



Microplastic Pollution in Nearshore Sediment from the Bohai Sea Coastline

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Abstract

Microplastics are recognized as an emerging global issue in marine environments. In this study, microplastic pollution in subtidal sediments from nine typical stations in the Bohai Sea was investigated. The mean concentration was 458.6 ± 150.0 items/kg of dry weight, varying from 280.0 to 773.4 items/kg. All of the microplastics were categorized according to shape, color and size. Among these microplastics, fiber (77.1%), white/blue/black (85.0%) and small microplastics ($< 1500 \mu\text{m}$) (82.9%) were the most abundant types. Seven polymer types were identified and were, in decreasing order of abundance, rayon > PE > PS > PP > PET > ABS > PA. The microplastics abundance was of the same order of magnitude as that of other similar areas. The microplastic characteristics suggest that tourism, maritime activities and sewage discharge are possible sources. Our results provide useful information for performing an environmental risk assessment of microplastic pollution in this area.

Keywords Microplastic · Coast · Nearshore sediment · Bohai sea

Since the invention of plastic in 1907, plastic products have been used in various fields of life and production. The total global production of plastics has been increased hundreds of times since its invention to the current level of 348 million tons (Plastics Europe 2018). Because of their durability and difficult degradation, plastics tend to exist in the ocean for a long time (Geyer et al. 2017). Microplastics are commonly

defined as plastic particles smaller than 5 mm (Thompson et al. 2009; Cole et al. 2011) and are recognized as an emerging global issue in both freshwater and marine environments (Avio et al. 2017). Most microplastics are derived from the degradation of larger plastic debris through solar radiation, biological degradation or mechanical forces (Rummel et al. 2017) and are also called secondary microplastics (Auta et al. 2017). Additionally, plastics can also be initially manufactured at a microscopic size as primary microplastics, which are used as detergents in some personal care products or as fiber in clothing (Pan et al. 2019; Wright and Kelly 2017). Microplastics have been found in every corner of the ocean, including along coastlines, in the water column, in sediments, in beaches and even in remote areas such as the deep sea and polar regions (Yu et al. 2016; Waller et al. 2017). As the ultimate destination of most marine microplastics, sediments have become the principle focus of studies assessing microplastic pollution (Claessens et al. 2011).

Additives such as preservatives and surfactants are added in the production of plastic products, so these additives are released in the process of degradation (Pikuda, et al. 2019). It has also been reported that microplastics can be ingested by bivalves, crustaceans and fish (Wright et al. 2013; Roch and Brinker 2017), which pose serious hazards to organisms

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and may carry ecological risks. Therefore, microplastics present potential threats to marine environments and animals. To better comprehend and evaluate the problems caused by microplastics, it is necessary to conduct in-depth analysis of the characteristics of microplastics in the environment.

The Bohai Sea is the largest inland sea in China, which is also a semienclosed shelf on the edge of the sea. With the development of the economy, the Bohai Rim region has become one of the most densely populated and industrially developed regions in China in recent decades. At the same time, this area is also facing greater environmental pressure. Therefore, we collected subtidal sediments from the Bohai sea to reveal the microplastic pollution level through quantitatively and qualitatively analyzing the characteristics of microplastics.

Materials and Methods

Surface sediments were collected from 9 sampling sites from the subtidal zone of the Bohai Rim in this study, including Dalian (S1), Gaizhou (S2), Huludao (S3), Qinhuangdao (S4), Caofeidian (S5), Dongjiang (S6), Huanghua (S7), Dongying (S8) and Yangkou (S9) (Fig. 1). In June 2017, three sediment samples were collected from the subtidal zone at each sampling site using a stainless steel box sampler, and approximately 500 g sediments from the top 5-cm depth were carefully collected using a pre-cleaned stainless steel shovel. All samples were stored

in an aluminum foil bag and refrigerated at $-20\text{ }^{\circ}\text{C}$ before further processing and analysis.

Microplastics were separated from sediment samples using the density separation method by Thompson et al. (2004). Briefly, all sediment samples were dried to constant weight at $60\text{ }^{\circ}\text{C}$ for 72 h. Then, 50 g of dried sediment was weighed and placed in a pre-rinsed glass beaker each time. 200 mL of saturated NaCl solution ($\rho = 1.2\text{ g/mL}$) was added and stirred with a clean glass rod for 5 min and then allowed to settle for 60–90 min. After the solution is stationary, transfer the supernatants to another clean beaker. The NaCl separation step was repeated at least three times to increase the recovery rate of microplastics. Afterwards, 5 mL H_2O_2 (30%) was added to each supernatant to achieve a final concentration of 0.73% to degrade the organic substance (Nuelle et al. 2014). After settling at room temperature for approximately 24 h, the supernatants were collected and passed through $1.0\text{ }\mu\text{m}$ glass fiber filter paper (Whatman GF/B) under vacuum filtration. Finally, the filter paper with microplastics was placed in a clean Petri dish and dried at room temperature before microscopic inspection. According to Zhao et al. (2018), the recovery rate of this approach was up to 92.5% on average.

The plastic-like particles on the filters were optically analyzed and photographed using a stereomicroscope (Olympus, SZX10, Japan). Images were taken by a stereomicroscope equipped with a camera (CnoptecTP510, Chongqing, China). Suspected microplastics were characterized (shape and color) and measure the maximum length of microplastics as its particle size. The polymer composition of plastic-like particles from sediments was determined using Fourier transform infrared microspectroscopy ($\mu\text{-FT-IR}$). Two or three plastic-like particles randomly selected from the filter of all sampling sites were placed on an ultra-fast motorized stage of a Nicolet™ iN10 infrared microscope (Thermo Fisher Scientific, USA) and then measured in transmittance mode using a liquid nitrogen cooled MCT detector. The $\mu\text{-FT-IR}$ spectrum of each plastic item was $4000\text{--}650\text{ cm}^{-1}$ by co-adding 128 scans at a resolution of 8 cm^{-1} . The spectra of each particle were obtained by using OMNIC software (Thermo Scientific, Madison, USA) and compared to the OMNIC polymer spectral library. Only a match of more than 70% can be accepted as microplastic (Zhao et al. 2018).

To avoid potential microplastic contamination in the laboratory, all of the utensils were thoroughly pre-cleaned and rinsed with Milli-Q water three times before and after use. Cotton lab coats and polymer-free gloves were always worn to ensure sterility during the experiments. All the experimental solutions were adopted after vacuum extraction with $1\text{ }\mu\text{m}$ glass fiber membranes (GF/B Whatman). Three procedural blanks were set to adjust the results obtained during laboratory analysis.

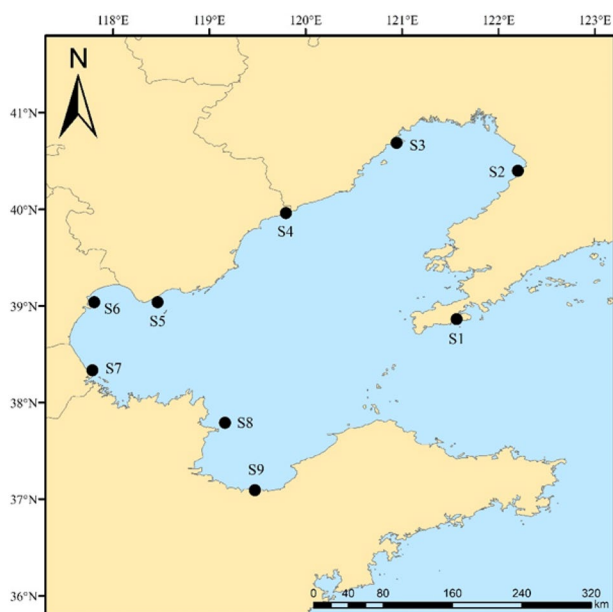


Fig. 1 Subtidal sediment sampling sites along the coast of the Bohai Sea

Statistical analyses were conducted using SPSS 16.0 software (SPSS Inc., Chicago, USA). The non-parametric Kruskal–Wallis H test was used to analyze multiple comparisons. If there was a significant difference ($p < 0.05$), the pair-wise Mann–Whitney U test was used.

Results and Discussion

No plastic polymer was detected in procedural blanks, indicating the contamination control procedure was successful. The distribution of microplastics in sediment samples collected from the subtidal zone of the Bohai Sea is shown in Fig. 2, with a total of 619 microplastics. The microplastic abundances at different sampling sites varied from 280.0 to 773.4 items/kg dry weight (d.w.) sediment, with an average abundance of 458.6 ± 150.0 items/kg d.w. The average value of microplastic abundance from site S1 to site S9 were 773.3 ± 72.9 , 373.3 ± 29.6 , 373.3 ± 66.9 , 466.7 ± 61.5 , 466.7 ± 42.2 , 586.7 ± 86.3 , 480.0 ± 78.7 , 280.0 ± 26.5 and 326.7 ± 70.7 items/kg d.w., respectively. The highest microplastics concentration was found at the S1 site; furthermore, microplastic abundance at this site was significantly higher than at other sites ($p < 0.05$). The S1 site was in a tourist district where many people traverse, which would produce a large amount of plastic garbage. The results were consistent with the view of other studies (Nor and Obbard 2014; Qiu et al. 2015). The S6 and S7 sites were selected in Dongjiang Port and Huanghua Port respectively, which were important regional ports and energy export ports in China. Heavy shipping activity have resulted in a high concentration of microplastics in the region. Similar results have been indicated that the abundances of microplastics near the port area were much higher (Chouchene et al. 2019). Sites S2, S3 and S4 are within a tourist district which is visited

by tourists and local residents. These three sites show high microplastic abundance due to human activities (Zhu et al. 2018; Zhao et al. 2018). S5 was located in a reclamation industrial zone, where there is input of industrial wastewater and plastic waste. Sites S8 and S9 are river estuary sites; abundance at these sites was of the same order of magnitude as reported from Changjiang Estuary (Peng et al. 2017). It has been shown that river input is the primary source for microplastics entering the sea (Zheng et al. 2019). However, it is interesting to note that microplastic abundance in sediment of the S8 site was significantly lower than at the other sites ($p < 0.05$) because this sampling site is in the Yellow River Delta Nature Reserve and has been less affected by human activities.

We compared our finding with previous results that use similar sampling methods and quantification units. Early research found that microplastic pollution was significantly positively correlated with river input, population density, and extent of industrial areas (Browne et al. 2011; Andrady 2011). Because of the unique characteristics of the semi-enclosed Bohai Sea, surrounded by dense population and many industrial factories, the microplastics abundance in sediments in these areas was significantly higher than that of the Yangtze River estuary (Peng et al. 2017). Our results suggested that microplastic abundance in the sediments of the subtidal zone of Bohai Sea is similar to that of sediments from China's Sishili Bay (Zhang et al. 2019), the Maowei Sea (Li et al. 2019) and Canada's Humber Bay (Corcoran et al. 2015).

Four different shapes of microplastics (fragments, pellets, films and fibers) were detected among all sampling sites (Fig. 3). As illustrated in Fig. 4a, fibers were the most chief composition among all sample types, with a proportion of 77.1%, followed by fragments (16.8%). This level is similar to those reported from the coastal mangrove sediments in Singapore (72%) (Nor and Obbard 2014) and coastal sediments in Slovenia (75%) (Laglbauer et al. 2014). In fact, it has been reported that fiber is the principal type of microplastic pollution (Frias et al. 2016); frequent human activities can produce a large number of microplastic fibers. Textile production and washing processes are considered to constitute a principle source of microplastic fibers (Browne et al. 2011). Moreover, fishing nets and other plastic products widely used in marine activities are also important sources of microplastic fibers (Zhao et al. 2014). Pellets and films constituted only a small portion of microplastics. The lowest proportion (58.0%) of fibers was found at the S8 site, which had the highest proportion of fragments (29.0%). This is because fragment microplastics are produced by aging and cracking of hard plastics or engineering plastics. There are many sources of hard plastics, mainly daily necessities, such as plastic bottles and tableware. Field investigations have revealed that hard plastics are widespread in Shandong

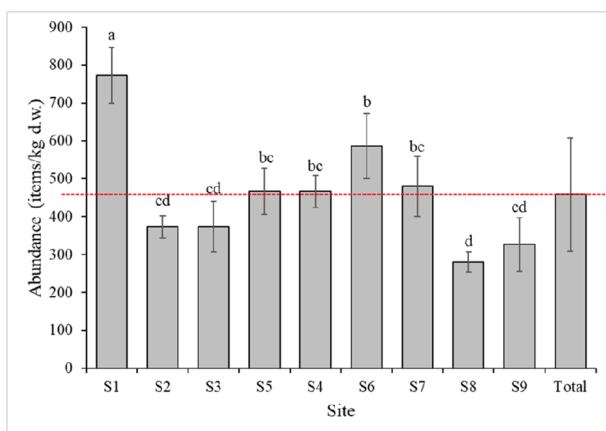


Fig. 2 Abundances of microplastics in subtidal sediments collected from the Bohai Sea. Means that do not have the same letter are significantly different at $p < 0.05$. Vertical bars represent the mean \pm SD

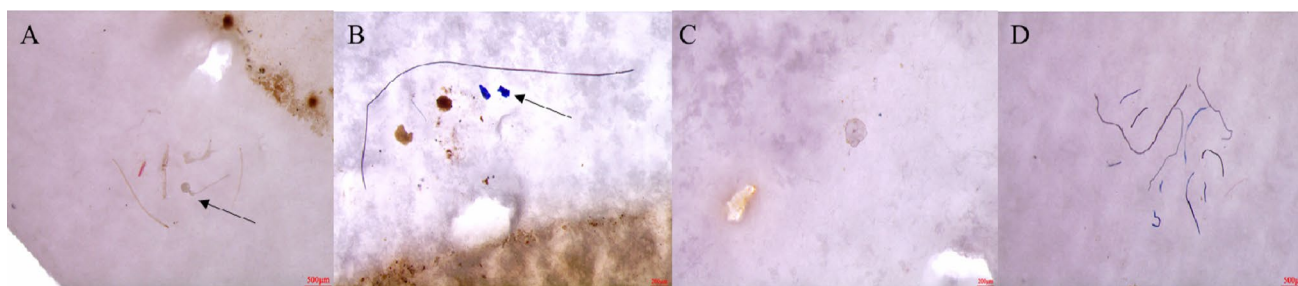


Fig. 3 Different shapes of microplastics in subtidal sediments collected from the coast of the Bohai Sea. **a** Pellet, **b** fragment, **c** film, **d** fiber. Arrow indicates the microplastic

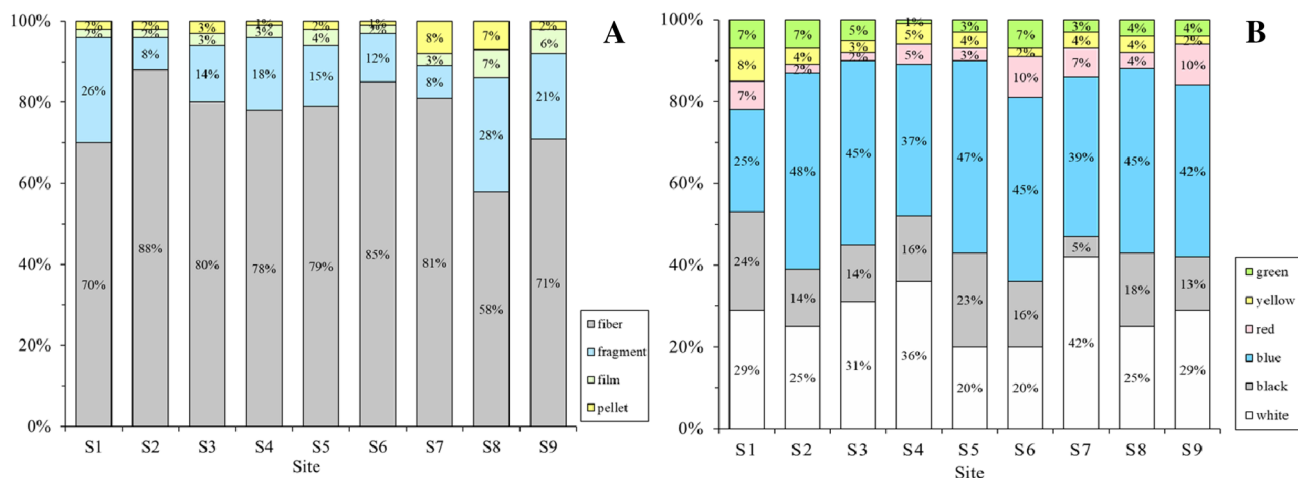


Fig. 4 Percentages of shapes (**a**) and colors (**b**) of microplastics in subtidal sediments collected from the coast of the Bohai Sea

coastal areas, such as coastal areas that need flood control, tidal flats, and salt fields (Zhou 2016). This type of plastic is prone to aging, leading to chipping and entry into the surrounding environment.

The colors of particles were mainly white (39.9%), black (16.6%) and blue (28.6%), accounting for 85.0% of all microplastic particles. In addition to the above three colors, green, yellow, and red were also found in the sediment samples (Fig. 4b). The variety of colors of microplastics indicates a wide range of sources. Hidalgo-Ruz et al. (2012) reported that white microplastics are the most common in the marine environment, presumably due to the weathering and fading of plastics in the marine environment. Because blue nylon rope is widely used in fishing activities, the proportion of blue microplastics is also high. The particle size range of sediment microplastics ranged from 22.2 to 4692.2 μm , and the average particle size was $1101.7 \pm 1033.2 \mu\text{m}$. Microplastics with particle size less than 500 μm constituted the largest proportion, accounting for 44.9%, followed by 500–1000 μm (25.5%). Microplastic abundance tended to decrease with increasing particle size. Some previous studies have reported similar results (Zhu et al. 2018; Lots et al.

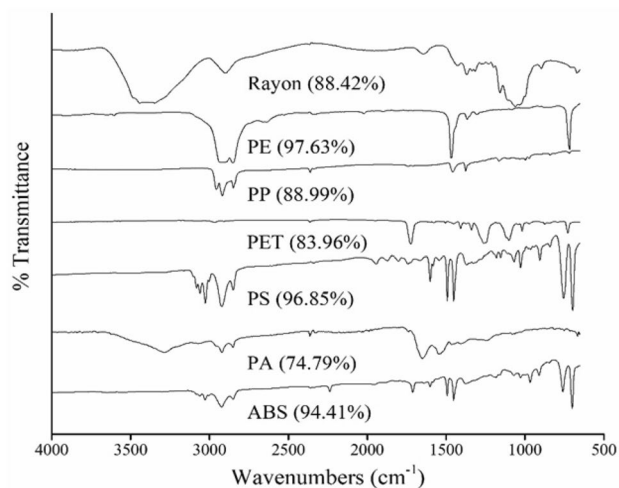


Fig. 5 The μ -FT-IR spectrum of microplastics in subtidal sediments collected from the coast of the Bohai Sea. The value in parenthesis is the average matching ratio with the standard spectrum for each polymer

Table 1 Relative abundances, percentages and densities of identified microplastic

Type of polymers	Number	%	Density (g/cm ³)	Library
Rayon	43	41.0	1.50	Hummel polymer samplly library
PE	18	17.1	0.89–0.91	Aldrich condensed phase samplly library
PS	17	16.2	1.04–1.08	Hummel polymer samplly library
PP	11	10.5	0.89–0.91	HR nicolet samplly library
PET	9	8.6	1.29–1.40	Hummel polymer samplly library
ABS	6	5.7	1.01–1.08	Hummel polymer samplly library
PA	1	1.0	1.07–1.08	Hummel polymer samplly library
Total	105	100	–	–

The plastic density is referred to Nuelle et al. (2014) and Peng et al. (2017)

2017), indicating that microplastics in sediment are mainly small in particle size. Smaller sizes of microplastics are usually more toxic to marine organisms (Wang et al. 2019); it is therefore essential to evaluate the risk of small plastics in the marine environment.

A total number of 120 particles were randomly selected, accounting for more than 10% of the total isolated particles, and the chemical composition was identified by μ -FT-IR. Among them, seven polymer types were identified in the sediment samples (Fig. 5), including Rayon (41.0%), polyethylene (PE, 17.1%), polystyrene (PS, 16.2%), polypropylene (PP, 10.5%), polyethylene terephthalate (PET, 8.6%), Acrylonitrile–butadiene–styrene (ABS, 5.7%) and polyamide (PA, 1.0%). In addition, approximately 12.5% could not be identified as ordinary synthetic polymers. The quantity of each microplastic polymer type observed in the samples is presented in Table 1. Rayon was one of the first commercially available man-made fibers, mainly used in personal hygiene products and textiles (Kauffman 1993). Strictly speaking, rayon it is not a plastic. But since rayon cannot be easily separated from other microplastics by microscopic observation, and it is likely to be ingested by marine animals (Peng et al. 2017). Meanwhile, it is widely reported in the marine environment (Woodall et al. 2014), so we included rayon in our results. It is suggested that sanitary wastewater might be the origin of rayon in the ocean (Woodall et al. 2014). However, Comnea-Stancu et al. (2017) reported that FT-IR transmission analysis could not distinguish between man-made fibers and natural fibers, which would result in an overestimation of rayon abundance. Relatively high abundances of PE, PS, PP and PET microplastics were also detected in this study, similar to levels observed in sediments from the South China Sea (Qiu et al. 2015). PE is currently the most widely used plastic in food-packaging film and agricultural film; while PP and PET are used in food packaging, plastic containers, carpets and pipes. Additionally, PP and PET are also increasingly used industrially to make clothes and textile products (Park et al. 2004). PS is not only used to make parts such as gears and electronic cases

but also to make optical glass or optical instruments. When they are degraded into microplastics, PE, PS, PP and PET should be abundant in sediments. The results are similar to polymer compositions of microplastics in sediments from tourist areas in Singapore (Nor and Obbard 2014). Accordingly, maritime activities and land-based sources might be the main sources of microplastics in nearshore sediments collected from the Bohai Sea coastal area.

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Compliance with Ethical Standards

Conflict of interest The authors declare that they have no conflict of interest.

Human and Animal Rights and Informed Consent This article does not contain any studies with human or animal subjects performed by any of the authors.

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