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Microplastics in sediment cores as indicators of temporal trends in microplastic pollution in Andong salt marsh, Hangzhou Bay, China

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ABSTRACT

Microplastics are widely dispersed in the coastal and marine environment, and sediments are considered to be sinks for microplastics. In this study, a total of 53 sediment samples were removed from different depths of two sediment cores collected in Andong salt marsh, Hangzhou Bay. The microplastics were extracted from the sediment cores through a density floating separation method. The shape, size, and color of the microplastics was then observed under a microscope. Polymer types were identified using micro-Fourier transform infrared spectroscopy. The average abundance of microplastics was 13.2 ± 6.0 items 50 g^{-1} dry weight. The main components of microplastics were polystyrene, cellophane, polyethylene and polypropylene. The quantities of microplastics at different depths in the sediment cores were consistent with the history of microplastic pollution.

1. Introduction

The global production of plastics has increased rapidly over the past 60 years, reaching almost 348 million tons in 2017 (PlasticsEurope, 2018). Plastics in the sea eventually breakdown into small particles (< 5 mm) via ultraviolet radiation, biodegradation and mechanical disruption (Shim et al., 2017). In the early years, these small particles were reported on, but received little detailed attention. Afterwards, the plastic particles were described using the term "microplastics" (Thompson et al., 2004), which aroused concern in the scientific community. The distribution and morphology of microplastics in the marine environment were subsequently extensively analyzed (Claessens et al., 2011; Zhou et al., 2018).

Microplastics are observed in a variety of shapes, such as fragments, fibers, pellets, and films (Zhao et al., 2014). Ocean currents and hydrodynamic processes may distribute microplastics widely (Ng and Obbard, 2006). They have been detected in the marine environment worldwide (Andrady, 2011). Microplastics are small in size and may be mistakenly ingested by marine organisms (Davison and Asch, 2011; Lazar and Gracan, 2011), potentially resulting in physical or toxicological effects (Waller et al., 2017), and have a serial negative effect on the organisms throughout the food web (Lusher et al., 2013). A recent study reported that indigestible microplastics were discovered in tiny shrimp living in the six deepest ocean trenches in the world (Peng et al., 2018). This suggests that the pollution by microplastics has already touched the deepest part of the world's ocean. Thus, microplastics present a growing pollution problem on account of their widespread and increasing threat to the environment, with potential risks to human health (Andrady, 2011; Laist, 1987; Zhang et al., 2019a).

At present, microplastics have been reported in sediments of beaches, estuaries, nearshore and offshore marine habitats (Laglbauer et al., 2014; Zhang et al., 2019a,b; Zhao et al., 2018, 2014). China produces and consumes a large amount of plastic each year, accounting for 26% of the world's entire plastic yield (Zhao et al., 2018). However, research on the distribution and abundance of microplastics in sediments of Hangzhou Bay is absent. Hangzhou Bay embodies one of the largest gulfs in the world (Zhang et al., 2014). The main sources of sediment to this triangular-shaped bay are the Yangtze River (Yuan et al., 2017), the erosion of the seabed of the outer bay, and the Qiantang River and its surrounding rivers. In the last century, the coasts of Hangzhou Bay have been subjected to considerable anthropogenic pressures from engineering constructions, local aquaculture, and industrial activities from adjacent towns and

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cities (Fang et al., 2016), which resulted in the large scale intrusion of plastic debris into the coast and seas (Bulletin of Marine ecological Environmental Quality of China, 2017). Andong salt marsh is located in a hydrodynamic turning point on the south coast of Hangzhou Bay (Wang et al., 2008). Sediment cores from the Andong salt marsh can be used to rebuild the local pollution history of microplastics.

The current study is aimed at investigating the abundance and distribution of microplastics in sediment cores from Andong salt marsh, Hangzhou Bay. The suspected microplastics were observed under a stereoscopic microscope. The size, shape, color and abundance were classified. The polymer types were then identified using micro-Fourier transform-infrared (μ-FTIR) spectroscopy to determine the components of each microplastic. This is a preliminary study to demonstrate the utility of sediment cores for understanding the history and trends of microplastic pollution, and contributes to the database of microplastic distribution in marine sediments in China.

2. Materials and methods

2.1. Sampling

Two sediment cores were collected from Andong salt marsh in October 2015, using a columnar gravity sampler lined with a 60-mm diameter polyvinyl chloride (PVC) core tube. The core positions are 30°22′41″N, 121°10′19″E and 30°22′26″N, 121°12′20″E (Fig. 1). The cores, referred to as C1 and C2, had a length of 127.5 cm and 162.5 cm respectively (Fig. 1). The cores were sliced at 5 cm intervals in a contamination-restricted laboratory. The wet sediment samples were placed in clean glass dishes covered with aluminum foil for oven-drying at 70 °C for 24 h to a constant weight. The dry sediments were lightly compressed using a stainless-steel spoon to disaggregate the sediment. Clean stainless-steel spoons were used to transfer sediment from the glass dishes to glass jars. The jars were stored at room temperature until analysis.

2.2. Age determination of samples

Sediment from the cores was dated based on the ^{210}Pb activity, using the constant initial concentration (CIC) model to assess the sedimentation rate. Since the activities of ^{210}Pb were determined by their natural decay over time, the ^{210}Pb activities gradually and linearly increased with depth. The calculated mean sedimentation rates were estimated at 5.03 cm/year for C1 and 4.89 cm/year for C2 (Loh et al., 2018); therefore, the period represented by cores C1 (127.5 cm) and C2 (162.5 cm) was 25 and 33 years respectively. The age of each sediment portion of the cores was then calculated by multiplying the average sedimentation rate with the average depth of each sediment portion.

2.3. Microplastic extraction from sediment

Microplastics were extracted from the sediment samples following the method described by Zhao et al. (2018), but with certain modifications. Fifty grams of dried sediment was weighed into a 250 mL glass beaker. The sediment was mixed with 200 mL of a saturated $CaCl_2$ salt solution ($\rho = 1.40$ g/cm^3) and manually stirred using a clean glass rod for 2 min. The sediment–water mixture was allowed to settled for 24 h. The supernatant was then carefully transferred to another glass beaker. This isolation step was repeated three times for each sample. Approximately 10 mL of 30% H_2O_2 (Sinopharm, China) was added to 100 mL aliquots of the samples to degrade organic matter. After 24 h of settlement, the supernatant was transferred onto 8 μm pore-size, 47-mm diameter cellulose nitrate filter (Shanghai Xingya, China) with the assistance of a vacuum pump. All filtration devices were rinsed with Milli-Q water several times. The filter was placed into a clean petri dish and air dried at room temperature. Procedural blanks were prepared using the same procedures.

2.4. Microscopic inspection

Microplastics on the filter papers were observed using a z-shaped pattern from left to right with a digital microscope (Sunny Optical Technology, China) equipped with MvImage software at up to × 160 magnification. The shape, color and size of suspected microplastics were recorded. Particle shape was classified into four categories: fibers, fragments, films and pellet. In this study, foamed microplastics are not included in the classification as foamed fragments were hardly detected in the sediment cores. The color was categorized into six groups: transparent, black, blue, red, yellow and white. The longest side of the particles was measured, and then classified into five groups: $<$ 1000 μm, 1000–2000 μm, 2000–3000 μm, 3000–4000 μm, 4000–5000 μm. The $<$ 1000 μm group was subdivided into four groups: 100–300 μm, 300–500 μm, 500–700 μm, 700–1000 μm. Microplastics smaller than 100 μm were not analyzed because experience during this study showed that it was difficult to remove particles of a smaller size from the filters.

2.5. Identification of microplastics with μ-FTIR

Suspected particles on the filter papers were randomly removed with forceps or needles for validation using μ-FTIR (Nicolet™ iN10; Thermo Scientific, USA) in reflection mode. The detector was cooled with liquid nitrogen before use. Particles were placed onto the surface of a gold plated slide. The detector was operated in 675–4000 cm^{-1} wave range, with a collection time of 3 s and co-addition of 16 scans at a resolution of 8 cm^{-1}. The aperture was set to 16-20 μm. The spectra were obtained through OMNIC™ Picta™ software (Thermo Fisher Scientific) and compared with the OMNIC polymer spectra library. Analysis of the composition of some large sized suspected microplastic particles was conducted under a μ-FTIR (Nicolet iS10) microscope in attenuated total reflection (ATR) mode. All spectra were collected at a resolution of 4 cm^{-1} using a diamond MicroTip accessory at 675–4000 cm^{-1}, with a collection time of 24 s for 32 co-scans. The spectra were compared to FTIR spectral libraries (see Table 1) to verify the polymer type. The spectrum analysis followed the method of (Woodall et al., 2014), where a quality index $\geq$ 70% matched with the standard database were acceptable.

2.6. Procedural contamination controls

All apparatus was rinsed three times with Milli-Q water and then dried before use. Plastic equipment was replaced with non-plastic equipment where possible. If not possible, plastic equipment was rinsed three times with Milli-Q water and checked to make sure no plastic debris was produced. Investigators that handled the samples wore 100% cotton laboratory coats and polymer-free white gloves. To ensure the $CaCl_2$ solution was free of plastic particles, anhydrous calcium chloride was dissolved in Milli-Q water and passed through filter paper before density floating separation. The laboratory window remained closed during the study. The procedural blanks, comprising 200 mL of saturated $CaCl_2$ solution, underwent the same procedure.

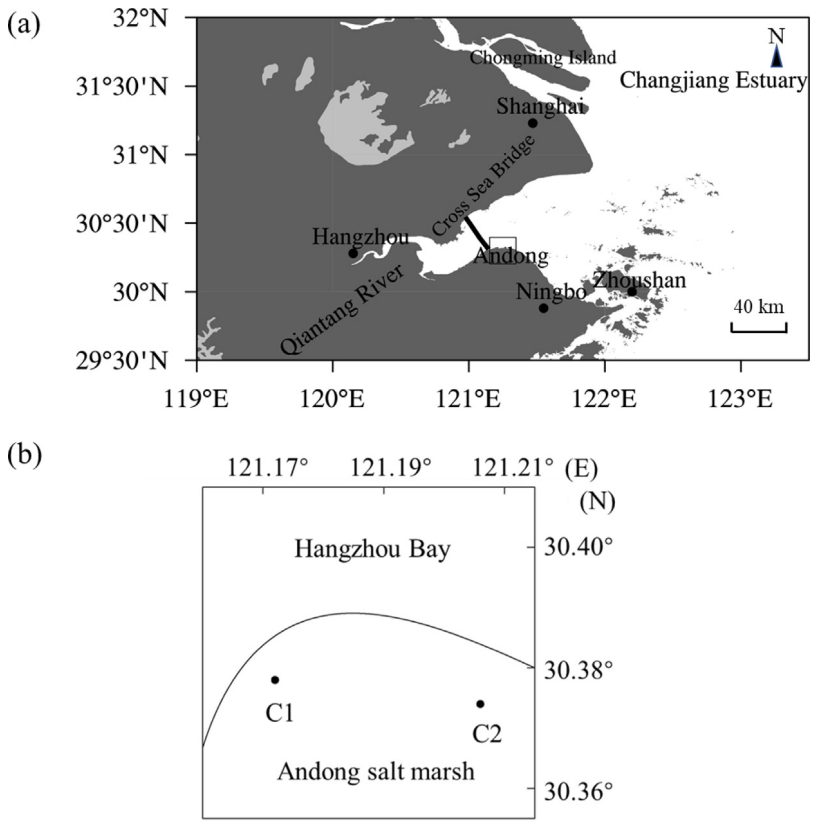

Fig. 1. (a) Map of Andong salt marsh in Hangzhou Bay, and (b) the locations where sediment cores C1 and C2 were collected.

Table 1
Information on the microplastics identified in sediment from Andong salt marsh, Hangzhou Bay.

Type of polymer	Number	Proportion of total particles (%)	FTIR Library
Polystyrene	87	40.1	HR Nicolet Sampler Library
Cellophane	36	16.6	Hummel Polymer Sample Library
Polyethylene	30	13.8	Hummel Polymer Sample Library
Polypropylene	28	12.9	Polymer Laminate Films
Polyethylene terephthalate	15	6.9	Wizard Library
Cellulose	13	6.0	Cross Sections Wizard
Polyacrylonitrile	8	3.7	HR Nicolet Sampler Library
Total	217	–	–

3. Results

3.1. Procedural blanks assessment

Five procedural blanks were prepared in parallel with five groups of samples, and 1.4 ± 1.0 items/sample was found in the procedural blanks. These microplastic particles were all in the form of fibers, which agreed with the results of Zhang et al. (2019a). This showed minor contamination from airborne microplastics. This number of fibers were subtracted from each sediment sample. Contamination from laboratory sources (air, sampling instruments, clothes, etc.) can thus be largely excluded.

3.2. Abundance and distribution of microplastics in sediment

Microplastics were detected in 53 sediment samples from cores C1 and C2 from Andong salt marsh, for a total of 701 pieces altogether. Microplastic abundance at different depths in cores C1 and C2 varied from 4.0–21.0 and 2.0–24.0 items per 50 g^{-1} dry weight of sediment respectively, with an average abundance in cores C1 and C2 of 14.8 ± 4.9 and 12.2 ± 6.3 items 50 g^{-1} dry weight sediment respectively. The quantity of microplastics with decreasing depth through the cores reveals a growing trend. There was marked difference in microplastic abundance over time (Fig. 2). The quantity of microplastics generally decreased with depth.

3.3. Shape, size and color of microplastics

Fibers (63%) were the most common shapes of microplastics, followed by fragments (16%), pellets (15%), and films (6%) (Fig. 3a). With regards to the color of the microplastics, transparent (84%) particles were dominant, while blue, black, red, yellow and white microplastics were observed less frequently (Fig. 3b). About 78% of the microplastic particles were < 1000 μm, with the most frequently observed microplastics in the size range of 100–500 μm, which takes up 66% of microplastics < 1000 μm. The results revealed that the proportion of observed microplastics increased with decreasing plastic particle size (Fig. 4).

3.4. Identification of microplastics using μ-FTIR

A total of 217 particles were identified as microplastics through μ-FTIR. The polymer types, and the number of microplastics of each type are provided in the Table 1. Polystyrene (PS) was the most abundant type, accounting for 40.1% of the total identified particles, followed by cellophane (16.6%), PE (13.8%), PP

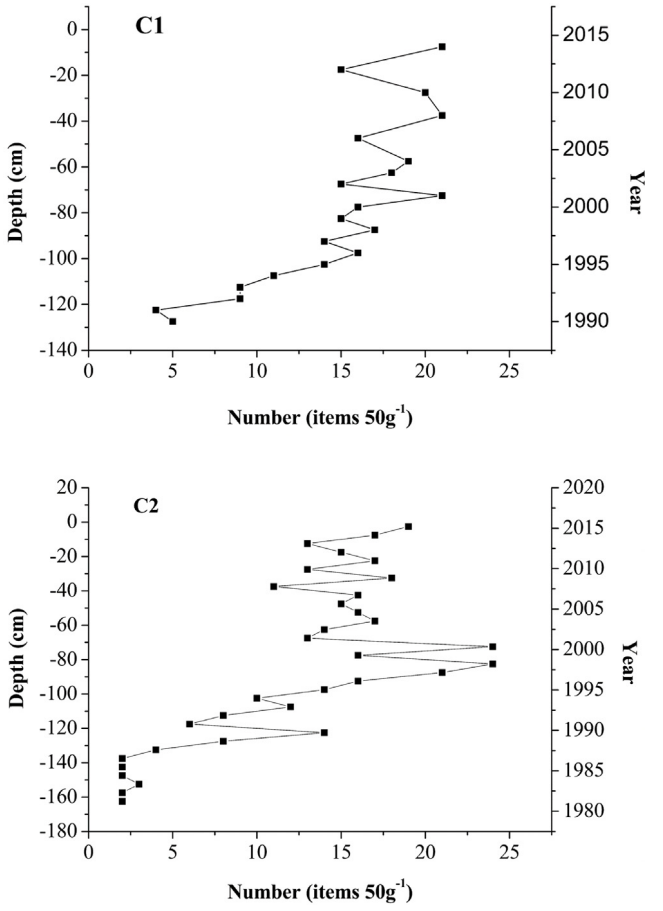

Fig. 2. Abundance of microplastics at different depths in sediment cores (C1 and C2) from Andong salt marsh, Hangzhou Bay.

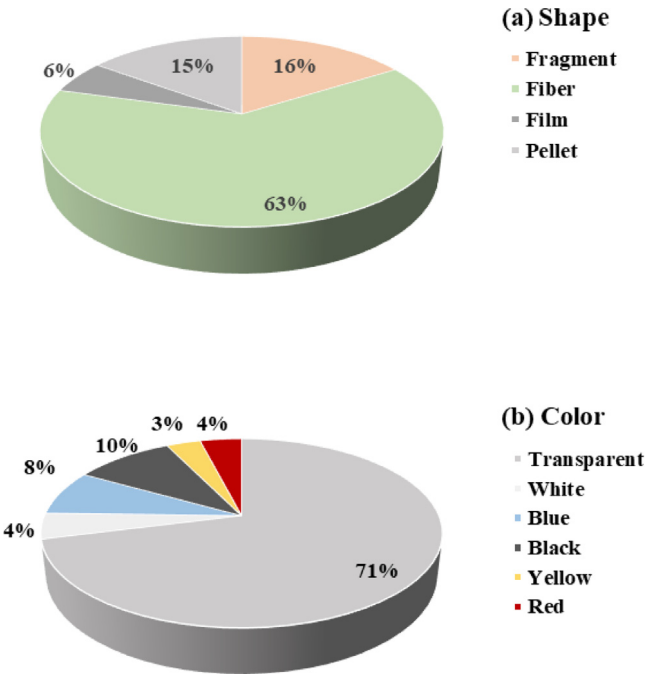

Fig. 3. Microplastic abundance in sediments from Andong salt marsh, Hangzhou Bay, categorized by (a) shape, (b) color . (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

(12.9%), PET (6.9%), cellulose (6.0%), and polyacrylonitrile (3.7%). The μ-FTIR spectrum of each type of microplastic is provided in Fig. 5. In addition, no polyvinyl chloride was detected, which meant that the polyvinyl chloride core tube had little influence on the experiment.

4. Discussion

Fifty-three sediment samples extracted at different depths from two sediment cores (C1 and C2) collected in Andong salt march, Hangzhou Bay, were examined for the presence of microplastics. The average abundance of microplastics was 13.2 ± 6.0 items 50 g^{-1} dry weight, with total microplastic concentrations of 265 items kg^{-1} dry sediment, which was in between the results of Claessens et al. (2011) (91.9 items kg^{-1} dry sediment) for the coast of Belgium and of Matsuguma et al. (2017) (300 items kg^{-1} dry sediment) for the Malaysian coast.

This study demonstrates the utility of sediment cores for understanding the history and trends in microplastic pollution. There was a temporal trend, showing that the abundance of microplastics in the sediment cores increased over time, but with fluctuations. The reason may include a combination of temporally variable inputs of microplastics, the rapid rate of sedimentation, and the natural (e.g., bioturbation) and anthropogenic (e.g., dredging) disturbance of sediment (Matsuguma et al., 2017). Two extreme climate events occurred within the time span of the cores. Fig. 2 shows an increase, especially in core C2 samples, in 1994, 1998 and 2001, when the number of microplastics was even more abundant than in 2015. This might be attributable to the super typhoon "Winnie" in 1997, which brought an extreme high tide in Hangzhou Bay and the Changjiang Estuary. Increased discharge from the river after the super typhoon can bring a lot of plastic waste to the tidal area, resulting in a higher concentration. The high tide event caused plastic waste to be washed ashore and possibly resulted in an apparent increase in the quantity of microplastics in the sediment cores. Corresponding to the layer of 42.5 cm, another distinctive event was the dry year of 2006, which led to a slight decrease of microplastics. The results related to these events were consistent with the temporal trends show in Fig. 2. In the deep layer (132.5–162.5 cm), dated to 1980s, few microplastics were observed. Plastics were invented before 1900, but it was not until the 1940s and 1950s that mass production of everyday plastic items really commenced (Thompson et al., 2009). At that time, China was less developed and most plastics were imported. The consumption of plastics would thus have been limited. Claessens et al. (2011) also used vertically sectioned sediment cores from beaches at Groenendijk and Koksijde-Bad on a time base of 15 years (1993–2008) to study the evolution of microplastic concentrations with time. The increase in microplastic concentrations was compared to the increase in annual global plastic production and similar trends were observed. A study at Kamilo Beach, a heavily polluted beach in the Hawaiian Archipelago, showed that plastic fragments occurred mostly (95%) in the top 15 cm of sediment cores (Carson et al., 2011). This finding suggests that sediment is a significant sink for microplastics in the marine environment, and the abundance of microplastics increases from deeper (older) to surface (younger) layers of the sediment.

As reported in other studies, fibers represented the highest proportion (63%) compared to other shapes of microplastic particles in this study (Fig. 3a). This was also observed in sediment collected from other similar sampling sites, like Changjiang Estuary in China (93%) (Peng et al., 2017), bay sediments in South Africa (90.07%) (Matsuguma et al., 2017) offshore sediments from the Bohai and the Yellow Seas in China (93.88%) (Zhao et al., 2018), and along the south-eastern coastline of South Africa (>

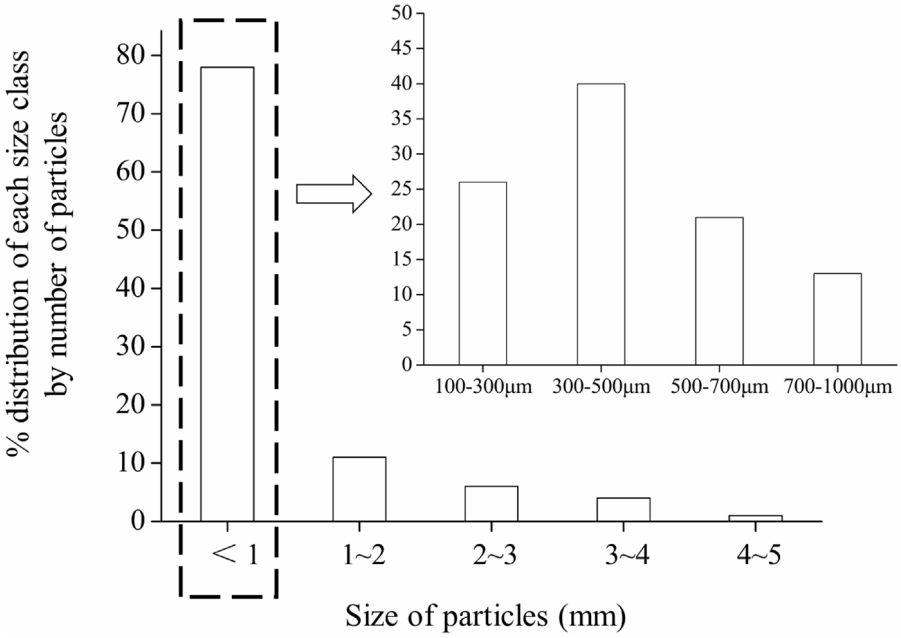

Fig. 4. Proportion (%) of microplastics in different size classes in Andong salt marsh, Hangzhou Bay. Embedded image shows the distribution of microplastics smaller than 1 mm subdivided into four groups: 100–300 μm, 300–500 μm, 500–700 μm, 700–1000 μm.

90%) (Nel and Froneman, 2015). The coastal areas with high human population density receive anthropogenic pressures from engineering constructions, local aquaculture, as well as industrial activities from adjacent towns and cities, which can generate enormous amounts of fiber-type microplastics. In addition, a portion of microfibers may originate from marine activities, such as fisheries, because fishing nets are mostly made from fibers (Zhang et al., 2019a).

With regard to the size of microplastic particles, 78% were < 1000 μm, and particles 300–500 μm in size were most commonly observed under stereo-microscope, accounting for 40% of particles < 1 mm (Fig. 4). As shown in the distribution histogram of all size fractions, the proportion of the observed microplastic particles increased with decreasing particle size, with the major quantity < 1 mm. Other studies also found that plastic particles < 1 mm in size are dominant among identified microplastics (Nor and Obbard, 2014; Zhao et al., 2018). The small size of microplastics mean they can easily be mistakenly ingested by marine organisms and may pose a potential threat to these biota (Lusher et al., 2015). The main potential harm relevant to ingestion of microplastics by birds, fish and invertebrates is attenuated foraging ability and feeding stimulus, loss of nutrition, intestinal blockage, and even mortality (Lu et al., 2016; Peda et al., 2016; Watts et al., 2015). Furthermore, microplastics have large surface area-to-volume ratios, and can act as carriers of harmful compounds from the surroundings, such as chemical additives, toxic metals and persistent organic pollutants (Andrady, 2011; Frias et al., 2010; Holmes et al., 2012; Mato et al., 2001; Zhang et al., 2018). Therefore, ingestion of microplastics can influence the organisms through both physical blockage and biochemical process by adsorption of contaminants from the environment (Brennecke et al., 2016; Koelmans et al., 2013).

Seven types of polymers were detected in this study, including PS, cellophane, PE, PP, PET, cellulose, and polyacrylonitrile (Fig. 5). Among them, PS, which is most commonly used in packaging and industrial insulation (Mathalon and Hill, 2014), was especially abundant in this study. PS was also most readily detected in sediments in Singapore's coastal marine environment (Ng and Obbard, 2006). The harbors of the Belgian coastal zone have

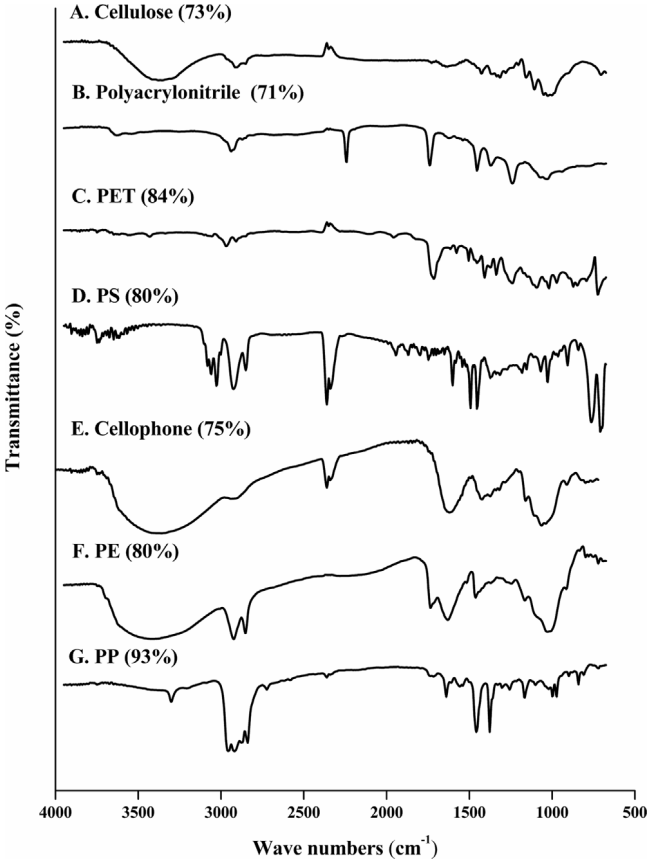

Fig. 5. μ-FTIR spectra and degrees of match for microplastics in sediments from Andong salt marsh, Hangzhou Bay.

high concentrations of PS pellets, which may originate from the plastics industry (Claessens et al., 2011). Certain types of hand sanitizers, cosmetic preparations and some jet cleaning media

may be the source of these particles, as these products may contain granular PE, PP or PS particles to act as a detergent (Gregory, 1996; Zitko and Hanlon, 1991). Cellophane, an organic cellulose-based polymer, often appears in food packaging and is used as a releasing agent in fiberglass and rubber production, was the second most dominant polymer type (16.6%) in this study. It has also been found as the main microplastic type in some sampling sites in the Solent estuary complex in the United Kingdom (Gallagher et al., 2016) and in offshore sediments in the Southern Yellow Sea and East China Sea (Zhang et al., 2019a).

5. Conclusions

This study investigated microplastic pollution in 53 sediment samples extracted from two sediment cores collected in Andong salt marsh, Hangzhou Bay. The microplastic abundance in sediment layers from cores C1 and C2 varied from 4.0–21.0 and 2.0–24.0 items 50 g^{-1} dry weight respectively, with an average abundance in cores C1 and C2 of 14.8 $\pm$ 4.9 and 12.2 $\pm$ 6.3 items 50 g^{-1} dry weight. The quantity of microplastics decreased with increasing depth, showing an evolution of microplastic abundance with time that is consistent with the increase in the production and use of microplastics over time. Extreme climate events and human activities would have affected the deposition and accumulation of microplastics. Fibers were the most frequently observed shape, and particles $<$ 1 mm were the most common size. PS, cellophane, PE and PP based microplastics were common, with PS most abundant in the cores. This is the first study to report the distribution and abundance of microplastics in the sediments of Andong salt marsh, Hangzhou Bay. The results provide useful information on microplastic pollution in sediments of the salt marsh, and contribute to the database of microplastic pollution in China.

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.

CRediT authorship contribution statement

Jingjing Li: Software, Validation, Formal analysis, Investigation, Data curation, Writing - original draft. **Wei Huang:** Investigation, Project administration. **Yongjiu Xu:** Methodology, Project administration. **Aimin Jin:** Resources, Visualization. **Dongdong Zhang:** Conceptualization, Visualization, Writing - review & editing. **Chunfang Zhang:** Conceptualization, Writing - review & editing, Supervision, Project administration, Funding acquisition.

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References

Andrady, A.L., 2011. Microplastics in the marine environment. Mar. Pollut. Bull. 62, 1596–1605.
Brennecke, D., Duarte, B., Paiva, F., Caçador, I., Canning-Clode, J., 2016. Microplastics as vector for heavy metal contamination from the marine environment. Estuar. Coast. Shelf Sci. 178, 189–195.
Bulletin of Marine ecological Environmental Quality of China, 2017. State oceanic adminstration.
Carson, H.S., Colbert, S.L., Kaylor, M.J., McDermid, K.J., 2011. Small plastic debris changes water movement and heat transfer through beach sediments. Mar. Pollut. Bull. 62, 1708–1713.
Claessens, M., De Meester, S., Van Landuyt, L., De Clerck, K., Janssen, C.R., 2011. Occurrence and distribution of microplastics in marine sediments along the Belgian coast. Mar. Pollut. Bull. 62, 2199–2204.
Davison, P., Asch, R.G., 2011. Plastic ingestion by mesopelagic fishes in the North Pacific Subtropical Gyre. Mar. Ecol. Prog. Ser. 432, 173–180.
Fang, H., Huang, L., Wang, J., He, G., Reible, D., 2016. Environmental assessment of heavy metal transport and transformation in the Hangzhou Bay, China. J. Hazard. Mater. 302, 447–457.
Frias, J.P., Sobral, P., Ferreira, A.M., 2010. Organic pollutants in microplastics from two beaches of the Portuguese coast. Mar. Pollut. Bull. 60, 1988–1992.
Gallagher, A., Rees, A., Rowe, R., Stevens, J., Wright, P., 2016. Microplastics in the Solent estuarine complex, UK: An initial assessment. Mar. Pollut. Bull. 102, 243–249.
Gregory, M., 1996. Plastic 'scrubbers' in hand cleansers a further (and minor) source for marine pollution identified. Mar. Pollut. Bull. 86, 7–871.
Holmes, L.A., Turner, A., Thompson, R.C., 2012. Adsorption of trace metals to plastic resin pellets in the marine environment. Environ. Pollut. 160, 42–48.
Koelmans, A.A., Besseling, E., Wegner, A., Foekema, E.M., 2013. Plastic as a carrier of POPs to aquatic organisms: a model analysis. Environ. Sci. Technol. 47, 7812–7820.
Laglbauer, B.J.L., Franco-Santos, R.M., Andreu-Cazenave, M., Brunelli, L., Papadatou, M., Palatinus, A., Grego, M., Deprez, T., 2014. Macrodebris and microplastics from beaches in Slovenia. Mar. Pollut. Bull. 89, 356–366.
Laist, D.W., 1987. Overview of the biological effects of lost and discarded plastic debris in the marine environment. Mar. Pollut. Bull. 18, 319–326.
Lazar, B., Gracan, R., 2011. Ingestion of marine debris by loggerhead sea turtles, Caretta caretta, in the Adriatic Sea. Mar. Pollut. Bull. 62, 43–47.
Loh, P.S., Cheng, L.-X., Yuan, H.-W., Yang, L., Lou, Z.-H., Jin, A.-M., Chen, X.-G., Lin, Y.-S., Chen, C.-T.A., 2018. Impacts of human activity and extreme weather events on sedimentary organic matter in the Andong salt marsh, Hangzhou Bay, China. Cont. Shelf Res. 154, 55–64.
Lu, Y., Zhang, Y., Deng, Y., Jiang, W., Zhao, Y., Geng, J., Ding, L., Ren, H., 2016. Uptake and accumulation of polystyrene microplastics in Zebrafish (Danio rerio) and toxic effects in liver. Environ. Sci. Technol. 50, 4054–4060.
Lusher, A.L., Hernandez-Milian, G., O'Brien, J., Berrow, S., O'Connor, I., Officer, R., 2015. Microplastic and macroplastic ingestion by a deep diving, oceanic cetacean: The True's beaked whale Mesoplodon mirus. Environ. Pollut. 199, 185–191.
Lusher, A.L., McHugh, M., Thompson, R.C., 2013. Occurrence of microplastics in the gastrointestinal tract of pelagic and demersal fish from the English Channel. Mar. Pollut. Bull. 67, 94–99.
Mathalon, A., Hill, P., 2014. Microplastic fibers in the intertidal ecosystem surrounding Halifax Harbor, Nova Scotia. Mar. Pollut. Bull. 81, 69–79.
Mato, Y., Isobe, T., Takada, H., Kanehiro, H., Ohtake, C., Kaminuma, T., 2001. Plastic resin pellets as a transport medium for toxic chemicals in the marine environment. Environ. Sci. Technol. 35, 318–324.
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.
Nel, H.A., Froneman, P.W., 2015. A quantitative analysis of microplastic pollution along the south-eastern coastline of South Africa. Mar. Pollut. Bull. 101, 274–279.
Ng, K.L., Obbard, J.P., 2006. Prevalence of microplastics in Singapore's coastal marine environment. Mar. Pollut. Bull. 52, 761–767.
Nor, N.H., Obbard, J.P., 2014. Microplastics in Singapore's coastal mangrove ecosystems. Mar. Pollut. Bull. 79, 278–283.
Peda, C., Caccamo, L., Fossi, M.C., Gai, F., Andaloro, F., Genovese, L., Perdichizzi, A., Romeo, T., Maricchiolo, G., 2016. Intestinal alterations in European sea bass Dicentrarchus labrax (Linnaeus, 1758) exposed to microplastics: Preliminary results. Environ. Pollut. 212, 251–256.
Peng, X., Chen, M., Chen, S., Dasgupta, S., Xu, H., Ta, K., Du, M., Li, J., Guo, Z., Bai, S., 2018. Microplastics contaminate the deepest part of the world's ocean. Geochem. Perspect. Lett. 1–5.
Peng, G., Zhu, B., Yang, D., Su, L., Shi, H., Li, D., 2017. Microplastics in sediments of the Changjiang Estuary, China. Environ. Pollut. 225, 283–290.
PlasticsEurope, 2018. An analysis of european plastics production, demand and waste data.

Shim, W.J., Hong, S.H., Eo, S.E., 2017. Identification methods in microplastic analysis: a review. Anal. Methods 9, 1384–1391.
Thompson, R.C., Olsen, Y., Mitchell, R.P., Davis, A., Rowland, S.J., John, A.W.G., McGonigle, D., Russell, A.E., 2004. Lost at sea: where is all the plastics? Science 304, 838.
Thompson, R.C., Swan, S.H., Moore, C.J., vom Saal, F.S., 2009. Our plastic age. Phil. Trans. R. Soc. B 364, 1973–1976.
Waller, C.L., Griffiths, H.J., Waluda, C.M., Thorpe, S.E., Loaiza, I., Moreno, B., Pacherres, C.O., Hughes, K.A., 2017. Microplastics in the Antarctic marine system: An emerging area of research. Sci. Total Environ. 598, 220–227.
Wang, Z., Wang, Z., Vriend, H.J.d., 2008. Impact of water diversion on the morphological development of the Lower Yellow River. Int. J. Sedim. Res. 23, 13–27.
Watts, A.J., Urbina, M.A., Corr, S., Lewis, C., Galloway, T.S., 2015. Ingestion of plastic microfibers by the crab carcinus maenas and its effect on food consumption and energy balance. Environ. Sci. Technol. 49, 14597–14604.
Woodall, L.C., Sanchez-Vidal, A., Canals, M., et al., 2014. The deep sea is a major sink for microplastic debris. R. Soc. Open Sci. http://dx.doi.org/10.1098/rsos.140317.
Yuan, H.-W., Chen, J.-F., Ye, Y., Lou, Z.-H., Jin, A.-M., Chen, X.-G., Jiang, Z.-P., Lin, Y.-S., Chen, C.-T.A., Loh, P.S., 2017. Sources and distribution of sedimentary organic matter along the andong salt marsh, Hangzhou Bay. J. Mar. Syst. 174, 78–88.
Zhang, D., Cui, Y., Zhou, H., Jin, C., Yu, X., Xu, Y., Li, Y., Zhang, C., 2019b. Microplastic pollution in water, sediment, and fish from artificial reefs around the Ma'an Archipelago, Shengsi, China. Sci. Total Environ. 703, 134768.
Zhang, Y., Du, J., Zhao, X., et al., 2014. A multi-proxy study of sedimentary humic substances in the salt marsh of the Changjiang Estuary, China. Estuar. Coast. Shelf Sci. 151, 295–301.
Zhang, C., Zhou, H., Cui, Y., Wang, C., Li, Y., Zhang, D., 2019a. Microplastics in offshore sediment in the Yellow Sea and East China Sea, China. Environ. Pollut. 244, 827–833.
Zhang, H., Zhou, Q., Xie, Z., Zhou, Y., Tu, C., Fu, C., Mi, W., Ebinghaus, R., Christie, P., Luo, Y., 2018. Occurrences of organophosphorus esters and phthalates in the microplastics from the coastal beaches in north China. Sci. Total Environ. (1505), 616-617-1512.
Zhao, J., Ran, W., Teng, J., Liu, Y., Liu, H., Yin, X., Cao, R., Wang, Q., 2018. Microplastic pollution in sediments from the Bohai Sea and the Yellow Sea, China. Sci. Total Environ. 6640–41, 637–645.
Zhao, S., Zhu, L., Wang, T., Li, D., 2014. Suspended microplastics in the surface water of the Yangtze Estuary System, China: first observations on occurrence, distribution. Mar. Pollut. Bull. 86, 562–568.
Zhou, Q., Zhang, H., Fu, C., Zhou, Y., Dai, Z., Li, Y., Tu, C., Luo, Y., 2018. The distribution and morphology of microplastics in coastal soils adjacent to the Bohai Sea and the Yellow Sea. Geoderma 322, 201–208.
Zitko, V., Hanlon, M., 1991. Another source of pollution by plastics: skin cleaners with plastics scrubbers. Mar. Pollut. Bull. 22, 41–42.