



# Microplastic pollution in North Yellow Sea, China: Observations on occurrence, distribution and identification

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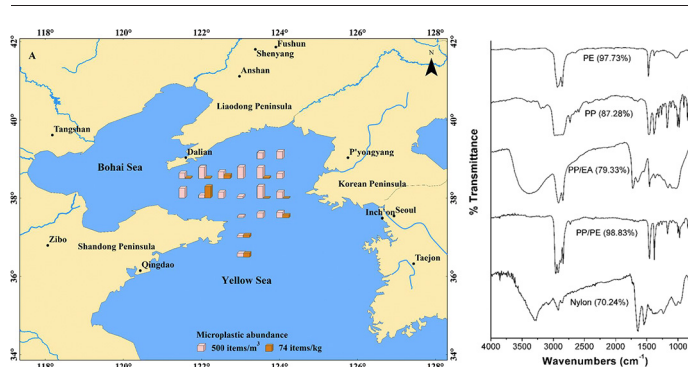
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## HIGHLIGHTS

- Microplastics were detected in the surface water and sediments of North Yellow Sea.
- Microplastics abundances were at medium level in comparison to other equivalent data.
- Polyethylene and polypropylene were the dominant compositions of microplastics.
- Threat of microplastics to fishery species should be focused on in future research.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Microplastics are emerging contaminants and have attracted widespread environmental concerns about their negative effects on the marine ecosystems. In this study, we investigated the abundances, distributions and characteristics of microplastics in surface seawater and sediments from the North Yellow Sea. The results showed that the abundance of microplastics was  $545 \pm 282$  items/m<sup>3</sup> in surface seawater and  $37.1 \pm 42.7$  items/kg dry weight in sediments, representing a medium microplastic pollution level compared with other sea areas. Small microplastics (<1 mm) made up >70% of the total microplastic numbers. Films and fibers were the dominant shapes of microplastics in both the surface seawater and sediments. Transparent microplastics were generally more common than microplastics of other colors. Based on the identification by a Fourier transform infrared microscope, polyethylene (PE) was the dominant composition of microplastics in surface seawater, while polypropylene (PP) was the most common polymer type in sediments. These results will improve our understanding of the environmental risks posed by microplastics to marine ecosystems.

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## 1. Introduction

As a consequence of their benefits, plastics have been extensively used in industry, agriculture and our daily lives. Global plastic

production has shown a steady increase and reached 322 million tons in 2015, of which China contributed to 27.8% (PlasticsEurope, 2016). It has been estimated that between 4.8 and 12.7 million tons of plastics have entered the ocean and formed marine litter due to inadequate disposal (Jambeck et al., 2015). A large number of plastics have long accumulated in the ocean because of their durability. Microplastics (defined as plastic particles <5 mm) dominate plastic debris (van Cauwenberghe et al., 2013) and originate from two sources, manufactured products

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(such as exfoliating facial scrubs, toothpastes and antifouling of boats) that contain microplastics (primary microplastics) and fragments released from larger plastic debris through photooxidation, mechanical action and biodegradation (secondary microplastics) (Auta et al., 2017).

Once entering into ocean, microplastics may pose serious hazards to marine organisms, including phytoplanktons (Bhattacharya et al., 2010), zooplanktons (Cole et al., 2013; Desforges et al., 2015), clams (Davidson and Dudas, 2016), mussels (Kolandhasamy et al., 2018), crabs (Watts et al., 2015), and fishes (Nadal et al., 2016; Ory et al., 2017). Microplastics not only can cause physical harm to marine organisms by contact, uptake and ingestion (Watts et al., 2015; Nadal et al., 2016; Kolandhasamy et al., 2018), but also provide the marine organisms with a potential pathway for exposure to organic pollutants, metals and pathogens adsorbed from the ambient environment (Zhang et al., 2015; Brennecke et al., 2016; Foulon et al., 2016; Wu et al., 2017) or to chemicals leached by themselves (Li et al., 2016; Hermabessiere et al., 2017). Microplastics may also pose risks to human health because they may be transferred through the food chain (Farrell and Nelson, 2013; Pellini et al., 2018). Therefore, the effects of microplastics on the marine environment have become major concerns.

To date, many efforts have been devoted to investigate the accumulation of microplastics in marine environments, including oceans (Desforges et al., 2014; Lusher et al., 2014; Maes et al., 2017), seas (Isobe et al., 2015; Aytan et al., 2016; Cincinelli et al., 2017; Gewert et al., 2017), bays (Song et al., 2015; Blašković et al., 2017; Frère et al., 2017), estuaries (Zhao et al., 2014, 2015; Peng et al., 2017) and coasts (de Lucia et al., 2014; Chae et al., 2015; Yu et al., 2016; Munari et al., 2017), as well as the deep seas (van Cauwenberghe et al., 2013; Bergmann et al., 2017; Courteney-Jones et al., 2017). Up to  $10^5$  particles/ $m^3$  of microplastics have been discovered in marine environments by global microplastic litter estimates (Dubaish and Liebezeit, 2013; Song et al., 2015). It has been assessed that China is not only the largest plastic producer and consumer in the world (PlasticsEurope, 2016), but also the largest source of mismanaged plastic litter that can enter the environment (Jambeck et al., 2015). A few studies on microplastic pollution are available for the Chinese marine environments (Zhao et al., 2014, 2015; Qiu et al., 2015; Yu et al., 2016; Fok et al., 2017; Peng et al., 2017; Zhang et al., 2017). Zhao et al. (2014) firstly reported the occurrences and distributions of microplastics in the surface water of Yangtze Estuary and East China Sea, China. Qiu et al. (2015) appraised the occurrences and abundances of microplastics in sediments from the South China Sea coast for the first time. However, little information on microplastic pollution in the North Yellow Sea is currently available.

The North Yellow Sea, a semi-enclosed epicontinental sea surrounded by the Shandong Peninsula, the Liaodong Peninsula and the Korean Peninsula, has attracted a lot of attentions not only because of its high biological production (Lin et al., 2005), but also owing to the ever-increasing environmental pressure from human activities on marine ecosystems (Huang et al., 2014). The potential threats posed by microplastics to marine organisms and human health mean that the abundances of microplastics in the North Yellow Sea need to be determined. The aims of this study were: (1) to determine the abundances and distributions of microplastics in surface seawater and sediments of the North Yellow Sea; (2) to characterize the isolated microplastics based on the size, shape, color and polymer type analysis; and (3) to discuss the potential sources of microplastics and the potential environmental risks posed by the microplastics.

## 2. Materials and methods

### 2.1. Study area

The North Yellow Sea is the north part of the Yellow Sea, which covers an area about  $8 \times 10^3$  km<sup>2</sup> with an average depth of 40 m. The sediments of North Yellow Sea mainly consist of sand, silt and clay.

The seawater temperature in the North Yellow Sea varies between 0 °C and 4 °C in winter, and in summer the seawater temperature approximates 25 °C at the surface and 5–20 °C at the bottom, respectively. The salinity in the North Yellow Sea ranges from 29.5‰ to 32.0‰. The surface circulation in the North Yellow Sea consists of three main currents, the Chinese Coastal Current, the Yellow Sea Warm Current and the Korean Coastal Current (Fig. S1). These currents accelerate the movement and mixture of the seawater in this area. Additionally, the North Yellow Sea is the ideal habitat (being the spawning grounds, feeding grounds, overwintering grounds and migration channels) for a great deal of fishery species (Liu, 1990) (Fig. S2).

### 2.2. Sample collection

During October 11–14, 2016, surface seawater samples were collected from 19 sites in the North Yellow Sea, while surface sediment samples were obtained from 10 sites due to restricted accessibility. Detailed information on the sampling sites is presented in Fig. 1 and Table S1. At each sampling station, 25 L of surface seawater was sampled at a 30 cm water depth by Niskin hydrophore (Courteney-Jones et al., 2017) and poured through a 30- $\mu$ m steel sieve. The sieve did not become blocked while any sample was collected. The retained particles on the sieve were rinsed with filtered ultrapure water into a clean glass bottle and preserved with 5% formalin solution (Zhao et al., 2014, 2015). Each sediment sample was gathered with a 0.1 m<sup>2</sup> Gray-O'Hara box corer and the topmost 5 cm of sediments (500–1000 g) were collected and stored at –20 °C until analysis (Frère et al., 2017).

### 2.3. Isolation of microplastics

In the laboratory, surface seawater samples were subjected to wet peroxide oxidation using 30% H<sub>2</sub>O<sub>2</sub> in the presence of a Fe(II) catalyst to digest organic matter (Zhao et al., 2014). Subsequently, plastic particles were isolated from the above mixture by density separation in a saturated NaCl solution (1.2 g/cm<sup>3</sup>) (Masura et al., 2015). The floating microplastics were passed through a 30- $\mu$ m steel sieve and then collected for further microscopic examination. For sediment samples, approximately 300 g of sediment was oven dried at 70 °C until sample dryness (Peng et al., 2017). The dry sample was weighed and resuspended within 250 mL of NaI solution (1.6 g/cm<sup>3</sup>) by manual stirring for 2 min with a glass rod in order to adequately separate the relatively light plastic particles from minerals (Nuelle et al., 2014). After 6 h of sedimentation, the supernatant was then poured through a 30- $\mu$ m sieve and the sieved solids were transferred to a pre-cleaned glass beaker and subjected to wet peroxide oxidation using H<sub>2</sub>O<sub>2</sub>. Then plastic particles were subject to density separation in NaCl solution (1.2 g/cm<sup>3</sup>) (Zobkov and Esiukova, 2017), which was in accord with the processing method used for the seawater samples in order to keep the data consistent.

To avoid potentially artificial and airborne plastic contamination, all apparatus were rinsed carefully with ultrapure water and wrapped tightly in aluminum foil. Cotton laboratory coat and latex gloves were worn during the entire process of sample collection and laboratory analysis. Moreover, blank experiments were conducted following the self-same procedures used for the samples but using filtered ultrapure water and glass microbeads ( $\phi = 2\text{--}3$  mm) instead of seawater and sediment samples, respectively, to detect any ambient microplastic contamination from the equipment and the laboratory. No piece of microplastic was found in the blank controls.

### 2.4. Identification of microplastics

Microplastics were counted and photographed using an optical microscope (Olympus BX-51, Japan) equipped with an AxioCam digital camera (Zeiss, German) according to the identification rules of Peng et al. (2017). Microplastics were manually put into six size classes

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