming that the potential laboratory pollution was negligible.

2.4. Statistic analysis

The microplastic content was expressed as microplastic number per kilogram dry sediment (items/kg). The average content was given as mean value ± standard deviation. All the experimental data and the difference of microplastic content between different sediment layers were tested using SPSS Statistics (22.0, IBM, USA). The homogeneity of variances and normality was checked by Levene's test and Kolmogorov-Smirnov test, respectively. The significance of different microplastic contents was tested by a one-way ANOVA followed by Tukey's test (significance level, $p < 0.05$).

3. Results and discussion

3.1. Microplastic content in Fuhe River estuary sediments

The microplastic content in the topmost sediment layer (0–5 cm) of 15 sampling sites along Fuhe River estuary is shown in Table 1. Obviously, microplastics were found in all sampling sites, and the average content was 558 ± 233 items/kg. The highest microplastic content was observed in F7 with 1049 ± 212 items/kg, significantly higher than that in F2, F5, F6, F8 and F11–F15 ($p < 0.04$), which might be attributed to the higher population density in villages near F7. The microplastic content in F15 was the lowest with 212 ± 14 items/kg due to lower human activities. The comparison of microplastic abundance in Fuhe River estuary sediments with that in other river sediments is shown in Table S3. The unit of microplastic content in sediments has not been unified in the relevant researches, because there is no uniform evaluation standard for microplastic pollution. The microplastic content was expressed as microplastic number or microplastic weight per kilogram dry or wet sediment in most studies. The microplastic pollution with the same unit of items/kg in this study was compared. Compared with those studies, the microplastic pollution in Fuhe River estuary sediments was of medium level. The microplastic content was significantly different in different river sediments around the world due to the different degrees of plastic use, different hydrodynamic and geographical conditions of studied areas (Bordós et al., 2019; Gray et al., 2018; Kataoka et al., 2019). Wang et al. (2018a) reported a high microplastic content of 32, 947 ± 15,342 items/kg in the sediments of Wenruitang River in Zhejiang Province, China, where population density is high and human activities are intensive. While Jiang et al. (2019) reported a very low microplastic content of 195 ± 64 items/kg in the sediment of a river in Tibet, where the population density is very low. Normally, the microplastic content is higher in the areas with denser population and more intense human activities (Peng et al., 2018; Wen et al., 2018; Cordova et al., 2019).

Table 1
Microplastic content in the topmost sediment layer (0–5 cm) of Fuhe River estuary sediments.

Sites	Microplastic content (items/kg)	Proportion (%)
F1	676 ± 290	8.1
F2	642 ± 246	7.7
F3	769 ± 194	9.2
F4	838 ± 230	10
F5	521 ± 126	6.2
F6	349 ± 88	4.2
F7	1049 ± 462	12.5
F8	322 ± 80	3.8
F9	683 ± 123	8.1
F10	270 ± 91	3.2
F11	241 ± 114	2.9
F12	550 ± 86	6.6
F13	597 ± 188	7.1
F14	659 ± 252	7.9
F15	212 ± 59	2.5
Total	8376	100
Average	558.4	

3.2. Microplastic content along sediment depth

The seven sampling sites F1, F4, F6, F7, F8, F12 and F15 covered different degrees of microplastic pollution were selected to represent the characteristics of vertical microplastic distribution in Fuhe River estuary sediments. Microplastic content in different depth sediments of seven sampling sites is shown in Table 2. The highest microplastic content was found in the 0–5 cm layer and the average content was 571 ± 281 items/kg, followed by that in the 5–10 cm layer (507 ± 236 items/kg). The microplastic number in the upper two layers accounted for more than 60% of the whole microplastic number in all layers (0–50 cm). The microplastic content in 10–15 cm, 15–20 cm layer was 76.4% and 54.7% of that in 0–5 cm layer, respectively. For the 20–40 cm layer, the microplastic content was less than 15% of that in the topmost layer (0–5 cm). No microplastic was detected when the depth of sediments exceeded 40 cm. The result was similar to that of other studies (Wang et al., 2019; Wu et al., 2019a). Wang et al. (2019) found that the microplastic content in 0–5, 5–10, 10–15, 15–20 cm sediment layer of South Yellow Sea was 2,143, 1,514, 1129 and 471 items/kg, respectively. Wu et al. (2019a) reported that the microplastic content in 2–5 cm and 5–10 cm sediment layer was 62.5% and 37.5% of that in the 0–2 cm sediment layer in a tidal flat of the Yangtze Estuary, respectively.

The relationship between the sediment depth and microplastic content was fitted by an exponential function (Fig. 2). The microplastic content in Fuhe River estuary sediments significantly decreased with the increase of sediment depth ($R^2 \geq 0.836$, $p \leq 0.002$). The fitted exponential showed that the decrease rate of microplastic content in 0–25 cm sediment layers was obviously greater than that in 25–40 cm sediment layers. The exponential variation trend of microplastic content in sediments was consistent with the exponential growth trend of global plastic production ($y = 1.385 \times 10\text{-}40\exp0.0475x$) reported by Brandon et al. (2019). The vertical microplastic distribution in sediments of the Derwent Estuary, Tasmania, Australia and the Santa Barbara Basin, California, USA was also found to be consistent with the growth rate of global plastic production (Willis et al., 2017; Brandon et al., 2019).

The formation of sediments is a long-term process. The formation period of different depth sediments can be analyzed through sedimentation rate, and the difference of vertical microplastic distribution might be attributed to the different plastic consumption levels in different periods. Guo et al. (2015) proved that the sedimentation rate of Baiyangdian Wetland was about 0.49 cm/year, therefore, it took about ten years to form a sediment depth of 5 cm. Microplastic accumulation in different periods in Fuhe River estuary sediments can be estimated based on this result. According to the record of China's annual plastic production (CPCIA, 1984–2014), the plastic production in China can be dated to the 1920s, and the plastic consumption was relatively low before the 1950s. Based on the plastic consumption around the Fuhe River, it is suggested that the sediments which exceeded 40 cm may form before the 1950s, therefore, there was no obvious microplastics in Fuhe River estuary sediments with a sediment depth larger than 40 cm. More microplastics were detected in 0–10 cm sediment layer of all seven sites, significantly higher than that in other layers ($p < 0.02$). This might be closely related to the rapid growth of population, development of plastic industries and increase of plastic consumption in recent decades. Grigore (2017) reported that the annual global yield of plastics increased by 222 million tons from 1993 to 2015. The microplastic content significantly reduced when the sediment depth was larger than 10 cm, which was possibly due to the low plastic production and use prior to 1999. Moreover, river erosion and heavy precipitation may lead to the disturbance of shallow sediment layer, which affected the vertical microplastic distribution in sediments. For example, there was a famous

Table 2
Microplastic content in different depth sediments of seven sampling sites.

Sites	Sediment depth (cm)							
	0–5	5–10	10–15	15–20	20–25	25–30	30–35	35–40
F1	676 ± 290	617 ± 246	239 ± 55	373 ± 70	49 ± 17	49 ± 16	0	0
F4	838 ± 230	593 ± 265	337 ± 94	360 ± 96	59 ± 11	77 ± 20	5 ± 2	14 ± 2
F6	349 ± 21	314 ± 140	197 ± 23	234 ± 45	41 ± 11	36 ± 10	0	0
F7	1049 ± 462	969 ± 319	429 ± 113	594 ± 98	85 ± 12	77 ± 12	38 ± 9	26 ± 6
F8	322 ± 80	381 ± 173	202 ± 84	103 ± 13	23 ± 8	0	0	0
F12	550 ± 86	487 ± 79	270 ± 9	149 ± 50	5 ± 1	0	12 ± 3	0
F15	212 ± 59	185 ± 83	92 ± 43	108 ± 66	0	0	0	0
Total	3996	3546	1766	1921	262	239	55	40
Mean	570.9	506.6	252.3	274.4	37.4	34.1	7.9	5.7
Standard deviation	280.7	236.1	100.1	166.3	28.1	32.5	13.0	9.6
Proportion (%)	33.8	30.0	15.0	16.2	2.2	2.0	0.5	0.3

Note: "Proportion" refers to the proportion of microplastic content in different sediment layers to the total microplastic content.

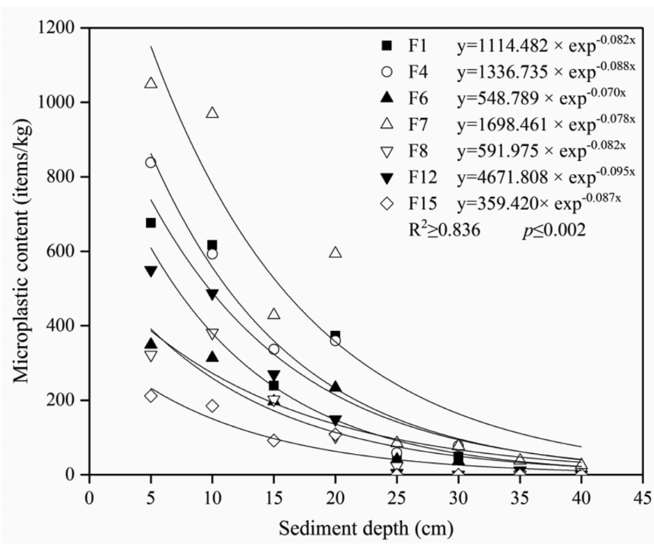

Fig. 2. Exponential relationship between sediment depth and microplastic content.

rainstorm in 1988, and the floods like this often occur in Baiyangdian area due to the heavy rain in a short time in summer. Wang et al. (2018b) calculated the microplastic content in sediments and sedimentation rate of the Yangtze River, and found that the vertical microplastic distribution in sediments was closely related to the formation period of sediments. Claessens et al. (2011) and Zalasiewicz et al. (2016) even suggested that the microplastic distribution could be a promising stratigraphic indicator for recently formed sediments.

3.3. Morphological characteristics of microplastics along sediment depth

3.3.1. Vertical distribution of microplastic shape

Four microplastic shapes were found in Fuhe River estuary sediments: fragment, fiber, film and pellet, and the photographs and SEM images of typical microplastics are shown in Fig. 3. The surface structure and texture of different microplastics differed. There were more protruding blocks on the surface of fragment microplastics, while in the surface of fiber microplastics more longitudinal cracks were observed; there were more small protruding particles on the surface of film microplastics, which may be due to the adsorption of sediment particles; and the surface of pellet microplastics showed obviously porous structure. These rough structures can increase the microplastic surface area, improving the adsorption capacity of other pollutants on microplastics and increasing the harm of microplastics to environment. The fragment microplastics were mostly the debris of woven bags, packaging materials and plastic containers, which might be from the fragmentation of large plastics by physical and chemical actions, such as weathering and ultraviolet light irradiation. The worn surface of microplastics may prove this speculation. The fiber microplastics had long and thin appearance, possibly came from clothing fibers in domestic sewage. Browne et al.

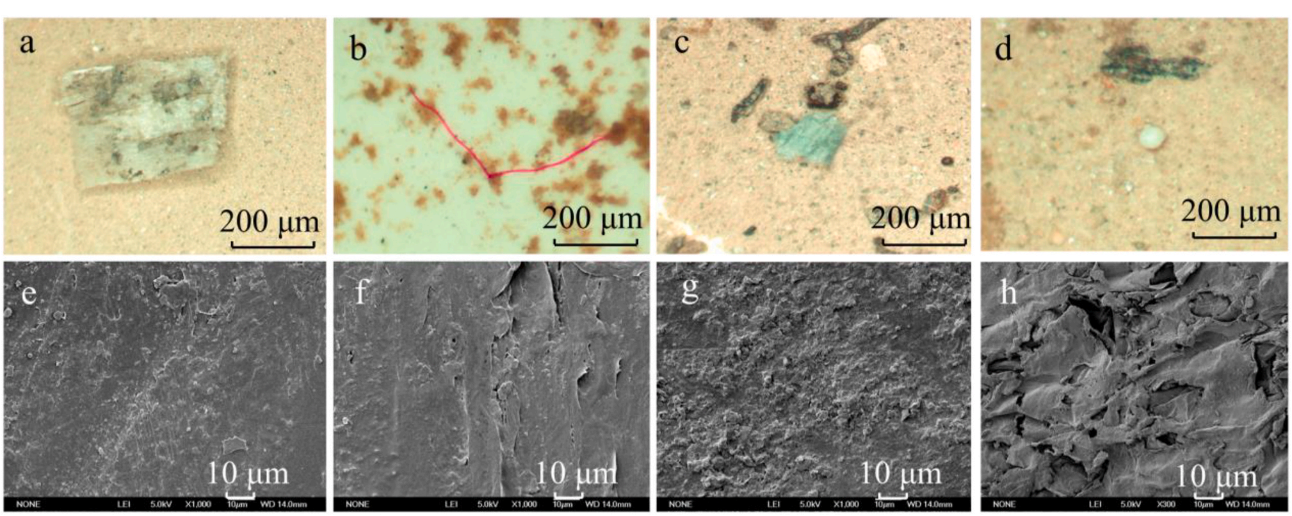

Fig. 3. Photographs of typical microplastics: (a) Fragment, (b) Fiber, (c) Film, (d) Pellet, and SEM images of typical microplastics: (e) Fragment, (f) Fiber, (g) Film, (h) Pellet.

(2011) suggested more than 1900 fabric fibers could be washed away from one wash of each piece of clothing. The film microplastics might be debris of discarded plastic packaging bags and agricultural plastic films with a very thin thickness. The pellet microplastics were ovoid, disc-shaped or cylindrical, which might be originated from industrial raw materials of daily personal care products, plastic products and resin particles (McDermid and McMullen, 2004; Hidalgo-Ruz et al., 2012). When a microplastic could not be classified as fiber, film or pellet, it was regarded as a fragment in this study.

The content of different shape microplastics in different depth sediments is shown in Fig. 4. The average content of fragment microplastics in seven sites was the highest in all sediment layers, followed by fiber microplastics. This was due to the high content of fragmented and fibrous microplastics in domestic wastes and wastewater (Jiang et al., 2019). Another possible reason was that most of plastic fragments were of polyethylene terephthalate (PET), and these plastic fibers were mostly of nylon (PA) and PET, which were easier to sink due to their relatively larger density (Wen et al., 2018). There was no film microplastics when the sediment depth was larger than 30 cm, which may be attributed to that the film microplastics were normally flake with large particle size and mostly of polyethylene (PE) and polypropylene (PP) with a low density. Piperagkas et al. (2019) reported that there were more light-weight microplastics (fibers, films and foamy plastics) in the surface sediments of Northern Crete, Greece. The particle shape is important, because it is closely related with the surface area of microplastics, which might significantly affect the adsorption behaviors of other pollutants on microplastics. Chubarenko et al. (2016) reported that the fouling adsorption by fiber and film microplastics was the largest, while that by pellet microplastics was the smallest. The texture, particle size and types of different shape microplastics are generally different, which can lead to different performances of microplastic pollution and their vertical distribution may be influenced by various factors.

3.3.2. Vertical distribution of microplastic size

There is no clear standard for the classification of microplastic particle size. Microplastics in Fuhe River estuary sediments were divided into four categories according to Peng et al. (2018): <0.1 mm, 0.1–0.5 mm, 0.5–1.0 mm and 1.0–5.0 mm, as shown in Fig. 5. Microplastics smaller than 0.1 mm in size accounted for the smallest proportion in all sediment samples, which may be related to the limitation of stereoscopic microscope. Microplastics smaller than 0.063 mm in size were difficult to be observed under the used stereoscopic microscope. The content of 0.1–0.5 mm microplastics was the most, followed by 0.5–1.0 mm microplastics in all sediment layers. The proportion of microplastics with particle size bigger than 1.0 mm decreased with the increase of sediment depth, and no such microplastic was found when the sediment depth was larger than 30 cm. Enders et al. (2015) simulated the vertical distribution of microplastic size and found that there were more microplastics with a particle size of 1 mm or larger in the layer less than 1 m in the Atlantic Ocean. Kunz et al. (2016) reported that there were more 1–2 mm microplastics in 0–5 cm sediment layer than that in 5–10 cm layer of Shalun Beach, Taiwan. Zheng et al. (2020) reported that the microplastics with smallest average size were mostly distributed in the bottom sediment (40–45 cm) due to more soils on the smaller microplastic surface. It seems that the particle size of microplastics normally decreased with the increase of sediment depth.

Microplastics with smaller particle size are usually easier to aggregate with natural colloids than that with larger particle size, which increases the sedimentation possibility of smaller microplastics (Besseling et al., 2017). When aquatic organisms and detrital materials accumulate on microplastic surface due to biofouling, the flotation and sedimentation of microplastics would be affected (Ye and Andrady, 1991; Long et al., 2015). Biological fouling and buoyancy of microplastics are normally related to their surface area and volume, respectively, and the microplastics in larger size settle slower than those in smaller size because of their smaller surface area to volume ratio (Ryan, 2015; Fazey and Ryan, 2016). Lower flow rate and eutrophic characteristics of water body in Fuhe River estuary may enhance the sedimentation of microplastics in smaller size, because such water environment may facilitate the development of fouling communities on microplastic surface. Therefore, the smaller microplastics with higher biological fouling would preferentially accumulate in sediments than larger microplastics (Wang et al., 2018a,b). Compared with larger microplastics, smaller ones are more easily swallowed by aquatic organisms, which may cause more smaller microplastics to enter deeper sediments along with aquatic organisms and their fecal pellets. The sedimentation process and water dynamics may also contribute to the complex distribution of microplastics indifferent particle sizes (Matsuguma et al., 2017; Zheng et al., 2020).

3.3.3. Vertical distribution of microplastic color

Various colors of microplastics were observed in Fuhe River estuary sediments, mainly including transparent, white, red, black and green, and the microplastics then were divided into three categories:

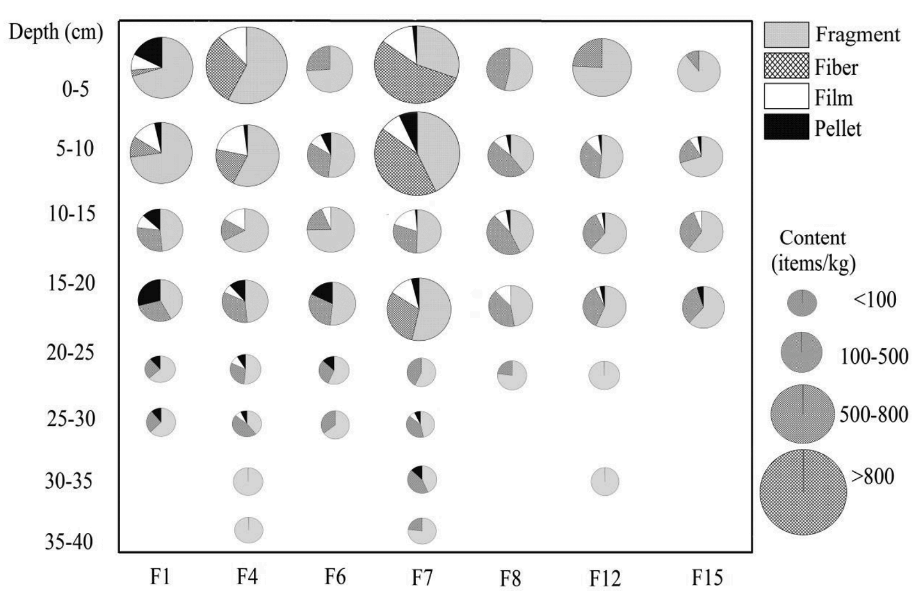

Fig. 4. Distribution of microplastic shape along sediment depth (Circle size represents the microplastic content).

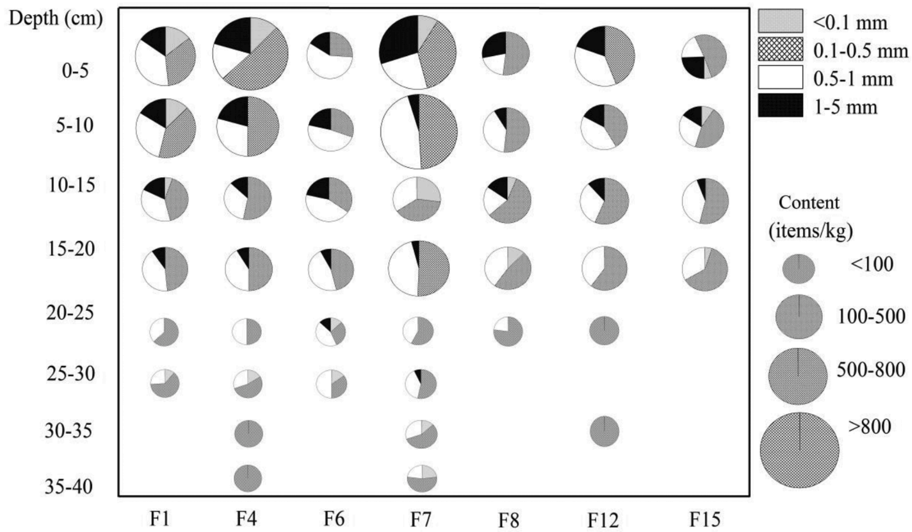

Fig. 5. Distribution of microplastic size along sediment depth (Circle size represents the microplastic content).

transparent, white and colored (Fig. 6). The proportion of transparent microplastics in 0–20 cm layers was larger than that in 20–40 cm layers. But the distribution of transparent microplastics in Jiaozhou Bay was the opposite of this study (Zheng et al., 2020). The difference between the two studies may be caused by the reginal conditions. There were colored microplastics in almost all layers, and the proportion of colored microplastics in 20–40 cm layers was larger than that in 0–20 cm layers. The density of most pigments is bigger than that of microplastics, which may increase the density of colored microplastics and make the colored microplastics more easily sink when the sediments are disturbed. Additionally, the aquatic organisms are more likely to swallow colored microplastics than transparent ones to cause possible harm to their health, which indicated that the deep study of microplastic color is necessary (Foekema et al., 2013; Huang et al., 2015; Qu et al., 2018).

3.4. Polymer identification of microplastics along sediment depth

There were five types of microplastic polymers to be observed in Fuhe River estuary sediments: PE, PP, PET, PA and polystyrene (PS). The proportion of different polymer types based on FTIR analysis is shown in Table S4, and the FTIR spectra of five typical microplastic polymers are shown in Fig. S1. With the increase of sediment depth, the proportion of non-plastic particles in microscopic examination decreased. Shallow sediments were mostly gray-black, contained more plant and biological debris, for example, shell and snail debris were often found in the 0–10 cm layers, which increased the difficulty of microplastic identification. When the sediment depth increased, the content of impurities decreased, and the accuracy of microplastic identification increased.

The proportion of different type microplastics in different depth sediments is shown in Fig. 7. In all sediment layers, PE and PP microplastics were observed and the sum of their proportions was larger than 50%. Although the density of PE and PP microplastics is lower than that of river water, it might significantly increase after biological fouling and impurity adsorption, particularly in eutrophic and low turbulent water in the Fuhe River estuary. Zettler et al. (2013) found that microorganisms were easy to be colonized on microplastics. PE has become the polymer type with the largest production since 1998, followed by PP, furthermore, PE and PP plastics are widely used in daily life, which may be the main reasons for the higher proportion of these two polymers. The proportion of PET microplastics in deeper sediment layers was higher than that in shallower layers due to its large density (1.38 g/cm^3). No PA microplastics were detected when the sediment depth exceeded 30 cm,

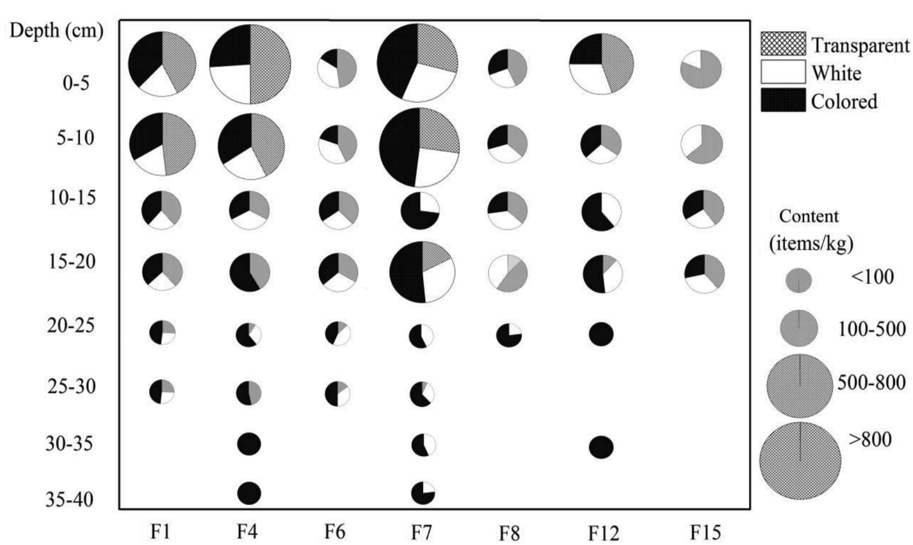

Fig. 6. Distribution of microplastic color along sediment depth (Circle size represents the microplastic content).

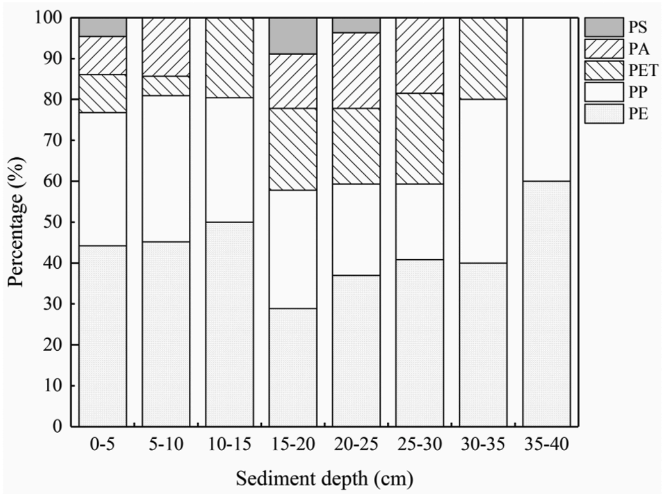

Fig. 7. Percentage of different type microplastics in different depth sediments.

which may be because that PA was not produced on a large scale in China until 2002. The content of PS microplastics in sediments is very low due to its low development. The demand for PS plastics has continuously declined since 2003. Matsuguma et al. (2017) also found more polyacrylate microplastics in 0–2 cm sediment layer than that in 38–40 cm layer of Sakurada-bori Moat, Japan due to its low density (0.95 g/cm^3). Many factors, including polymer composition of plastic products, sources and introduction routes of microplastics, might have led to the differences in microplastic types between different study areas.

Lipuma (2016) reported that the effluent of MWTPs is an important source of microplastics in many rivers. PE, PET and PA are the main microplastic types in the effluent of MWTPs, and fragment and fiber are their main existing shapes, which is consistent with the results of microplastics in Fuhe River estuary sediments (Li et al., 2019). This indicated that the microplastics in Fuhe River estuary sediments may mostly come from the effluent of MWTPs. In addition, imperfect treatment facilities and management regulations of wastewater and solid wastes in rural areas also contribute to microplastic pollution in these areas. For example, before the establishment of Xiong'an New Area, the rural domestic sewage was directly discharged into the Fuhe River and the solid wastes were randomly stacked around villages due to the lack of strict management. Therefore, the management of domestic sewage and solid wastes should be strengthened, and the treatment facilities should be improved in rural areas.

4. Indication of vertical microplastic distribution and research prospect

The variation of microplastic content and particle size along with sediment depth of oceans, rivers and wetlands is similar. However, the variation of microplastic shapes and colors along sediment depth is significantly different between the studied oceans and wetlands (Matsuguma et al., 2017; Dai et al., 2018; Näkki et al., 2019; Zheng et al., 2020). The vertical distribution characteristics of microplastics may reflect the long-term microplastic pollution of water body and the harm to water ecosystem, which can make the risk of microplastic pollution clearer (Dai et al., 2018). Therefore, more attention should be paid to the vertical distribution of microplastics in different water bodies.

On the other hand, not only the polymer type of microplastics in Fuhe River estuary sediments is complex, but also the real sedimentary environment is complicated and changeable. Heavy metals, such as Cd and Hg, were observed in Baiyangdian sediments (Guo et al., 2015), which may be adsorbed on microplastics, increasing the environmental risk. The adsorption of heavy metals on microplastics is related to many factors, including the microplastic type, heavy metal type, form and concentration, and surrounding environmental conditions. In addition, Yu et al. (2021) reported that the microplastics in soil promoted the transformation of heavy metals from bioavailable to stable organic-bound fractions, reducing the bioavailability of heavy metals related to the physicochemical parameters of soil and kind of heavy metals. Therefore, further researches are needed to demonstrate the interaction between microplastics and heavy metals in sediments and their impact on aquatic ecosystem.

Moreover, biofilm is often formed on the microplastic surface in environment, but there were few studies on the combined pollution of microplastic surface biofilm with other pollutants. The microplastic surface biofilm may affect the behaviors of nutrient conversion, metal and antibiotic adsorption (Leiser et al., 2020; Chen et al., 2021). Microplastic surface biofilm can also selectively enrich pathogens, which may produce more complex and serious toxic effect on aquatic organisms, especially on zoobenthos and whole food chain, which needs to be further studied (Wu et al., 2019b).

Currently, there is still no unified ecological risk assessment method for microplastic pollution, which is limited by the current research process. Microplastic pollution is not only affected by the natural environment, such as flow velocity, sedimentation process, adsorption behaviors, and so on, but also human activities, economic development and other factors play important roles in the microplastic distribution. The ecological risk of microplastic pollution is an important criterion for controlling microplastic pollution and formulating relevant policies. Therefore, the construction of scientific ecological risk assessment system is an inevitable requirement.

5. Conclusions

Vertical microplastic distribution in the estuary sediments of Fuhe River to Baiyangdian Wetland was investigated. The content, shape and particle size of microplastics presented significant differences in different depth sediments. With the increase of sediment depth, both total content and particle size of microplastics decreased obviously. The proportion of colored microplastics in a deeper sediment layer was larger than that in a shallower layer. PE and PP were the main plastic polymer types in Fuhe River estuary sediments, but there were more PET microplastics in 15–35 cm sediment layers. The main sources of microplastic pollution in Fuhe River estuary sediments are domestic wastewater and solid wastes, so the proper solutions controlling wastewater and solid waste discharge are of great significance on the control of microplastic pollution in Fuhe River estuary.

Credit author statement

Zeyan Zhou: Conceptualization, Writing, Visualization, Reviewing and Editing, **Panyue Zhang**: Supervision, Conceptualization, Reviewing and Editing, **Guangming Zhang**: Supervision, Conceptualization, Reviewing and Editing, **Siqi Wang**: Writing, Visualization and Editing, **Yaijing Cai**: Writing, Visualization and Editing, **Hongjie Wang**: Writing, Visualization and 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.

Acknowledgements

This work was supported by the Major Science and Technology Program for Water Pollution Control and Treatment (No. 2018ZX07110003).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2021.130800.

References

Besseling, E., Quik, J.T.K., Sun, M.Z., Koelmans, A.A., 2017. Fate of nano- and microplastic in freshwater systems: a modeling study. Environ. Pollut. 220, 540–548.
Bordós, G., Urbányi, B., Micsinai, A., Kriszt, B., Palotai, Z., Szabó, I., Hantosi, Z., Szoboszlay, S., 2019. Identification of microplastics in fish ponds and natural freshwater environments of the Carpathian basin, Europe. Chemosphere 216, 110–116.
Brandon, J.A., Jones, W., Ohman, M.D., 2019. Multidecadal increase in plastic particles in coastal ocean sediments. Sci. Adv. 5, 587.
Browne, M.A., Crump, P., Niven, S.J., Teuten, E., Tonkin, A., Galloway, T., Thompson, R., 2011. Accumulation of microplastic on shorelines worldwide: sources and sinks. Environ. Sci. Technol. 45 (21), 9175–9179.
Chen, H.B., Tang, M.G., Yang, X., Tsang, Y.F., Wu, Y.X., Wang, D.B., Zhou, Y.Y., 2021. Polyamide 6 microplastics facilitate methane production during anaerobic digestion of waste activated sludge. Chem. Eng. J. 408, 127251.
Chubarenko, I., Bagaev, A., Zobkov, M., Esiukova, E., 2016. On some physical and dynamical properties of microplastic particles in marine environment. Mar. Pollut. Bull. 108, 105–112.
Claessens, M., Meester, S.D., Landuyt, L.V., Clerck, K.D., Janssen, C.R., 2011. Occurrence and distribution of microplastics in marine sediments along the Belgian coast. Mar. Pollut. Bull. 62, 2199–2204.
Cordova, M.R., Purwiyanto, A.I.S., Suteja, Y., 2019. Abundance and characteristics of microplastics in the northern coastal waters of Surabaya, Indonesia. Mar. Pollut. Bull. 142, 183–188.
Cordova, M.R., Ulumuddin, Y.Y., Purbonegoro, T., Shiomoto, A., 2021. Characterization of microplastics in mangrove sediment of Muara Angke wildlife reserve, Indonesia. Mar. Pollut. Bull. 163, 112012.
Dai, Z.F., Zhang, H.B., Zhou, Q., Tian, Y., Chen, T., Tu, C., Fu, C.C., Luo, Y.M., 2018. Occurrence of microplastics in the water column and sediment in an inland sea affected by intensive anthropogenic activities. Environ. Pollut. 242, 1557–1565.
Díaz-Jaramillo, M., Islas, M.S., Gonzalez, M., 2021. Spatial distribution patterns and identification of microplastics on intertidal sediments from urban and semi-natural SW Atlantic estuaries. Environ. Pollut. 273, 116398.
Enders, K., Lenz, R., Stedmon, C.A., Nielsen, T.G., 2015. Abundance, size and polymer composition of marine microplastics $\geq$10 μm in the Atlantic Ocean and their modelled vertical distribution. Mar. Pollut. Bull. 100, 70–81.
Fazey, F.M., Ryan, P.G., 2016. Biofouling on buoyant marine plastics: an experimental study into the effect of size on surface longevity. Environ. Pollut. 210, 354–360.
Foekema, E.M., De Gruijter, C., Mergia, M.T., van Franeker, J.A., Murk, A.J., Koelmans, A.A., 2013. Plastic in north sea fish. Environ. Sci. Technol. 47 (15), 8818–8824.
Gerolin, C.R., Pupim, F.N., Sawakuchi, A.O., Grohmann, C.H., Labuto, G., Semensatto, D., 2020. Microplastics in sediments from Amazon rivers, Brazil. Sci. Total Environ. 749, 141604.
Geyer, R., Jambeck, J.R., Law, K.L., 2017. Production, use, and fate of all plastics ever made. Sci. Adv. 3, 25–29.
Gray, A.D., Wertz, H., Leads, R.R., Weinstein, J.E., 2018. Microplastic in two South Carolina Estuaries: occurrence, distribution, and composition. Mar. Pollut. Bull. 128, 223–233.
Grigore, M., 2017. Methods of recycling, properties and applications of recycled thermoplastic polymers. Recycling 2 (4), 24.
Guo, W., Huo, S.L., Ding, W.J., 2015. Historical record of human impact in a lake of northern China: magnetic susceptibility, nutrients, heavy metals and OCPs. Ecol. Indicat. 57, 74–81.
Guo, X., Wang, J.L., 2019. The chemical behaviors of microplastics in marine environment: a review. Mar. Pollut. Bull. 142, 1–14.
He, B.B., Goonetilleke, A., Ayoko, G.A., Rintoul, L., 2020. Abundance, distribution patterns, and identification of microplastics in Brisbane River sediments, Australia. Sci. Total Environ. 700, 134467.
Hidalgo-Ruz, V., Gutow, L., Thompson, R.C., Thiel, M., 2012. Microplastics in the marine environment: a review of the methods used for identification and quantification. Environ. Sci. Technol. 46 (6), 3060–3075.
Huang, D.L., Tao, J.X., Cheng, M., Deng, R., Yin, L.S., Li, R.J., 2021. Microplastics and nanoplastics in the environment: macroscopic transport and effects on creatures. J. Hazard Mater. 407, 124399.
Huang, D.L., Wang, R.Z., Liu, Y.G., Zeng, G.M., Lai, C., Xu, P., Lu, B.A., Xu, J.J., Wang, C., Huang, C., 2015. Application of molecularly imprinted polymers in wastewater treatment: a review. Environ. Sci. Pollut. Res. Int. 22 (2), 963–977.
Jiang, C.B., Yin, L.S., Li, Z.W., Wen, X.F., Lou, X., Hu, S.P., Yang, H.Y., Long, Y.N., Deng, B., Huang, L.Z., Liu, Y.Z., 2019. Microplastic pollution in the river of the Tibet plateau. Environ. Pollut. 249, 91–98.
Kataoka, T., Nihei, Y., Kudou, K., Hinata, H., 2019. Assessment of the sources and inflow processes of microplastics in the river environments of Japan. Environ. Pollut. 244, 958–965.
Kramm, J., Völker, C., 2018. Understanding the Risks of Microplastics: A Social-Ecological Risk Perspective. Freshwater Microplastics.
Kunz, A., Walther, B.A., Löwemark, L., Lee, L., 2016. Distribution and quantity of microplastic on sandy beaches along the northern coast of Taiwan. Mar. Pollut. Bull. 111, 126–135.
Lee, J., Lee, J.S., Jang, Y.C., Hong, S.Y., Shim, W.J., Song, Y.K., Hong, S.H., Jang, M., Han, G.M., Kang, D., 2015. Distribution and size relationships of plastic marine debris on beaches in South Korea. Arch. Environ. Contam. Toxicol. 69, 288–298.
Leiser, R., Wu, G.M., Katrin, W.P., 2020. Biofouling, metal sorption and aggregation are related to sinking of microplastics in a stratified reservoir. Water Res. 176, 115748.
Li, X.W., Ji, Y.Y., Mei, Q.Q., Chen, L.B., Zhang, X.L., Dong, B., Dai, X.H., 2019. Microplastics in wastewater and sludge from wastewater treatment plant: a review. Water Purific. Technol. 38 (7), 13–22.
Lipuma, L., 2016. Consider a Source: Microplastic in Rivers Is Abundant, Mobile, and Selects for Unique Bacterial Assemblages Marine Science Conference.
Long, M., Moriceau, B., Gallinari, M., Lambert, C., Huvet, A., Raffray, J., Soudant, P., 2015. Interactions between microplastics and phytoplankton aggregates: impact on their respective fates. Mar. Chem. 175, 39–46.
Lu, L., Luo, T., Zhao, Y., Cai, C.H., Fu, Z.W., Jin, Y.X., 2019. Interaction between microplastics and microorganism as well as gutmicrobiota: a consideration on environmental animal and human health. Sci. Total Environ. 667, 94–100.
Matsuguma, Y., Takada, H., Kumata, H., Kanke, H., Sakurai, S., Suzuki, T., Itoh, M., Okazaki, Y., Boonyatumanond, R., Zakaria, M.P., Weerts, S., Newman, B., 2017. Microplastics in sediment cores from Asia and Africa as indicators of temporal trends in plastic pollution. Arch. Environ. Contam. Toxicol. 73, 230–239.
McDermid, K.J., McMullen, T.L., 2004. Quantitative analysis of small-plastic debris on beaches in the Hawaiian Archipelago. Mar. Pollut. Bull. 48, 790–794.
Näkki, P., Setälä, O., Lehtiniemi, M., 2019. Seafloor sediments as microplastic sinks in the northern Baltic Sea-Negligible upward transport of buried microplastics by bioturbation. Environ. Pollut. 249, 74–81.
Peng, G.Y., Xu, P., Zhu, B.S., Bai, M.Y., Li, D.J., 2018. Microplastics in freshwater river sediments in Shanghai, China: a case study of risk assessment in mega-cities. Environ. Pollut. 234, 448–456.
Piperagkas, O., Papageorgiou, N., Karakassis, I., 2019. Qualitative and quantitative assessment of microplastics in three sandy Mediterranean beaches, including different methodological approaches. Estuar. Coast. Shelf S. 219, 169–175.
PlasticsEurope, Plastic-The Facts 2019: an Analysis of European Plastics Production, Demand and Waste Data (PlasticsEurope, 2019).
Possatto, F.E., Barletta, M., Costa, M.F., Ivar, J.A., Dantas, D.V., 2011. Plastic debris ingestion by marine catfish: an unexpected fisheries impact. Mar. Pollut. Bull. 65 (5), 1098–1102, 0.
Qu, X.Y., Su, L., Li, H.X., Liang, M.Z., Shi, H.H., 2018. Assessing the relationship between the abundance and properties of microplastics in water and in mussels. Sci. Total Environ. 621, 679–686.
Ryan, P.G., 2015. Does size and buoyancy affect the long-distance transport of floating debris? Environ. Res. Lett. 10, 084019.
Shen, M.C., Huang, W., Chen, M., Song, B., Zeng, G.M., Zhang, Y.X., 2020. (Micro)plastic crisis: Un-ignorable contribution to global greenhouse gas emissions and climate change. J. Clean. Prod. 254, 120138.
Tang, L., 2014. Study on water ecological characteristics and main evaluation factors of medium and small rivers in north area. Environ. Prot. Sci. 40 (1), 41–45, 0.
Tang, Y.Q., Liu, Y.G., Zhang, W., Zhao, J.M., He, S.Y., Yang, C.P., Zhang, T., Tang, C.F., Zhang, C., Yang, Z.S., 2020. A review: research progress on microplastic pollutants in aquatic environments. Sci. Total Environ. 142572.
Thompson, R.C., 2004. Lost at sea: where is all the plastic. Science 304 (5672), 838-838.
Wang, F., Nian, X.M., Wang, J.L., Zhang, W.G., Peng, G.Y., Ge, C., Dong, C.Y., Qu, J.G., Li, D.J., 2018a. Multiple dating approaches applied to the recent sediments in the Yangtze River (Changjiang) subaqueous delta. Quat. Geochronol.
Wang, Z.F., Su, B.B., Xu, X.Q., Di, D., Huang, H., Mei, K., Dahlgren, R.A., Zhang, M.H., Shang, X., 2018b. Preferential accumulation of small (<300 μm) microplastics in the sediments of a coastal plain river network in eastern China. Water Res. 144, 393–401.
Wang, J., Wang, M.X., Ru, S.G., Liu, X.S., 2019. High levels of microplastic pollution in the sediments and benthic organisms of the South Yellow Sea, China. Sci. Total Environ. 651, 1661–1669.
Wen, X.F., Du, C.Y., Xu, P., Zeng, G.M., Huang, D.L., Yin, L.S., Yin, Q.D., Hu, L., Wan, J., Zhang, J.F., Tan, S.Y., Deng, R., 2018. Microplastic pollution in surface sediments of urban water areas in Changsha, China: abundance, composition, surface textures. Mar. Pollut. Bull. 136, 414–423.
Willis, K.A., Eriksen, R., Wilcox, C., Hardesty, B.D., 2017. Microplastic distribution at different sediment depths in an urban estuary. Front. Mar. Sci. 4, 419.
Wu, F.R., Pennings, S.C., Tong, C.F., Xu, Y.T., 2019a. Variation in microplastics composition at small spatial and temporal scales in a tidal flat of the Yangtze Estuary, China. Sci. Total Environ. 699, 134252.
Wu, X., Pan, J., Li, M., Li, Y., Bartlam, M., Wang, Y., 2019b. Selective enrichment of bacterial pathogens by microplastic biofilm. Water Res. 165, 114979.
Ye, S., Andrady, A.L., 1991. Fouling of floating plastic debris under Biscayne Bay exposure conditions. Mar. Pollut. Bull. 22, 608–613.
Yu, H., Zhang, Z., Zhang, Y., Fan, P., Xi, B.D., Tan, W.B., 2021. Metal type and aggregate microenvironment govern the responsesequence of speciation transformation of different heavy metals tomicroplastics in soil. Sci. Total Environ. 752, 141956.
Zalasiewicz, J., Waters, C.N., Ivar, J.A., Corcoran, P.L., Barnosky, A.D., Cearreta, A., 2016. The geological cycle of plastics and their use as a stratigraphic indicator of the Anthropocene. Anthropocene 13, 4–17.

Zettler, E.R., Mincer, T.J., Amaral-Zettler, L.A., 2013. Life in the Plastisphere: microbial communities on plastic marine debris. Environ. Sci. Technol. 47, 7137–7146.

Zhang, K., Shi, H.H., Peng, J.P., Wang, Y.H., Xiong, X., Wu, C.X., Lamb, P.K.S., 2018. Microplastic pollution in China's inland water systems: a review of findings, methods, characteristics, effects, and management. Sci. Total Environ. 630, 1641–1653.

Zheng, Y.F., Li, J.X., Cao, W., Jiang, F.H., Zhao, C., Ding, H.B., Wang, M.H., Gao, F.L., Sun, C.J., 2020. Vertical distribution of microplastics in bay sediment reflecting effects of sedimentation dynamics and anthropogenic activities. Mar. Pollut. Bull. 115, 110885.