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# Pouring hot water through drip bags releases thousands of microplastics into coffee

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#### ABSTRACT

Microplastics (MPs) released from food packaging have attracted widespread attention. In this study, drip bags made from polyethylene (PE), polypropylene (PP), polyester (PET), and rayon selected from eight brands were employed to investigate MPs releasing. Fourier-transform infrared microspectroscopy (µ-FTIR), optical microscope and scanning electron microscope (SEM) were used to study the effects of brewing time and temperature on the release of MPs. The results showed that a single plastic coffee bag steeped at 95 °C for 5 min could release more than 10,000 MPs particles into a cup of coffee. Irregular blocks, long strips, and size range of 10-500 µm MPs were easier to be released, implying that consuming 3-4 cups of coffee will lead to an intake of 50 thousand MPs particles daily. Rayon was the primary type of released MPs, accounting for over 80% of the total amount of the released MPs. Our results are hoped to provide evaluation standards of material selection for processing coffee bags.

## 1. Introduction

Plastics have a broad application in the industrialized world for many advantages, including their convenience, low cost, anti-corrosion, insulation, and stable physio-chemical properties (Gao et al., 2021). According to the latest report, the utilization of plastics increased by 2.5% in 2019 compared with that in 2018 (Plastics Europe, 2020). However, only 10% of plastics are recycled or incinerated, with the rest being released into the environment or landfills (He et al., 2021). These wasted plastics are mostly converted into tiny particles through various external factors, such as mechanical abrasion, weathering, ultraviolet (UV) radiation, and biological metabolism (Liu et al., 2021). Among these, small particles with a size of 5 mm were defined as microplastics (MPs) (Thompson et al., 2004).

As widely used food packaging materials, plastics are extensively adopted in the food industry, which causes an increasing food safety concern (Bouwmeester et al., 2015). Kedzierski et al. (2020) found that the contamination level of extruded polystyrene MPs (MP-XPS) in packaged foods ranged from 4.0 to 18.7 MP-XPS/kg. The MPs content in eggs even reached 11.67  $\pm$  3.98 particles/egg (Liu et al., 2022). Mason et al. (2018) investigated the distribution of MPs in 27 different bottled water from 11 different brands in 9 other countries and found that 93% of the individual bottles (including all the brands and batches) were contaminated by MPs, 13% of which were microfibers particles. Surprisingly, bottled water was detected to have a two-fold amount of MPs particles compared with tap water on average. Diaz-Basantes et al. (2020) detected MPs with various sizes and quantities in honey, milk, soft drinks, and beer from Ecuador. The researchers reported that the number of MPs distributed was 20-80  $mL^{-1}$  for beers, 10  $mL^{-1}$  for bottled mineral water, and 200–500  $g^{-1}$  for teas (Li et al., 2022). Nowadays, it is convenient for white-collar workers to order take-out. However, the take-out containers have the risk of containing some released MPs. It was previously reported that MPs were found in all take-out containers, ranging from 3 to 29 pieces/container (Du et al., 2020). The weight of MPs released by take-out containers varies by shape (round-shaped:  $12 \pm 5.12$  mg; rectangular-shaped:  $38 \pm 5.29$  mg; disposable plastic cups:  $3 \pm 1.13$  mg) (Fadare et al., 2020). Notably, MPs could also be released from feeding bottles and silicone-rubber baby teats (Li et al., 2020; Su et al., 2021). MPs residues in foods may affect human health. Research revealed that infants within 12 months old might have an average consumption of 1.6 million polypropylene plastic particles per day (Li et al., 2020; Su et al., 2021). Zhang et al. reported the presence of PET and polycarbonate (PC) MPs in the feces of the newborn. Moreover, the concentration of PE in infant feces was significantly higher than that of adults (Zhang et al., 2021). Kannan and Vimalkumar (2021) found that MP exposure in laboratory animals was

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linked to various forms of disorders, including inflammation, immunological response, endocrine disruption, alteration of lipid and energy metabolism, etc.

Coffee and tea belong to the "three major beverages" in the world. Their consumption seems to be safe within usual intake levels. Because of the popularity of these two beverages, the health risk posed by their plastic packaging (e.g., non-woven) cannot be ignored. Non-woven materials not only provide the required gas/aroma barrier to maintain tea/coffee properties throughout the shelflife, but also can withstand certain brewing temperatures while allowing the coffee to pass through (Buntinx et al., 2014; Kellie, 2016). Teabags can release a large number of MPs during high-temperature immersion. A single plastic teabag could release approximately 11.6 billion microplastics and 3.1 billion nano plastics at 95 °C of brewing temperature (Hernandez et al., 2019). Coffee consumers have reached 350 million in China (https://www. coffinance.com/detail/4550). People around the world drink 1.6 billion cups of coffee every day. Considering the great demand for coffee, various kinds of coffee have sprung up, especially drip bag coffee, a convenient and popular beverage product among young people. However, the release of MPs from the coffee bags and the influence of brewing temperature and time are often overlooked.

This research aimed to explore whether MPs can be released from drip coffee bags into the water during brewing a cup of coffee. Drip coffee bags were steeped in 95 °C hot MilliQ water for different periods, and the tested particles were measured using microscopy techniques. The composition and morphology characterization of MPs particles were analyzed by  $\mu$ -FTIR and SEM. Moreover, the leaching components of the drip bag coffee steeped at different times were classified. The result of this research may provide evidence for food package or environmental risk assessment.

#### 2. Materials and methods

#### 2.1. Materials and chemicals

To examine the MPs abundance in different drip coffee bags, the drip bag coffee from eight popular commercial brands was purchased from JD.com on March 17-18th, 2021. These coffee bags were named A, B, C, D, E, F, G, and H (Table S1). Each experiment contained three parallel drip coffee bags. The main description on the label is the nutritional composition of the coffee, while the compositions of drip bags were rarely described. The standards of polyethylene (PE, 96%), polypropylene (PP, 97%), polyester (PET, 95%), and rayon (95%) were obtained from HuaChuang plastic and chemistry company (Dongguan, China). Ethanol (AR) was purchased from BoNuo company (Dalian, China). All chemicals were used without further purification.

## 2.2. Sample pretreatment

The drip bag coffee was ripped along the crease, and the coffee powder was removed. The plastic outer package was emptied to ensure that the detected MPs particles were released from the coffee bags without other interfering factors. The empty coffee bags (referred to as A-H) were thoroughly washed three times with ultrapure (MilliQ) water at room temperature to remove the residual coffee powder or other plastic debris. All samples were then dried with a suitable flow rate of nitrogen. Coffee bags (A-H) were put in the experimental coffee cup and brewed with hot MilliQ water (95 °C; pH  $\sim$  6.9, ultrapure water). According to the most typical coffee brew time (Maille et al., 2021), three different steeping periods, including 1 min, 3 min, and 5 min, were used to treat eight drip bags, respectively. The hot water was filtered through a metal filter membrane (Stainless steel, Beijing JiuDing, China) with a pore size of 10 µm. Subsequently, the membrane was put into a beaker containing 15 mL of absolute ethanol. As the density of ethanol is smaller than MPs and it cannot dissolve MPs, ethanol (GR) just acted as a solvent for dispersing and settling MPs without any effect on the

physical and chemical properties of MPs (Li et al., 2018) (Fig.S3). After ultrasonic treatment (60 w) for 1 h, the released MPs can be dispersed well in the ethanol solution. Then, ethanol extract was added into a 15 mL conical glass tube and stood for half an hour. Heated nitrogen was used to concentrate the ethanol to 200  $\mu$ L, which was then transferred into a 1 mL glass bottle and stored at 4 °C. Triplicate samples were prepared for all experiments.

### 2.3. Mps identification and characterization

Within the spectral range of 4000 to 750  $\text{cm}^{-1}$  in total reflection mode, all MPs were confirmed with a PerkinElmer Spectrum Spotlight 400 micro-Fourier transform infrared spectroscope (µ-FTIR; PerkinElmer Inc., USA) under the average of 15 scans for each spectrum. It is equipped with a liquid nitrogen-cooled mercury cadmium telluride (MCT) array detector attachment consisting of a germanium (Ge) crystal. The resolution and pixel size were set as 8  $cm^{-1}$  and 25  $\mu$ m, respectively. Four standards of plastic were scanned with the image mode of µ-FTIR, and the generated standard infrared spectra were imported into the software spectrum to establish a standard library for subsequent sample identification. The total absorbance infrared images of MPs extracted from different brands of drip coffee bags were then imported into Spectrum IMAGE and compared with standard infrared spectra to export corrected images for PE, PET, PP, and rayon. The detected match rates for PE, PET, PP, and rayon were obtained, respectively. When interpreting FTIR output, only readings with a match rate greater than 70% were considered to have a reliable spectral match (after visual inspection). Finally, the detected target plastic components were further counted with the ImageJ software (version 1.48v, National Institutes of Health, USA). The obtained total absorbance infrared image of the MPs was the indistinguishable components of the mixed MPs. By combining the total absorbance infrared with each standard infrared spectrum of MPs in the self-built database, each component image of MPs could be obtained. The particles with high response values were selected and counted with ImageJ to calculate the amount of each MP component.

## 2.4. Estimation of MPs exposure

The ethanol concentrate of MPs (200  $\mu$ L) was dispersed by the ultrasonic cleaner for 5 min until the particles in the bottle were dispersed evenly in the solution. Then 10  $\mu$ L solution from each group was sampled with a glass pipette three times, then placed on a coverslip waiting for evaporation. The types of plastics were identified by the  $\mu$ -FTIR method, and the number of corresponding MPs components was counted by Image J software. Each sample was measured three times, and the average was taken as the final result. Data were multiplied by 20 (corresponding multiple) to obtain the final result. The number of total MPs was obtained by summing up the amounts of the identified MPs.

## 2.5. Microstructure of MPs

A scanning electron microscope (SEM) (JSM-7800F, JEOL, Japan) was used to observe the fiber surface morphology adhered to the drip coffee bag before steeping, along with the size and morphology of the MPs released from the drip coffee bag after steeping at different times periods. Briefly, 10  $\mu$ L of water was drop-casted onto a coverslip and viewed under microscopy (Nikon Ti-S, Tokyo, Japan) and SEM. SEM observation provides information about the morphological characteristics of the MPs and IR spectra reflect the specific composition of the MPs. The combination of these two methods well determined the particular composition of the MPs and improved the accuracy of the experimental results. Subsequently, each component of MPs was particularly identified by  $\mu$ -FTIR, and its characteristics were observed. Briefly, each MP component was picked up with tiny tweezers and placed on a unique small glass slide, which was then measured by SEM. Triplicate sets of

experiments with parallel repetitions were carried out.

#### 2.6. Quality assurance and quality control

Nitrile gloves and cotton lab coats were used throughout the experiment to exclude the influence of exogenous pollution. All test tubes and beakers used in the experiments were glassware and washed with ultrapure water. A blank control group was also set to reduce the possible interference from the reagents used in the investigation and the air environment in the laboratory. All solutions (including MilliQ water) were filtered through glass fiber filters with a pore size of 1.6 µm before use. During the experiment, plastic and air exposure were limited to the greatest extent. In the experiment, glassware was used throughout to avoid any potential plastic contamination from the atmosphere. In addition, the pipette tips were also cleaned three times with ultrapure water and ethanol (GR) to ensure their cleanness. Before the experiment, the test-bed was also cleaned. The samples were covered by a clean glass beaker to prevent MPs pollution from the air. These measures significantly reduced the external interference factors of MPs and ensured the accuracy of the experimental results.

#### 2.7. Statistical analysis

The particle size and counts were established by using ImageJ software (version 1.48v, National Institutes of Health, USA) (Schneider et al., 2012). Data were represented as mean  $\pm$  standard deviation (SD) of three individual experiments. The figures were plotted and imported by Origin 2021pro (Originlab Corporation, USA) and Spectrum IMAGE. Principal component analysis (PCA), heat map analysis, and hierarchical cluster analysis (HCA) were conducted by MetaboAnalyst (Chong et al., 2018). P < 0.05 was considered statistically significant.

#### 3. Results and discussion

#### 3.1. Abundance of MPs in drip coffee bags

The total amounts of MPs in drip coffee bags from different commercial brands after steeping at different periods are shown in Fig. 1 and Table S2. As shown, the MPs abundance of eight brands of drip coffee bags increased in a time-dependent manner. After steeping for 1 min and 3 min, MPs abundance of brand C drip coffee bag was the highest compared with the other brands (5407  $\pm$  921 items and 7607  $\pm$  722 items, respectively). However, the brand B drip coffee bag released the highest MPs (12807  $\pm$  1139 items) when all brands of drip coffee bags were steeped for 5 min. In contrast, brand E and G drip coffee bags showed the lowest MPs release in the three periods. Overall, the total amount of MPs increased with the steeping time.

It has been reported that these polymer materials (e.g. PET) were degraded at 95 °C or even high temperatures, due to the destruction of their molecular structures (Holland and Hay, 2000). Hydrolysis of the polymer might be the cause of this degradation. The cleavage of chemical functional groups reacting with water was a hydrolytic degradation. As a reduction in the molecular weight of polymer macromolecules, chain scission could result in structure loosening. The abundance of MPs in drip coffee bags might vary with material properties (Du et al., 2020). Some drip bags were made from staple fibers, which might contain MPs, causing the release of MPs from drip bags.

As shown in Fig. S1, the components of MPs were detected from the bags before steeping. It means that under slight mechanical force, the MPs particles might easily be peeled off into the water and adhere to the coffee powder. However, the drip bag itself was equivalent to a small filter device, which could keep some non-edible substances (including MPs) in the drip bag and prevent them from entering the water. Therefore, we only considered the MPs generated on the outer surface of the drip bag under high-temperature immersion in water. In our daily life, the friction between plastic tableware and the outer surface of the drip coffee bags is consistent with the flushing of water used in our experiments, so the abundance of MPs in real life can also be considered to match with the above results. Non-disposable tableware may produce fewer "flaking MPs" as they are usually made of non-plastic materials including glass, ceramic, or stainless steel (Du et al., 2020). Moreover, the other parts of the MPs, such as atmospheric MPs pollution, should also be considered. Atmospheric MPs pollution is a global environmental problem. It is possible that the air MPs pollution was not only presented during the production process of drip coffee bags but also detected in some food packages. The fact is that some food packaging itself produces



Fig. 1. Total MPs in drip coffee bags of different commercial brands after different steeping time. Different letters indicate significant difference (P < 0.05).

MPs, as well as the other MPs pollution in the atmosphere during production and processing (Cai et al., 2017; Dris et al., 2017). In other words, the sources of MPs in coffee bags include the flaking of the coffee bag itself and atmospheric pollution.

During the production process of food packaging, although only a small amount of MPs can be mixed with the food, food packaging can release more MPs under high-temperature conditions. Both tea and coffee require high-temperature water to brew, which provides viable conditions for MPs release. Other beverage packages (such as cola or juice) release a limited amount of MPs under normal temperatures.

#### 3.2. Size and area distribution of MPs in drip coffee bags

Compared with the characteristics of MPs in the drip coffee bags, boxplot, and Gaussian distribution curve were used to describe the size, shape, and distribution of MPs. The size and area maximum, minimum, average value, and distribution of MPs in all commercial brands (A-H) are presented in Fig. 2a-f, respectively. The amount of MPs with the size of 10–500  $\mu$ m from the coffee bags (brands B and D) exceeded 5000 in each steeping stage. MPs with a size below 20  $\mu$ m accounted for more than 50% of the total. The content of < 20  $\mu$ m MPs in brand B drip coffee bag during steeping 5 min was the highest compared to the other seven brands. Similarly, for brand B and D, the amount of MPs with the main area (<2000  $\mu$ m<sup>2</sup>) was the highest. The size and area distributions are consistent with the Gaussian distribution, which also agrees with the report of Li et al. (Li et al., 2021). When MPs are exposed to humans and other organisms, a potential ecological risk in response to human and nutrient levels should be concerned (Hernandez et al., 2019).

MPs with a size of greater than 10  $\mu$ m could be accumulated in the digestive tract of animals, inducing oxidative stress, damaging intestinal, liver and gill tissues, and increasing the heart rate and swimming speed of animals. The percentage of animal mortality ranged from 6.7 to 21.6 caused by MPs with particle size greater than 10  $\mu$ m (Visalli et al., 2021). Therefore, most current research focused on the MPs (particle size >10  $\mu$ m) (Sun et al., 2021; Wang et al., 2022). More than 80% of MPs particles detected in the steeped drip coffee bag were smaller than 80  $\mu$ m, which poses a potential safety risk and should deserve the public's attention.

With further differentiation and degradation of MPs particles, the size of MPs will be further reduced to  $<10 \ \mu$ m. This study focused on MPs particles larger than 10  $\mu$ m, on the one hand based on the toxicity of these MPs (greater than10  $\mu$ m) and the other hand the accuracy of the detection instrument. In recent years, many researchers have explored the toxicity of these MPs ( $<10 \ \mu$ m) (Liang et al., 2021; Sun et al., 2021). These studies indicated that MPs ( $<10 \ \mu$ m) damaged the intestinal barrier and induced mild immune response in the colon. That is the limitation of our study which not focused on MPs < 10  $\mu$ m. However, MPs ( $<10 \ \mu$ m) can't be ignored and even more important in terms of toxicity. Therefore, we need to study and make a breakthrough in other toxicity problems of MPs with different sizes in the future.

## 3.3. Identification of MPs polymer types

The infrared imaging system is a microscopic analysis technology developed on the basis of FTIR microscope and step-and-scan interference technology, which can scan the mixed sample quickly and obtain a



Fig. 2. Frequency distribution of MPs released from different brands drip bags coffee during the different steeping time. (a-c) the size distribution and (d-f) the area distribution of MPs in all commercial brands (A-H), respectively.

large area infrared images containing spectral information (Oh and Koenig, 1998). As shown in Fig. 3, the total absorbance infrared images (Fig. 3a) of the MPs extraction solution of different drip coffee bags and the standard infrared spectrum of PE, PET, PP and rayon (Fig. 3b) were obtained. The detected similarity rate of PE, PET, PP and rayon was 91%, 86%, 93%, and 85%, respectively (Fig. 3c-f). In the component images, the correlation coefficient increased with the gradation of red. In Fig. 3, it can be seen that the color transition from purple-black to redpink suggested a gradual increase in the correlation coefficient. The larger the correlation coefficient, the higher the precision of the detected component. The results confirmed that the drip coffee bags were a blend of PE, PP, PET, and rayon, or a blend of PET and rayon.

Ding et al. (2019) and Lin et al. (2021) detected the components and characteristics of MPs in bivalves, fish, and coastal sediments of the East

China Sea using  $\mu$ -FTIR equipped with attenuated total reflection ( $\mu$ -ATR-FTIR), respectively. The transmission was also selected mode when using  $\mu$ -ATR-FTIR. Therefore, Fourier-transformed infrared coupled with microscopes ( $\mu$ -FTIR) is considered a convenient, fast, and non-destructive technology for MPs identification. It can not only provide accurate spectral maps with defined and characteristic peaks of each polymer but also exhibit the shape of MPs particles. However, when the amount of MPs increases, the detection of MPs components becomes extremely cumbersome and time-consuming, which reduces the accuracy of analyzing MPs distribution. Fast and large-area molecular level scanning can be performed and combined with the plastic standard infrared spectrum, which can analyze the composition of MPs expediently and accurately (Fig. 3).



Fig. 3. The detected method of MPs in drip bags coffee. (a) The total absorbance infrared image of the total MPs; (b) Standard infrared spectrum of PE, PET, PP and rayon; (c-f) Detected specific components of MPs in the sample and corresponding similarity, respectively.

#### 3.4. Percentage of MPs component

The thickness of the lines indicated the amounts of MPs (Fig. 4). The number of MPs detected from each treatment for each product is shown from the left column to the middle column. The number of MPs in different types of treatment and for each product is shown from the center column to the right column. The changes in the proportions of MPs types from various groups are shown in the Sankey diagram (Fig. 4a). The percentage of 4 different polymer types was detected in eight commercial brands drip coffee bags (Fig. 4b-d). The main components of brand G and H drip coffee bags were PET and rayon. Brand A-F coffee bags contained all types of polymer. The percentage of each plastic polymer varied in brands of coffee bags. Regardless of brand or

steeping time, the proportion of rayon was the highest among the tested samples. In all brands, the rayon released accounted for 97.38%, 95.73%, and 89.64% (brand H) after steeping for 1 min, 3 min, 5 min, respectively, followed by PE (26.17%, 25.41%, 18.91%, brand D), PP (15.63%, 13.66%, 11.87, brand D), PET(14.14%, 10.33%, 12.54%, brands G) after steeping different periods (Fig. 4b-d). However, PE and PP were not found in brand G and H.

Rayon is an artificial textile material with many advantages (Kauffman, 1993), which is often mixed with other fabrics (such as polyester and cotton) and applied in food packaging. It has been reported that rayon is defined as a primary component of MPs in food and the environment (Bessa et al., 2018; Lusher et al., 2015; Mohsen et al., 2022). According to previous research, rayon was mainly concentrated in



Fig. 4. The linked network among types of MPs and variation of the relative percentages (%) of four MPs components in 8 commercial brands drip coffee bag after different steeping time periods. (a) Sankey diagram; (b) 1 min; (c) 3 min; (d) 5 min.

aquatic products. Lusher et al. (2015) also reported that 53% of the polymers found in the stomachs and intestines of deep-diving *Mesoplodon mirus* on the northern and western coasts of Ireland were rayon. Rayon was also found in the deep sea (Woodall et al. 2014). Recently, rayon was mainly detected in plastic products, such as food packaging, fast food boxes, or plastic containers (Du et al., 2020). Although the use of rayon is widespread nowadays, it has disadvantages in the properties of low crystallinity, low degree of orientation, and standard machine washing (Park et al., 2004), which is easy to stretch, shrink, or bleed onto other media (https://www.whowhatwear.com/what-is-rayon). Therefore, rayon products could be disintegrated rapidly and leached out the higher amounts of rayon, which also verifies our research.

# 3.5. Micro-image of MPs in drip coffee bags

SEM, Nikon microscopy, and  $\mu$ -FTIR spectra observations of the collected MPs particles presented different shapes in drip coffee bags (Fig. 5). The morphology of MPs could be satisfactorily presented using microscopic techniques, and the morphology discrepancies of different types of polymers were also observed. The results showed that fragmented MPs accounted for the majority. Before steeping, SEM was used to observe the filament interpenetrations of the drip bag, and the particles of different sizes adhered to the fibers (Fig. S2). MPs were dispersed in water by steeping for different periods. The small pieces of MPs attached to the drip bag filaments were easy to elute, and the drip bags had strong heat resistance stability. The filaments were not readily broken by boiling water in a short time. However, a few parts of

filaments could also be soaked fracture due to the poor processing technology of the drip bag fabrication. The MPs particles released after steeping appeared as an irregular block-like structure attached to a wrinkled strip (Fig. 5d). The surface of the MPs was wrinkled and rough, which possibly could be coffee bags deformed under hot water brewing (Fig. 5a-c).

## 3.6. Different classification methods analysis

Principal component analysis (PCA) is a mathematical tool to represent the variation in the dataset using a small number of factors (Granato et al., 2018). As a result, four plastic components (PE, PET, PP, and rayon) were detected in drip coffee bags from eight different commercial brands. The first three principal components were exported and highly contributed to the total variance: 82.9%, 10.7%, and 4.4%, respectively. The data implicated that PC1 was the main factor in differentiating the difference between drip coffee bags. The sum of these three contribution rates was 98% (higher than 85%). As shown in Fig. 6a, circles of different colors indicated the same time of steeping drip coffee bags from different brands. The distance between the points reflected the similarity of their detected MPs components. It could be seen from the score scatter diagram (Fig. 6a) that the extracts of eight brands of drip coffee bags steeped at different times could be well divided into group one (brand A-F) and group two (brand G, H). The biplot of different brands steeped times and MPs components were shown in Fig. 6b, as the loading plot showed the location distribution for the MPs components. Group one was related to PE and PP, and group



Fig. 5. Infrared spectra and microstructure of the most frequently observed MPs. (a) PE fragment; (b) PET fragment; (c) PP fragment; (d) Rayon; (a-c) Scanning electron microscope (SEM) and (d) Nikon microscopy images of the extracted MPs, respectively.



**Fig. 6.** Different classification methods to analyze four MPs components in drip bag coffee of different commercial brands. (a) PCA scores plot; (b) PCA biplot; (c) Dendrogram of HCA of MPs from the 8 commercial brands; (d) Heatmap of MPs components in drip bag coffee of different commercial brands.

two was associated with PET and rayon. Hence, different drip coffee bags could be distinguished by the various components of MPs.

Based on the result of PCA, hierarchical cluster analysis (HCA) was used to classify the eight different drip bags, which illustrated a more intuitive and straightforward result of the grouping. As shown in Fig. 6c, the two clusters suggested brand G&H and brand (A  $\sim$  F), respectively. HCA was the same as the distribution of different brands in Fig. 6a, assuming that PC1 and PC2 played an essential role in the model. The HCA and PCA mutually proved the precision of the classification of eight different brands of drip coffee bags with varying times of steeping.

The heat map is another method to express different profiles (Bergkvist et al., 2010). As shown in Fig. 6d, plastic compounds from various brands of drip bags were analyzed at different steeping times with a clustering heat map. Four major plastic compounds were shown on the heat map. The difference in color depth represented the number of detected compounds on the heat map. Four main compounds were also increased with the extension of the steeping time. The eight different brands of drip bags could be divided into two groups according to the component clustering, which was proved by the PCA and HCA analysis.

#### 3.7. Estimation of MPs intake by humans drinking coffee

According to previous research, humans drink 3–4 cups of coffee with an advantage to their health (Poole et al., 2017). Based on the results of this study, we could make the following calculations: if people brew three to four cups of coffee through a drip bag every day, they would at least intake 18,140 MPs (at most 51227) every day on average. Inferred from this, more than millions of MPs particles would be ingested each year. As a new brewing material, coffee bags could better release the flavor of coffee and were favored by consumers. Therefore, it is also vital to replace the fabricated drip bags with other harmless materials.

# 4. Conclusions

In the current research, we quantitatively investigated MPs releasing from drip coffee bags with eight brands at different steeping periods. The total absorbance infrared image of MPs generated by µ-FTIR combined with the standard plastics spectrum could generate a distribution map of each detected MP, which was convenient and fast to quantify the content of the components. At least four types of plastic polymers (PE, PET, PP and rayon) were detected in the six brands of drip coffee bags. The other two brands mainly contained PET and rayon. According to the plastic components released from drip coffee bags, all the experimental brands could be classified into two clusters. More than 10,000 MPs were found in the drip coffee bag after steeping for 5 min. The loose structure and the rough surface of drip coffee bags were likely to result in more MPs. Based on these preliminary studies, it is also crucial to further investigate the potential toxic mechanisms of MPs from drip bags of different sizes and structures.

# CRediT authorship contribution statement

Hao-Peng Wang: Methodology, Investigation, Formal analysis, Data curation, Writing – original draft. Xu-Hui Huang: Formal analysis. Jia-Nan Chen: Formal analysis. Meng Dong: Formal analysis. Yu-Ying Zhang: Supervision, Formal analysis. Lei Qin: Supervision, Project administration.

#### **Declaration of Competing Interest**

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

## Data availability

Data will be made available on request.

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.foodchem.2023.135717.

#### References

- Bergkvist, A., Rusnakova, V., Sindelka, R., Garda, J. M., Sjogreen, B., Lindh, D., ... Kubista, M. (2010). Gene expression profiling-Clusters of possibilities. *Methods*, 50 (4), 323–335. https://doi.org/10.1016/j.ymeth.2010.01.009
- Bessa, F., Barria, P., Neto, J. M., Frias, J., Otero, V., Sobral, P., & Marques, J. C. (2018). Occurrence of microplastics in commercial fish from a natural estuarine environment. *Marine Pollution Bulletin, 128*, 575–584. https://doi.org/10.1016/j. marpolbul.2018.01.044
- Bouwmeester, H., Hollman, P. C., & Peters, R. J. (2015). Potential health impact of environmentally released micro- and nanoplastics in the human food production chain: Experiences from nanotoxicology. *Environmental Science & Technology*, 49(15), 8932–8947. https://doi.org/10.1021/acs.est.5b01090
- Buntinx, M., Willems, G., Knockaert, G., Adons, D., Yperman, J., Carleer, R., & Peeters, R. (2014). Evaluation of the thickness and oxygen transmission rate before and after thermoforming mono- and multi-layer sheets into trays with variable depth. *Polymers*, 6(12), 3019–3043. https://doi.org/10.3390/polym6123019
- Cai, L., Wang, J., Peng, J., Tan, Z., Zhan, Z., Tan, X., & Chen, Q. (2017). Characteristic of microplastics in the atmospheric fallout from Dongguan city, China: Preliminary research and first evidence. *Environmental Science and Pollution Research, 24*, 24928–24935. https://doi.org/10.1007/s11356-017-0116-x
- Chong, J., Soufan, O., Li, C., Caraus, I., Li, S., Bourque, G., ... Xia, J. (2018). MetaboAnalyst 4.0: Towards more transparent and integrative metabolomics analysis. *Nucleic Acids Research*, 46(W1), W486–W494. https://doi.org/10.1093/ nar/gky310
- Diaz-Basantes, M. F., Conesa, J. A., & Fullana, A. (2020). Microplastics in honey, beer, milk and refreshments in Ecuador as emerging contaminants. *Sustainability*, 12(14). https://doi.org/10.3390/su12145514
- Ding, J. F., Li, J. X., Sun, C. J., Jiang, F. H., Ju, P., Qu, L. Y., ... He, C. F. (2019). Detection of microplastics in local marine organisms using a multi-technology system. *Analytical Methods*, 11(1), 78–87. https://doi.org/10.1039/c8ay01974f
- Dris, R., Gasperi, J., Mirande, C., Mandin, C., Guerrouache, M., Langlois, V., & Tassin, B. (2017). A first overview of textile fibers, including microplastics, in indoor and

outdoor environments. Environmental Pollution, 221, 453-458. https://doi.org/ 10.1016/j.envpol.2016.12.013

- Du, F., Cai, H. W., Zhang, Q., Chen, Q. Q., & Shi, H. H. (2020). Microplastics in take-out food containers. Journal of Hazardous Materials, 399, Article 122969. https://doi. org/10.1016/j.jhazmat.2020.122969
- Fadare, O. O., Wan, B., Guo, L. H., & Zhao, L. (2020). Microplastics from consumer plastic food containers: Are we consuming it? *Chemosphere*, 253, Article 126787. https://doi.org/10.1016/j.chemosphere.2020.126787
- Gao, F. L., Li, J. X., Hu, J., Sui, B. L., Wang, C. X., Sun, C. J., ... Ju, P. (2021). The seasonal distribution characteristics of microplastics on bathing beaches along the coast of Qingdao, China. *Science of The Total Environment*, 783, Article 146969. https://doi.org/10.1016/j.scitotenv.2021.146969
- Granato, D., Santos, J. S., Escher, G. B., Ferreira, B. L., & Maggio, R. M. (2018). Use of principal component analysis (PCA) and hierarchical cluster analysis (HCA) for multivariate association between bioactive compounds and functional properties in foods: A critical perspective. *Trends in Food Science & Technology*, 72, 83–90. https:// doi.org/10.1016/j.tifs.2017.12.006
- He, D. F., Zhang, X. T., & Hu, J. N. (2021). Methods for separating microplastics from complex solid matrices: Comparative analysis. *Journal of Hazardous Materials, 409*, Article 124640. https://doi.org/10.1016/j.jhazmat.2020.124640
- Hernandez, L. M., Xu, E. G., Larsson, H. C. E., Tahara, R., Maisuria, V. B., & Tufenkji, N. (2019). Plastic teabags release billions of microparticles and nanoparticles into tea. *Environmental Science & Technology*, 53(21), 12300–12310. https://doi.org/ 10.1021/acs.est.9b02540
- Holland, B. J., & Hay, J. N. (2000). Thermal degradation of nylon polymers. *Polymer International*, 49(9), 943–948. https://doi.org/10.1002/1097-0126(200009)49: 9<943::AID-PI400>3.0.CO:2-5
- Kannan, K., & Vimalkumar, K. (2021). A review of human exposure to microplastics and insights into microplastics as obesogens. *Frontiers in Endocrinology*, 12, Article 724989. https://doi.org/10.3389/fendo.2021.724989
- Kauffman, G. B. (1993). Rayon The first semi-synthetic fiber product. Journal of Chemical Education, 70(11), 869–956. https://doi.org/10.1021/ed070p887
- Kedzierski, M., Lechat, B., Sire, O., Le Maguer, G., Le Tilly, V., & Bruzaud, S. (2020). Microplastic contamination of packaged meat: Occurrence and associated risks. *Food Packaging and Shelf Life*, 24. https://doi.org/10.1016/j.fpsl.2020.100489
- Kellie, G. (2016). Developments in the use of nonwovens in packaging. Advances in Technical Nonwovens, (pp. 423-442). Doi: 10.1016/b978-0-08-100575-0.00016-4.
- Li, C. J., Zhu, L. X., Wang, X. H., Liu, K., & Li, D. J. (2021). Cross-oceanic distribution and origin of microplastics in the subsurface water of the South China Sea and Eastern Indian Ocean. Science of The Total Environment, 805, Article 150243. https://doi.org/ 10.1016/j.scitotenv.2021.150243
- Li, D. Z., Shi, Y. H., Yang, L. M., Xiao, L. W., Kehoe, D. K., Gun'ko, Y. K., Boland, J. J., & Wang, J. J. (2020). Microplastic release from the degradation of polypropylene feeding bottles during infant formula preparation. *Nature Food*, 1(11), 746–754. https://doi.org/10.1038/s43016-020-00171-y
- Li, L., Li, M., Deng, H., Cai, L., Cai, H., Yan, B., ... Shi, H. (2018). A straightforward method for measuring the range of apparent density of microplastics. *Science of the Total Environment*, 639, 367–373. https://doi.org/10.1016/j.scitotenv.2018.05.166
- Li, Y., Peng, L., Fu, J., Dai, X., & Wang, G. (2022). A microscopic survey on microplastics in beverages: The case of beer, mineral water and tea. *Analyst, 147*, 1099–1105. https://doi.org/10.1039/d2an00083k
- Liang, B., Zhong, Y., Huang, Y., Lin, X., Liu, J., Lin, L., ... Huang, Z. (2021). Underestimated health risks: polystyrene micro- and nanoplastics jointly induce intestinal barrier dysfunction by ROS-mediated epithelial cell apoptosis. *Particle and Fibre Toxicology*, 18(1), 20. https://doi.org/10.1186/s12989-021-00414-1
- Lin, J., Xu, X. M., Yue, B. Y., Xu, X. P., Liu, J. Z., Zhu, Q., & Wang, J. H. (2021). Multidecadal records of microplastic accumulation in the coastal sediments of the East China Sea. *Chemosphere*, 270, Article 128658. https://doi.org/10.1016/j. chemosphere.2020.128658
- Liu, Q., Chen, Z., Chen, Y., Yang, F., Yao, W., & Xie, Y. (2021). Microplastics and nanoplastics: emerging contaminants in food. *Journal of Agricultural and Food Chemistry*, 69(36), 10450–10468. https://doi.org/10.1021/acs.jafc.1c04199
- Liu, Q., Chen, Z., Chen, Y., Yang, F., Yao, W., & Xie, Y. (2022). Microplastics contamination in eggs: Detection, occurrence and status. *Food Chemistry*, 397, Article 133771. https://doi.org/10.1016/j.foodchem.2022.133771
- Lusher, A. L., Hernandez-Milian, G., O'Brien, J., Berrow, S., O'Connor, I., & Officer, R. (2015). Microplastic and macroplastic ingestion by a deep diving, oceanic cetacean: The True's beaked whale Mesoplodon mirus. *Environmental Pollution*, 199, 185–191. https://doi.org/10.1016/j.envpol.2015.01.023
- Maille, M. J., Sala, K., Scott, D. M., & Zukswert, H. (2021). Critical examination of particle swelling during wetting of ground coffee. *Journal of Food Engineering*, 295. https://doi.org/10.1016/j.jfoodeng.2020.110420
- Mason, S. A., Welch, V. G., & Neratko, J. (2018). Synthetic polymer contamination in bottled water. *Frontiers in Chemistry*, 6, 407. https://doi.org/10.3389/ fchem.2018.00407
- Mohsen, M., Lin, C., Liu, S., & Yang, H. (2022). Existence of microplastics in the edible part of the sea cucumber Apostichopus Japonicus. *Chemosphere*, 287(Pt 1), Article 132062. https://doi.org/10.1016/j.chemosphere.2021.132062
- Oh, S. J., & Koenig, J. L. (1998). Phase and curing behavior of polybutadiene diallyl phthalate blends monitored by FTIR imaging using focal-plane array detection. *Analytical Chemistry*, 70(9), 1768–1772. https://doi.org/10.1021/ac971290d
- Park, C. H., Kang, Y. K., & Im, S. S. (2004). Biodegradability of cellulose fabrics. Journal of Applied Polymer Science, 94(1), 248–253. https://doi.org/10.1002/app.20879
- Plastics Europe, 2020. Plastics-the facts 2020. An analysis of European plastics production, demand and waste data. Association of Plastic Manufacturers, Brussel.

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- Poole, R., Kennedy, O. J., Roderick, P., Fallowfield, J. A., Hayes, P. C., & Parkes, J. (2017). Coffee consumption and health: Umbrella review of meta-analyses of multiple health outcomes. *British Medical Journal*, 359, Article j5024. https://doi. org/10.1136/bmj.j5024
- Schneider, C. A., Rasband, W. S., & Eliceiri, K. W. (2012). NIH Image to ImageJ: 25 years of image analysis. *Nature Methoeds*, 9(7), 671–675. https://doi.org/10.1038/ nmeth.2089
- Su, Y., Hu, X., Tang, H. J., Lu, K., Li, H. M., Liu, S. J., ... Ji, R. (2021). Steam disinfection releases micro(nano)plastics from silicone-rubber baby teats as examined by optical photothermal infrared microspectroscopy. *Nature Nanotechnology*, 17, 76–85. https://doi.org/10.1038/s41565-021-00998-x
- Sun, H., Chen, N., Yang, X., Xia, Y., & Wu, D. (2021). Effects induced by polyethylene microplastics oral exposure on colon mucin release, inflammation, gut microflora composition and metabolism in mice. *Ecotoxicology and Environmental Safety, 220*, 112340. https://doi.org/10.1016/j.ecoenv.2021.112340
- Sun, J., Zhu, Z. R., Li, W. H., Yan, X., Wang, L. K., Zhang, L., ... Ni, B. J. (2021). Revisiting microplastics in landfill leachate: unnoticed tiny microplastics and their fate in treatment works. *Water Research*, 190, 116784. https://doi.org/10.1016/j. watres.2020.116784

- Thompson, R. C., Olsen, Y., Mitchell, R. P., Davis, A., Rowland, S. J., John, A. W., ... Russell, A. E. (2004). Lost at sea: Where is all the plastic? *Science*, 304(5672), 838. https://doi.org/10.1126/science.1094559
- Visalli, G., Facciola, A., Pruiti Ciarello, M., De Marco, G., Maisano, M., & Di Pietro, A. (2021). Acute and sub-chronic effects of microplastics (3 and 10 microm) on the human intestinal cells HT-29. *International Journal of Environmental Research and Public Health*, 18. https://doi.org/10.3390/ijerph18115833
- Wang, C., Jiang, L., Huang, W., Wang, C., & He, M. (2022). Light availability modulates the responses of the microalgae Desmodesmus sp. to micron-sized polyvinyl chloride microplastics. *Aquatic Toxicology*, 249, 106234. https://doi.org/10.1016/j. aquatox.2022.106234
- Woodall, L. C., Sanchez-Vidal, A., Canals, M., Paterson, G. L., Coppock, R., Sleight, V., ... Thompson, R. C. (2014). The deep sea is a major sink for microplastic debris. *Royal Society Open Science*, 1(4), Article 140317. https://doi.org/10.1098/rsos.140317
- Zhang, J., Wang, L., Trasande, L., & Kannan, K. (2021). Occurrence of polyethylene terephthalate and polycarbonate microplastics in infant and adult feces. *Environmental Science & Technology Letters*, 8, 989–994. https://doi.org/10.1021/acs. estlett.1c00559