

## Microplastic Pollution in Table Salts from China

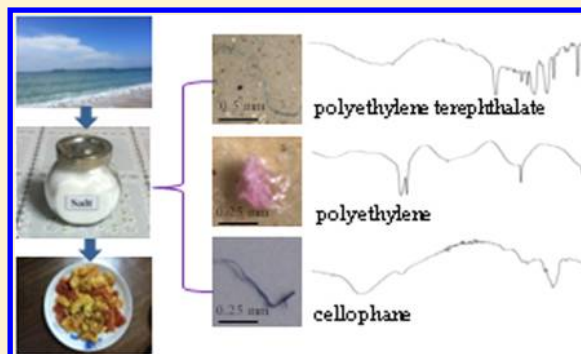
Dongqi Yang,<sup>†</sup> Huahong Shi,<sup>\*,†</sup> Lan Li,<sup>‡</sup> Jiana Li,<sup>†</sup> Khalida Jabeen,<sup>†</sup> and Prabhu Kolandhasamy<sup>†</sup>

<sup>†</sup>State Key Laboratory of Estuarine and Coastal Research, East China Normal University, Shanghai 200062, China

<sup>‡</sup>Research Center for Analysis and Measurement, Donghua University, Shanghai 201620, China

**S** Supporting Information

**ABSTRACT:** Microplastics have been found in seas all over the world. We hypothesize that sea salts might contain microplastics, because they are directly supplied by seawater. To test our hypothesis, we collected 15 brands of sea salts, lake salts, and rock/well salts from supermarkets throughout China. The microplastics content was 550–681 particles/kg in sea salts, 43–364 particles/kg in lake salts, and 7–204 particles/kg in rock/well salts. In sea salts, fragments and fibers were the prevalent types of particles compared with pellets and sheets. Microplastics measuring less than 200  $\mu\text{m}$  represented the majority of the particles, accounting for 55% of the total microplastics, and the most common microplastics were polyethylene terephthalate, followed by polyethylene and cellophane in sea salts. The abundance of microplastics in sea salts was significantly higher than that in lake salts and rock/well salts. This result indicates that sea products, such as sea salts, are contaminated by microplastics. To the best of our knowledge, this is the first report on microplastic pollution in abiotic sea products.



### 1. INTRODUCTION

Marine debris has become a global concern due to its vast and growing threat to the marine and coastal environment.<sup>1,2</sup> The degradation of large, individual plastic items ultimately leads to millions of microplastic pieces, which are likely the most numerous plastic debris in the ocean today.<sup>3</sup> Microplastics are defined as plastic materials or fragments measuring less than 5 mm and have been found not only in the marine environments but also in rivers, lakes, and even in ice.<sup>4–9</sup> Several recent studies showed that the coast of China is a hotspot of microplastic pollution.<sup>2,10–14</sup>

Sea products are among the main sources of food for human beings. The pollution of microplastics in the sea will undoubtedly lead to the pollution of sea products. Microplastics can even be transferred from sea products to humans through the food chain, which increases the potential health risks to humans. Recently, reviews by Seltenrich<sup>15</sup> and Bouwmeester et al.<sup>16</sup> highlighted the importance of investigating the potential risk of transferring microplastics from the food chain to humans. Microplastics have been detected in a large variety of marine organisms, such as mussels and fish.<sup>7,14</sup> To date, however, there has been no report on microplastic pollution in abiotic sea products. Table salts provide essential elements for humans. Salt materials primarily come from the sea, saline lakes, saline rocks, and saline wells. Subsequently, table salts can be classified into sea salts, lake salts, rock salts, and well salts according to their sources. However, rock and well salts are typically labeled as rock/well on the packages of table salts and are regarded as the same type in markets in China.

Sea salt is typically produced by crystallization due to the combined effects of wind and sunlight. Before sea salt

crystallizes, seawater circulates along a series of successive ponds with increasing levels of salinity due to the continuous evaporation of water. Sea salts may contain anthropogenic contaminants present in the seawater if they remain after the concentration and crystallization processes.<sup>17</sup> Therefore, it is necessary to monitor the presence of contaminants in sea salts. Seawater is widely polluted by microplastics; thus, we hypothesize that sea salts might contain microplastics.

To test this hypothesis, we collected different brands of sea salts from random supermarkets throughout China. We also collected lake salts and rock/well salts for comparisons. The abundance, type, and composition of the microplastics were measured and analyzed.

### 2. MATERIALS AND METHODS

**2.1. Collection of Table Salts.** Fifteen brands of table salts were collected from supermarkets in China during October and November of 2014. These salts represent three main types of table salts according to their source in China. An average package with a weight range from 240 to 500 g was chosen. Each type of salt originates from a different location (Supporting Information (SI) Figure S1). One blank extraction group without salt was tested simultaneously to correct the potential procedural contamination. Three replicate packages were used to compare among different brands of the same type

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of salt. Five replicate brands were used for the comparison among the different types.

**2.2. Quality Control of the Experiments.** To avoid contamination, all of the liquid (tap water and hydrogen peroxide) was filtered using 1  $\mu\text{m}$  pore size filter paper prior to use. All containers and beakers were rinsed three times with filtered water. The samples were immediately covered when they were not in use. All the experimental procedures were finished as soon as possible.

**2.3. Hydrogen Peroxide Treatment.** Approximately 240–250 g of table salts from one package of salts was directly placed into a 1 L glass bottle at a height of 35 cm. The sample in one bottle was regarded as a replicate, and four replicate bottles were prepared for each brand. Approximately 100 mL of 30%  $\text{H}_2\text{O}_2$  was added to each bottle to digest the organic matter. The bottles were covered and placed in an oscillation incubator at 65 °C at 80 rpm for 24 h and then at room temperature for 48 h.

**2.4. Floatation and Filtration.** Approximately 800 mL of filtered water was added to each bottle. A glass rod was used to stir the salts in the bottle until they were completely dissolved. The salt solution in one bottle of the four replicates was immediately transferred onto a piece of 5  $\mu\text{m}$  pore size, 47 mm cellulose nitrate filter paper using a vacuum system. The filter paper was then placed into a clean Petri dish with a cover and was dried at room temperature to observe the total number of particles. The three other replicate bottles containing the salt solution were covered with glass lids and held overnight. The supernatants of the salt solutions were transferred onto 5  $\mu\text{m}$  pore size, 47 mm cellulose nitrate filter papers. The filter papers were placed in clean Petri dishes with covers and were dried at room temperature for further microplastic analysis. The material deposited at the bottom of the bottles was also transferred into other Petri dishes for microscopic observation.

**2.5. Visual Observation of Microplastics under a Microscope.** The filters were observed under a Carl Zeiss Discovery V8 Stereomicroscope (MicroImaging GmbH, Göttingen, Germany), and images were obtained with an AxioCam digital camera. A visual assessment was performed to identify the types and colors of microplastics according to the physical characteristics of the particles. Some particles were randomly selected for verification using micro-Fourier Transform Infrared Spectroscopy ( $\mu\text{-FT-IR}$ ). The abundance of microplastics was calculated based on the microscopic observation and was confirmed with  $\mu\text{-FT-IR}$ .

**2.6. Identification of Microplastics with  $\mu\text{-FT-IR}$ .** The plastic-like particles on the filter paper were randomly selected for  $\mu\text{-FT-IR}$  analysis (Thermo Nicolet iN10 MX) in transmittance mode. The spectrum range was set to 4000–675  $\text{cm}^{-1}$  with a collection time of 3 s and with 16 coscans for each measurement. The spectral resolution was 8  $\text{cm}^{-1}$  for all samples, and the aperture size ranged from 50  $\times$  50  $\mu\text{m}$  to 150  $\times$  150  $\mu\text{m}$ , depending on the size of the particles. The 15 table salt packages were also identified using a  $\mu\text{-FT-IR}$  microscope in attenuated total reflection mode. All spectra were collected at a resolution of 8  $\text{cm}^{-1}$  using a diamond MicroTip accessory from 4000 to 675  $\text{cm}^{-1}$ , with a collection time of 3 s and with 16 coscans.

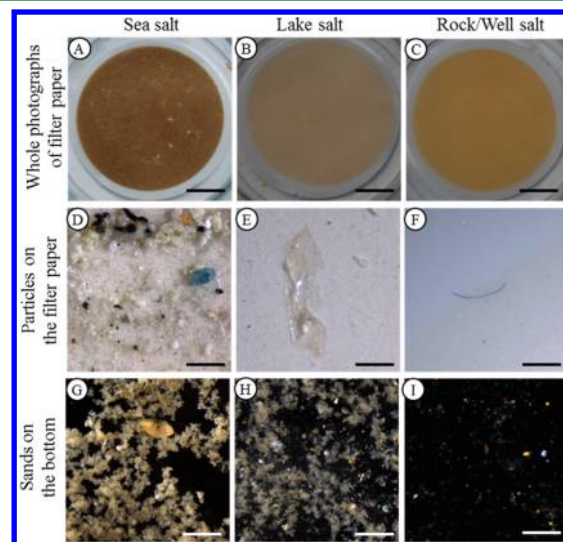
All the spectra were then compared with the library (Hummel Polymer and Additives, Polymer Laminate Films) to verify the polymer type. The spectrum analysis followed the method of Woodall et al.<sup>18</sup> Briefly, matches with a quality index  $\geq 0.7$  were accepted. Matches with a quality index  $< 0.7$  but  $\geq 0.6$

were individually inspected and interpreted based on the proximity of their absorption frequencies to those of chemical bonds in the known polymers. Matches with a quality index  $< 0.6$  were rejected.<sup>18</sup>

**2.7. Data Analysis.** The mean differences of the abundance of microplastics among groups were determined by one-way analysis of variance (ANOVA) followed by Tukey's HSD test (homogeneous variances) or the Tamhane-Dunnnett test (heterogeneous variances), along with multiple comparisons. A 0.05 significance level was chosen.

### 3. RESULTS

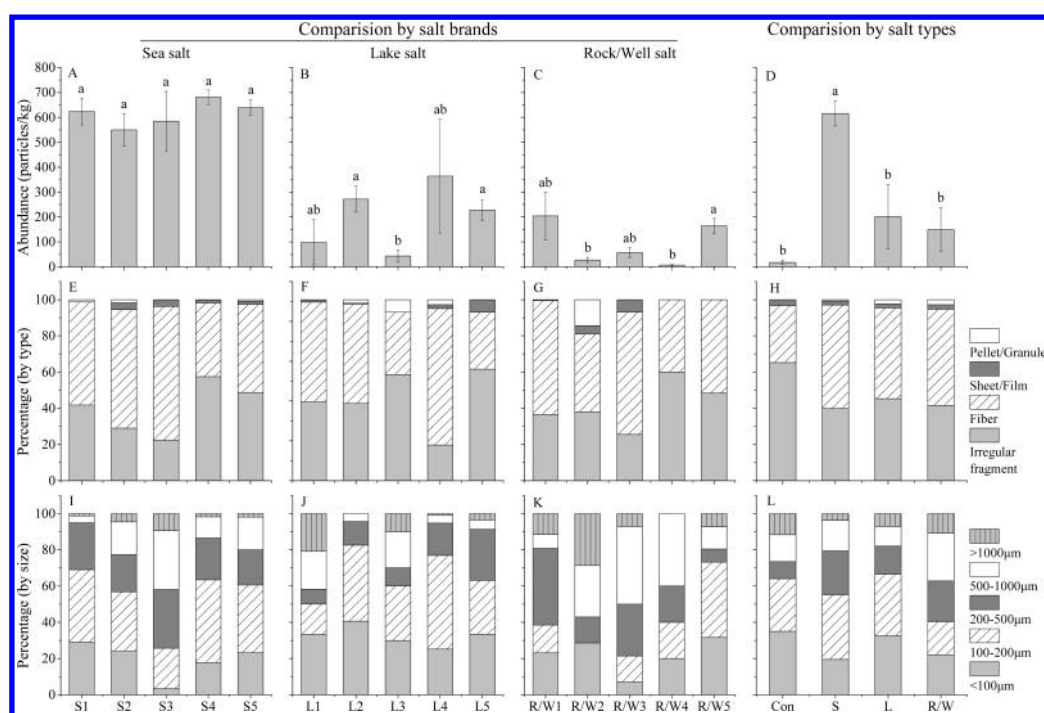
**3.1. Particles in Table Salts.** In the process of handling the samples, contamination with airborne microplastics was prevented. The procedural blanks only contained  $4.4 \pm 2.1$  particles/filter of microplastics, which is equal to 18 particles/kg when the average weight of salt (240–250 g) in each bottle is considered in the control group. When the entire salt solution was filtered, the color of the filter paper for sea salts was much darker than the filter papers for lake salts and rock/well salts (Figure 1A–C). When the supernatants of the salt



**Figure 1.** Photographs of the total particles isolated from table salts. A–C, the particles in the salt solution without separation; D–F, the particles in the supernatant of the salt solutions. More particles were observed in sea salts (D) than lake salts (E) and rock/well salts (F); G–I, the particles at the bottom of the bottle after removal of the supernatant. Scale bar = 10 mm (A–C) or 0.2 mm (D–I).

solutions were filtered, the particle density on the filter papers for sea salts was higher than the particle density on the filter papers for lake salts and rock/well salts (Figure 1D–F). Multiple types of particles, including fibers, fragments, and pellets, occurred in table salts (Figure 1D–F). The most diverse colors were observed in the fibers followed by the fragments. The most common colors were black, red, blue, and white. Some sand particles were also found at the bottom of the bottles (Figure 1G–I).

**3.2. Abundance, Type, and Size of Microplastics in Table Salts.** The number of microplastics was 550–681 particles/kg in sea salts, 43–364 particles/kg in lake salts, and 7–204 particles/kg in rock/well salts (Figure 2A–C). Different brands showed no significant differences in the abundance of microplastics in sea salts ( $p > 0.05$ ) (Figure 2A). Microplastic



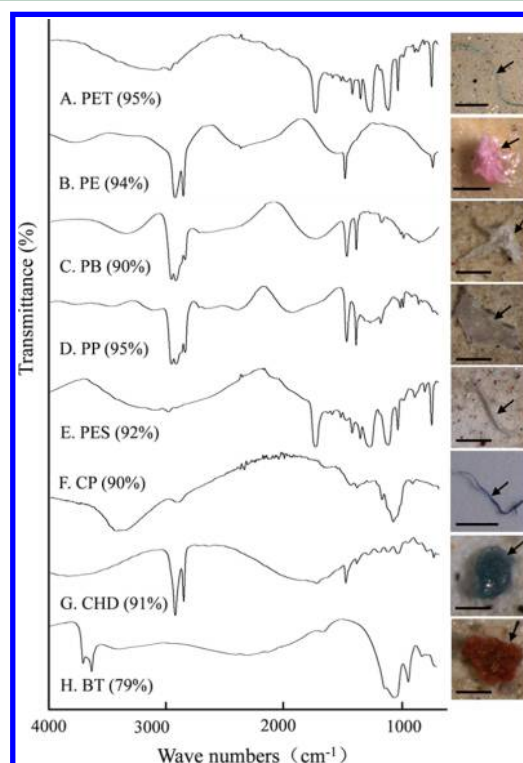
**Figure 2.** Comparison of the abundance (A–D), type (E–H), and size (I–L) of microplastics in table salts ( $n = 3$  for A–C, E–G, and I–J;  $n = 5$  for D, H, and L). Each value represents the mean  $\pm$  standard deviation. The letters above the bars indicate significant differences ( $p < 0.05$ ). If two arbitrary groups have the same letter, then they are not significantly different. The significant differences are not marked in E–L due to limited space. The abbreviations on the X-axis are as follows: Con, control group; L, lake salt; R/W, rock/well salt; S, sea salt.

content was less in L3 than in L2 and L4 in lake salts (Figure 2B), and it was less in R/W2 and R/W4 than in R/W5 in rock/well salts group ( $p < 0.05$ ) (Figure 2C). The average concentration of microplastics in sea salts was approximately 3 times higher than that in lake salts and 7 times higher than that in rock/well salts (Figure 2D) ( $p < 0.05$ ). No significant differences for the abundance were observed among the control, lake salt, and rock/well salt groups ( $p > 0.05$ ).

The composition of microplastic types varied among different brands for the same type of table salts. However, fragments and fibers were the more prevalent types of microplastic particles in sea salts ( $p < 0.05$ ) (Figure 2E). Pellets and sheets accounted for less than 6% of the total number of microplastics in each of the three types of salts (Figure 2E–H). The sizes of the microplastic particles ranged from 45  $\mu\text{m}$  to 4.3 mm in all of the table salts. The microplastics measuring less than 200  $\mu\text{m}$  represented the majority, accounting for 55% of the total number of microplastics in sea salts ( $p < 0.05$ ) (Figure 2I). The proportion of microplastics  $>1$  mm in size reached 28.6% in R/W2 and 20.8% in L1 (Figure 2).

**3.3. Types of Microplastics in Table Salts Identified with  $\mu$ -FT-IR.** One hundred and fifty-two plastic-like particles were selected and identified using  $\mu$ -FT-IR. Various types of microplastics were identified, including polyethylene terephthalate (PET), polyester (PES), polyethylene (PE), poly(1-butene) (PB), polypropylene (PP), and cellophane (CP) (Figure 3A–F). Some nonplastic particles, such as cyclohexane derivatives (CHD) and bentonite (BT), were also identified (Figure 3G–H). The spectrum matches were at least 90% for most of the identified particles.

For the 152 selected particles, 84.9% were identified as microplastics, 6.6% as nonmicroplastics, and 8.6% as unidentified particles. In sea salts, the most common micro-



**Figure 3.** Analysis of microplastics with micro-FT-IR. Abbreviations: BT, bentonite; CP, cellophane; CHD, cyclohexane derivatives; PB, poly(1-butene); PE, polyethylene; PP, polypropylene; PES, polyester; PET, polyethylene terephthalate. The value in the brackets indicates the matches of the spectra with the standards. The black arrows in the photographs indicate the particles that were identified. Scale bar = 0.5 mm (A) or 0.25 mm (B–H).

Table 1. Types of Microplastics Identified with Micro-FT-IR for the Particles Randomly Selected from Table Salts

composition of particles	sea salt		lake salt		rock/well salt		total	
	no. <sup>a</sup>	%	no.	%	no.	%	no.	%
particles measured	52	100	50	100	50	100	152	100
plastic particles	44	84.6 <sup>b</sup>	44	88	41	82	129	84.9
polyethylene terephthalate (PET)	12	27.3 <sup>c</sup>	5	11.4	4	9.8	21	16.3
polyethylene (PE)	9	20.5	0	0	2	4.9	11	8.5
polyester (PES)	6	13.6	2	4.5	1	2.4	9	7
poly(1-butene) (PB)	4	9.1	5	11.4	2	4.9	11	8.5
polypropylene (PP)	1	2.3	3	6.8	2	4.9	6	4.7
PE and PP copolymer (PE-PP)	0	0	2	4.5	1	2.4	3	2.3
polymerized, oxidized material(POM)	1	2.3	4	9.1	1	2.4	6	4.7
polyalkene (PAK)	0	0	4	9.1	0	0	4	3.1
polyacrylonitrile (PAN)	1	2.3	0	0	1	2.4	2	1.6
poly methyl acrylate (PMA)	1	2.3	0	0	0	0	1	0.8
poly(vinyl chloride) (PVC)	0	0	0	0	1	2.4	1	0.8
ethylene vinyl acetate (EVA)	0	0	0	0	1	2.4	1	0.8
poly(vinyl acetate:ethylene) 3:1	1	2.3	0	0	0	0	1	0.8
cellophane (CP)	8	18.2	19	43.2	24	58.5	51	39.5
cellulose (CL)	0	0	0	0	1	2.4	1	0.8
nonplastic particles	6	11.5	2	4	2	4	10	6.6
unidentified particles	2	3.8	4	8	7	14	13	8.6

<sup>a</sup>Number of particles. <sup>b</sup>The percentage of plastic particles in all the measured particles from each type of salt. <sup>c</sup>The percentage of each type in all the plastic particles from each type of salt.

plastic was PET, followed by PE and cellophane (Table 1). In lake salts, however, cellophane was the most common microplastic, accounting for 43.2% of all identified microplastics. In rock/well salts, the most common microplastic was also cellophane. Eight types of nonplastics were identified (SI Table S1). The plastic packages for all salts were determined as PE, except for that of L1 (PET) in this study (SI Figure S2).

## 4. DISCUSSION

**4.1. The Identification of Microplastics.** In the present study, we measured the microplastic pollution in 15 brands of three types of table salts. In the control groups, we found very low concentrations of microplastics. The identification of microplastics was based on a microscopic method and was partially confirmed with  $\mu$ -FT-IR,<sup>9</sup> which is one of the most popular methods used to confirm the composition of microplastics. The spectrum matches reached 98% in a study by Frias et al.<sup>19</sup> The plastic packages containing the salts were analyzed using  $\mu$ -FT-IR in this study, and the spectrum matches were also higher than 98% for 14 of the 15 samples. Due to the complexity of environmental samples, however, the matches are occasionally not high and even show great variations for the same or a similar sample. The factors affecting the matches include environmental degradation and weathering of the microplastic surfaces, inefficient particle recovery, and misidentification of particles.<sup>20</sup> Woodall et al.<sup>18</sup> established a match of 70% as an acceptable value. In the present study, we combined microscopic and spectroscopic methods, which was an effective technique that provided relatively reliable results.<sup>21</sup>

Although there is no uniform definition of microplastics, it has been accepted that some nonplastic synthetic organic particles are also classified as microplastics.<sup>4,20,22</sup> In the present study, we followed this accepted classification for microplastics. For example, cellophane is an organic cellulose-based polymer used in food packaging and cigarette wrappers as well as a release agent in the manufacture of fiberglass and rubber products. Nowadays, cellophane is often used as coatings

combined with synthetic polymers. The shape of fiber cellophane found in this study might originate from the fiberglass products of cellophane or the weathering of broken cellphone films in the environments. In a previous study, cellophane and cellulose were regarded as microplastics.<sup>20</sup> In combination with the morphological features, the corresponding particles matching cellophane and cellulose were classified as microplastics. Although some particles were identified as nonplastics, they were closely related to the compositions or additives of plastics. For example, cyclohexane derivatives are one of the characteristic compounds added to polymers such as PP.<sup>23</sup> Some particles, such as 1-bromooctadecane, might originate from laboratory organic synthesis. However, other particles, such as bentoite and calcium carbonate, are mostly naturally occurring inorganic compounds. Therefore, it is very important to investigate the composition of particles in the identification of microplastics.

**4.2. Microplastic Pollution in Sea Salts.** The much higher microplastic contents in sea salts strongly suggest that they have been contaminated by microplastics. In contrast, lake salts and rock/well salts were less contaminated by microplastics. This difference might be due to the different pollution levels of microplastics in salts from different sources. Currently, many studies have found microplastics in seawater and in lakes.<sup>9,24</sup> In China, the sources of sea salts are from the coastal waters in locations where the population density is very high (SI Figure S1). In recent studies, microplastics have been found in water and in sediments of the coastal and estuarine environments, as well as in commercial bivalves from local markets in China.<sup>10–14</sup> The density of microplastics reached 4137.3 particles/m<sup>3</sup> in the Yangtze Estuary.<sup>10</sup> The average abundance of microplastics reached 5595 particles/m<sup>2</sup> on the beaches of Hong Kong, which was higher than international averages.<sup>12</sup> A high content of microplastics (up to 8714 particles/kg) was also found in the coastal sediments from Hainan Island in China.<sup>13</sup> These results suggest that microplastics are widespread in the coastal and estuarine environ-

ments of China, which might serve as the primary sources of sea salts.

The sources of lake salts are remote mountain lakes located in northwestern China (SI Figure S1). The population density on the east coast of China, where the sea salts in this study originate, was 559 people/km<sup>2</sup> in 2013.<sup>25</sup> However, the population density in the northwest, where the lake salts in this study originate, was only 12 people/km<sup>2</sup>. Undoubtedly, the population density and economic structures will greatly affect the pollution levels of microplastics in the local environments.<sup>2,11</sup> Although there are no available data of microplastics in salt lakes worldwide, including those in China, it is expected that the microplastic pollution level should be much lower in salt lakes in the northwest than in the coastal waters along the east coast of China due to the great differences in the population density and the developing levels of economy.

In contrast to sea salts or lake salts, rock/well salts are typically collected from the underground in China, even as deep as hundreds of meters. It is difficult for synthetic polymers to penetrate into these deep areas. The lowest concentrations of microplastics in rock/well salts suggest that the transport to surface water is the primary route of microplastics introduced into the environment. However, microplastic pollution has other routes, such as atmospheric transport.<sup>16</sup> Microplastic pollution has also been found in honey.<sup>26</sup> The occurrence of microplastics in rock/well salts suggests that microplastics might be introduced into table salts during the production stages of collecting, wind drying, and packaging.

It is reasonable to deduce that the microplastics in sea salts primarily arise from the seawater. However, due to the different methods of investigation, it is difficult to construct precise relationships of the microplastic types, size, and chemical composition between the sea salts and seawater. Plankton nets are typically used in microplastic investigations in seawater, and particles less than 333  $\mu\text{m}$  are not available due to the size of the nets. However, we found that the microplastics measuring less than 200  $\mu\text{m}$  represented the majority of those in sea salts. These results were in accordance with those found in commercial bivalves from markets in China.<sup>14</sup> In nine investigated bivalve species, the microplastics measuring less than 250  $\mu\text{m}$  consisted of 33–84% of the total microplastics calculated by species.<sup>14</sup> In most studies, PE is one of the most common polymers in microplastics in coastal environments.<sup>27,28</sup> However, we found that PET was the most common polymer in sea salts. This might be due to the high density of PET (1.38 g/cm<sup>3</sup>), indicating that it is likely to settle in the raw salts during the specialized processes of sea salt production, such as crystallization.

It is interesting that cellophane was the dominant microplastic in lake salts and rock/well salts in the present study. As a type of packaging debris, cellophane wrappers have even been found in Lake Hovsgol, Mongolia.<sup>4</sup> Cellophane wrappers account for 52% of the total debris ingested by dolphins living in a fully marine environment and 58% in an estuarine environment.<sup>22</sup> Cellophane has also been found as the predominant microplastic in two sample points of the Solent estuarine complex in the UK.<sup>20</sup> Further studies should be conducted to elucidate the reasons leading to the high proportion of cellophane in table salts.

**4.3. The Risk of Microplastics in Sea Salts to Human Health.** Microplastics are a pervasive pollutant present in marine environments worldwide and tend to increase in concentration over time due to plastic fragmentation.<sup>2,3</sup> Adults

should consume less than 5 g of salts according to new guidelines issued by the World Health Organization (WHO). Therefore, for the maximum consumption of sea salts, adults will ingest approximately 1000 microplastic particles each year just from table salts. Humans will also ingest microplastics by consuming other sea products, such as mussels and fish, as well as other microplastic-contaminated food and water.<sup>29</sup> Although the amount of microplastics ingested through salt consumption is much less than that through mollusk consumption by the top European consumers (11,000 microplastic particles per year),<sup>30</sup> more individuals will be affected because table salts are required and consumed in our daily diet.

Microplastics are a particular threat to organisms due to their small size and their capacity to absorb persistent organic pollutants.<sup>31</sup> The constituents of plastics, as well as the chemicals and metals they absorb, may ultimately be ingested by humans through the consumption of seafood. Due to the pollution of seawater, many contaminants have been found in sea salts, including plasticizers, such as di(2-ethylhexyl) adipate and benzyl butyl phthalate.<sup>15</sup> Plastic might be the direct sources of these contaminants. However, plastics might absorb contaminants from the seawater and transfer them to the sea products. Therefore, the presence of marine microplastics in sea salts might pose a threat to food safety.

In sum, we measured the microplastic pollution in sea salts, lake salts, and rock/well salts in China. Our results strongly suggest that the sea salts were contaminated by microplastics, which mostly arise from the seawater columns. To the best of our knowledge, this is the first report on microplastic pollution in abiotic sea products.

## ■ ASSOCIATED CONTENT

### 📄 Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.5b03163.

Figure S1, sources of table salts tested in this study; Figure S2, spectra of the 15 packages of table salts; Table S1, eight nonplastic particles identified with micro-FT-IR (PDF)

## ■ AUTHOR INFORMATION

### Corresponding Author

\*Phone: 86 21 62455593. E-mail: hhshi@des.ecnu.edu.cn.

### Notes

The authors declare no competing financial interest.

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## ■ REFERENCES

- (1) Cózar, A.; Echevarría, F.; González-Gordillo, J. I.; Irigoien, X.; Úbeda, B.; Hernández-León, S.; Palma, Á. T.; Navarro, S.; García-de-Lomas, J.; Ruiz, A. Plastic debris in the open ocean. *Proc. Natl. Acad. Sci. U. S. A.* **2014**, *111*, 10239–10244.
- (2) Jambeck, J. R.; Geyer, R.; Wilcox, C.; Siegler, T. R.; Perryman, M.; Andrady, A.; Narayan, R.; Law, K. L. Plastic waste inputs from land into the ocean. *Science* **2015**, *347*, 768–771.
- (3) Law, K. L.; Thompson, R. C. Microplastic in the seas. *Science* **2014**, *345*, 144–145.
- (4) Free, C. M.; Jensen, O. P.; Mason, S. A.; Eriksen, M.; Williamson, N. J.; Boldgiv, B. High-levels of microplastic pollution in a large, remote, mountain lake. *Mar. Pollut. Bull.* **2014**, *85*, 156–163.

- (5) Yonkos, L. T.; Friedel, E. A.; Perez-Reyes, A. C.; Ghosal, S.; Arthur, C. D. Microplastics in four estuarine rivers in the Chesapeake Bay, U.S.A. *Environ. Sci. Technol.* **2014**, *48*, 14195–14202.
- (6) Obbard, R. W.; Sadri, S.; Wong, Y. Q.; Khitun, A. A.; Baker, I.; Thompson, R. C. Global warming releases microplastic legacy frozen in Arctic Sea ice. *Earth's Future* **2014**, *2*, 315–320.
- (7) Gall, S. C.; Thompson, R. C. The impact of debris on marine life. *Mar. Pollut. Bull.* **2015**, *92*, 170–179.
- (8) Klein, S.; Worch, E.; Knepper, T. P. Occurrence and spatial distribution of microplastics in river shore sediments of the Rhine-Main area in Germany. *Environ. Sci. Technol.* **2015**, *49*, 6070–6076.
- (9) Song, Y. K.; Hong, S. H.; Jang, M.; Han, G. M.; Rani, M.; Lee, J.; Shim, W. J. A comparison of microscopic and spectroscopic identification methods for analysis of microplastics in environmental samples. *Mar. Pollut. Bull.* **2015**, *93*, 202–209.
- (10) Zhao, S.; Zhu, L.; Wang, T.; Li, D. Suspended microplastics in the surface water of the Yangtze Estuary System, China: First observations on occurrence, distribution. *Mar. Pollut. Bull.* **2014**, *86*, 562–568.
- (11) Zhao, S.; Zhu, L.; Li, D. Microplastic in three urban estuaries, China. *Environ. Pollut.* **2015**, *206*, 597–604.
- (12) Fok, L.; Cheung, P. K. Hong Kong at the Pearl River Estuary: A hotspot of microplastic pollution. *Mar. Pollut. Bull.* **2015**, *99*, 112–118.
- (13) Qiu, Q.; Peng, J.; Yu, X.; Chen, F.; Wang, J.; Wang, J.; Dong, F. Occurrence of microplastics in the coastal marine environment: First observation on sediment of China. *Mar. Pollut. Bull.* **2015**, *98*, 274–280.
- (14) Li, J.; Yang, D.; Li, L.; Jabeen, K.; Shi, H. Microplastics in commercial bivalves from China. *Environ. Pollut.* **2015**, *207*, 190–195.
- (15) Seltenrich, N. New link in the food chain? Marine plastic pollution and seafood safety. *Environ. Health Perspect.* **2015**, *123*, A34–A41.
- (16) Bouwmeester, H.; Hollman, P. C. H.; Peters, R. J. B. Potential health impact of environmentally released micro- and nanoplastics in the human food production chain: Experiences from nanotoxicology. *Environ. Sci. Technol.* **2015**, *49*, 8932–8947.
- (17) Serrano, R.; Nacher-Mestre, J.; Portolés, T.; Amat, F.; Hernández, F. Non-target screening of organic contaminants in marine salts by gas chromatography coupled to high-resolution time-of-flight mass spectrometry. *Talanta* **2011**, *85*, 877–884.
- (18) Frias, J. P.; Otero, V.; Sobral, P. Evidence of microplastics in samples of zooplankton from Portuguese coastal waters. *Mar. Environ. Res.* **2014**, *95*, 89–95.
- (19) Gallagher, A.; Rees, A.; Rowe, R.; Stevens, J.; Wright, P. Microplastics in the Solent estuarine complex, UK: An initial assessment. *Mar. Pollut. Bull.* **2015**, DOI: 10.1016/j.marpollbul.2015.04.002.
- (20) 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. The deep sea is a major sink for microplastic debris. *R. Soc. Open Sci.* **2014**, *1*, 140317.
- (21) Song, Y. K.; Hong, S. H.; Jang, M.; Kang, J.-H.; Kwon, O. Y.; Han, G. M.; Shim, W. J. Large accumulation of micro-sized synthetic polymer particles in the sea surface microlayer. *Environ. Sci. Technol.* **2014**, *48*, 9014–9021.
- (22) Denuncio, P.; Bastida, R.; Dassis, M.; Giardino, G.; Gerpe, M.; Rodríguez, D. Plastic ingestion in Franciscana dolphins, *Pontoporia blainvilliei* (Gervais and d'Orbigny, 1844), from Argentina. *Mar. Pollut. Bull.* **2011**, *62*, 1836–1841.
- (23) Dekiff, J. H.; Remy, D.; Klasmeyer, J.; Fries, E. Occurrence and spatial distribution of microplastics in sediments from Norderney. *Environ. Pollut.* **2014**, *186*, 248–256.
- (24) Eriksen, M.; Mason, S.; Wilson, S.; Box, C.; Zellers, A.; Edwards, W.; Farley, H.; Amato, S. Microplastic pollution in the surface waters of the Laurentian Great Lakes. *Mar. Pollut. Bull.* **2013**, *77*, 177–182.
- (25) National Bureau of Statistics of China. Population at year-end of 2013 by region. *China Statistical Yearbook*; Beijing, 2014.
- (26) Liebezeit, G.; Liebezeit, E. Non-pollen particulates in honey and sugar. *Food Addit. Contam., Part A* **2013**, *30*, 2136–2140.
- (27) Hidalgo-Ruz, V.; Gutow, L.; Thompson, R. C.; Thiel, M. Microplastics in the marine environment: A review of the methods used for identification and quantification. *Environ. Sci. Technol.* **2012**, *46*, 3060–75.
- (28) Vianello, A.; Boldrin, A.; Guerriero, P.; Moschino, V.; Rella, R.; Sturaro, A.; Da Ros, L. Microplastic particles in sediments of Lagoon of Venice, Italy: First observations on occurrence, spatial patterns and identification. *Estuarine, Coastal Shelf Sci.* **2013**, *130*, 54–61.
- (29) Van Cauwenberghe, L.; Claessens, M.; Vandegehuchte, M. B.; Janssen, C. R. Microplastics are taken up by mussels (*Mytilus edulis*) and lugworms (*Arenicola marina*) living in natural habitats. *Environ. Pollut.* **2015**, *199*, 10–17.
- (30) Van Cauwenberghe, L.; Janssen, C. R. Microplastics in bivalves cultured for human consumption. *Environ. Pollut.* **2014**, *193*, 65–70.
- (31) Engler, R. E. The complex interaction between marine debris and toxic chemicals in the ocean. *Environ. Sci. Technol.* **2012**, *46*, 12302–12315.