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Microplastics from consumer plastic food containers: Are we consuming it?

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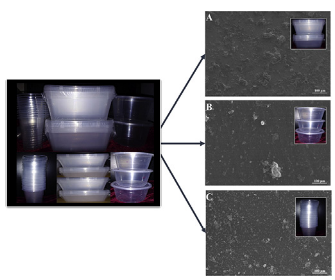
HIGHLIGHTS

- Microplastics in new plastic food containers were isolated and characterized.
- Majority of the microplastic particles isolated are in nano size.
- 188 tonnes of MPs are estimated to be released annually for human consumption.

GRAPHICAL ABSTRACT



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ABSTRACT

Microplastic (MP) accumulation in the environment has become an issue of human and environmental importance. Great efforts were made recently to identify the sources of MP exposure to humans and their release into the environment. Here, we employed spectroscopic techniques to identify and characterize MP in consumer plastic food containers that are, in huge quantity, used for food delivery and disposable plastic cups for daily drinking. We determined the average weight of isolated MP per pack to be 12 ± 5.12 mg, 38 ± 5.29 mg, and 3 ± 1.13 mg for the round-shaped, rectangular-shaped plastic container and disposable plastic cups, respectively, with various morphological features including cubic, spherical, rod-like as well as irregular shapes, which may either be consumed by humans or released into the environment. This study demonstrates that new plastic containers can be an important source of direct human and environmental exposure to microplastics. Most importantly, our results indicated that necessary attention must be given to morphological features of realistic MPs when evaluating their risks to humans and the environment.

1. Introduction

The knowledge of the impacts of microplastics (MPs) on human health and food safety is vital to its risk evaluation and a major panacea to this global menace. To achieve this, detection and identification of MPs source(s), quantification of the environmental concentrations, assessment of the exposure level, and route(s), as well as its possible effects, must be investigated. Recent studies

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suggest that plastic materials and plastic particles (micro/nano size), as well as their associated chemicals, can be detrimental to human well-being (Rist et al., 2018). Plastic particles intentionally produced for consumer care products or industrial uses are referred to as primary MPs while the degradation products of bulky plastics materials under natural processes in the environment are called secondary MPs. However, plastic particles unintentionally produced and released during the production of plastics materials/products are yet to receive adequate attention from researchers. This is of great concern as plastic products have become an integral part of our daily life. For instance, many food products are majorly transported in plastic packaging materials, which have been identified as a significant contributor to the proliferation of plastics in the environment (Gallego-Schmid et al., 2019; Geueke et al., 2018; Rodrigues et al., 2019). Moreso, plastics have found numerous uses in households, agriculture, medicine, pharmacy, engineering as well as research fields. Hence, there is a need to give more attention to MPs pollution in our daily lives.

Identification of sources of human exposure to MPs is crucial to understanding its risk. A number of studies have reported the routes by which MP particles are introduced into the environment, to include wastewater treatment plants (WWTP), effluents, and stormwater drains (Lattin et al., 2004; van Wezel et al., 2016). Other sources are the degradation of mismanaged plastic materials in the form of abrasion of plastic products and paints (Rist et al., 2018), abandoned fishing nets in oceans (Andrady, 2011), MP fibres from textile materials (Browne et al., 2011), and microbeads used in hygiene and personal care products (Wu et al., 2016). All these may constitute a significant contribution to human MP exposure directly or indirectly.

Of critical concern are the recent studies that demonstrated human exposure and presence of MPs in food products intended for human consumption such as table salts (Gündoğdu, 2018; Iñiguez et al., 2017; Karami et al., 2017; Kim et al., 2018; Yang et al., 2015), canned sardine (Karami et al., 2018), beer (Liebezeit and Liebezeit, 2014), sea fish (Li et al., 2015; Santillo et al., 2017; Van Cauwenberghe and Janssen, 2014), honey, sugar (Liebezeit and Liebezeit, 2013), teabags (Hernandez et al., 2019), mineral and drinking water (Cermakova et al., 2018; Eerkes-Medrano et al., 2018; Mintenig et al., 2019; Welle and Franz, 2018). These findings estimated between 37 to as high as billion plastic particles consumption from various food products per individual annually (Hernandez et al., 2019; Karami et al., 2018; Van Cauwenberghe and Janssen, 2014; Yang et al., 2015). MPs in seafood products, for instance, have been attributed to the massive pollution of the marine with plastic wastes. Moreover, MPs have been reported in human stools due to food in-take (Schwabl et al., 2019). Even though no scientific evidence has emerged to date on the direct or indirect hazard resulting from human exposure to plastics or MPs (Rist et al., 2018), the concern, however, is that of health vigilance on the possible disastrous effects.

Different studies have been initiated globally to understand the health implications of human exposure to MPs. However, most investigations to date have based their observations on synthetic microbeads, usually referred to as primary MPs, which are deliberately manufactured for industrial or domestic uses. While this line of thought and research is not out of place and indeed an initial approach to stimulate our understanding of these emerging pollutants. It is, however, important to pay equal attention to the unintentional sources of secondary MPs which formed the larger percentage of plastics particles in the environment. Moreso, it is well known that the physicochemical properties of nanomaterials play important role in their toxic nature and degree of lethality (Albanese et al., 2012). Primary MPs as presently been used in the toxicological studies are mostly of uniform sizes and shapes, while secondary MPs exist in various sizes and shapes therefore, it is difficult to evaluate its actual health risk.

The global food delivery market was valued at $89 billion in 2015, a subsequent rise of 2.7% is expected yearly to over $102 billion by 2020 (Gallego-Schmid et al., 2019). In emerging economies like China, the rapid development and industrial revolution have ushered in an era of food delivery industries, resulting in extensive usage of plastic containers for food packaging and delivery every single day. Plastics consumption in consumer products stands at 8.2 million tonnes in 2003 alone, with a 7% projected annual growth (Rosato, 2005). This is as a result of changing work lifestyles and social status, which may provide more channels for human contact with plastics products more often than in the past years.

Therefore, the aim of the present study is to determine if newly manufactured plastic food containers are a source of human exposure to unintentionally produced MPs. In addition, to evaluate and compare the physicochemical properties of the conventional experimental model of MPs to that of secondary MPs derived from plastics food containers. Hence, we carried the extraction of plastic particles from two commercial plastic containers and disposable plastic cups, usually used for food packaging and water consumption, purchased from local supermarkets in Beijing. Simple water extraction was carried out. Fourier transform infrared spectroscopy and scanning electron microscope were used for the characterization of the particles. We demonstrated that newly manufactured plastic materials in the market are a potential direct source of human exposure to MPs. Also, the experimental MPs are not morphologically accurate representations of secondary MP particles that humans encounter daily.

2. Methods

Materials. In order to investigate the presence of plastic particles (M/NP particles) in newly manufactured consumer plastic food containers, two types of commonly used plastic containers (round (CPC) and rectangular (RPC) molded) and a disposable plastic cup (DPC) were purchased. These containers are known to be used for quick food delivery and packaging, which is commonly referred to as “take away pack” (sealed in 10/5 pieces per pack) while the disposable cups contained 50 pieces in a pack. All were purchased from local supermarkets in Beijing. Three packs each were used for the experiment, and the information on plastic packs was collected from the package labels. The polymeric composition of the materials is polypropylene (PP) inscribed on each of the containers. The weight of each of the plastic containers with their lids were determined. Precautionary measures were taken to prevent any particle contamination from indoor air, and this was achieved by carrying out the experimental procedure inside the laminar flow chamber (KLCZ-1220A, Yataikelong, Beijing, China).

Extraction of plastic particles. The purchased food containers were unsealed inside the hood to prevent any contamination from ambient air particles during the extraction process. Each container was quickly covered with the lids to prevent any form of external contamination. 10 ml of ultrapure water was dispensed into each of the plastic containers. The containers were shaken for 2–3 min using a mechanical shaker in order to wash the inner of the container effectively. The procedure was repeated twice. Thereafter, the extracted solutions were vacuum dried using Christ freeze drier (Alpha 1–4 LDplus, An der UnterenSose 50, Germany) to remove the water leaving the extracted particles in the vail. The weight of the extracted particles was determined. Two controls were set up, which are: ultrapure water used for the extraction of the microplastics (SC1), and ultrapure water left open in the laboratory experimental hood through the period of extraction (SC2).

Characterization of particles. The extracted plastic particles were analyzed using Fourier-transformed infrared (FT-IR) spectrometry (FT/IR-6100, JASCO Corporation, Tokyo, Japan) to identify the functional groups. The sample spectra were recorded as 64 scans in the spectral range of 500–4000 cm^{-1} at a resolution 4 cm^{-1} and were compared to the polymer library (KnowItAll, Bio-Rad) to verify the polymer type. Approximately 1 mg ml^{-1} concentration of the obtained particles was prepared and 1 μl was placed on a glass disk, fixed onto the SEM aluminum sample disk. The samples were characterized using the scanning electron microscopy (SU 8020, HITACHI, Tokyo, Japan) to obtain the surface morphology of the particles. No plastic particle was observed in any of the controls set up for the experiment, when observed under the microscope, except for fibres which have not similar features with our samples.

Particle size measurement and image analysis. Twenty different images each of the samples were obtained and a minimum of five hundred particles were measured using ImageJ software (NIH, USA) to obtain different particle sizes distribution that were observed. OriginPro 8 software (Northampton, MA, USA) was used for the plot.

3. Results and discussion

Isolation and Characterization. It has been estimated that about 40% of manufactured plastics globally (PlasticsEurope, 2019) is produced for single-use and majorly food packaging, after which a significant percentage ends up in the environment. The degradation of this mismanaged plastics in the environment due to natural conditions such as mechanical, UV depolymerization, biodegradation, thermal-oxidation, and moisture absorption/hydrolysis (Mattsson et al., 2015), results in secondary MPs. However, during plastics production, plastic debris areleft on the surface of plastics products which eventually get to the environment. To the best of our knowledge, no attention has been given to this crucial source of direct human exposure to MPs.

Here, we isolated, characterized and quantified the presence of MPs in three differently shaped plastic containers purchased from local supermarkets, manufactured to convey foods and water for human consumption. Fig. 1 shows the FT-IR analysis of the MPs extracted from the three containers under study. The spectra were compared with the Bio-Rad library database and were found to be

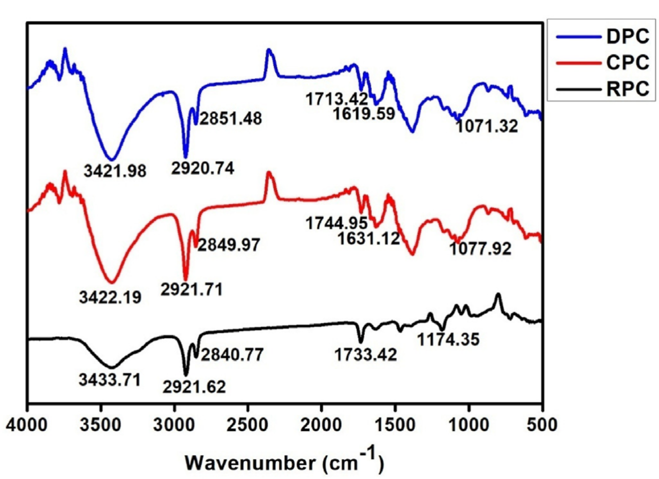

Fig. 1. The FT-IR spectrum of extracted plastic particles from CPC (red), RPC (black) and DPC (blue) showing their characteristic peaks. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

most consistent with a typical polypropylene spectrum, which was also in line with the plastics type inscription on the containers by the manufacturers.

From the spectra, we identified some characteristic peaks which are peculiar to PP for all the sampled MPs at 1077, 1174, and 1071 cm^{-1}, which correspond to isotactic polypropylene band. The absorption band of the carbonyl group in the PP spectrum is broad, and this may denote the carbonyl group presence in various products of oxidation during processing and manufacturing. The absorption band within the wavenumber range of 1631–1744, 1733, and 1619-1713 cm^{-1} corresponds to carbonyl absorption typical of (>C = O) and (-CCH_3 –CH_2 –CO- CH_3) (Urbaniak-Domagala, 2012). The peaks observed at 2849–2921, 2840–2921, and 2851-2920 cm^{-1} denote the aromatic C–H asymmetric and symmetric tension for ethylene sequence polymeric materials while the peaks at 3422, 3433, and 3421 cm^{-1} correspond to the stretching vibration frequency of hydroxyl group. However, the observed variation in absorption peaks of the sampled containers can be attributed to the changes in the layer structure which may result from possible variations in product chemical composition, electrochemical factors, UV radiation or low temperature (cooling) on the polymeric materials (Urbaniak-Domagala, 2012).

The SEM was employed to obtain the morphology of the plastic particles before and after its extraction from the surface of the containers. Fig. 2A, C, and 2E show the plastic particles on the surfaces of each container in varied abundance, which are easily detached through washing (Fig. 2B, D and F). Various sizes and shapes were observed in all the three types of containers and the particle distribution patterns were similar in all the containers. Fig. 3A, C and E (high magnification) and Fig. 3B, D and F (low magnification) represent the different types of MP sizes and shapes obtained from all the plastic containers under study. Particle sizes range between 0 and 210 nm, with a larger percentage of the particles below 50 nm (3G, H and I). The particles shapes that were observed include cubic, spherical, rod-like as well as irregular shapes (Fig. 3A, C and E red arrows).

The knowledge of M/NPs toxicity to human health and food safety can be significantly improved through the understanding of the veracious identity of the particles in the environment. Most studies conducted to date used model MPs, which are mainly spherical and with a specific uniform size range. In contrast, different reports have reiterated the importance of size and shape in the toxicological study of nanoparticles (Albanese et al., 2012; Chithrani et al., 2006; Gratton et al., 2008). For instance, Gratton et al. demonstrated that rod-like particles show the highest uptake in HeLa cells, followed by spheres, cylinders, and cubes (Gratton et al., 2008). In the same view, recent studies have as well demonstrated the implications of size and shape of MPs, using aquatic animals. Smaller plastic particles exhibit more toxic effects compared to the bigger ones (Frydkjær et al., 2017; Jaikumar et al., 2019; Jemec et al., 2016; Ma et al., 2016; Ziajahromi et al., 2017). Chen et al. used two different sizes of spherical plastic particles with 50 nm and 45 μm size to investigate the direct and indirect toxic effects of plastic particles toward zebrafish (Danio rerio) larvae locomotor activity. The 50 nm size was found to inhibit zebrafish larvae locomotion while no inhibition was observed in the 45 μm treated group (Chen et al., 2017). Our result showed an average diameter of 50 nm in the plastic debris in food containers, which may induce stronger toxicity of the plastic particles derived for food containers. Likewise, irregularly shaped particles are retained longer in the body of an aquatic animal than uniformly shaped particles (Frydkjær et al., 2017). These observations suggest that the conventional experimental MPs do not seem to represent the larger proportion of possible realistic MPs in human exposure in terms of its morphological features, as observed in the

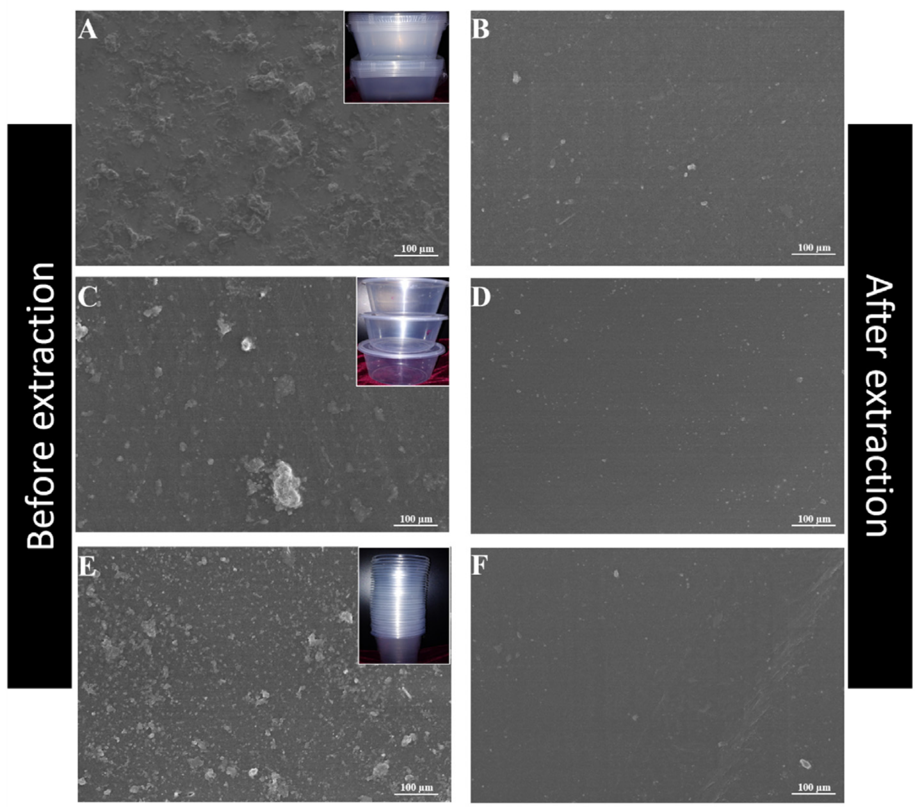

Fig. 2. SEM images (A, C, E) of plastic particles on the surface of CPC, RPC andDPC, respectively, before extraction and (B, D, F) after extraction. Insets: stacked individual plastic containers.

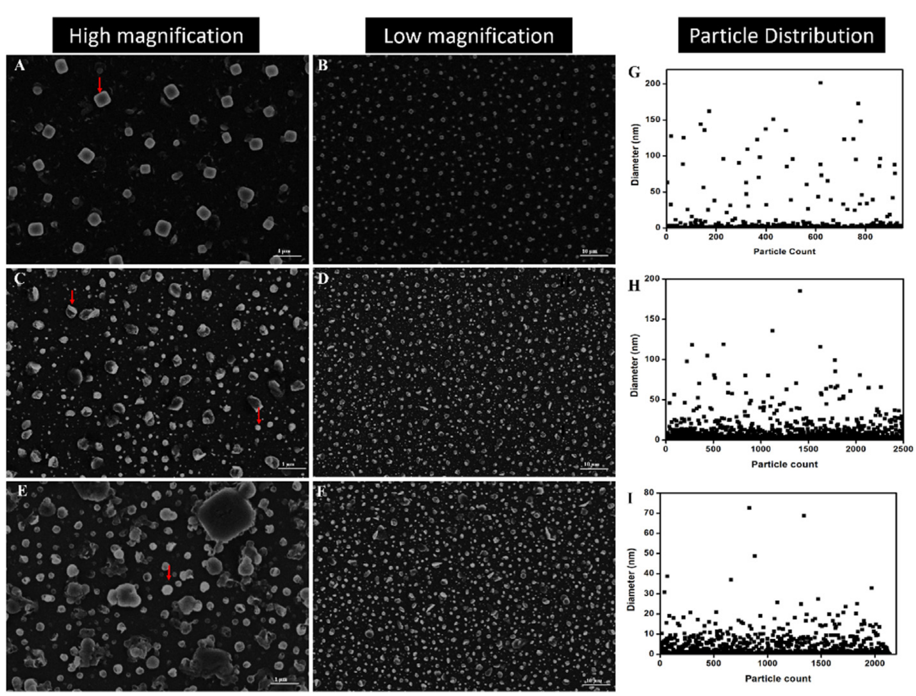

Fig. 3. SEM images (A, C, and E) and (B, D, and F) represent different types of plastic particles sizes and shapes observed in all the three sampled containers both at high and low magnification. (G, H, and I) represent the particle distribution observed across different shapes, computed using ImageJ software, and plotted on OriginPro 8 software. Red arrows indicate some of the shapes observe from all the plastic containers. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

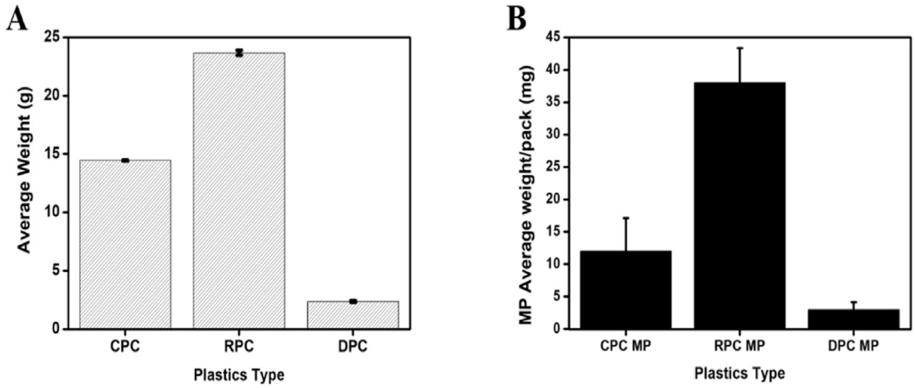

Fig. 4. **(A)** illustrates the average weight (g) for each plastic container; CPC-round shaped, RPC -rectangular shaped, and DPC- disposable plastics cups. **(B)** represents the average weight (mg) of microplastics obtained from each pack of the containers in a similar order. N = 3packs.

unintentionally released secondary MPs from food containers. As a result, there is a high possibility that the present studies may underestimate the impacts of MPs therefore, the resulting data may not be accurate enough to predict the risk associated with MPs exposure in humans as well as the environment.

Quantification. we quantified the extracted MPs from consumer plastic containers. Fig. 4 shows the average weight of each plastic container and the average weight of plastic particles extracted from each container pack. The average weights of the round (CPC) and rectangular (RPC) shaped containers, as well as the plastic cups (DPC), are 14.46 ± 0.04 g, 23.67 ± 0.21 g, and 2.38 ± 0.08 g, respectively (Fig. 4A). The weights of the MP isolated from each container pack (of 10, 5, and 50 pieces) are 12 ± 5.12 mg, 38 ± 5.29 mg, and 3 ± 1.13 mg in the same order (Fig. 4B).

Identification of human direct exposure route to MPs in the environment is imperative to the body of knowledge required in MPs risk assessment. Our results supported the reports of various studies that have observed the presence of MPs in food products (Karami et al., 2018), human stool (Schwabl et al., 2019), cat and dog foods and feces (Zhang et al., 2019) as well as indoor and outdoor air (Liu et al., 2019). For instance, Schymanski et al., in their report on the presence of plastic particles in bottled water, recorded 14 particles/L in single-use plastic bottles while 118 particles/L were observed for reusable plastic bottles (Schymanski et al., 2018). Recently, Hernandez et al. reported billions of MP particles in teabags, which are consumed by humans. Furthermore, Hwang et al. in their study using human cells (HDFs, PBMCs, Raw 264.7) observed that PP particles stimulated the immune system and enhanced potential hypersensitivity to PP particles via an increase in the levels of cytokines and histamines at 1000 μg ml^{-1} concentration (Hwang et al., 2019). From our results, approximate average weights of 1.2 mg, 7.6 mg and 0.06 mg of plastic particles for a single CPC, RPC, and DPC, respectively were obtained. Hence, with the accumulation of MPs from these containers to a certain degree, it is sufficient to induce allergy if possibly consumed whenever a new container is used without adequate washing. Nevertheless, even if washed, these particles still go into the environment, wherein their fate and transport in the environment are yet to be ascertained. Furthermore, for every single ton of these plastic food containers produced, an approximate quantity of 75.3, 291.3 and 22.9 g of MPs are released either for human consumption or into the environment respectively, from each container under study (see SI). By implication, taking the least contaminated DPC for instance, an annual production of plastic food containers of about 8.2 M tones will release an average of 187,780,000 g (approx. 188 tonnes) MPs for human consumption or discharged into the environment, mostly of which are in nano size.

In addition, a recent review estimated an annual MPs consumption ranges from 39000 to 52000 particles in approximately 15% of Americans' caloric intake, depending on age and sex. An increase in exposure rate of 10.8–57% when inhalation of MP polluted air was considered and an additional 4000–90000 particles depending on individual water intake (Cox et al., 2019). All these points to the fact that humans are exposed to MPs more than we ever thought and their impacts on human health are yet to be unrevealed.

4. Conclusion

This work investigated the presence and quantity of M/NPs in new plastics products for food packaging, fast food delivery, and water consumption. Spectroscopic analysis showed that the plastic debris were from the plastic materials which were unintentionally produced during manufacturing. SEM scanning revealed various shapes of particles that may indicate the model MPs presently used in data collection for MP impacts does not reflect the real MPs. Also, this study shows that M/NPs are readily dislocated upon simple water wash, releasing particles with size mostly less than 50 nm. Our data provides another evidence that human might be more exposed to MPs than it is known as of yet.

Declaration of competing interest

The authors declare no competing interests.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2020.126787.

Author contributions

Oluniyi O. Fadare: Conceptualization, Methodology, Investigation, Writing- Original draft **Wan Bin:** Conceptualization and Supervision, Writing-Reviewing and Editing **Lixia**

Zhao and **LiangHong Guo:** Resources and Fund Acquisition.

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