ELSEVIER

Contents lists available at ScienceDirect

# **Environmental Pollution**

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



# Microplastics in offshore sediment in the Yellow Sea and East China Sea. China<sup>★</sup>



Chunfang Zhang  $^{a, b, 1}$ , Hanghai Zhou  $^{a, 1}$ , Yaozong Cui  $^b$ , Chunsheng Wang  $^c$ , Yanhong Li  $^b$ , Dongdong Zhang  $^{a, *}$ 

- <sup>a</sup> Institute of Marine Biology, Ocean College, Zhejiang University, Zhoushan, 316021, Zhejiang, China
- <sup>b</sup> College of Environmental Science and Engineering, Guilin University of Technology, Guilin, 541006, China
- <sup>c</sup> Second Institute of Oceanography, State Oceanic Administration, Hangzhou, China

#### ARTICLE INFO

Article history:
Received 5 August 2018
Received in revised form
23 October 2018
Accepted 24 October 2018
Available online 25 October 2018

Keywords: Marine pollution Microplastics Offshore sediments Southern Yellow Sea East China sea

#### ABSTRACT

Microplastic particles are a global concern due to their widespread and growing threat to marine and coastal environments. To improve knowledge of microplastic pollution in China, we investigated 25 sediment samples collected with a box corer in the Southern Yellow Sea and East China Sea off the coast of China. The microplastics were extracted from sediments via density separation, after which they were observed under a microscope and characterized according to shape, color, and size, while polymer type identification was performed using micro-Fourier transform infrared spectroscopy. The abundance of microplastics in the offshore region of the Southern Yellow Sea and East China Sea was mapped. The mean concentration of microplastics at the 25 sites was  $13.4 \pm 0.6$  particles  $100 \, \text{g}^{-1}$  dry weight (range: 6.0 - 24.0 particles  $100 \, \text{g}^{-1}$  dry weight). Based on the categorization according to shape, color, and size, fiber (77%) was the most abundant shape, while blue (35%) and transparent (29%) were the most prevalent colors. In addition, the dominant size of microplastics was smaller than  $1000 \, \mu m$  which accounted for 89%. Finally, polyethylene, polyethylene terephthalate, acrylic, polyester, cellulose, and cellophane were the most abundant types of microplastics identified. Our result highlighted the presence of microplastics in offshore sediments from the Yellow Sea and East China Sea, and provided useful information for evaluating the environmental risks posed by microplastics in China.

© 2018 Elsevier Ltd. All rights reserved.

# 1. Introduction

Microplastic particles are ubiquitous in marine environments, as well as in soils, rivers, lakes, and even ice (Gall and Thompson, 2015; Klein et al., 2015; Obbard et al., 2014; Yonkos et al., 2014). Thus, they have become a global concern due to their widespread and growing threat to the environment, with the potential for detrimental impacts on human health.

Microplastics in marine environments are potentially toxic and can adversely affect organisms throughout the food web, although their ecological and public health effects have not been fully elucidated. Studies suggest that microplastic toxicity might be attributable to their physical and chemical properties. For example,

microplastics may release monomers (e.g., bisphenol A, styrene, and vinyl chloride) or additives mixed with monomers during the manufacturing process to improve the physical and chemical properties of plastics (e.g., plasticizers, stabilizers, and flame retardants) (Ivar do Sul and Costa, 2014), some of which have endocrine-disrupting effects (Meeker et al., 2009; Yang et al., 2011). In addition, since microplastics have large surface area-to-volume ratios, they can readily absorb pollutants from the surrounding environment, especially trace metals and persistent organic pollutants (POPs), acting as a pollutant transport medium (Andrady, 2011; Cole et al., 2011; Frias et al., 2010; Ivar do Sul and Costa, 2014; Teuten et al., 2009). Meanwhile, microplastics support microbial colonization and can serve as vectors for microbial species dispersal since plastics can migrate rapidly among marine habitats. This could also result in the spread of pathogens or invasive species (McCormick et al., 2014). Most importantly, microplastic particles are small and can be readily ingested by marine organisms, including plancton, bivalves, polychaetes, crustaceans, fish, and

<sup>\*</sup> This paper has been recommended for acceptance by Eddy Y. Zeng.

<sup>\*</sup> Corresponding author.

E-mail address: zhangdongzju@zju.edu.cn (D. Zhang).

<sup>&</sup>lt;sup>1</sup> These authors contributed equally to this study and are joint first authors.

seabirds, which may lead to adverse effects (lvar do Sul and Costa, 2014; Lourenço et al., 2017; Setala et al., 2014; Tanaka et al., 2013; Teuten et al., 2009). Overall, microplastics and their associated risk potential could have greater detrimental effects on ecosystems and human health than large plastic debris, since they tend to bioaccumulate and biomagnify along the food web (lvar do Sul and Costa, 2014; Wright et al., 2013).

The widespread occurrence of microplastics in various marine environments is well documented, although their presence in sediments has mainly been investigated in beaches, coastal areas, estuaries, harbors/ports, and deep-sea sediments (Browne et al., 2011; Laglbauer et al., 2014; Mathalon and Hill, 2014; Peng et al., 2017; Woodall et al., 2014). By contrast, little research (Zhao et al., 2018) is available on offshore sediments, even though floating plastic debris and associated microplastics tend to accumulate far from continental margins. Hence, the presence and distribution of microplastics in offshore sediments should be clarified, since surface plastic material can be rapidly transported to further seafloor sediments. Moreover, even though the coastal region of China along the Southern Yellow Sea and East China Sea is one of the most densely urbanized and industrialized zones in China, the study by Zhao et al. (2018) is the only report that investigate the abundance and distribution of microplastics in sediments in offshore regions of Bohai Sea and Yellow Sea. Until now, the microplastics pollution in sediments from offshore regions of East China Sea has not been reported yet.

Therefore, the aim of this study was to investigate the occurrence and distribution of microplastics in sediments from offshore regions in the Southern Yellow Sea and East China Sea. The extracted microplastics were characterized according to abundance, shape, color, and size via observations under a stereoscopic microscope. Finally, the polymer types were identified using micro-Fourier transform-infrared ( $\mu$ -FTIR) spectroscopy.

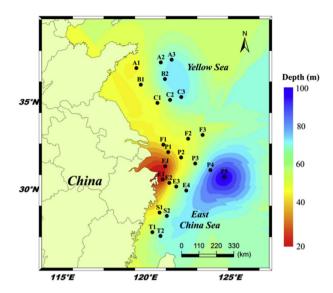
#### 2. Materials and methods

#### 2.1. Sampling

Samples were collected from 25 locations using a box corer in March 2017 during a cruise along the coast of the Southern Yellow Sea and East China Sea, China (Fig. 1). Table 1 presents the depth information of the sampling sites, along with the sediment fractions and types. Sediments in the upper 10 cm were collected from each location and stored in precleaned bottles. All high-density polyethylene sample bottles were rinsed with Milli-Q water before use to migrate the contamination and carefully stored in boxes before boarding the vessel. The bottles containing samples were stored at room temperature until analysis, and clean stainless-steel spoons were used for removing samples from the bottles. Two replicates were collected from each sampling site.

# 2.2. Microplastic extraction from sediments

Microplastics were extracted from the sediment samples using the method employed by (Thompson et al., 2004), with some modifications. The sediment samples were dried at 70 °C for 24 h to a constant weight, and an aliquot of 100 g was taken from each site. Then, the samples were transferred into clean glass beakers. Approximately 150 mL of 30%  $\rm H_2O_2$  (Sinopharm, China) was added to each bottle to digest organic matter (e.g., diatoms). Samples were dried again at 70 °C for 24 h to achieve a constant weight. Since no visible plastics were observed in the sediment, the samples were not passed through sieves before filtration. For the density separation, NaI (Sinopharm, China) was dissolved in Milli-Q water to prepare a concentrated solution with a density of 1.6 g cm $^{-3}$ , as



**Fig. 1.** Locations of the 25 sampling sites in the offshore regions of Southern Yellow Sea and East China Sea. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

described by (Van Cauwenberghe et al., 2013b). The solution was transferred to a beaker and manually stirred with a clean glass rod for 2 min. For filtration, the supernatant was settled for 24 h before transferring onto a piece of 8-µm pore-size, 47-mm cellulose nitrate filter paper (Shanghai Xingya, China) with the aid of a vacuum pump and rinsed with Milli-Q water. Finally, the filter paper was dried at 40 °C for 24 h before microscopic inspection. The filter paper was then placed in a clean Petri dish with an aluminum cover and dried at room temperature to observe the total number of particles. Control experiment was carried out by filtrating the ultrapure water through nitrate cellulose paper filters and no microplastics was observed under microscopy. Thus, the contamination from the nitrate cellulose paper filters could be excluded.

# 2.3. Microscopic inspection

The microplastic particles on the filters were optically analyzed and photographed using a digital microscope (Sunny Optical Technology, China) equipped with MvImage software at up to  $\times$  160 magnification. A visual assessment was performed to identify the shape, color, and size of microplastics according to the physical characteristics of the particles. Particle shape was categorized into four groups: fibers (fibrous or thin uniform plastic strands), fragments (hard, jagged-edged particles), films (thin, two-dimensional plastic films), and micro-pellets (hard, rounded particles).

Particle color was divided into six categories: transparent. white, blue, black, yellow, and red. The blue category included deep blue, light blue, deep green, and light green particles. The black category included transparent black, gray, and white-striped black particles. The yellow category also included orange and brown particles. The transparent category included colorless particles, while the white category included silver particles. The red category also included pink and purple particles. Some particles were randomly selected for verification using μ-FTIR. Mesoplastic particles longer than 5000 µm were not included in the results. The microplastic density was expressed as the number of particles per 100 g of dry sediment to enable comparison with other studies. Particles in the size range of  $60-200 \,\mu\text{m}$ ,  $200-500 \,\mu\text{m}$ , 500-1000 μm and 1000-5000 μm were analyzed since size larger than 60  $\mu m$  was necessary for the  $\mu$ -FTIR analysis according to our experience.

**Table 1**Sampling site descriptions, sediment fractions, and sediment types.

Site	Latitude (E)	Longitude (N)	Depth (m)	Sand (%)	Mud (%)	Clay (%)	Mean grain size (ø)	Sediment type
A1	36.00°	122.15°	45	35.07	40.18	24.75	5.49	Silty mud
A2	36.00°	123.95°	72	12.12	55.21	32.67	7.06	Silty mud
A3	36.00°	124.75°	78	68.71	20.07	11.22	4.08	Pelitic silt
B1	35.00°	122.20°	54	12.61	55.73	31.65	6.79	Silty mud
B2	35.00°	123.95°	80	66.24	19.74	14.02	3.49	Pelitic silt
C1	33.75°	123.05°	50	41.53	41.61	15.28	5.03	Sandy silt
C2	33.75°	123.95°	71	35.15	39.60	24.54	5.79	Silty mud
C3	33.75°	124.75°	80	28.75	41.79	28.88	5.78	Silty mud
E1	29.40°	122.25°	10	14.39	57.13	28.48	6.74	Sandy silt
E2	29.12°	122.62°	52	24.93	48.84	26.22	5.92	Clay-silty sand
E3	28.83°	123.00°	64	33.51	41.26	25.23	5.99	Clay-silty sand
E4	28.48°	123.57°	74	62.80	23.08	14.11	4.15	Pelitic silt
P1	30.85°	123.00°	45	33.64	43.57	22.79	5.71	Silty mud
P2	30.38°	123.75°	51	72.91	18.56	8.52	3.93	Clay-silty sand
P3	29.85°	124.55°	60	71.79	17.55	10.66	3.87	Pelitic silt
P4	29.27°	125.40°	85	66.23	20.54	13.22	4.12	Pelitic silt
P5	28.70°	126.15°	115	65.81	22.23	11.95	3.93	Pelitic silt
F1	31.33°	122.80°	48	27.24	48.92	23.84	5.78	Sandy silt
F2	31.33°	124.50°	52	42.65	35.68	16.64	4.96	Sandy silt
F3	31.33°	125.50°	59	21.44	51.38	27.18	6.01	Clay-silty sand
FJ	30.10°	122.60°	17	31.06	45.06	23.88	5.83	Clay-silty sand
S1	27.60°	121.60°	41	0.00	63.82	36.18	7.51	Mud
S2	27.33°	122.00°	81	67.94	21.65	10.41	4.05	Clay-silty sand
T1	26.60°	120.90°	60	40.61	41.13	18.27	5.23	Sandy silt
T2	26.30°	121.35°	79	63.25	28.14	8.61	4.10	Clay-silty sand

# 2.4. Identification of microplastics with $\mu$ -FTIR

Plastic-like particles on the filter paper were randomly selected for verification using μ-FTIR (Nicolet 6700; Thermo Scientific, USA) in transmittance mode. Microplastic particles were placed onto a KBr surface. The detector spectral range was 650–4000 cm<sup>-1</sup>, with a collection time of 28 s and co-addition of 64 scans at a resolution of  $8 \,\mathrm{cm}^{-1}$ . The aperture was set at  $100-100 \,\mathrm{\mu m}$ . The spectra were processed with OMNICTM PictaTM software (Thermo Fisher Scientific) and compared with the OMNIC polymer spectra library. Some suspected microplastic particles were also identified under a μ-FTIR microscope in attenuated total reflection (ATR) mode. All spectra were collected at a resolution of 4 cm<sup>-1</sup> using a diamond MicroTip accessory at 675–4000 cm<sup>-1</sup>, with a collection time of 24 s and with 20 co-scans. The spectra were compared with FTIR spectral libraries (see Table 2) to verify the polymer type. The spectrum analysis followed the method of (Woodall et al., 2014), where matches with a quality index > 0.7 were accepted and those with a quality index < 0.7 were rejected.

# 2.5. Grain size distribution

The grain size distribution of the sediment was analyzed with a laser particle-size analyzer (MS300; Malvern Instruments, Malvern, UK). The relationship between grain size and microplastic

abundance was calculated using Spearman's correlation analysis (Nor and Obbard, 2014; Peng et al., 2017; Thompson et al., 2004).

# 2.6. Contamination control

All instruments used during the extraction process were rinsed with Milli-Q water and dried before the experiments. The use of plastic equipment was avoided or replaced with non-plastic utensils where possible. Cotton lab coats and polymer-free gloves were always worn to ensure sterility during sample handling. In addition, clothing made from synthetic fibers were avoided during the experiments (Browne et al., 2011; Cozar et al., 2015). Procedural blanks were included to check for background contamination from laboratory sources via the air, clothes, sampling tools, vessels, etc. The blanks were set up for 2, 5, and 10 weeks over the full course of the laboratory work. The entire extraction process was performed in a clean fume hood to prevent contamination with airborne microplastics.

# 3. Results

#### 3.1. Procedural blanks assessment

The contamination control steps were strictly followed during the process of sample handling to prevent the contamination with

**Table 2** Information on the identified microplastics.

Type of polymer	Number	Proportion of total particles (%)	Density (g cm <sup>-3</sup> )	FTIR Library
Cellophane	19	37.2	1.50-1.52	Synthetic Fibers by Microscope
				Hummel Polymer and Additives
Polyethylene terephthalate	11	21.6	1.37	Cross Section Wizard
				Synthetic Fibers by Microscope
Polyethylene	9	17.6	1.05	Aldrich Linked IR
Polyester	6	11.8	1.37	Synthetic Fibers by Microscope
Acrylic	5	9.8	1.18	Synthetic Fibers by Microscope
Cellulose	1	2.0	1.22	Hummel Polymer and Additives
Total	51	_	_	_

airborne microplastics. In the procedural blanks,  $1.2 \pm 1.0$  items/sample was found which were all in the form of fibers. This result indicated minor contamination from airborne microplastics.

#### 3.2. Microplastic abundance and distribution in offshore sediments

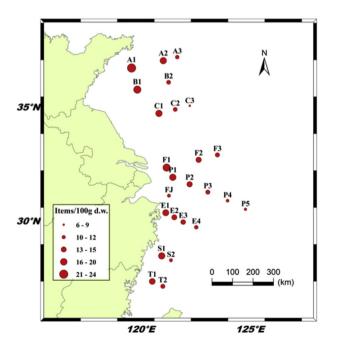
Microplastic particles were detected in all 25 sediment samples from the Southern Yellow Sea and East China Sea, yielding a total of 366 particles. As shown in Fig. 2, the amount of microplastics in the sites near shoreline were higher than that in the further places, indicating that microplastics abundance decreases with distance from the shore. The microplastic abundances of different sampling sites varied from 6.0 to 24.0 particles  $100\,\mathrm{g}^{-1}$  dry weight (d.w.) sediment (Fig. 2), with an average abundance of  $15.5\pm6.1$  and  $14.2\pm3.8$  particles  $100\,\mathrm{g}^{-1}$  d.w. sediment for Yellow Sea and East China Sea, respectively.

# 3.3. Microplastic shape, color, and size

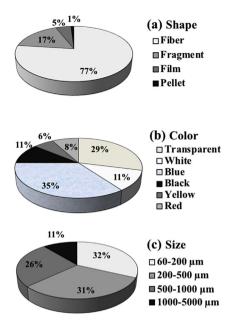
Among the 366 microplastic particles identified, fiber was the most commonly observed shape, accounting for 77% of total particles, followed by fragments (17%), films (5%), and pellets (1%) (Fig. 3a). Regarding particle color, blue and transparent microplastic particles were relatively abundant, accounting for 64% in total (Fig. 3b), followed by black, white, red, and yellow particles (Fig. 3b). Overall, 89% of microplastic particles were <1000  $\mu m$  (Fig. 3c). Among them, particles in the size range of 60–200  $\mu m$ , 200–500  $\mu m$ , and 500–1000  $\mu m$  were relatively evenly distributed, accounting for 32%, 31%, and 26% of total particles, respectively. By contrast, particles in the size range of 1000–5000  $\mu m$  only accounted for 11% of particles (Fig. 3c). Overall, particle size ranged from 19.5 to 4953.2  $\mu m$  (mean: 439.5  $\pm$  298.4  $\mu m$ ).

#### 3.4. Sediment grain size distribution and microplastic abundance

Table 1 presents information on the sediments, including grain



**Fig. 2.** Microplastic abundance in sediments in the Southern Yellow Sea and East China Sea off the coast of China. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)



**Fig. 3.** Microplastic abundance in sediments from the Southern Yellow Sea and East China Sea categorized by (a) shape, (b) color, and (c) size. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

size distribution. The statistical analysis indicated that microplastic distribution was significantly correlated with the grain size of sediment (Spearman's correlation, r=0.454, p=0.024<0.05, n=25).

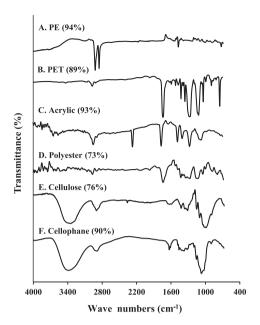
Five different sediment types were identified among the 25 sediment samples, i.e., mud (1/25), silty mud (6/25), pelitic silt (6/25), sandy silt (5/25), as well as clay-silty sand (7/25) (Table 1). The average abundance of microplastics (particles  $100 \, \text{g}^{-1}$  dry weight) differed among sediment types, with the highest value for mud (19 particles, n=1), followed by sandy silt (18.2  $\pm$  1.9 particles, n=5), silty mud (16.8  $\pm$  6.6 particles, n=6), and clay-silty sand (13.0  $\pm$  2.5 particles, n=7), with the lowest for pelitic silt (10.7  $\pm$  1.9 particles, n=6).

# 3.5. Identification of microplastics using $\mu$ -FTIR

In total, 51 particles were identified as microplastics, representing six polymer types: polyethylene (PE), polyethylene terephthalate (PET), acrylic, polyester, cellulose, and cellophane (Fig. 4). Table 2 presents the total number of each microplastic polymer type identified in the samples. Cellophane was the most abundant polymer type, accounting for 37.2% of the total identified particles. Meanwhile, the proportion of PET, PE, polyester, acrylic, and cellulose decreased in order (Table 2).

#### 4. Discussion

In this study, microplastic particles were detected in all 25 offshore sediment samples from the Southern Yellow Sea and East China Sea, indicating the extensive dispersion of microplastics in these areas. Moreover, the microplastics abundance decreases with distance from the shore as the microplastic abundance in sampling sites close to the shore were higher than those at distant sites (Fig. 2). Meanwhile, the significance was tested by Spearman's correlation analysis, which revealed that the sediment grain size distribution was correlated with microplastic distribution (r = 0.454, p = 0.024, n = 25).



**Fig. 4.** Identification of microplastics in sediments using  $\mu$ -FTIR. Abbreviations: PE, polyethylene; PET, polyethylene terephthalate. Values in brackets indicate the matches of the spectra with the standards.

The recent study by (Zhao et al., 2018) is the only report to have investigated microplastic pollution in offshore sediments. Interestingly, they found no discernible difference between microplastic abundance between nearshore and distant sites in 11 of 13 sections examined in the Bohai Sea and Yellow Sea off the coast of China. However, these findings were not comparable with the present study, because the areas investigated differed and the distance between adjacent sampling sites in each section was further in the current study. From the perspective of microplastic concentration expressed as average particle abundance per 100 g of dry sediment, the results in this study were similar to those from sediments from Slovenian beaches (Laglbauer et al., 2014), Belgian harbors (Claessens et al., 2011), and Chinese beaches, estuaries, and offshore areas (Peng et al., 2017; Yu et al., 2016; Zhao et al., 2018). However, the microplastic abundances in an Italian lagoon (Vianello et al., 2013) and Canadian beach sediments (Mathalon and Hill, 2014) were one order of magnitude higher than those of this study, while the abundances on a beach in Singapore (Ng and Obbard, 2006) and at the high-tide line (Van Cauwenberghe et al., 2013a) and on a beach (Van Cauwenberghe et al., 2015) in Belgium were two orders of magnitude lower.

Among the shapes of microplastic particles observed in this study, fibers were the most abundant, accounting for 77% of the total particles (Fig. 3a). Similar results have been observed in coastal mangrove sediments collected from Singapore (72%) (Nor and Obbard, 2014) and Slovenia (75%) (Laglbauer et al., 2014; Mohamed Nor and Obbard, 2014). Some studies have reported even higher proportions of fibers in sediments, for example in coastal wetlands on the Easter Atlantic (99.8%) (Lourenço et al., 2017), nearshore sediments of South Africa (90%) (Nel and Froneman, 2015), and bay sediments of Croatia (90.07%) (Blašković et al., 2017). In China (Peng et al., 2017), reported that fibers accounted for 93% of the total microplastics in sediments from the Changjiang Estuary, while (Zhao et al., 2018) demonstrated that fiber-type microplastics were observed at all sediment samples in the Bohai Sea and Yellow Sea, accounting for 93.88% of all particles. In fact, it has been extensively reported that fibers account for the majority of microplastic pollution (Frias et al., 2016; Ivar do Sul and Costa, 2014), including in the deep sea (Taylor et al., 2016; Woodall et al., 2014). Human populations have always congregated in coastal areas, and frequent human activity can generate large amounts of fiber-type microplastics, such as fibers originating from the production, washing, and natural aging of textiles. Moreover, marine activities such as fisheries can represent a source of microfibers, because fishing nets are mostly made from fibers. Therefore, the high proportion of microfibers in this study was reasonable, as fibers cannot be completely removed during sewage treatment (Browne et al., 2011; Mason et al., 2016; Zhao et al., 2018).

Regarding particle size, 89% of microplastic particles were <1000 µm (Fig. 3c). The abundance of small-sized microplastics has been demonstrated in many studies (Mohamed Nor and Obbard, 2014; Peng et al., 2017; Zhao et al., 2018). Moreover, in the present study, particles <500 µm were the most frequently observed particle size (63%) (Fig. 3c), which was higher than that reported in a Chinese estuary (Peng et al., 2017) and offshore sediments in the Bohai Sea (33%) and Yellow Sea (37.87%) off the coast of China (Zhao et al., 2018). However, (Mohamed Nor and Obbard, 2014) reported that microplastics with a diameter < 1000 μm accounted for > 90% of particles in Singapore's coastal mangrove ecosystems, where 58% of the observed microplastics were <40 µm. In addition, the size of all microplastics recovered from deep-sea sediments of the Atlantic Ocean and Mediterranean Sea were within 1000 µm, with particles ranging from 75 μm to 161 μm at their largest diameter (Van Cauwenberghe et al., 2013b).

Interestingly, large particles (>5000  $\mu$ m) were rarely observed, with only one large plastic particle identified (12,200  $\mu$ m) among all sediment samples. Similarly, the large proportion of smaller-sized microplastics in Singapore mangroves was speculated to have been driven by the degradation of macroplastics, since the high temperatures and insolation associated with Singapore's tropical climate tend to accelerate the degradation of plastic debris (Mohamed Nor and Obbard, 2014; Ng and Obbard, 2006). In the case of offshore or deep-sea sediments, the prevalence of smaller-sized microplastics might be attributable to the fragmentation and degradation of larger plastic particles during the long-distance and deep-sea transport.

We used  $\mu$ -FTIR to identify suspected microplastics. Through the application and comparison using three different modes (i.e., ATR, transmission, and reflection mode), we found that ATR mode offered a better spectral response for larger microplastic particles, transmission mode showed a good response for fiber-type microplastics, and reflection mode exhibited poor performance for all selected microplastic particles. Sixty-five particles on the filter papers were selected for  $\mu$ -FTIR analysis. Among them, 78.5% were identified as microplastics, 12.3% as nonmicroplastics, and 9.2% as unidentified particles. Additionally, the overall matches are not high and even show great variations for the same or a similar sample due to the complexity of sediment samples. This could attributable to the environmental degradation and weathering of the microplastic surfaces, inefficient particle recovery, as well as misidentification of particles.

Six types of polymers were identified: PE, PET, acrylic, polyester, cellulose, and cellophane (Fig. 4). Among the identified microplastics, cellophane was the most abundant polymer type, followed by PET, polyethylene, polyester, acrylic, and cellulose (Table 2). Studies have found that PE and polypropylene (PP) are the most common polymer types in microplastics in coastal environments (Hidalgo-Ruz et al., 2012; Vianello et al., 2013; Zbyszewski et al., 2014). However, we found that cellophane and PET were the most common polymers in offshore sediments in the Southern Yellow Sea and East China Sea, while PE accounted for a relatively minor proportion and PP was undetected. This might be

due to the high density of cellophane  $(1.50-1.52\,\mathrm{g\,cm^{-3}})$  and PET (1.38 g cm<sup>-3</sup>), making them more likely to settle into sediment. Furthermore, we used a dense solution (NaI) to facilitate the separation of denser polymers. Cellophane is an organic cellulosebased polymer used widely in food packaging and as a releasing agent in fiberglass rubber production, and has also been found as the predominant microplastic type in several sampling points in the Solent estuarine complex in the United Kingdom (Gallagher et al., 2016) and in lake salts and rock/well salts from China (Yang et al., 2015). The East China Sea and Southern Yellow Sea are shallow seas on the edge of the western Pacific Ocean, the continental shelf of which are the largest and widest in China, forming a fishery with vast areas and superior conditions. Nowadays, the reports on the toxic effects of microplastics to marine organisms are accumulating, including intestinal blockage, tissue abrasion, interference with metabolism and so on (Lu et al., 2016; Pedà et al., 2016; Watts et al., 2015). The prevalence of microplastic pollution as observed in the sediments of East China Sea and Southern Yellow Sea might threaten fishery species, especially in their vulnerable early life stages, resulting in the decline of fisheries resources. Meanwhile, marine sediments act as important "sink" for pollutants, such as POPs and heavy metals, and microplastics are ready to adsorb these pollutants from surrounding environments (Brennecke et al., 2016; Zhang et al., 2015). In addition, the small size of microplastics makes it easy to be ingested by marine organisms and the toxic substance inherent and/or carried by microplastics could be transferred to various marine organisms along the food wed, eventually posing threat to human health. To better understand potential effects of microplastics, further studies should be carried out to evaluate the combined effects of microplastics and toxic substances in the East China Sea and Southern Yellow Sea. It is also important to investigate the microplastic pollution in fishery species of the East China Sea and Southern Yellow Sea as well as to evaluate the potential risks posed to human health through consuming microplastics in sea products.

# 5. Conclusions

We observed microplastic particles in all 25 offshore sediment sampling sites in the Southern Yellow Sea and East China Sea. The abundance of microplastics in sediments varied from 6.0 to 24.0 particles  $100\,\mathrm{g}^{-1}$  d.w. sediment, with an average abundance of  $15.5 \pm 6.1$  and  $14.2 \pm 3.8$  particles  $100 \,\mathrm{g}^{-1}$  d.w. sediment for Yellow Sea and East China Sea, respectively. The distribution of microplastics was positively correlated with sediment grain size (r = 0.454, p = 0.024, n = 25), and the microplastics abundance decreases with distance from the shore. Fibers were the most common microplastic shape, while microplastics with a particle size <500 μm were the most commonly observed size fraction. Six polymer types were identified, and their frequency followed the order (from highest to lowest) cellophane (19/51), PET (11/51), polyethylene (9/51), polyester (6/51), acrylic (5/51), and cellulose (1/51). The widespread occurrence of microplastics in various marine environments is generally well documented; however, few studies are available on offshore marine sediments. Therefore, our results provide useful information on the microplastic distribution in the sediments of offshore areas in the Southern Yellow Sea and East China Sea, and provide fundamental data on microplastic pollution in the sediment of offshore marine areas. The findings could also support future study in the modeling of micropastics mitigation from coastal area to remote marine environments, as well as the evaluation of the environmental risks posed by microplastics.

#### **Declarations of interest**

None.

### Acknowledgements

Funding: This study was supported by the National Natural Science Foundation of China [grant numbers 31400096, 41701346], and by the Open Foundation from Key Laboratory of Health Risk Factors for Seafood of Zhejiang Province [grant number 201702].

# Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2018.10.102.

#### References

- Andrady, A.L., 2011. Microplastics in the marine environment. Mar. Pollut. Bull. 62, 1596–1605
- Blašković, A., Fastelli, P., Čižmek, H., Guerranti, C., Renzi, M., 2017. Plastic litter in sediments from the Croatian marine protected area of the natural park of Telaščica bay (Adriatic Sea). Mar. Pollut. Bull. 114, 583–586.
- 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.
- 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, 9175–9179.
- 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.
- Cole, M., Lindeque, P., Halsband, C., Galloway, T.S., 2011. Microplastics as contaminants in the marine environment: a review. Mar. Pollut. Bull. 62, 2588.
- Cozar, A., Sanz-Martin, M., Marti, E., Gonzalez-Gordillo, J.I., Ubeda, B., Galvez, J.A., Irigoien, X., Duarte, C.M., 2015. Plastic accumulation in the Mediterranean Sea. PloS One 10.
- Frias, J.P.G.L., Gago, J., Otero, V., Sobral, P., 2016. Microplastics in coastal sediments from Southern Portuguese shelf waters. Mar. Environ. Res. 114, 24–30.
- Frias, J.P.G.L., Sobral, P., Ferreira, A.M., 2010. Organic pollutants in microplastics from two beaches of the Portuguese coast. Mar. Pollut. Bull. 60, 1988–1992.
- Gall, S.C., Thompson, R.C., 2015. The impact of debris on marine life. Mar. Pollut. Bull. 92, 170—179.
- 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.
- 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, 3060–3075.
- Ivar do Sul, J.A., Costa, M.F., 2014. The present and future of microplastic pollution in the marine environment. Environ. Pollut. 185, 352–364.
- Klein, S., Worch, E., Knepper, T.P., 2015. Occurrence and spatial distribution of microplastics in river shore sediments of the rhine-main area in Germany. Environ. Sci. Technol. 49, 6070.
- 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.
- Lourenço, P.M., Serra-Gonçalves, Ferreira, J.L., Catry, T., Granadeiro, J.P., 2017. Plastic and other microfibers in sediments, macroinvertebrates and shorebirds from three intertidal wetlands of southern Europe and west Africa. Environ. Pollut. 231, 123–133.
- 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.
- Mason, S.A., Garneau, D., Sutton, R., Chu, Y., Ehmann, K., Barnes, J., Fink, P., Papazissimos, D., Rogers, D.L., 2016. Microplastic pollution is widely detected in US municipal wastewater treatment plant effluent. Environ. Pollut. 218, 1045–1054.
- Mathalon, A., Hill, P., 2014. Microplastic fibers in the intertidal ecosystem surrounding halifax harbor, nova scotia. Mar. Pollut. Bull. 81, 69–79.
- McCormick, A., Hoellein, T.J., Mason, S.A., Schluep, J., Kelly, J.J., 2014. Microplastic is an abundant and distinct microbial habitat in an urban river. Environ. Sci. Technol. 48, 11863.
- Meeker, J.D., Sathyanarayana, S., Swan, S.H., 2009. Phthalates and other additives in plastics: human exposure and associated health outcomes. Phil. Trans. Biol. Sci. 364, 2097–2113.
- 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.
- Obbard, R.W., Sadri, S., Wong, Y.Q., Khitun, A.A., Baker, I., Thompson, R.C., 2014. Global warming releases microplastic legacy frozen in Arctic Sea ice. Earth's Future 2, 315–320.
- Pedà, 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, 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.
- Setala, O., Fleming-Lehtinen, V., Lehtiniemi, M., 2014. Ingestion and transfer of microplastics in the planktonic food web. Environ. Pollut. 185, 77–83.
- Tanaka, K., Takada, H., Yamashita, R., Mizukawa, K., Fukuwaka, M.-a., Watanuki, Y., 2013. Accumulation of plastic-derived chemicals in tissues of seabirds ingesting marine plastics. Mar. Pollut. Bull. 69, 219–222.
- Taylor, M.L., Gwinnett, C., Robinson, L.F., Woodall, L.C., 2016. Plastic microfibre ingestion by deep-sea organisms. Sci. Rep. 6, 33997.
- Teuten, E.L., Saquing, J.M., Detlef, R.U.K., Barlaz, M.A., Jonsson, S., Björn, A., Rowland, S.J., Thompson, R.C., Galloway, T.S., Yamashita, R., Ochi, D., Watanuki, Y., Moore, C., Viet, P.H., Tana, T.S., Prudente, M., Boonyatumanond, R., Zakaria, M.P., Akkhavong, K., Ogata, Y., Hirai, H., Iwasa, S., Mizukawa, K., Hagino, Y., Imamura, A., Saha, M., Takada, H., 2009. Transport and release of chemicals from plastics to the environment and to wildlife. Phil. Trans. Biol. Sci. 364. 2027—2045.
- Thompson, R.C., Olsen, Y., Mitchell, R.P., Davis, A., Rowland, S.J., Anthony, W.G.J., McGonigle, D., Russell, A.E., 2004. Lost at sea: where is all the plastic? Science 304, 838-838.
- Van Cauwenberghe, L., Claessens, M., Vandegehuchte, M.B., Janssen, C.R., 2015. Microplastics are taken up by mussels (Mytilus edulis) and lugworms (Arenicola marina) living in natural habitats. Environ. Pollut. 199, 10–17.
- Van Cauwenberghe, L., Claessens, M., Vandegehuchte, M.B., Mees, J., Janssen, C.R., 2013a. Assessment of marine debris on the belgian continental shelf. Mar. Pollut. Bull. 73, 161–169.

- Van Cauwenberghe, L., Vanreusel, A., Mees, J., Janssen, C.R., 2013b. Microplastic pollution in deep-sea sediments. Environ. Pollut. 182, 495.
- Vianello, A., Boldrin, A., Guerriero, P., Moschino, V., Rella, R., Sturaro, A., Da Ros, L., 2013. Microplastic particles in sediments of Lagoon of Venice, Italy: first observations on occurrence, spatial patterns and identification. Estuar. Coast Shelf Sci. 130, 54–61.
- Watts, A.J.R., 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., Paterson, G.L.J., Coppock, R., Sleight, V., Calafat, A., Rogers, A.D., Narayanaswamy, B.E., Thompson, R.C., 2014. The Deep Sea Is a Major Sink for Microplastic Debris, vol. 1. Royal Society Open Science, 140317-140317.
- Wright, S.L., Thompson, R.C., Galloway, T.S., 2013. The physical impacts of microplastics on marine organisms: a review. Environ. Pollut. 178, 483–492.
- Yang, C.Z., Yaniger, S.I., Jordan, V.C., Klein, D.J., Bittner, G.D., 2011. Most plastic products release estrogenic chemicals: a potential health problem that can Be solved. Environ. Health Perspect. 119, 989–996.
- Yang, D.Q., Shi, H.H., Li, L., Li, J.N., Jabeen, K., Kolandhasamy, P., 2015. Microplastic pollution in table salts from China. Environ. Sci. Technol. 49, 13622–13627.
- Yonkos, L.T., Friedel, E.A., Perez-Reyes, A.C., Ghosal, S., Arthur, C.D., 2014. Microplastics in four estuarine rivers in the Chesapeake Bay, U.S.A. Environ. Sci. Technol. 48, 14195.
- Yu, X., Peng, J., Wang, J., Wang, K., Bao, S., 2016. Occurrence of microplastics in the beach sand of the Chinese inner sea: the Bohai Sea. Environ. Pollut. 214, 722–730.
- Zbyszewski, M., Corcoran, P.L., Hockin, A., 2014. Comparison of the distribution and degradation of plastic debris along shorelines of the Great Lakes, North America. J. Great Lake. Res. 40, 288–299.
- Zhang, W., Zhang, Z., Ma, D., Ma, X., Wang, Y., Wang, J., Wang, J., 2015. Persistent organic pollutants carried on plastic resin pellets from two beaches in China. Mar. Pollut. Bull. 99, 28–34.
- Zhao, J., Ran, W., Teng, J., Liu, H., Liu, Y., Yin, X., Cao, R., Wang, Q., 2018. Microplastic pollution in sediments from the Bohai Sea and the Yellow Sea, China. Sci. Total Environ. 640–641, 637–645.