



Microplastics in take-out food containers

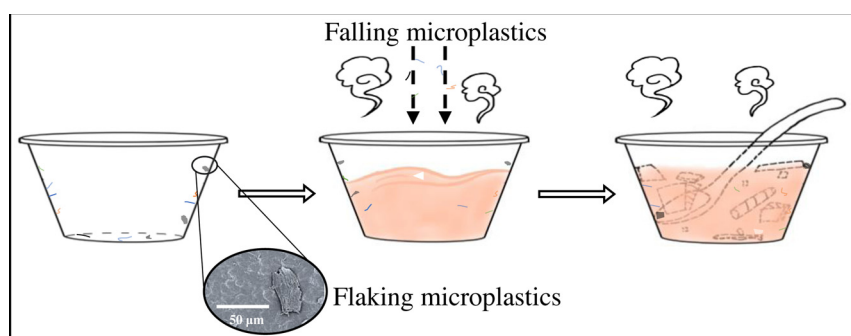
Fangni Du^a, Huiwen Cai^a, Qun Zhang^{a,b}, Qiqing Chen^a, Huahong Shi^{a,*}

^a State Key Laboratory of Estuarine and Coastal Research, East China Normal University, Shanghai 200241, China

^b School of Ecological and Environmental Sciences, East China Normal University, Shanghai 200241, China



GRAPHICAL ABSTRACT



ARTICLE INFO

Editor: R. Teresa

Keywords:

Microplastics
Take-out container
Human ingestion
Human health

ABSTRACT

Microplastics have been detected in various media including water, sediment, and seafood, whereas there are few studies focusing on microplastics in take-out containers. In this study, we collected take-out containers made of common polymer materials (polypropylene, PP; polystyrene, PS; polyethylene, PE; polyethylene terephthalate, PET) from five cities in China. Microplastics in the containers were analyzed after different treatments (direct flushing and flushing after immersing with hot water). Our results showed that microplastics were found in all take-out containers and abundance ranged from 3 to 29 items/container. The highest abundance occurred in PS containers with rough surface. The polymer types of some detected particles were the same as those of original containers, accounting for 30% of the total microplastics; other types included polyester, rayon, acrylic, and nylon. Treating the containers with hot water did not influence microplastic abundance. Our study indicates that microplastics in take-out containers come from atmospheric fallout and flakes from container's inner surfaces. Under slight mechanical force, loose structure and rough surface of PS containers can flake off microplastics, entering water more easily. Based on the microplastic abundance in take-out containers, people who order take-out food 4–7 times weekly may ingest 12–203 pieces of microplastics through containers.

1. Introduction

Plastic production has significantly increased from 1.7 million tons in the 1950s to over 348 million tons in the 2010s (PlasticsEurope, 2018). Due to the lack of strict regulations on their disposal, a large

number of plastic wastes have ended up in aquatic and terrestrial environments, causing serious pollution (Derraik, 2002; Chae and An, 2018). Plastic wastes pose even greater dangers when they break into small pieces, called microplastics (size < 5 mm) (Thompson et al., 2004). The environmental and ecological risks of microplastics have

* Corresponding author.

E-mail address: hshshi@des.ecnu.edu.cn (H. Shi).

<https://doi.org/10.1016/j.jhazmat.2020.122969>

Received 19 February 2020; Received in revised form 14 May 2020; Accepted 14 May 2020

Available online 26 May 2020

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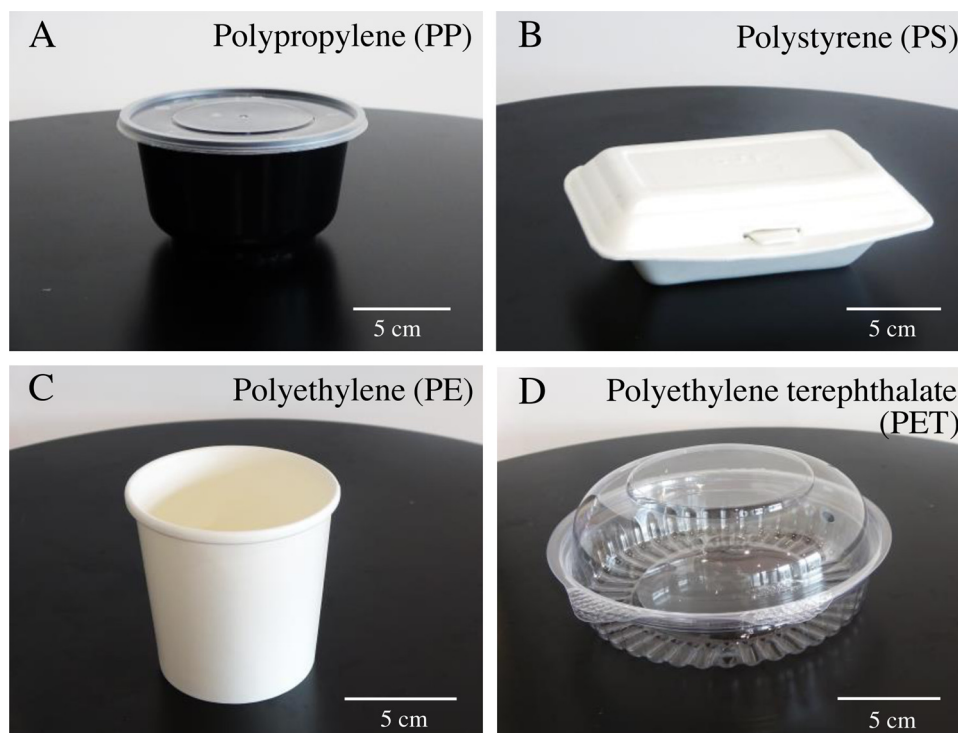


Fig. 1. Four types of take-out container.

been well documented using various matrices (Andrady, 2011; He et al., 2018). In recent years, risks of potential human health have attracted more attention since microplastics have been found in various human foods and environmental compartments, such as drinking water, table salt, sugar, and air (Liu et al., 2019; Li et al., 2016; Liebezeit and Liebezeit, 2013; Yang et al., 2015; Pivokonsky et al., 2018). It has been estimated that human consumption of microplastics via air and commonly consumed foods ranged from 203 to 332 particles per person per day (Cox et al., 2019).

Microplastics can enter human body via digestive tract through food consumption, which has been confirmed by microplastics found in human feces (Yan et al., 2020). Among food varieties, seafood has been widely studied using the total soft tissues of some species that are esculent, such as shellfish (Li et al., 2015; Van Cauwenberghe and Janssen, 2014). This increases the possibility of microplastics transferring from food to humans. In addition to the inner-source pollution caused by biological ingestion of microplastics, microplastic contamination in food also comes from external-source pollution, such as atmosphere deposition. Microplastics have been found both in indoor and outdoor air (Dris et al., 2015; Wright et al., 2020; Zhang et al., 2019). Catarino et al. (Catarino et al., 2018) found that household fibers fallout can cause contamination of food. In addition to air contamination, food package is another important external source of microplastics. For instance, plastic package was deduced to be a potential source of microplastics in table salt (Iniguez et al., 2017); plastic bottles cap can release microplastics into bottled water (Winkler et al., 2019); plastic teabags can release both microplastics and nanoplastics into steeped tea (Hernandez et al., 2019). Policy makers attempt to control proliferation of plastic pollution by enacting laws of plastic bag restriction; yet, plastics are still widely used by the food package industry, providing microplastic sources (Trevor, 2018; Rochman et al., 2015).

With the development of social economy and online business, take-out industry develops rapidly (NDRC (National Development and Reform Commission), 2019). It is convenient and low-cost to order take-out foods, especially for white-collar workers. However, food safety of plastic package cannot be ignored because the package is in contact with food directly and related to human intake (Marsh and

Bugusu, 2007). In fact, human exposure to microplastics via plastic food packages has been reported in previous studies (Winkler et al., 2019; Hernandez et al., 2019). Among the plastic food packages, take-out containers are widely used and act as an external source for human exposure to microplastics, especially considering their poor heat resistances and often being used for containing hot food for delivery. Meanwhile, microplastics from polypropylene food containers have been isolated and characterized in the latest report (Fadare et al., 2020). However, there has been no study focusing on the influence of different plastic materials used for making these containers, as well as the high temperature on microplastic abundance.

In this study, we collected four types of take-out containers from five cities in China. To simulate custom of eating food from such containers and delivery process, the containers were treated with two methods: directly flushing the interior of each container, or flushing it after putting hot water in the container for 30 min. The microplastics and the surfaces of containers after different treatments were characterized. Human intake of microplastic via take-out containers was then estimated based on the microplastic abundances and take-out ordering frequency of high-risk consumers.

2. Materials and methods

2.1. Collection of take-out containers

Before the experiment, in order to understand the components of the take-out containers commonly used, we ordered 100 take-out food items from different restaurants in Shanghai, and kept the containers. The chemical compositions of the containers were identified by the Attenuated Total Reflection Fourier Transform Infrared spectroscopy (ATR-FTIR) (Thermo Scientific, Nicolet iS5N, USA). The spectrum range was set to $4000\text{--}600\text{ cm}^{-1}$ with 16 scans for each measurement, and the spectral resolution was 4 cm^{-1} for all samples. After the identification, four commonly used types of take-out containers (polypropylene, PP; polystyrene, PS; polyethylene, PE; polyethylene terephthalate, PET) were chosen and purchased from February to May 2019 (Fig. 1).

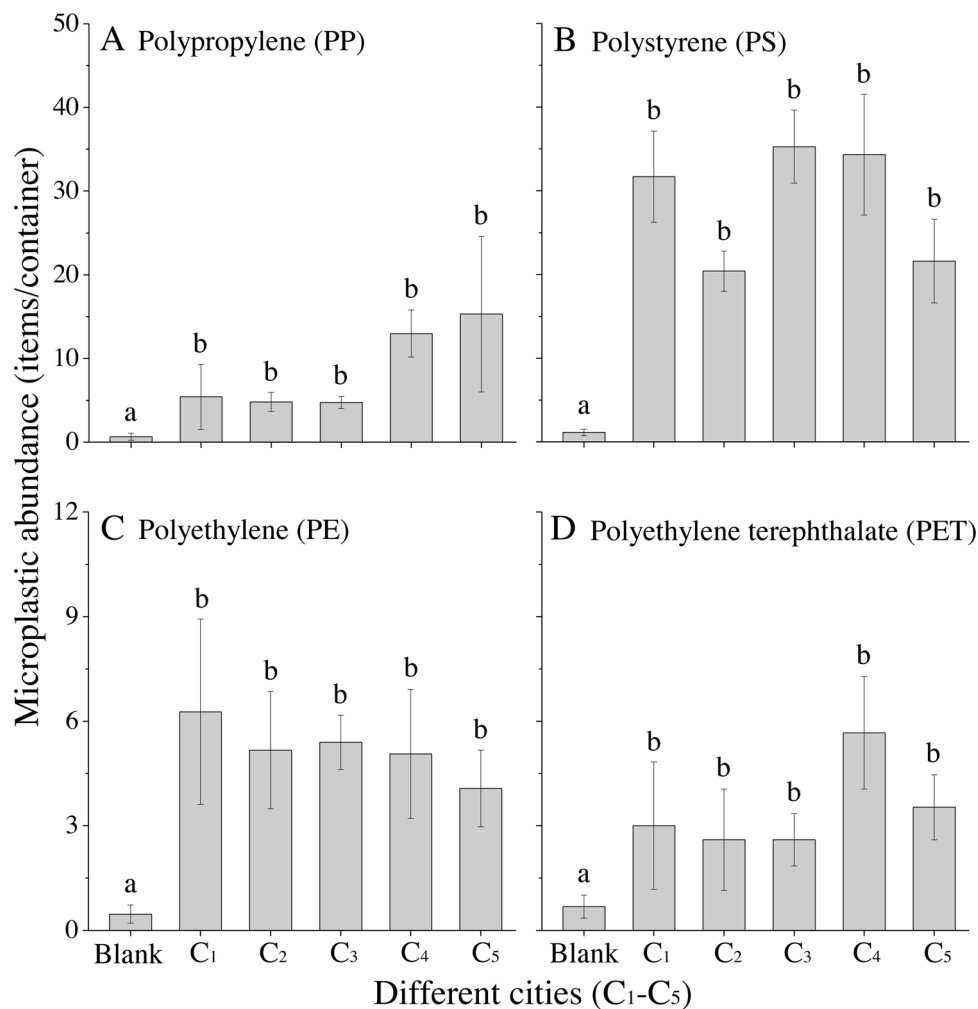


Fig. 2. Microplastic abundance of take-out containers in different cities. Each group included three replicate ($n = 3$).

The take-out containers came from several suppliers of five cities in China: Shijiazhuang, Qingdao, Chengdu, Hangzhou, and Xiamen. The containers were purchased online and mailed to our school by express delivery. They were packed and sealed in cardboard containers before entering the laboratory, with 100 containers stacked together in a group. To avoid microplastic contamination from the air, we chose containers in the middle of each stack as our samples.

2.2. Different treatments of containers and separation of microplastics

Before the formal experiment, pre-experiment was conducted to simulate various eating conditions including direct flushing, flushing after immersion with hot water, flushing after microwave heating for one minute, and flushing after refrigeration for six hours. Except for PP containers, the other three types of containers were not suitable for microwave or cryopreservation due to their poor tolerance of heat or refrigeration. Combined with the commonly use rules in real life, containers were treated with direct flushing and flushing after immersion with hot water. The first treatment was to simulate custom of eating take-out food (the friction of food with tableware) by flushing the inner surface wall of each container three times with Milli-Q water. Then, the water was filtered through 5 μm membrane (MF-Millipore™) using glass filters (T-50, JinTeng, China) and vacuum pumps (GM-1.0A, JinTeng, China). For the second treatment, hot water (100 °C) was added to the untreated containers. The containers were then covered by lids and placed in incubators with 20 rpm for 30 min to simulate the delivery process. Then, the containers were flushed with Milli-Q water; the

flushed water was filtered through membrane filters, as the steps in the first treatment. After filtration, the filter membranes were put into glass dishes ($D = 60$ mm) immediately; the dishes were then kept in the cabinet of a clean room at room temperature for future analysis. The experiment for each type of take-out container for each treatment was repeated three times. Since PET containers have bad heat resistance and are not used as hot food containers, they were not treated with hot water.

Control groups (glass beakers) were also treated in the two ways to correct the potential procedural contamination. In addition, air quality of the laboratory was strictly controlled using three air purifiers (6000 V, Allerair, Canada). Both samples and control groups were covered with aluminized papers or lids when not being processed.

2.3. Observation and identification of microplastics

The particles on the membranes were observed under a Carl Zeiss Discovery microscope (Micro Imaging GmbH, Gottingen, Germany). Their physical characteristics including shapes and colors were recorded. Image J software was used to measure particle sizes. Three size classes were taken into account: ≤ 500 , 501–1000, and ≥ 1001 μm . Chemical compositions of the particles were identified by $\mu\text{-FTIR}$ (Thermo Scientific, Nicolet iN10, USA) using the transmission mode. The spectrum range was set to 4000–600 cm^{-1} with 16 scans for each measurement, and the spectral resolution was 4 cm^{-1} for all samples. The spectra were compared with the Thermo Scientific Infrared spectra library and our semi-synthetic celluloses database. A quality index of at

least 70% match was considered acceptable (Li et al., 2018).

2.4. Observation of inner surface wall of take-out containers

In order to observe the characteristics of inner surfaces of the containers after the two treatments, scanning electron microscope (SEM) (SYST TA PRO 1156) was adopted, which can provide high resolution imaging of material surface. SEM operating parameters were as follows: Accelerating Voltage 3 kV, Emission Current 13,300 nA, and Working Distance varying from 10.9 to 11.7 mm. SEM images were taken from different areas of each sample with various magnifications (30–100 x).

2.5. Data analysis

Statistical analysis was performed using SPSS 18.0 (IBM Corp, Armonk, NY, USA). Microplastic abundance of different types of containers and after different treatments were tested by one-way ANOVA, followed by Fisher's LSD post hoc test for multiple comparisons. The Kruskal-Wallis test was applied to test the differences in microplastic abundances of the take-out containers from different cities, as well as abundances of microplastics in the same city after different treatments. We took $p < 0.05$ as a significant difference.

3. Results

3.1. Microplastics in take-out containers from different cities

In order to analyze the differences in production and transport process of the containers by different manufacturers, as well as the effects on microplastic characteristics in the containers, the collected microplastics were analyzed and compared. According to the data of suspected particles measured, microplastic abundance of the take-out containers ranged from 1 to 41 items/container (Fig. 2). The physical characteristics of microplastics were also observed. Fiber was the dominant shape, accounting for more than 50% of the total microplastics (Table S1). High proportion (55%) of microplastics with sizes smaller than 500 μm was found in the samples, with the smallest size being 43 μm (Table S2). The main colors of microplastics were transparent and white, with the total proportion ranging from 63% to 98% (Table S3). The abundances and characteristics (size, shape, and color) of microplastics showed no significant differences among different cities.

3.2. Microplastics in take-out containers of different types

The abundances and characteristics of microplastics in the containers made of different materials were compared. Microplastic abundances were 9 items/container in PP containers, 29 items/container in PS containers, 5 items/container in PE containers, and 3 items/container in PET containers; the highest value occurred in PS containers (Fig. 3A). In PS containers, the proportion of fiber was higher than those of the other three types of containers (Fig. 3B). The main sizes of the microplastics in PS containers ($> 500 \mu\text{m}$) were longer than those in the other three types of containers ($< 500 \mu\text{m}$) (Fig. 3C). The proportions of different microplastic colors were similar in the four types of containers (Fig. 3D).

A total of 819 suspected particles were identified using $\mu\text{-FTIR}$. Natural polymers accounted for the majority of the total particles (57%), and the main polymer type was cotton. The identified microplastics included rayon, acrylic, nylon, polyester, PP, PS, PE, and PET (Fig. 4). Among them, some were in the same color and component with their original containers (PP, PS, PE, and PET); thus, they were considered as microplastics flaking from the take-out containers. Other types of microplastics (rayon, acrylic, nylon, and polyester) were considered to be introduced from another source. In different types of the containers, the proportion of flaking microplastics showed its highest

value in PS containers (62 %), followed by PP containers (32 %), PE containers (22 %), and PET containers (3 %).

3.3. Microplastics in take-out containers after different treatments

In the pre-experiment, four treatments including direct flushing, flushing after immersion with hot water, flushing after microwave heating for one minute, and flushing after refrigeration for six hours were used to treat PP containers from Hangzhou and Xiamen cities. The abundances of microplastics showed no significant difference in PP containers among the four treatments (Table S4). Considering the material characteristics of the containers and the commonly used rules, the two treatments of direct flushing and flushing after immersion with hot water were used in the formal experiments. Eating food from take-out container was simulated through direct flushing the inner surface of the container (treatment 1), and the take-out food delivery process was simulated through treating the container with hot water and shaking (treatment 2). PET containers were excluded in treatment 2 because they would deform by hot water. In general, the abundances of total microplastics showed no significant differences between treatments 1 and 2 in the three types of containers (Fig. 5). To determine the possible effects of hot water treatment on take-out containers themselves, abundances of flaking microplastics were analyzed, which showed no change after hot water treatment. Total of eight types (rayon, acrylic, nylon, polyester, PP, PS, PE, and PET) of synthetic polymers were detected after treatment 2 (Fig. S1). The proportions of flaking microplastics were 25 % (PP), 77 % (PS) and 24 % (PE) (Fig. S1).

3.4. Characteristics of container surface after different treatments

SEM was adopted to observe the inner surfaces of the take-out containers after the two treatments. After treatment 1, the surfaces of PP and PET containers were relatively smooth, with several protrusions on them (Fig. 6A₁ and D₁). In contrast, the PS surface was rough with many broken places, with a size range of 20–100 μm (Fig. 6B₁). For the PE surface, there were folds with size of 5–15 μm (Fig. 6C₁). All containers showed a few particles adhered or detached on the surface (Fig. 6A₁, C₁, and D₁).

After the hot water treatment, obvious changes occurred on the surfaces of PP and PE containers while there was almost no change on the PS surfaces (Fig. 6A₂–C₂). Irregular objects with diameters of about 1–5 μm appeared on the surface of PP containers after the hot water treatment (Fig. 6A₂). The PE container surface was more uneven, with PE sprayed film separating clearly from the base (Fig. 6C₂).

4. Discussion

4.1. Characteristics of microplastics in take-out containers of different types

In this study, we collected take-out food containers from five cities in order to detect the influence of production and transport process of the containers by different manufacturers on microplastic abundances. As expected, microplastic characteristics showed no significant differences among different cities. Therefore, the abundance data we obtained are representative for general microplastic level of take-out food containers in China.

To detect the influence of container component on microplastic abundance, four types of commonly used take-out containers were collected. The highest abundance of total microplastics (29 items/container) and flaking microplastics (18 items/container) were presented in PS containers, while much lower abundances were found in other three types of containers. The differences of microplastic abundances among different types of take-out containers may be associated with different material characteristics caused by different manufacturing processes. For instance, PS containers are formed through injecting gas into PS masterbatch melts, which results in relatively loose

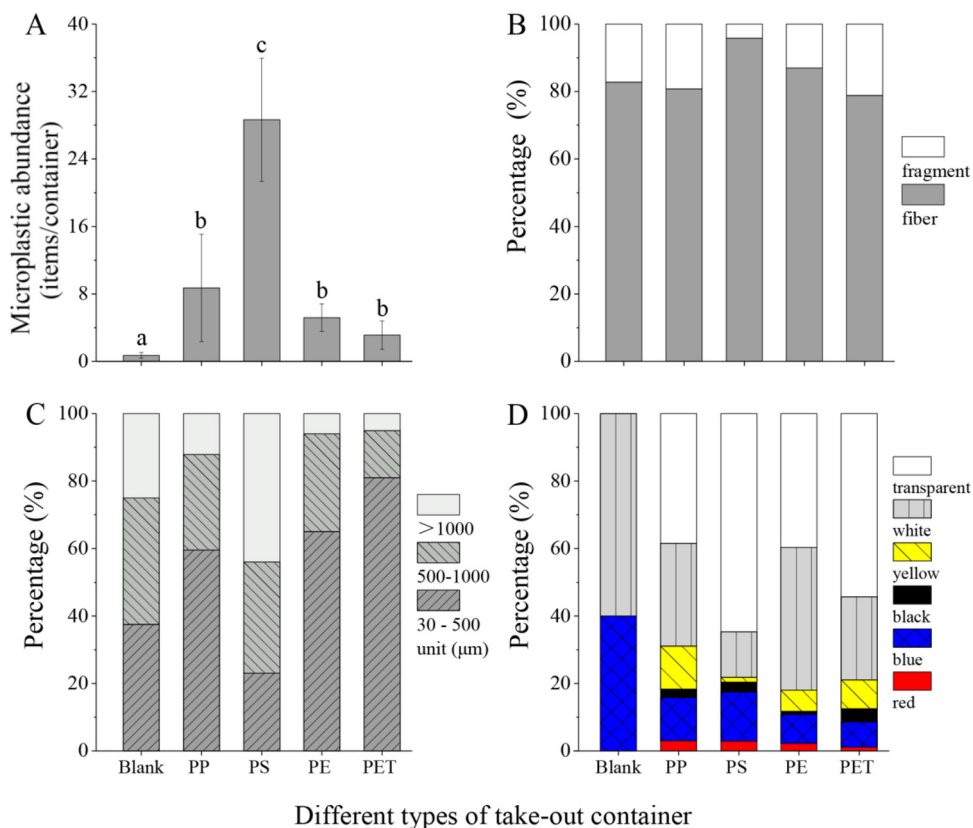


Fig. 3. Characteristics of microplastics in take-out containers of different types. A, abundance; B, shape; C, color; D, size. Abbreviations: PP, polypropylene; PS, polystyrene; PE, polyethylene; PET, polyethylene terephthalate. Each group included fifteen replicates (n = 15).

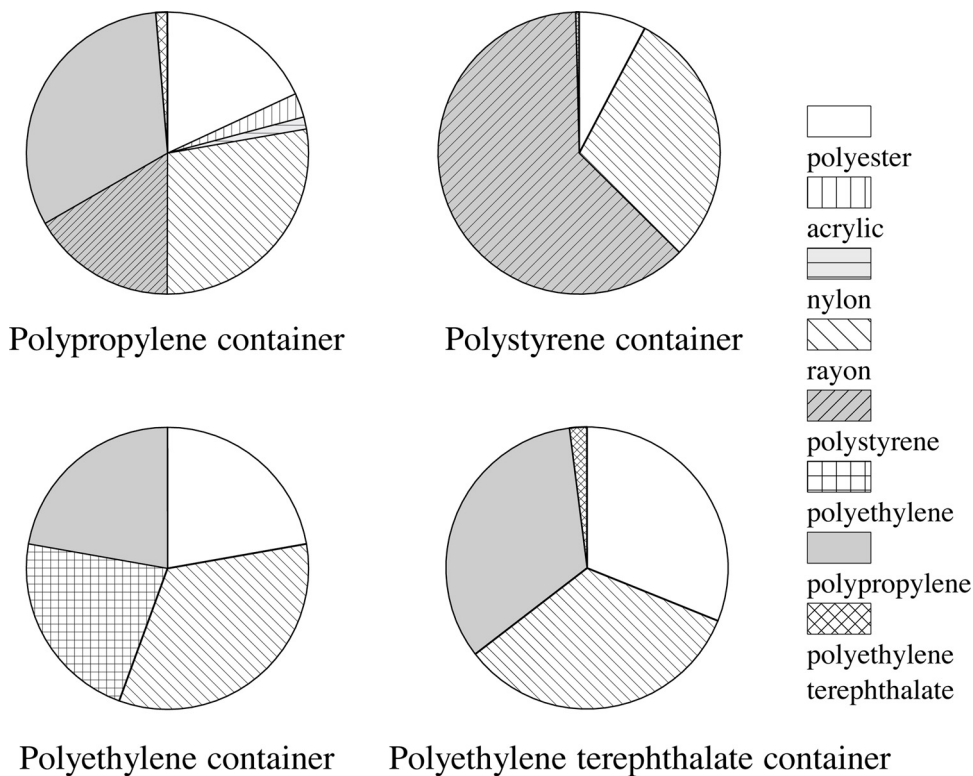


Fig. 4. Chemical compositions of microplastics in take-out containers of different types.

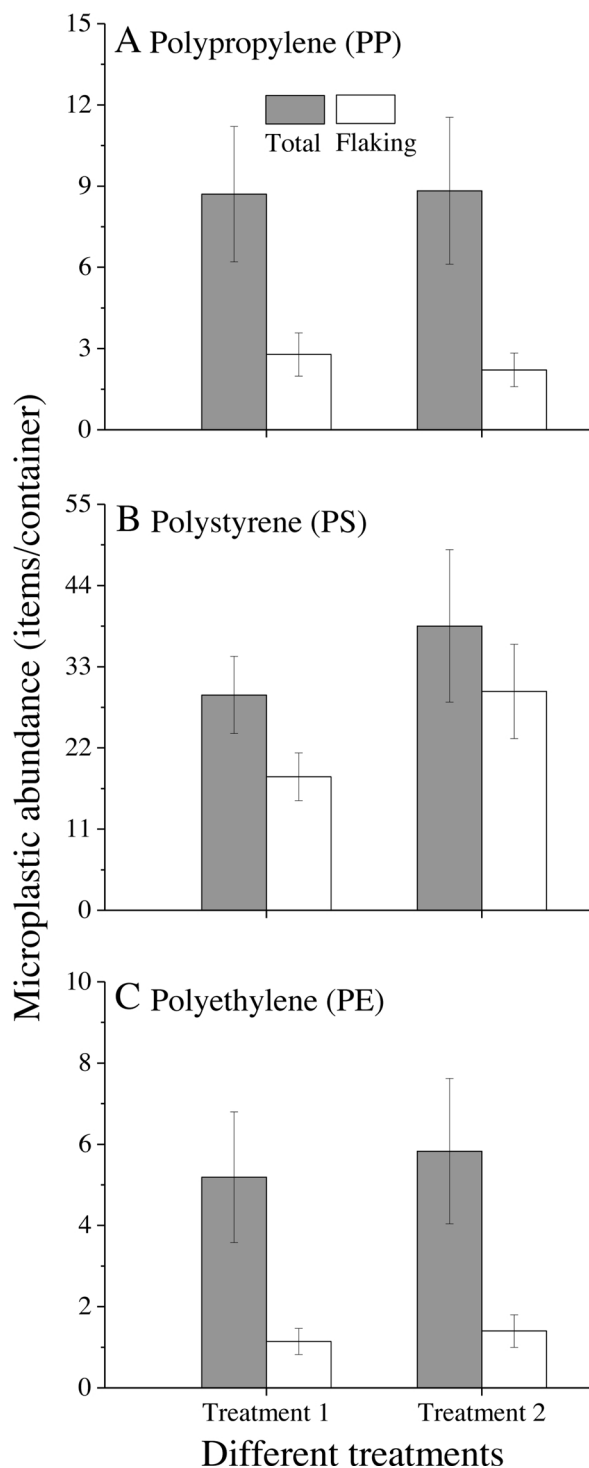


Fig. 5. Abundance of total and flaking microplastics from containers after different treatments. Treatment 1: Direct flushing; Treatment 2: Flushing after immersion in hot water for 30 min. Each group included fifteen replicates ($n = 15$).

structure and rough surface of the container. The manufacturing processes of PP and PET containers are similar; they are both formed through pressurized injection of the melted masterbatch into mold cavity, leading to smooth surfaces. As for PE containers, they were made by spraying PE film on the inner surface of the paper container. The different surface characteristics of the containers were clearly reflected in SEM images (Fig. 6). There was a large number of damaged parts on the surfaces of PS containers, whereas the surfaces of PE, PP,

and PET containers remained smooth. Taking the microplastic abundance, manufacturing process and surface characteristics of different types of containers into consideration, we speculate that the loose structure and rough surface of PS container may cause microplastics flaking from the inner surface more easily than the other three types of containers. Therefore, denser materials with smooth surfaces are more suitable for take-out food packaging, to reduce microplastics flaking from the container.

The polymer types and color of microplastics we separated are the same as those of their original containers (PP, PS, PE, and PET), which may be resulted from flaking microplastics from the containers themselves. Polyester, nylon, acrylic, and rayon were also found. These particles, accounting for 70% of the total microplastics, likely came from atmospheric microplastic pollution. In other words, apart from the flaking microplastics caused by the containers themselves, microplastics from the air should not be ignored, either. We considered that the air contamination was from the process of production, storage and transport of the take-out containers. Atmospheric microplastic pollution is widespread in both indoor and outdoor environments. In a previous study, 175–313 items/m²/day of microplastics have been found in Dongguan city, China (Cai et al., 2017). More serious pollution was found in indoor environments, ranging from 523 to 3673 items/m²/day (Dris et al., 2017). The contamination of microplastics from air is not only limited to take-out containers, but also occurs in other food packages. Thus, it is necessary to reduce atmospheric microplastic pollution in workshops during production process of various food or food packages. Similarly, air contamination also occurs in laboratory. In the present study, air quality was strictly controlled during experiments, and glass containers were used as control. As a result, small number of microplastics were found in control groups. However, air contamination is extremely hard to eliminate even in strictly controlled conditions (Woodall et al., 2015; Torre et al., 2016). More types of controls (e.g., negative control and positive control) can improve the reliability of results, and therefore, should be adopted in our future studies (Koelmans et al., 2019).

4.2. Influences of treatments on microplastic abundance in take-out containers

Microplastic abundances of the containers were analyzed to detect the influences of the two treatments. Microplastics were detected after we flushed the inner surface of the container, which means that these particles can flake from take-out containers into food easily through slight mechanical force. In real life, the friction of food or tableware with the inner surface of a take-out container is similar to that of the water flushing used in our experiments, so the microplastic abundance in reality may be similar to the microplastics abundance in this study. Compared with take-out containers, non-disposable tablewares do not involve “flaking microplastics” since they are commonly made of non-plastic materials, such as stainless steel and ceramic. Also, microplastics from the air can also be removed by washing them before using them for meals. In other words, disposable take-out containers increase the risk of human exposure to microplastics in two ways.

Unexpectedly, microplastic abundances did not show significant differences between the two treatments. It indicates that the treatment of hot water and shaking, which is similar to what happens during the food delivery, has no significant influence on microplastic abundance of take-out containers. This means that the temperature (cold or hot) and the shaking of food containers will not increase human microplastic intake via take-out food.

SEM was adopted to observe the influence of hot water on the surfaces of the containers. The images show that surface changes occurred in PP and PE containers after hot water treatment, but no obvious changes of PS containers' surfaces are seen (Fig. 6). Although the surface changes of PP and PE containers did not increase the microplastic abundance in the corresponding containers (Fig. 5), it may cause

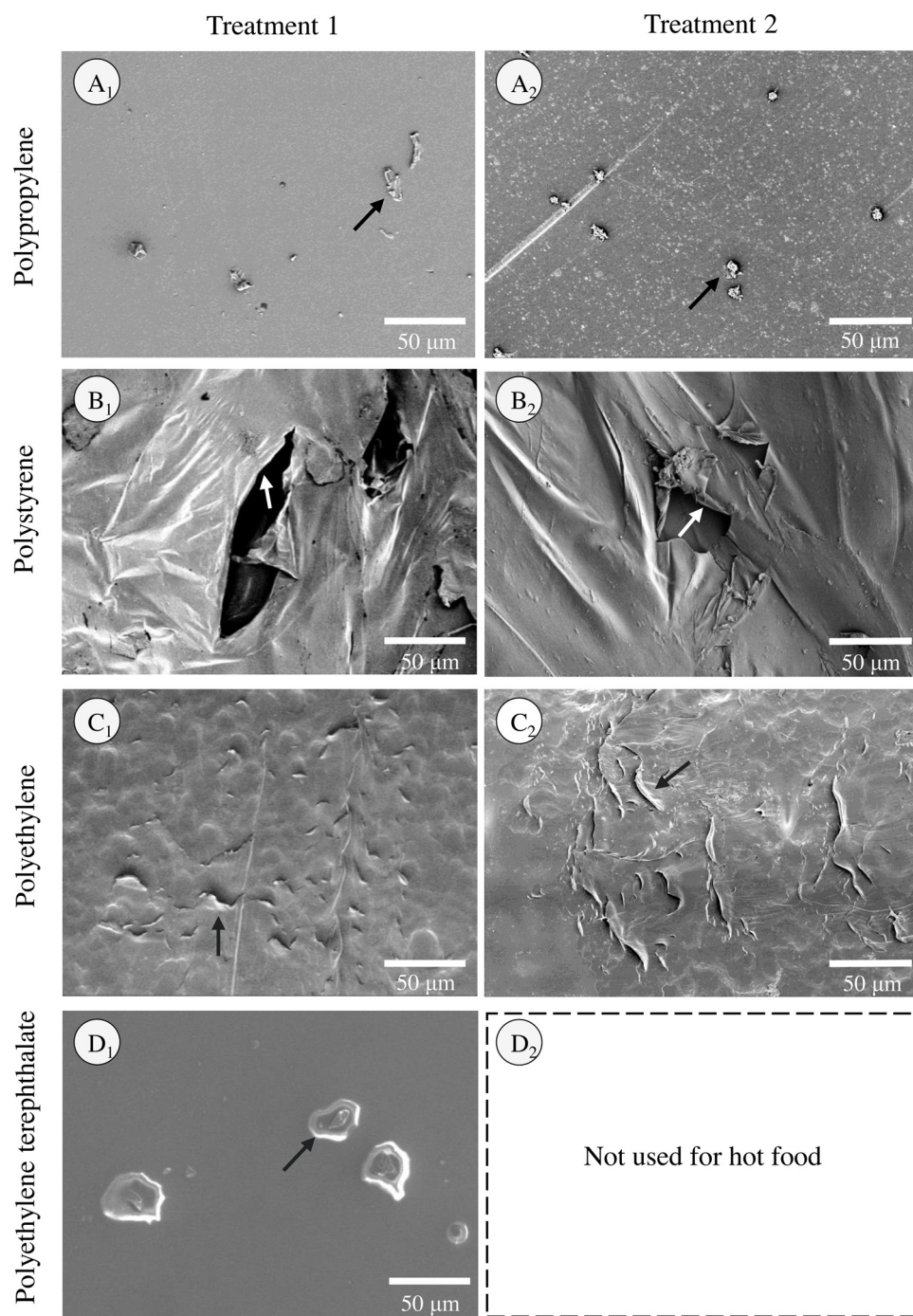


Fig. 6. SEM images of container surfaces after different treatments. Treatment 1: Direct flushing; Treatment 2: Flushing after immersion in hot water for 30 min. The black arrows in the photographs indicate the protrusions and broken places on the container surfaces.

some additives releasing from the plastic containers. Multiple types of additives are used in plastics to ensure their performance (Fries and Zarfl, 2012). Cao et al. (Cao and Corriveau, 2008) reported that migration of bisphenol A from polycarbonate bottles into water increased with the immersion time when being heated at 70 °C. Moreover, many studies found that fluorescent additives, bisphenol A, and endocrine disrupting chemicals could also be released from plastic products (Fikarová et al., 2019; Luo et al., 2019; Teuten et al., 2009; Chen et al., 2019). The additives from microplastics may affect organisms. It has been reported that leachates from plastic products could cause immobility to *Daphnia magna* and toxic effects to coral reef fish (Hamlin et al., 2015; Lithner et al., 2009).

In fact, many other factors are involved in take-out food

consumption. For instance, most food contain oil, especially the fried food. Besides, our pre-experiment showed that except for PP containers, the other three types of containers were not suitable for microwave or cryopreservation due to their poor tolerance of heat or refrigeration. However, not all plastic products are used according to the rules (especially some products are not marked with materials at all). Plastics that are not suitable for microwave heating, or for cryopreservation, may release microplastics in large quantities after these treatments and increase human exposure risk. Therefore, the influence of oil, improper use of plastic containers, and other factors on microplastic flaking should also be considered in future research.

4.3. Estimation of microplastic intake by humans

The take-out industry is growing rapidly, in terms of its economic value and market. According to the data of National Development and Reform Commission Research (NDRC (National Development and Reform Commission), 2019), the number of Chinese take-out customers reached 358 million people in 2018 (NDRC (National Development and Reform Commission), 2019); about 70 % of them are under the age of 30. Among them, 60 % are white-collar workers, with an average ordering frequency of 4–7 times per week.

Human intake of microplastics via take-out food containers was studied, and then calculated based on the average abundance of microplastics in take-out containers and the order frequency of high-risk people (white-collar workers). Accordingly, a person may ingest 12–203 microplastics per week. The highest value (203 items/week) was calculated based on the data of the PS containers. However, considering that the most widely used take-out containers are made of PP instead of PS, the range of 12–63 items/week calculated based on the data of the PP containers is likely to be closer to the reality. Note that only one container being calculated for each order and the detection limitations (20 μm) of the instruments may result in underestimate of human intake of microplastics.

As discussed earlier, atmospheric microplastics can cause container contamination during manufacturing process, which is also likely to happen during eating take-out food. A previous study reported that the potential for human ingestion of fibers resulting from atmospheric microplastic fallouts is higher than that via mussel consumption (Catarino et al., 2018). To compare human intake of microplastics via one take-out container and that via atmospheric deposition during eating take-out food, the atmospheric microplastic fallout data (523–3973 items/day/m²) of Dris et al. (Dris et al., 2017) was adopted. The mealtime was assumed to be 20 min. Based on the average open area of the containers (87 cm²) in our study, the number of microplastics from air deposition ranges from 0.1 to 0.5 items/container during a meal, which is much lower than that from one take-out container (11 items/container). Therefore, in the whole process of take-out food consumption, from ordering to eating, microplastics from containers contribute the most to human exposure.

Human exposure to microplastics via different pathways has been estimated in previous study, among which inhalation (25,575 items/capita/year) contributes to 80 % of the total particle intake. Other pathways include table salt (3000 items/capita/year), tap water (2784 items/capita/year), bivalves (2163 items/capita/year), and dust (1063 items/capita/year) (Kim et al., 2018; Dehghani et al., 2017). According to our results, 2977 microplastics may be ingested through take-out containers per person per year, which is similar to the intake via food. Moreover, besides take-out containers, plastic bags are also used for storing food. It has been reported that about 11.6 billion microplastics and 3.1 billion nanoplastics could be released after steeping one teabag at brewing temperature (95 °C) (Hernandez et al., 2019). In addition, a bottle cap made of HDPE was found to release microplastics as many as 1,225,500 particles after opened/closed 100 times (Winkler et al., 2019). Therefore, further in-depth studies on microplastics in food packages should be conducted. Meanwhile, it is important to reduce the use of plastic products, by using other materials as substitutes for food packages.

5. Conclusions

In the present study, we quantitatively investigated microplastics in take-out containers. The highest microplastic abundance was found in the containers made of PS. There were two major sources of microplastics in take-out containers, i.e., atmospheric fallout and particles flaking from the container inner surface. Loose structure and rough surface of PS containers are likely to result in more microplastics in containers. Based on the microplastic abundance of the take-out

containers and the take-out ordering frequency of white-collar workers, human microplastic intake ranges from 12 to 203 items per week, assuming 20 min mealtime and one take-out container. Further research is needed, focusing on microplastics from various food packages.

Declaration of Competing Interest

The authors declare that they have no competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

CRediT authorship contribution statement

Fangni Du: Conceptualization, Writing - original draft, Methodology, Formal analysis, Visualization. **Huiwen Cai:** Writing - review & editing, Methodology. **Qun Zhang:** Writing - review & editing, Formal analysis. **Qiqing Chen:** Writing - review & editing. **Huahong Shi:** Writing - review & editing, Supervision, Project administration.

Acknowledgements

This work was supported by grants from the National Key Research and Development (2016YFC1402204) and the Natural Science Foundation of China (41776123).

Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:<https://doi.org/10.1016/j.jhazmat.2020.122969>.

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