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Microplastic risk assessment in surface waters: A case study in the Changjiang Estuary, China



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ARTICLE INFO	A B S T R A C T
<i>Keywords:</i> Microplastics Changjiang Estuary Risk assessment Polymer Pollution load index	The rapid development of plastic industry has resulted in a series of environmental problems caused by microplastics originating from larger plastics. Microplastic pollution risk in surface waters of the Changjiang Estuary was explored based on risk assessment models. The average microplastic concentration was $23.1 \pm 18.2 \text{ n/100 L}$. Shape, size, color and composition types of microplastics were examined. The risk assessment models were developed using data on both the concentration and chemical hazard of microplastic polymers. Assessment results indicated that polyvinyl chloride exhibited a critical concern for microplastic risk. Areas around aquaculture farms were regarded as "hotspots" of microplastic pollution due to the accumulation of microplastics and the presence of hazardous microplastic. This risk assessment of microplastics bridged gaps

1. Introduction

Plastics are synthetic polymers widely used in various industries, which largely originate from the polymerization of crude oil (Lithner et al., 2011). Given the wide range of use of plastic products, plastic waste contributes to a large amount of waste in the environment. A large amount of plastic waste is directly discarded into the ocean and it is estimated that will increase to 30 million tonnes by 2040 (Neufeld et al., 2016). Ocean dynamics and anthropogenic factors can result in the crushing of plastic debris and the formation of microplastics (MPs) (Thompson et al., 2004).

Most available information on the distribution of MPs in the environment comes from research in offshore areas, estuaries and the open ocean (Collignon et al., 2014; Doyle et al., 2011; Law et al., 2010; Moore et al., 2001). Ubiquitous MPs pose potential dangers to aquatic organisms, due to a number of characteristics. Various types of MPs, including fibers, pellets and fragments, are now commonly found in organisms (Lusher et al., 2013; Li et al., 2015; Jabeen et al., 2017). It has been reported that MP particles are more likely to be mistakenly ingested if they are similar in color to prey items (Laurier and Mason, 2007). MPs with a median particle size of 0.8 mm have been widely detected in the bodies of fish (Foekema et al., 2013). Hazardous MP polymers are known to induce detrimental biological effects on plankton, fish and birds (Mato et al., 2001; Ogata et al., 2009; Henry, 2013).

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Current research in this area has included some risk assessments applying different methods and indicators. Wilcox et al. (2013) developed oceanic drift models to make predictions of the probability of a marine turtle being endangered by abandoned fishing gears. Based on chemical compositions of plastic polymers, Lithner et al. (2011) incorporated the chemical hazard of additives, monomers, polymers and polymerization, and developed a hazard ranking model of plastic polymers to assess the hazard effects on human health and organisms. Thermodynamic features of plastics have been selected as important factors in determining the adsorption capacity of MPs (Gouin et al., 2011). A MP risk assessment framework for a mega-city was constructed combining the distribution characteristics of MPs in an urban river (Peng et al., 2018). Severe ecological risk of MPs in mussels were correlated with high MP concentrations in the surrounding water, which suggested that mussels could be used as biological indicators to monitor MP pollution levels in coastal waters (Qu et al., 2018).

in understanding between field research and policy-making for surface waters. This research provides baseline

data for assessing the environmental risk of microplastics in this growing area of research.

The MP risk assessment process remains difficult, although researchers have recently incorporated environmental influences. To date the actual risk posed by MPs has not been determined, as that laboratory exposure concentrations are not compared to environmentally relevant thresholds (Koelmans et al., 2017). Ambiguous statements like "potential", or "could" are often used to mislead policy making (Koelmans et al., 2017). Some studies may focus on a certain aspect of MP pollution, and thus cannot fully describe the risk of MPs (Bencze and Alsop, 2014). In terms of field research, there is no standardized

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method to monitor of MPs in the environment (Hidalgo-Ruz et al., 2012).

A previous study suggested that China may have the largest output of plastic waste from terrestrial sources entering the ocean globally (Jambeck et al., 2015). High levels of MPs have also been widely observed in surface waters of estuaries in China (Zhao et al., 2014, 2015). In addition, seafood collected from Shanghai, China had abundant MPs (Li et al., 2015; Jabeen et al., 2017). Therefore, the Changjiang Estuary might experience potential MP risks (Peng et al., 2017).

In the present study, an initial assessment in surface waters of the Changjiang Estuary and the adjacent East China Sea was performed adopting the hazard score of plastic polymers created by Lithner et al. (2011) and the pollution load index (PLI). The aim of the study was to provide a primary understanding of the risk of MP pollution in surface waters. It is hoped that this research will assist policy making for marine MP pollution by providing preliminary data on MP assessment.

2. Materials and methods

2.1. Data collection

The Changjiang Estuary is a funnel-shaped estuary. Chongming Island divides the estuary into the North Branch and the South Branch (Zhao et al., 2018). Runoff from Changjiang is mainly discarded via the South Branch (Gao et al., 2008). The Changjiang Estuary is the gateway where the Changjiang River flows into the East China Sea, which represents multiple characteristics affected by interactions between marine and terrestrial environments (Liu et al., 2001; Liu et al., 2018). Its ecological status plays an important role in the development of local industry, agriculture and trading in Shanghai (Liu et al., 2015). Therefore, the ecological protection of the Changjiang Estuary and the adjacent area has received worldwide attention.

Samples were collected at 29 sampling stations in the Changjiang Estuary (C stations) and the adjacent East China Sea (ECS) (E stations) in August 2017 on board the Runjiang No.1. (Fig. 1). At each sampling station, 100 L of water was pumped through a stainless steel sieve with a mesh size of 70 μ m at a depth of 50 cm. Residues on the sieve were washed into clean 250 mL high – density polyethylene bottles with purified water. Three replicate samples were collected at each sampling station. All samples were persevered at 5 °C until further analysis. Due to diversity in the field, it was difficult to avoid aerial contamination during sampling. Measures were taken during field work to reduce aerial contamination as much as possible, including washing the sieve and sampling bottles in advance.

In the laboratory, samples were digested with a 30% $\rm H_2O_2$ solution for at least 12 h to remove biological materials. Using an excessive amount of digestive solution over a sufficient time period ensures a complete reaction during this process. Samples were then heated in water bath at 50 °C to speed up the reaction before they were filtered onto a Sartorius filter (0.45 μm pore size). Residual material on the filters was carefully transferred to clean glass petri dishes and dried to a constant weight.

The particles on the filter were counted and photographed under a Leica M165 FC microscope. Suspected particles were then transferred onto another clean filter for chemical analysis of polymers. A small amount of pure water was dropped on to the filter beforehand to prevent particle loss during this process. The morphological descriptions (fiber, fragment, film and spherical pellet) of particles were selected based on criteria used in previous studies (Zhao et al., 2014, 2015). The particles were also classified into four size groups: < 0.07 mm, 0.07–1.0 mm, 1.0–5.0 mm, and > 5.0 mm. In addition, the particles were described according to their color (black, transparent or colored).

 μ -FTIR (micro-Fourier transform infrared spectroscopy, Bruker LUMOS, Germany) was used to determine all suspected particles on the filters. Contaminants attached to the surface of the particles, such as sediments, were cleaned in an ultrasonic bath. The ATR (Attenuated

Total Reflection) mode is generally adopted for surface analysis of particles. This mode measures changes in infrared beam, which exhibits total internal reflection when the ATR crystal comes into contact with a sample. Therefore, it is important to obtain accurate contact between the crystal and the sample. By determining the absorbance of the selected infrared beam, it is possible to differentiate between different types of polymers. A list of potential compounds with hit quality was contributed by FTIR library (based on Nature Fiber Library and Polymer Library). The components with the highest matching values were identified as the main types of suspected particles. In order to check the matching rate of spectra from samples and data libraries, plastic products of known materials, including food packaging (polyethylene), bottles (polypropylene), tableware (polystyrene) and fishing gear (polyamide), were tested. The polymer types of all plastic products tested were accurately detected under ATR mode. Materials with a spectra matching rate of over 60% were accepted as being the main components, but matches below 60% were not rejected. This is an effective and useful method to determinate the characteristics of resultant peaks with µ-FTIR observations.

Correct detection of MP polymer types in samples is important in the risk assessment, therefore airborne pollution must be strictly controlled during experimental procedures. Precautions to avoid aerial contamination following Zhao et al. (2017) were undertaken during the whole procedure. The stainless steel sieve used for sampling was cleaned repeatedly three times to ensure that no impurities remained. All solutions and purified water were pre-filtered using Sartorius filters (0.45 µm pore size). Glass ware was cleaned with purified water and placed in an autoclave at 450 °C for 2 h. Clothes and plastic containers made of unknown materials used in the lab were detected with µ-FTIR to exclude any suspected particles. In order to avoid potential air pollution during laboratory work, air was sucked through three 0.45 µm filter papers where any residues could be observed on the filter membrane.

2.2. MP risk assessment approach

Both the concentration and the chemical composition of MPs should be considered in order to evaluate the potential risks of MPs in surface waters of the Changjiang Estuary and the adjacent ECS. The risk of MPs in surface waters based on two indicators was assessed separately. Following Lithner et al. (2011), the chemical toxicity of MP polymers was used as an important index to evaluate its ecological harm. Our study utilized the hazard scores of plastic polymers from Lithner et al. (2011) and used the polymer types of MPs as indexes to assess the risk of MPs (see Table 1). The following formula was used:

$$H = \sum P_n \times S_n \tag{1}$$

where *H* is the calculated polymer risk index caused by MP, P_n is the percent of MP polymer types collected at each sampling station, and S_n is the score from Lithner et al. (2011) for the polymer compound that comprised MP particles.

In a larger area, it is not possible to complete the assessment solely on a single indicator. In such case, the pollution load index (PLI) which refers the MP concentration in Changjiang Estuary, was used. Tomlinson et al. (1980) proposed the pollution load index (PLI) to assess the level of pollution in estuaries. The PLI is regarded as a standardized rule for monitoring the degree of pollution between different areas (Angulo, 1996). The assessment model was as follows (Tomlinson et al., 1980):

$$CF_{i} = C_{i}/C_{oi}$$

$$PLI = \sqrt{CF_{i}}$$

$$PLI_{zone} = \sqrt{PLI_{1}PLI_{2}\cdots PLI_{n}}$$
(2)

The pollution load index of MPs at each station (*PLI*) is related to MP concentration factors (CF_i). The CF_i of MPs is the quotient of the MP



Fig. 1. Location of sampling stations in the Changjiang Estuary and adjacent East China Sea in August 2017. Black circles represent the location of the sampling stations and the Bailonggang Wastewater Treatment Plant is represented by a triangle. Red arrows illustrate current paths (the Changjiang Diluted Water, CDW and the Taiwan Warm Current, TWC) in summer. Land elevation (m) and water depth (m) are shown on the right of the map. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

concentration at each station (C_i) and the minimal MP concentration (C_{oi}). The C_{oi} (0.05 n/100 L) is defined as the minimum average concentration based on the available literature (Isobe et al., 2014). In order to compare the degree of MP risk in study areas, a specific value was assigned to C_{oi} . In fact, the value (C_{oi}) did not affect the comparison results using the formula (2). Following a literature review, and considering the study methods, objectives and geographical location, the average concentration of microplastics in surface waters of the Japan Sea was chosen as the standard level. With more research in the area of microplastics, it is possible that a lower concentration will be discovered. More reliable technology is required to determine the standard level of microplastic pollution. For example, a mathematic model would simulate the environmental carrying capacity of microplastic

pollution. However, this was not the priority of this study. The PLI in the Changjiang Estuary (PLI_{zone}) was obtained by the n-root from the n-PLI that was obtained for all the stations.

Previous studies have given no consistent expression for risk level following MP pollution. The criteria for the risk level of MP pollution presented in Table 2 provides a definition of the degree of MP risk, providing support for risk management of MP pollution.

3. Results and discussion

3.1. Abundance and distribution

In this study, no aerial plastic-like particles were found. Natural

Table 1

Detailed information for microplastic polymers detected in this	is study, including monomer, density,	usage and score (Data from	Plastics Europe, 2016).
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Polymer	Abbreviation	Monomer	Density (g/cm ³)	Main applications	Score ^b
Polyethylene	PE	Ethylene	0.91-0.96	Toys, bottles, pipes, house ware, etc.	11
Polypropylene	PP	Propylene	0.85-0.94	Food packaging, microwave-proof containers, etc.	1
Polyvinyl chloride	PVC	Vinyl chloride	1.41	Pipes, cable insulation, garden hoses, etc.	10,551
Polyamide (nylon)	PA	Adipic acid	1.14-1.15	Bearings, automotive applications, etc.	47
Acrylonitrile-butadiene-styrene	ABS	Styrene	1.02-1.08	Automotive applications, pipes, etc.	6552
Polyurethane	PUR	Propylene oxide	0.40-0.60	Upholstery, sports mats, packaging bags, etc.	7384
Polystyrene	PS	Styrene	1.05	Spectacle frames, plastic cups, packaging, etc.	30
Polycarbonate	PC	Bisphenol A	1.19	Information storage discs, security windows, etc.	1177
Styrene acrylonitrile	SAN	Styrene	1.06-1.10	Cosmetic containers, ballpoint pens, lighters, etc.	6788
Acrylate-styrene-acrylonitrile ^a	ASA	styrene	1.05	Sporting goods, antennae, sanitary products, etc.	-

^a Acrylate – styrene – acrylonitrile lacks ecological toxicity data, therefore its hazard score cannot be determined.

^b The value for the score of each polymer is taken from Lithner et al. (2011).

Table 2

Risk level criteria for microplastic pollution.

Value of the polymer index	<10	10-100	100-1000	> 1000
Value of the pollution load index	~10	10–20	20–30	> 30
Risk category	I	II	III	IV

materials (e.g. rayon) were not included in this result. A total of 2014 MP particles were found at all sampling stations, with an average concentration of 23.1 ± 18.2 n/100 L (Kruskal-Wallis test, p = 0.001 < 0.05). In summer 2013, the density of MPs in the Changjiang Estuary was reported to be 4137.3 n/m³ (413.7 n/100 L) (Zhao et al., 2014). The results obtained in the current study are significantly lower than those from 2013. The difference may be due to differences in sampling methodology, neustonic trawls were used in 2013 while a pump was used in the current study. In addition, the water depth and the pore size of the steel sieve used by Zhao et al. were deeper and smaller, respectively. Furthermore, there was a lack of steps to identify particle components by μ -FTIR in the 2013 study, which could lead to the inclusion of suspected non-plastic particles in the results, potentially providing a higher density estimate of MPs.

There was a distinct variation in MP distribution between sampling stations. Highest densities were recorded at sampling stations E3 (43.8 \pm 21.3 n/100 L), E9 (50.6 \pm 31.5 n/100 L), E17 (66.3 \pm 21.6 n/100 L) and E21 (52.1 \pm 29.4 n/100 L) (Fig. 2). MP concentrations at some offshore sampling stations were higher than those at inshore locations. In particular, large quantities of MPs accumulated in the northeast of the study area. The distribution of MPs may be explained by complex water movements. Water circulation patterns in the study area in the summer are mainly influenced by the Changjiang Diluted Water (CDW), the Taiwan Warm Current (Zhu et al., 2003). The CDW initially moves southeastwards near the mouth of the estuary. It then divides into two branches, one flowing southeastwards and the other

flowing northeastwards (Zhu et al., 2003). A consistent trend of increasing MP concentrations was recorded along the CDW transport route flowing northeastwards. This trend suggests that the CDW, together with other water masses, may have brought MP particles from the Changjiang Estuary to the northeastern region. Additionally, the strong hydrological force of both the Changjiang Estuary and Hangzhou Bay carries plastic waste from the land to the East China Sea, thus causing the area in the vicinity of station E17 to be highly polluted.

3.2. Physical properties

MP particles can be classified into four groups based on shape, i.e., fiber, fragment, film and sphere (Hidalgo-Ruz et al., 2012). MP concentrations in the Changjiang Estuary and the adjacent ECS were highly variable between sampling stations, but the two areas shared similar physical properties of MPs (Fig. 3). The majority of MPs were fibers (82.8%), followed by fragments (15.1%) and films (2.1%) (Fig. 3A). No spherules were found at any station. This is consistent with the findings of Zhao et al. (2014), who also reported that spherules were rarely observed in samples in the Changjiang Estuary. MP spherules in domestic sewage mainly originate from facial scrub usage and the majority can be largely removed during sewage treatment (Browne et al., 2011). Fishing lines (PA) existing in the shape of fibers accounted for one fifth (Fig. 3A). Maritime activities (e.g. packaging, fishing gear) utilize a wide variety of plastics, which can enter the ocean accidentally or deliberately (UNEP, 2016). Extreme weather events are frequent in the summer and can accelerate the drifting of MPs. A total of 238 fragment particles were found from all samples in this study with 14.3% in the ECS and 17.4% in the Changjiang Estuary, respectively. When the composition of these fragments was identified by µ-FTIR, it was found that all fragments had a similar material quality to large plastic products on the market (Fig. 3E), implying that some MPs may have the same origin as large plastic waste.



Fig. 2. The distribution of microplastics in surface waters. Areas of low microplastic concentration are represented by blue while areas of high microplastic concentration are shown in red. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)



Fig. 3. Shape (A), color (B), size (C) and polymer category (D) of microplastics. Compositions of microplastics of different shape classifications, polyethylene (PE), polypropylene (PP), polyamide (PA), polystyrene (PS) and polyvinyl chloride (PVC).

In this study, colored particles (including blue, pink and gray) comprised the majority of MPs in collected samples (76.7%). The remainder of the samples were comprised of black (18.2%) and transparent (5.1%) MPs (Fig. 3B). Colored MP particles are commonly used as packaging, clothing materials and many other applications. Color can be used to identify the source of MP particles. MPs lose their original color due to weathering in the aquatic environment (Davis and Sims, 1983).

Particles < 5.0 mm accounted for over 90% of the total plastic particles found in this study. Particles in the 0.07–1.0 mm and 1.00–5.0 mm ranges accounted for 68.4% and 26.2%, respectively. Plastic particles > 5.0 mm were seldom found (Fig. 3C). It is possible that some particles > 5.0 mm may not have been collected due to size limitations of the pump pipe and small sampling volumes. Some smaller particles were difficult to identify by visual inspection using microscopic analysis.

3.3. MP risk assessment

3.3.1. Polymer properties

All MP particles were measured for their polymer compositions, which was determined by μ -FTIR spectroscopy and the detected spectrum is shown in Fig. 4. The detected natural fibers (e.g. rayon) were not included in the results. Polyethylene (PE), polypropylene (PP) and polyvinyl chloride (PVC) accounted for 82.4%, 9.1% and 6.5%, respectively (Fig. 3D). Polyamide (PA), acrylonitrile-butadiene-styrene

(ABS), polystyrene (PS), polycarbonate (PC), acrylate-styrene-acrylonitrile (ASA), polyurethane (PUR) and styrene acrylonitrile (SAN) accounted for < 3% of the total MP polymers identified. In this study on surface waters, 8% of the MPs were collected had densities ranging from 1.02–1.41 g/cm³. Theoretically, the density of MPs is a pivotal factor that affects its vertical distribution in the water. MPs which are more dense than seawater (1.02 g/cm³) (Aliani et al., 2003) are likely to sink in the water (Di and Wang, 2018). Dense MP types in our samples may be circulated by the vertical structure of tidal currents in shallow water near the Changjiang Estuary. PVC MPs with a density of 1.41 g/cm³ in surface waters can result from strong upward and downward (vertical mixing) of water.

To determine the difference between MP polymers at sampling stations, principal component analysis (PCA) was performed to describe the principal components (PC) of primary MP polymer types. The distance between stations represents the different degree of occurrence of MP polymers. The PCA result can reflect most of the sample information with the contribution of PC1 (44.7%) and PC2 (20.8%) (Fig. 5). MP polymer types varied greatly at all stations and showed no obvious clustering. Some sampling stations that were closely located had similar MP polymer compositions, such as E12 and E15.

3.3.2. Chemical risk assessment of MPs

The surface waters in the Changjiang Estuary and the adjacent ECS face serious MP pollution according to the risk index based on the chemical properties of MPs. A rank partition of risk index of MP



Fig. 4. Polymer composition of microplastics obtained using the µ-FTIR. Each spectrogram corresponds to a microscope image of the microplastic polymer type.



Fig. 5. Principle component analysis (PCA) plot for microplastic polymer categories at all sampling stations.

pollution is shown in Table 2. As seen in Fig. 6, non-negligible MP risk was found at both landward sites of the inner Changjiang Estuary and at offshore sites in the ECS. Areas had a high chemical risk index due to the presence of polymers with high hazard scores, such as PVC. Unlike other suspended particles, MPs are not equally distributed in the water column. Shape, size and composition are significant factors which can affect the distribution of MPs. In the present study, 90% of MPs shared similar physical characteristics, allowing for a comparison of MP risk in different areas. Similar methods were used in urban river sediments in Shanghai, which also showed that high chemical toxicity would result in a high environmental risk of MPs (Peng et al., 2018).

The chemical properties of MPs as polymers have been studied by many researchers. Some polymers are regarded as biologically inert, and tend not to have a negative effect on the aquatic environment (Matlack, 2001). However, the polymerization reaction is not always completed during the production process (Lithner et al., 2011). Unreacted residual monomers and other polymerization impurities are released into the environment posing a threat to ecosystems and human health (Araújo et al., 2002). Polymers in MP particles can also exhibit environmental risks. PVC, considered to be the most harmful polymer in this study, is the third most popularly used plastic and its global production is estimated to be 37 million tonnes annually (Lithner et al., 2011). Once PVC enters the environment, it may release carcinogenic monomers and intrinsic plasticizers resulting in lethal effects for marine animals (Green et al., 2016). Some contaminants, such as POPs, can be easily absorbed onto the surface of PVC molecules producing composite ecological effects (Pedà et al., 2016). Given the chemical toxicity of the MP polymers, PE, PP, PS and PVC are generally used as stressors presenting in toxicological test systems (Canesi et al., 2015). During plastic production, some additives are mixed to improve the freshness and maintain the functionality of products (OECD, 2004). For instance, pigments of different functions are utilized in the shipbuilding industry, where alkyd resin is frequently used as a fiber-reinforced plastic matrix on ships. These additives may be easily released from MPs into marine environments due to degradation by sunlight and heat (Davis and Sims, 1983).

3.3.3. The pollution load index of MPs

Based on the PLI, the MP pollution load at each station is given in Fig. 6. Six stations were highly polluted, ten stations were moderately polluted, eight stations had light pollution, and only five stations had negligible levels. The risk within the region was determined using PLIzone and the ratio of high risk stations. According to Pearson's test, there was a significant correlation between polymer risk index and PLI (p = 0.02 < 0.05). It was revealed that areas with a high MP pollution load not only had a high concentration of MPs, but also exhibited a large variety of hazardous polymer. Therefore, increased MP pollution risk is dependent on high levels of accumulation in the environment and the existence of harmful microplastic polymers. According to the assessment result of chemical risk and the PLI, the risk pollution of MPs in surface waters of the Changjiang Estuary and the adjacent ECS is not too serious, with medium pollution (Fig. 6). The level of MP pollution in the ECS was greater than that in the Changjiang Estuary (PLI: 20.4 > 18.4).



Fig. 6. Risk degree of microplastics in the Changjiang Estuary (CE) and the adjacent East China Sea (ECS). Different colors represent different polymer risk indexes and circles with different sizes represent different pollution load indexes. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

The risk of MP pollution is not only due to accumulation of MPs caused by hydrological dynamics, but also frequent human activities which increase the risk of MPs in the environment. The highest PLI of MPs was recorded at sampling stations near the Zhoushan Islands areas (E17 and E21). This area is a large fishing ground and popular visitor location in China, which receives high amounts of wastes from aquaculture and tourism. According to the Bulletin of China Marine Environmental Status in 2016, the waste around Zhoushan was mostly small garbage and beach trash, with over 85% comprising plastic waste (SOA, 2016). Therefore, the Zhoushan Islands are both a source of MPs and a possible risk zone for MP exposure. It is considered that the areas surrounding the Zhoushan Islands are "hotspots", which would benefit from resources allocated to continuous monitoring.

In this study, the risk level of the estuarine area which is considered to be seriously disturbed was lower than that in the open sea. This may be explained by the presence of a sewage treatment plant. Located near the Changjiang Estuary, the Bailonggang Wastewater Treatment Plant, is one of the largest sewage treatment plants in the world, treating $200 \times 10^4 \,\text{m}^3/\text{d}$ (Wang et al., 2012). It has been found that over 90% of MP can be removed with sewage treatment technology (Talvitie et al., 2017), controlling MP pollution from the land into the sea. In the open sea of the ECS, direct dumping in the absence of effective supervision aggravates the MP pollution load. Our understanding of the risks posed by MPs remains limited. The risk of MPs will not cease until the accumulation of plastic waste stops (Wilcox et al., 2013). For this reason, several countries, including Switzerland, Germany, Austria and Belgium, have adopted policies to ban or reduce the production and usage of plastic (Plastics Europe, 2015). China has issued a series of laws to strictly control plastic pollution since the 1980s (Peng et al., 2018). Thanks to these efforts, the amount of plastic waste has been gradually reduced. However, the lack of regulations regarding MP pollution has led to a lack of awareness amongst the general public regarding MP pollution. An option of managing MP pollution worth considering, is to

consult and follow policies relating to plastic pollution, such as the Toxic Substances Control Act and the Marine Strategy Framework Directive (Koelmans et al., 2017).

To date MP pollution in aquatic systems has not been part of conventional environmental assessments. As the problem of MPs becoming increasingly serious, assessing MP risk will be contribute to a full understanding of the status of MP pollutants, migration pathways, the potential harm to human health and help to improve the safety of aquatic environments.

4. Conclusion

An initial assessment of the risk of MP pollution in surface waters was performed in the Changjiang Estuary and the adjacent East China Sea. The environmentally relevant value of MPs provided solid data for risk assessment. High MP concentrations occurred both in the inner Changjiang Estuary and in the open sea of ECS. These two research areas had MPs of similar size, shape and color. Fiber, colored and 0.7-1.0 mm MP particles accounted for a large percentage of all samples. Using polymer category to assess MP risk, the main types were PE and PP based on µ-FTIR technology. PVC was an important source of high MP pollution risk. The assessment of the pollution load index of MPs showed that the study area was moderately polluted. The risk level of MP pollution was related to hydrological dynamics and human activities. The areas surrounding fishing grounds was identified "hotspots" and displayed the highest MP risk. Those areas should be intensively monitored. Increasing evaluation indicators will form the basis of risk assessment in the future. As emphasized in this study, fully understanding the characteristics and distribution of MPs will assist efforts to enact and improve legislation and regulations to reduce MP risk. It is recommended that work on the environmental risk of MPs be carried out as early as possible, to aid MP pollution control.

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References

- Aliani, S., Griffa, A., Molcard, A., 2003. Floating debris in the Ligurian Sea, north-western Mediterranean. Mar. Pollut. Bull. 46, 1142-1149. http://dx.doi.org/10.1016/S0025-326X(03)00192-9.
- Angulo, E., 1996. The Tomlinson pollution load index applied to heavy metal, 'Mussel-Watch' data: a useful index to assess coastal pollution. Sci. Total Environ. 187, 19-56. http://dx.doi.org/10.1016/0048-9697(96)05128-5.
- Araújo, P.H.H., Sayer, C., Poco, J.G.R., Giudici, R., 2002. Techniques for reducing residual monomer content in polymers: a review. Polym. Eng. Sci. 42, 1442–1468. http://dx.doi.org/10.1002/pen.11043.
- Bencze, J.L., Alsop, S. (Eds.), 2014. Activist Science and Technology Education. Springer, Berlin, pp. 652.
- Browne, M.A., Crump, P., Niven, S.J., Teuten, E., Tonkin, A., Galloway, T.S., 2011. Accumulation of microplastic on shorelines worldwide: sources and sinks. Environ.
- Sci. Technol. 45, 9175–9179. http://dx.doi.org/10.1021/es201811s. Canesi, L., Ciacci, C., Bergami, E., Monopoli, M.P., Dawson, K.A., Papa, S., Canonico, B., Corsi, I., 2015. Evidence for immunomodulation and apoptotic processes induced by cationic polystyrene nanoparticles in the hemocytes of the marine bivalve Mytilus. Mar. Environ. Res. 111, 34-40. http://dx.doi.org/10.1016/j.marenvres.2015.06.008.
- Collignon, A., Hecq, J., Galgani, F., Collard, F., Goffart, A., 2014. Annual variation in neustonic micro- and meso-plastic particles and zooplankton in the Bay of Calvi (Mediterranean-Corsica). Mar. Pollut. Bull. 79, 293-298. http://dx.doi.org/10.1016/ i.marpolbul.2013.11.023.
- Davis, A., Sims, D., 1983. Weathering of Polymers. Applied Science Publishers Ltd, United Kingdom (Nov 30).
- Di, M., Wang, J., 2018. Microplastics in surface waters and sediments of the Three Gorges Reservoir, China. Sci. Total Environ. 616, 1620-1627. http://dx.doi.org/10.1016/j. citotenv.2017.10.150
- Doyle, M.J., Watson, W., Bowlin, N.M., Sheavly, S.B., 2011. Plastic particles in coastal pelagic ecosystems of the Northeast Pacific Ocean. Mar. Environ. Res. 71, 41–52. http://dx.doi.org/10.1016/j.marenvres.2010.10.001.
- Foekema, E.M., De, G.C., Mergia, M.T., Van Franeker, J.A., Murk, A.J., Koelmans, A.A., 2013. Plastic in North Sea fish. Environ. Sci. Technol. 47, 8818-8824. http://dx.doi. org/10.1021/es400931b.
- Gao, L., Li, D.J., Ding, P.X., 2008. Nutrient budgets averaged over tidal cycles off the Changjiang (Yangtze River) Estuary. Estuar. Coast. Shelf Sci. 77, 331-336. http://dx. doi.org/10.1016/j.ecss.2007.09.018.
- Gouin, T., Roche, N., Lohmann, R., Hodges, G., 2011. A thermodynamic approach for assessing the environmental exposure of chemicals absorbed to microplastic. Environ. Sci. Technol. 45, 1466-1472. http://dx.doi.org/10.1021/es103202
- Green, D.S., Boots, B., Sigwart, J., Jiang, S., Rocha, C., 2016. Effects of conventional and biodegradable microplastics on a marine ecosystem engineer (Arenicola marina) and sediment nutrient cycling. Environ. Pollut. 208, 426-434. http://dx.doi.org/10. 1016/j.envpol.2015.10.010.
- Henry, S.C., 2013. The incidence of plastic ingestion by fishes: from the prey's perspective. Mar. Pollut. Bull. 74, 170-174. http://dx.doi.org/10.1016/j.marpolbul.2013.07.
- 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. http://www.biomedsearch.com/nih/ Microplastics-in-Marine-Environment-Review/22321064.html.
- Isobe, A., Kubo, K., Tamura, Y., Kako, S., Nakashima, E., Fujii, N., 2014. Selective transport of microplastics and mesoplastics by drifting in coastal waters. Mar. Pollut. Bull. 89, 324-330. http://dx.doi.org/10.1016/j.marpolbul.2014.09.041
- Jabeen, K., Su, L., Li, J.N., Yang, D.Q., Tong, C.F., Mu, J.L., Shi, H.H., 2017. Microplastics and mesoplastics in fish from coastal and fresh waters of China. Environ. Pollut. 221, 141-149. http://dx.doi.org/10.1016/j.envpol.2016.11.055.
- Jambeck, J.R., Geyer, R., Wilcox, C., Siegler, T.R., Perryman, M., Andrady, A., Narayan, R., Law, K.L., 2015. Plastic waste inputs from land into the ocean. Science 347, 768-771. http://science.sciencemag.org/content/347/6223/768.
- Koelmans, A.A., Besseling, E., Foekema, E., Kooi, M., Mintenig, S., Ossendorp, B.C., Redondo-Hasselerharm, P.E., Verschoor, A., Van Wezel, A.P., Scheffer, M., 2017. Risks of plastic debris: unravelling fact, opinion, perception, and belief. Environ. Sci. Technol. 51, 11513-11519. http://dx.doi.org/10.1021/acs.est.7b02219.
- Laurier, F., Mason, R., 2007. Mercury concentration and speciation in the coastal and open ocean boundary layer. J. Geophys. Res. Atmos. 112. http://dx.doi.org/10.1029/ 2006JD007320.
- Law, K.L., Moret-Ferguson, S., Maximenko, N.A., Proskurowski, G., Peacock, E., Hafner, J., Reddy, C.M., 2010. Plastic accumulation in the North Atlantic subtropical gyre. Science 329, 1185-1188. http://dx.doi.org/10.1126/science.1192321.
- Li, J.N., Yang, D.Q., Li, L., Jabeen, K., Shi, H.H., 2015. Microplastics in commercial bivalves from China. Environ. Pollut. 207, 190-195. http://dx.doi.org/10.1016/j envpol.2015.09.018.
- Lithner, D., Larsson, Å., Dave, G., 2011. Environmental and health hazard ranking and assessment of plastic polymers based on chemical composition. Sci. Total Environ. 409, 3309-3324. http://dx.doi.org/10.1016/j.scitotenv.2011.04.038.

- Liu, M., Hou, L.J., Yang, Y., Zou, H., Lu, J., Wang, X., 2001. Distribution and sources of polycyclic aromatic hydrocarbons in intertidal flat surface sediments from the Yangtze estuary, China. Environ. Geol. 41, 90-95. http://dx.doi.org/10.1007 \$002540100347
- Liu, G.P., Lei, G., Zhu, L.X., Li, D.J., 2015. Research on active fluorometer and remote sensing technology of Changjiang Estuary in winter. Mar. Environ. Sci. 34, 245-250. http://dx.doi.org/10.13634/j.cnki.mes.2015.02.015
- Liu, J.N., Du, J.Z., Wu, Y., Liu, S., 2018. Nutrient input through submarine groundwater discharge in two major Chinese estuaries: the Pearl River estuary and the Changjiang River Estuary. Estuar. Coast. Shelf Sci. 203, 17-28. http://dx.doi.org/10.1016/j.ecss. 2018 02 005
- 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. http://dx.doi.org/10.1016/j.marpolbul.2012.11.028.
- Matlack, A.S., 2001. Introduction to Green Chemistry. Marcel Decker Inc., New York. 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. http://dx.doi.org/10.1021/es0010498. Moore, C.J., Moore, S.L., Leecaster, M.K., Weisberg, S.B., 2001. A comparison of plastic and plankton in the North Pacifc Central Gyre. Mar. Pollut. Bull. 42, 1297-1300. http://dx.doi.org/10.1016/S0025-326X(01)00114-X.
- Neufeld, L., Stassen, F., Sheppard, R., Gilman, T., 2016. In the New Plastics Economy: Rethinking the Future of Plastics. World Economic Forum. https:// newplasticseconomy.org/
- OECD, 2004. Emission Scenario Document on Plastic Additives. Series on Emission Scenario Documents, No. 3. Paris: Environmental Directorate, OECD Environmental Health and Safety Publications.
- Ogata, Y., Takada, H., Mizukawa, K., Hirai, H., Iwasa, S., et al., 2009. International Pellet Watch: global monitoring of persistent organic pollutants (POPs) in coastal waters. 1. Initial phase data on PCBs, DDTs, and HCHs. Mar. Pollut. Bull. 58, 1437-1446. http://dx.doi.org/10.1016/j.marpolbul.2009.06.014
- 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. http://dx.doi.org/10.1016/j.envpol.2016.01.083.
- Peng, G.Y., Zhu, B.S., Yang, D.Q., Su, L., Shi, H.H., Li, D.J., 2017. Microplastics in sediments of the Changjiang Estuary, China. Environ. Pollut. 225, 283-290. http://dx. doi.org/10.1016/j.envpol.2016.12.064
- Peng, G.Y., Xu, P., Zhu, B.S., Bai, M.Y., Li, D.J., 2018. Microplastics in freshwater river sediments in Shanghai, China: a case study of risk assessment in mega-cities. Environ. Pollut. 234, 448–456. http://dx.doi.org/10.1016/j.envpol.2017.11.034. Plastics Europe, 2015. Plastics – The Facts 2015, an Analysis of European Plastics
- Production, Demand and Waste Data. Plastics Europe, Brussels, Belgium.
- Plastics Europe, 2016. Plastics The Facts 2016, an Analysis of European Plastics Production, Demand and Waste Data. Plastics Europe, Brussels, Belgium.
- Qu, X.Y., Su, L., Li, H.X., et al., 2018. Assessing the relationship between the abundance and properties of microplastics in water and in mussels. Sci. Total Environ. 621 679–686. http://dx.doi.org/10.1016/j.scitotenv.2017.11.284. SOA, 2016. Bulletin of China Marine Environmental Status. State Oceanic Administration,
- People's Republic of China. pp. 37-38.
- Talvitie, J., Mikola, A., Koistinen, A., Setälä, O., 2017. Solutions to microplastic pollution - removal of microplastics from wastewater effluent with advanced wastewater treatment technologies. Water Res. 123, 401-407. http://dx.doi.org/10.1016/j watres.2017.07.005
- Thompson, R.C., Olsen, Y., Mitchell, R.P., et al., 2004. Lost at sea: where is all the plastic? Science 304, 838. http://science.sciencemag.org/content/304/5672/838
- Tomlinson, D.L., Wilson, J.G., Harris, C.R., Jeffrey, D.W., 1980. Problems in the assessment of heavy-metal levels in estuaries and the formation of a pollution index. Helgoländer Meeresun. 33, 566-575. https://link.springer.com/article/10. 1007%2FBF02414780.
- UNEP, 2016. Marine Plastic Debris and Microplastics: Global Lessons and Research to Inspire Action and Guide Policy Change. United Nations Environment Programme, Nairobi.
- Wang, L.H., Zha, X.Q., Shao, Q., 2012. Design and commissioning of sludge anaerobic digestion system in Bailonggang wastewater treatment plant. China Water Wastewater 28, 43-48.
- Wilcox, C., Hardesty, B.D., Sharples, R., Griffin, D.A., Lawson, T.J., Gunn, R., 2013. Ghostnet impacts on globally threatened turtles, a spatial risk analysis for northern Australia. Conserv. Lett. 6, 247–254. http://dx.doi.org/10.1111/conl.12001. Zhao, S.Y., Zhu, L.X., Wang, T., Li, D.J., 2014. Suspended microplastics in the surface
- water of the Yangtze Estuary System, China: first observations on occurrence, distribution. Mar. Pollut. Bull. 86, 562-568. http://dx.doi.org/10.1016/j.marpolbul. 2014.06.032
- Zhao, S.Y., Zhu, L.X., Li, D.J., 2015. Microplastic in three urban estuaries, China. Environ. Pollut. 206, 597-604. http://dx.doi.org/10.1016/j.envpol.2015.08.02
- Zhao, S.Y., Danley, M., Evan, W.J., Li, D.J., Tracy, J.M., 2017. An approach for extraction, characterization and quantitation of microplastic in natural marine snow using Raman microscopy. Anal. Methods 9, 1470-1478. http://dx.doi.org/10.1039/ C6AY02302A.
- Zhao, J., Guo, L.C., He, Q., Wang, Z.B., van Maren, D.S., Wang, X.Y., 2018. An analysis on half century morphological changes in the Changjiang Estuary: spatial variability under natural processes and human intervention. J. Mar. Syst. 181, 25-36. http://dx. doi.org/10.1016/j.jmarsys.2018.01.007.
- Zhu, J., Ding, P.X., Hu, D., 2003. Observation of the diluted water and plume front off the Changjiang River estuary during august 2000. Oceanol. Limnol. Sinica 34, 249-255. https://www.researchgate.net/publication/285033794_Observation_of_the_diluted_ water_and_plume_front_off_the_Changjiang_River_estuary_during_August_2000.