



# Analysis of microplastics in various foods and assessment of aggregate human exposure via food consumption in Korea<sup>☆</sup>

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## ABSTRACT

Evidence of microplastics in humans has recently been demonstrated. The primary route of human exposure to microplastics is consumption of contaminated food and water. However, quantitative estimations of exposure to microplastics are limited, which hinders human health risk assessments. In this study, abundances of microplastics were measured in eight food types, comprising 90 products of table salts, soy sauces, fish sauces, salted seafood, seaweed, honey, beer, and beverage. Aggregate human exposure to microplastics via food consumption was assessed based on the number and mass of microplastics, using deterministic calculations and Monte Carlo simulations. The determinations revealed that average adult Koreans likely ingest  $1.4 \times 10^{-4}$  and  $3.1 \times 10^{-4}$  g of microplastics per week, respectively. These results are orders of magnitude smaller than earlier estimates of 0.1–5 g of microplastics per week that likely chose experimental outliers. Therefore, careful selection of literature data and estimation methods is needed to provide more realistic exposure estimations from microplastic counts. This study extends our understanding of MP occurrence in food and provides a more thorough estimate of aggregate microplastic exposure via food consumption.

## 1. Introduction

Environmental pollution by microplastics (MPs) has become extensive during the last two decades due to increased production, ubiquitous usage, uncontrolled disposal, and extremely slow natural degradation of plastics (Prata, 2018). MPs are synthetic particles smaller than 5 mm that are intentionally manufactured (primary MPs) or generated from larger plastics (secondary MPs). Both are released into the environment through various anthropogenic and natural pathways, contaminating ecosystems and entire food webs (Rahman et al., 2021). MPs accumulate in seawater (Alfaro-Núñez et al., 2021; Kanhai et al., 2017), freshwater (Bordós et al., 2019; Eo et al., 2019; Rodrigues et al., 2018), soil, and sediment (Horton et al., 2017; Mani et al., 2019).

Due to their ubiquitous occurrence in the environment, MPs have been found in various aquatic organisms that are important food sources, including fishes (Avio et al., 2020; Cheung et al., 2018; Markic et al., 2018; Oliveira et al., 2020; Song et al., 2022) and shellfishes (Catarino

et al., 2018; Gong et al., 2021; Hossain et al., 2020; Phuong et al., 2018; Waite et al., 2018). More MPs could be accumulated in aquatic products from aquaculture areas than the open ocean due to aquacultural activities and the common usage of fishing nets, foam buoys, and other plastics products (Lin et al., 2022). This could significantly increase the human exposure to MPs through the ingestion of aquatic products because 49.2% of aquatic food production come from aquaculture in 2020 (FAO, 2022). Therefore, global contamination by MPs is likely to return to our dinner table through the ingestion of food (Kwon et al., 2020). Human exposure to MPs is inevitable. Ingestion of MPs may lead to intestinal blockage and oxidative stress, increase the inflammatory response, reduce intestinal mucus secretion, damage the intestinal barrier function, trigger an imbalance of gut microbiota, and alter metabolism (Jin et al., 2019; Lu et al., 2018).

With growing concerns of human dietary exposure to MPs, many studies reported the occurrence of MPs in seafood and food from land-based sources, such as processed foods (Bouwmeester et al., 2015). The accumulation of MPs has been described in beers (Kosuth et al.,

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## Abbreviations

KNHANES	Korea National Health and Nutrition Examination Survey
ANM	The average number of microplastics intake
ARM	The average rate of microplastics intake
AMM	The average mass of microplastic particle

2018; Liebezeit and Liebezeit, 2014), honey (Liebezeit and Liebezeit, 2015; Mühlischlegel et al., 2017a), soft drinks (Shruti et al., 2020), and milk (Kutralam-Muniasamy et al., 2020). MPs could be from original food or from atmospheric fallout, manufacturing, and packaging (Karami et al., 2018; Li et al., 2020a; Udovicki et al., 2022). Further research is needed to clarify the abundance of MPs in different food types as well as the effects of the manufacturing/cooking process on the level of MPs in cooked food.

Humans are exposed to MPs through ingestion of food and water, inhalation of indoor and outdoor air, and direct dermal contact of the particles through personal care products, textiles, or indoor dust (Prata, 2018; Revel et al., 2018). Among exposure routes, ingestion of food and water is considered the primary route of human exposure to MPs (Rahman et al., 2021). As MPs have also been found in human feces (Yan et al., 2022) and blood (Leslie et al., 2022), food consumption is a very important route for MPs accumulation in humans. However, the extent of exposure via food consumption remains largely unknown. Although a few studies have quantitatively estimated the number of MP particles consumed by humans from seafood (Akoueson et al., 2020; Barboza et al., 2020; Catarino et al., 2018) and salt (Kim et al., 2018; Yang et al., 2015), the mass of aggregate exposure to MPs is not well-defined. There has been a significant disagreement in the estimated human dietary exposure among different studies (Mohamed Nor et al., 2021; Senathirajah et al., 2021) likely due to the uncertainties in the sources of experimental data and estimation methods (Pletz, 2022). Thus, further investigation is required to provide more quality data on the occurrence of MPs in commercially available food and beverages (Sewwandi et al., 2023).

In this study, we analyzed the size and composition of MPs in 90 foods comprising eight food types that were suspected to contain MPs to provide an extensive dataset for human dietary exposure to MPs in Korea and to give a more precise estimate of human dietary exposure. Aggregate human exposure was estimated using deterministic calculation and Monte Carlo simulation based on the abundances of MPs in the various food items we examined, literature data, and food intake statistics from the Korea National Health and Nutrition Examination Survey (KNHANES). Although the dataset was limited to Korean population, this more precise estimation was compared with earlier estimations.

## 2. Material and methods

### 2.1. Chemicals and materials

Concentrated hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>, 35% w/w) was purchased from Junsei Chemicals (Tokyo, Japan). Potassium hydroxide (KOH) was purchased from Daejung Chemicals (Siheung, Republic of Korea). Ferrous sulfate heptahydrate (FeSO<sub>4</sub>·7H<sub>2</sub>O) was obtained from Wako Chemicals (Osaka, Japan). Stainless steel filters with a pore size of 20 µm (KF-STC4720) were provided by the Korea Institute of Analytical Science and Technology (Seoul, Republic of Korea).

Food groups with high intake and sales that were suspected to contain substantial amounts of MPs were selected. Eight types of food were selected: table salt (12 products), soy sauce (10 products), fish sauce (eight products), salted seafood (15 products), seaweed (15

products), honey (six products), domestic and imported beer (18 products), and processed drinks (six products). Salt, honey, and beverages are commonly consumed in the world. Meanwhile, fish sauce, soy sauce, salted seafood, and seaweed were selected since fermentation techniques are examples of authentic Korean cuisine (Kim et al., 2016). Fish sauce and soy sauce are among the most basic seasoning to make the food savory, while salted seafood and seaweed are also traditional Korean foods (Kim et al., 2016). All products were purchased from e-commerce platforms considering their market share and availability. Detailed information on the food types, product types, and abbreviations for each product is provided in Table S1 (Supplementary Material).

### 2.2. Microplastic pretreatment methods

Pretreatment methods established in previous studies and existing literature have been reviewed (Kwon et al., 2020). A pretreatment method was developed to qualitatively and quantitatively evaluate the abundances of MPs (>20 µm) in each food type (Fig. 1). Method validation of pre-treatments for salt, soy sauce, and salted pollock roe was conducted in four independent laboratories based on blind tests in our previous study (Kim et al., 2022), achieving recoveries of reference materials (~100 µm) at 73, 77, and 86%, respectively. Soy sauce and salted pollock roe were assumed as representative sample matrices for solid (salted seafoods, seaweeds, and honey) and liquid (beer, fish sauce, and processed drinks) foods, respectively. The workflow of the pollock roe and seaweed is shown in Fig. S1 (Supplementary Material). Two-way analysis of variance (ANOVA) with Tukey's post hoc test was conducted to evaluate the differences in the abundances of MPs in the various products and manufacturing processes.

### 2.3. Dried seaweed and kelp washing scenarios

Effects of washing on MPs contamination were investigated in dried seaweed and kelp. In Korea, seaweed and kelp are mainly used for making soup and broth, respectively. They can be cooked either directly or after being washed. Therefore, four washing scenarios for each product were established as shown in Table S3, Supplementary Material. In the case of dried seaweed, 50 g of samples was soaked into 2 L of distilled water for 24 h at room temperature and then washed from one to three times by dipping samples into soaked water in a mixing bowl (10 times per wash). In the case of dried kelp, 50 g of products was prepared and washed from one to three times under running distilled water (2 L per wash). Afterward, the pretreatment of seaweed was performed and the reduction of MPs in dried seaweed and dried kelp after each scenario was investigated.

### 2.4. Chemical and size identification

The material composition of MPs was identified by FTIR spectroscopy using Nicolet™ iN10 MX (Thermo Fisher Scientific, Waltham, MA, USA), LUMOS II, or Hyperion 2000 (both from Bruker, Billerica, MA, USA) infrared imaging microscopes. The embedded imaging detectors were used to survey the spectrum of every pixel of the entire filter, creating a spectrum map of particles deposited on a 20 µm stainless steel filter. After the analysis, the plastic material type was determined as particles having infrared spectra that matched ≥70% reference spectra databases. The coordination of each MP was recorded, and its size was determined using FTIR microscopy images and a ruler tool as the longest dimension. The detailed FTIR settings are listed in Table S2 (Supplementary Material). The FTIR workflows for identifying MPs are shown in Fig. S2 (Supplementary Material).

### 2.5. Human exposure analysis

#### 2.5.1. Deterministic calculation

MPs exposure via food consumption was calculated using deter-

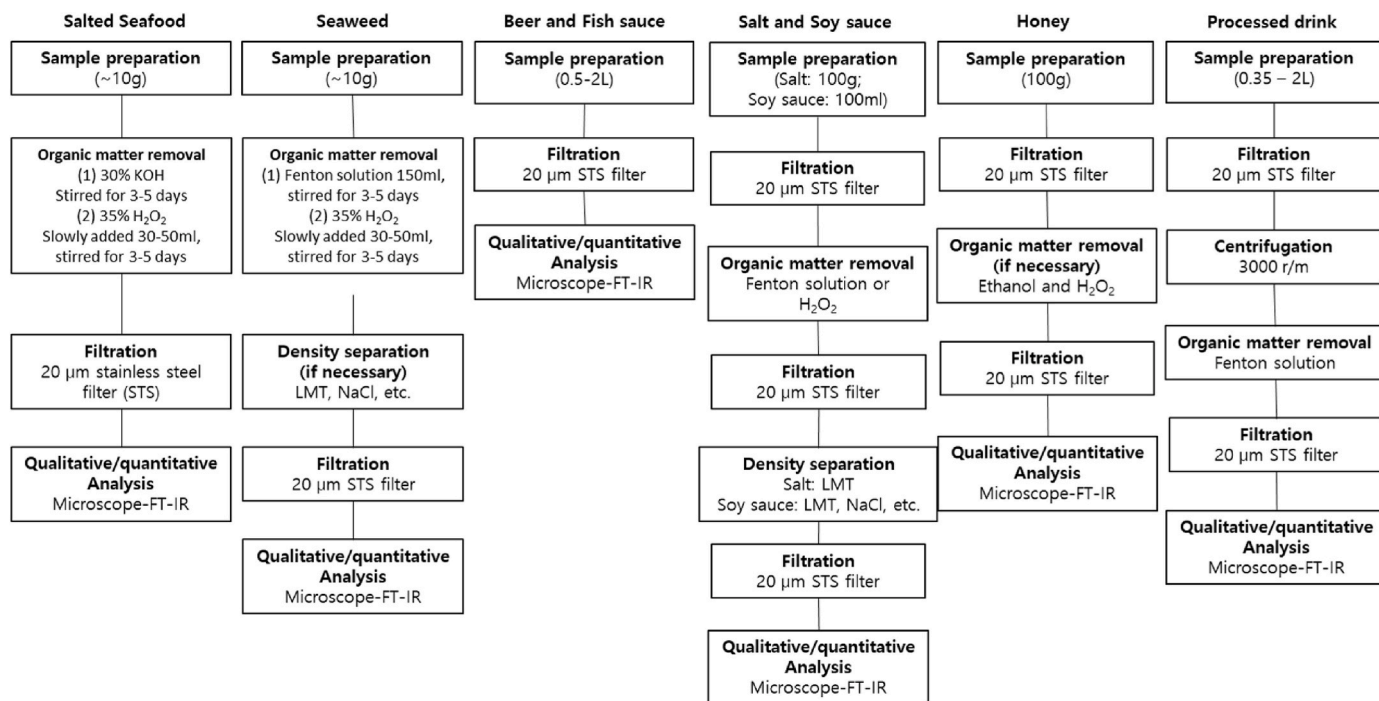


Fig. 1. Flow charts of the pretreatment methods for the eight food types.

ministic calculation and Monte Carlo simulation. The former used the arithmetic values of MPs abundance in this study and the food intake frequency from 2016 KNHANES data (KNHANES 2016). KNHANES 2016 includes data of the frequency and intake of 116 food groups surveyed from 20,671 people of all ages nationwide in Korea. Face-to-face interviews were conducted under external quality control, internal quality assurance, and control procedure programs in all steps (Kweon et al., 2014). Therefore, the ingestion rate in this study may be more reliable than the recommended dietary intake in other studies (Barboza et al., 2020; Cox et al., 2019). From this data, the mean and 95th percentile intakes of eight food groups for the entire Korean population were extracted. The average number of MPs intake (ANM) was calculated by multiplying the mean abundances of MPs by the mean intake of each food type. Values reported in particles per liter (p/L) for soy sauce, beverages, and beer were converted to p/g, assuming a density of 1 g/mL for further measurements. The ANM was calculated as follows:

$$\text{ANM (p / week / person)} = \text{number of particles per weight (p / g)} \times \text{food intake (g / week)} \quad (1)$$

The average rate of MP ingestion (ARM) (g/week/person) was calculated using a previously described method (Senathirajah et al., 2021) with some modifications. A representative estimated average mass of individual MP particles (AMM) was derived to calculate the ARM using particle size distribution. The MP size was classified into sections (e.g., 20–50 μm). The geometric mean and geometric standard deviation of particle size were derived from log-normal distributions instead of a uniform distribution assumed in Senathirajah et al. (2021). The AMM was calculated by assuming that MPs had an average spherical shape with an average density ( $\rho$ ) of 0.98 g/mL. A spherical shape was assumed instead of a cube or film shape because it was considered as an appropriate solution (Senathirajah et al., 2021)

$$\text{AMM (g / particles)} = \frac{\pi L^3 \rho}{6 \times 10^{12}} \quad (2)$$

$$\text{ARM (g / week / person)} = \text{ANM (p / week / person)} \times \text{AMM (g / particles)} \quad (3)$$

### 2.5.2. Monte Carlo simulation

The Monte Carlo simulation was performed using Crystal Ball from Oracle (Redwood Shores, CA, USA) using geometric means and geometric standard deviations of particle size and MPs abundances in this study, as well as KNHANES food intake data. A Monte Carlo simulation ( $n = 100,000$  iterations) was performed to obtain the ANM and ARM for each food type. The geometric mean and standard deviation of abundances of MPs were derived from the log-normal distribution of the abundances. The geometric mean and geometric standard deviation of food intake were also generated using the log-normal distribution conversion function of Crystal Ball software from the arithmetic mean and standard deviation of individual food intake in the KNHANES data. The ANM and ARM estimated by the Monte Carlo simulations were compared with those of deterministic calculations and other studies.

### 2.5.3. Aggregated human exposure via food consumption

Aggregate human exposure via food consumption was estimated as aggregate ANM and aggregate ARM by adding ARM or ANM of food items in this study with the values of other foods from previous studies. Only the categories of consumables with robust data were utilized, therefore, fishes, shellfishes, and water were chosen. The boundary of literature studies was set within Korea to minimize geographical variations. The abundance and size distribution of MPs were extracted from previous studies on MPs analysis in fishes (Kim et al., 2019), shellfishes (Cho et al., 2019; Kim et al., 2019), and water (Jung et al., 2022; Ministry of Environment of Korea, 2017). The intake frequency of fishes, shellfishes, and water was extracted from a 24-h dietary recall survey in KNHANES 2020. ARM, ANM, and AMM values of fish, shellfish, and water were estimated using deterministic calculations and Monte Carlo simulations.

We assumed that there was no change in the abundances of MPs during preparation and cooking. This assumption was considered reasonable for instant drinks, such as beer and beverages, or ingredients, such as soy sauce and fish sauce, that are directly added to food. If the cooking process is complicated and MPs are likely to be lost or introduced into food, there may be limitations to the application of the human exposure method.

## 2.6. Quality control

All labware used in the sampling and post-sampling processes were made of metal or glass to prevent possible contamination by MPs. All the sample preparation, pretreatment, and filtration processes were performed inside a laminar flow box to prevent contamination by indoor airborne MPs. All solutions, including ultrapure water and chemical reagents, were filtered using a metal filter (20 or 5  $\mu\text{m}$ ) before use. Filtered water and ethanol were used to clean all glassware prior to the laboratory experiments. The samples were covered with aluminum foil if they were moved outside the laminar flow hood. Experimenters wore cotton coats and nitrile gloves during all steps. The analysis of each product was repeated two or three times to evaluate the uncertainty in sampling and analysis. Blank samples (400–1000 mL deionized and filtered water) were analyzed per batch and data were reported only when no MPs greater than 20  $\mu\text{m}$  in the blank were observed for a given batch. No MPs were found in the blank samples, except for one polyethylene (PE) in the blank sample for honey (Table S8; Supplementary Material).

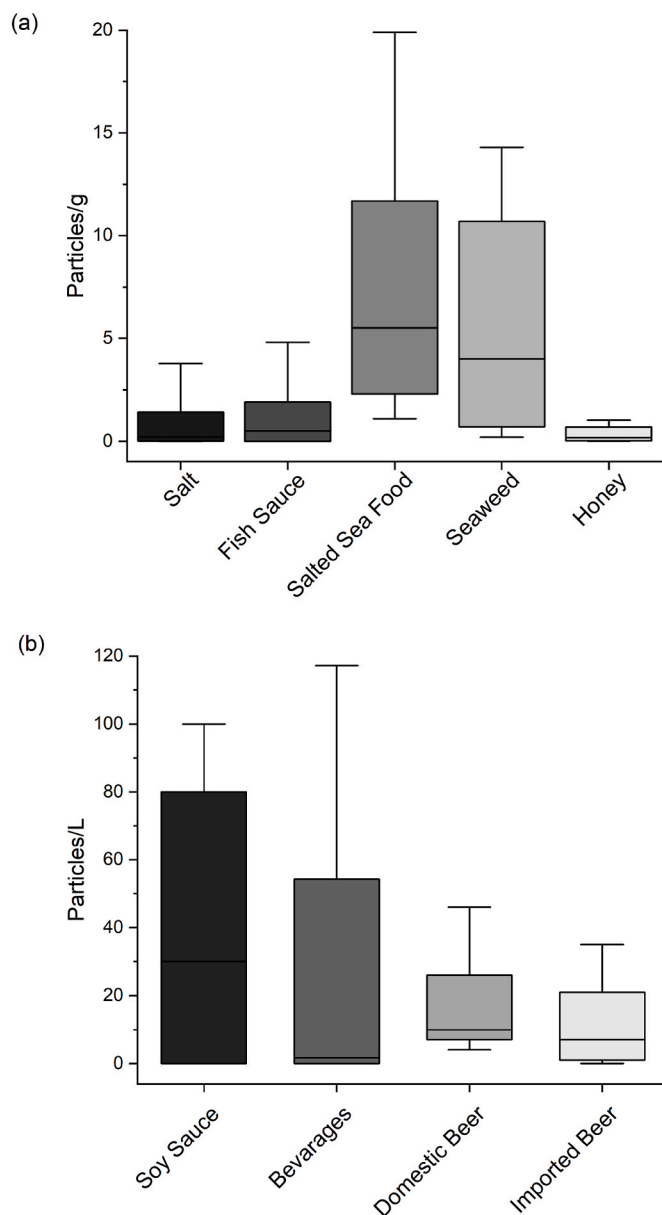
## 3. Results and discussion

### 3.1. Microplastics (p/g) in salt, fish sauce, salted seafood, seaweed, and honey

Abundances of MPs in salt ranged from non-detectable (n.d.) (SB-2, SC-5) to 3.77 (SA-3) p/g (median 0.22 p/g) (Fig. 2a). Sea salt (1.19 p/g) had the highest median number of detected MPs, followed by deep sea water salt (0.23 p/g), bamboo salt (0.15 p/g), and refined salt (0.11 p/g) (Table S4, Supplementary Material). The variation in the abundances by salt type can be explained by additional refining steps. Sea salts are generally produced by filtering and evaporating seawater; thus, the larger pore size of the filter allows for higher MP abundances. The slightly lower median value of MPs in deep seawater salts than in sea salts is limited by the sinking of low-density MPs in deep seawater. Baking at high temperatures to produce bamboo salts could further remove residual MPs. Refined salts are prepared by electrodialysis, which prohibits the migration of undissolved particles and further lowers the median MP abundance. The monitored levels of MPs in salts were close to the values reported in other studies (Iñiguez et al., 2017; Kim et al., 2018; Renzi et al., 2019). However, several studies reported a much greater occurrence of MPs in salts (Kosuth et al., 2018; Renzi and Blašković, 2018). This can be explained by the difference in the pre-treatment methods used, such as Rose Bengal staining (Kosuth et al., 2018) and the methods used to select and count MPs. In previous studies (Kosuth et al., 2018; Renzi and Blašković, 2018), suspected particles were visually selected, partially confirmed by FTIR/Raman spectroscopy, and extrapolated to estimate the abundance of MPs in the entire sample. This process inevitably involves measurement errors, resulting in uncertainties in the reported values. In this study, qualitative/quantitative analysis of MPs was performed using the mapping function of FTIR on the entire filter. The spectra of all suspected particles were recorded and analyzed to provide more reliable analysis results.

This is the first study to report the abundance of MPs in fish sauce. The abundance of MPs in fish sauce varied from n. d. to 4.80 p/g (median 0.60 p/g) (Fig. 2a). The median value was 1.40 p/g in anchovy fish sauce, 0.50 p/g in sand lance fish sauce and imported fish sauce, and 0.05 in imported fish sauce (Table S5, Supplementary Material). ANOVA suggested that the MPs abundance of anchovy fish sauce was significantly different from those of sand lance and imported fish sauce ( $p < 0.05$ ).

Abundances of MPs in five types of seaweed were analyzed, including dried kelp, salted kelp, dried seaweed, raw seaweed, and laver. The abundance of MPs in seaweed varied from 0.20 (KP-2) to 14.30 p/g (SWD-1), with a median value of 4.00 p/g (Fig. 2a). The highest median value was in laver (6.20 p/g), followed by salted kelp



**Fig. 2.** Abundances of microplastic in (a) salt, fish sauce, sea food, seaweed, and honey and (b) soy sauce, beverages, domestic beer, and imported beer. The data are expressed as median and interquartile range, with the bars representing the 10 and 90th percentiles and the whisker indicating the minimum and maximum range. Each of duplicate or triplicate analyses of a sample was included in the whisker plot to describe the entire range of measured values.

(3.60 p/g), dried seaweed (3.30 p/g), raw seaweed (2.60 p/g), and dried kelp (0.90 p/g) (Table S6, Supplementary Material). Comparing the results of dry and salted kelp, revealed a higher number of MPs in salted products, suggesting MPs pollution from external sources, including salt. Only one study (Li et al., 2020b) reported the mean value of MPs in commercial seaweed nori. The level ( $1.8 \pm 0.7$  p/g) was lower than the average level of MPs detected in laver ( $6.67 \pm 2.20$  p/g) in this study. NaCl solution was used for MPs flotation and isolation by Li et al. (2020b) instead of the lithium metatungstate solution used in the present study. This may have been the reason for the lower value previously reported.

Analysis of MPs of the salted seafood group was performed on five products, including salted pollock roe, anchovy, clam, pollock innards, and squid. The abundance of MPs in pollock roe and pollock innards is reported for the first time. Overall, the number of detected MPs ranged

from 1.30 (SQ-1) to 19.90 (AC-1) p/g (median 5.30 p/g) (Fig. 2a). The median values were 5.70 p/g in the pollock roe, 6.30 p/g in anchovy, 4.80 p/g in clam, 4.90 p/g in pollack innards, and 7.70 p/g in (Table S7, Supplementary Material). Two previous studies (Abbasi et al., 2018; Akhbarizadeh et al., 2018) have reported the MPs abundances (p/g) in the edible flesh of fishes. They were 0.16–1.50 p/g (Abbasi et al., 2018) and  $1.00 \pm 0.96$  p/g (Akhbarizadeh et al., 2018), which were smaller than the mean value in salted anchovy ( $6.49 \pm 5.59$  p/g) in this study. Previous studies (Cho et al., 2019; Hermabessiere et al., 2019; Su et al., 2018, 2016; Wu et al., 2020) have investigated the number of MPs (p/g) in the soft tissue of clams using similar pretreatment methods to those used in this study. The number of detected MPs was 0.03–1.08 (Cho et al., 2019),  $0.21 \pm 0.05$  (Wu et al., 2020), and  $0.74 \pm 0.35$  (Hermabessiere et al., 2019). These values were smaller than that in salted clams (2.10–8.70 p/g) in this study. Su et al. (2016, 2018) reported ranges of 0.30–4.90 and 0.20–12.50 p/g, respectively, which are more comparable with the abundances of MPs determined presently. Contamination of squid by MPs has been examined on only one previous study (Gong et al., 2021). The reported values were  $0.74 \pm 0.77$  p/g in the intestine,  $0.20 \pm 0.24$  p/g in gill, and  $0.30 \pm 0.24$  p/g in stomach tissues. The mean value in salted squid flesh determined in this study ( $7.77 \pm 5.37$  p/g) was significantly higher than the values previously reported, bolstering the literature concerning MPs in salted seafood. Overall, the abundances of MPs in salted seafood were higher than the values in raw seafood. The detected MPs were possibly from the salt added, manufacturing, and/or packaging processes. Besides, lower water content can be also an important factor contributing to the higher MPs abundances in dry products compared to raw one.

There is a lack of evidence of MPs contamination in honey (Mühlschlegel et al., 2017b). In this study, the abundances of MPs in honey ranged from 0.01 (HC-1) to 1.02 (HA-1) p/g, with a median value of 0.18 p/g (Fig. 2a). The median values were 0.33 p/g of domestic flower honey, 0.17 p/g of imported flower honey, and 0.09 p/g of domestic sugar feeding honey (Table S8, Supplementary Material). There was a significant difference in the mean number of MPs detected among the three groups ( $p < 0.05$ ). Post hoc Tukey analysis indicated a significant difference in the number of MPs detected between domestic flower honey and domestic sugar-fed honey. Three previous studies reported the presence of synthetic particles in honey. Synthetic particles in honey varied from 0.40–0.70 p/g (Liebezeit and Liebezeit, 2013) and 0.01–0.42 p/g (Liebezeit and Liebezeit, 2015). However, their composition was not verified as plastic, leading to difficulties in comparison. Mühlshlegel et al. (2017b) concluded that honey was not significantly contaminated with MPs, as only one polyethylene terephthalate (PET) was found in three 250-g honey samples. MPs were visually counted by Mühlshlegel et al. (2017b), possibly leading to underestimation of MPs in honey.

### 3.2. Microplastics (p/L) in soy sauce, beverage, and beer

MPs were detected in all purchased products, except for the DC-1 bottled tea sample. The MP abundance in soy sauce ranged from n. d. (FA-8, 10) to 100.0 (FA-2) p/L, with a median of 30.0 p/L (Fig. 2b). The median values were 30.0 p/L of Korean-style soy sauce, 50.0 p/L of brewed soy sauce, and 10.0 p/L of imported soy sauce (Table S9; Supplementary Material). There was no difference in the number of MPs detected between Korean-style soy sauce (FA-1–4) and brewed soy sauce (FA-5–8) ( $p > 0.05$ ). However, there was a significant difference in the number of MPs detected between domestic (FA-1–8) and imported soy sauce (FA-9, 10) ( $p < 0.05$ ). Fewer foreign products may have limited the comparison. Additional data are required to compare the MP abundances between domestic and imported soy sauces. To explore the source of the contaminating MPs, the manufacturing process was investigated. Although filtration was included, soy sauce still contained a greater level of MPs than other filtered foods, such as beer. MPs are suspected to originate from external sources, such as sea salt added

during the fermentation process, which requires further investigation.

The MP abundance in beverages ranged from n. d. (DC-1) to 41.00 (DB-1) p/sample (n.d.–117.14 p/L, median 1.75 p/L) (Fig. 2b). The median values were 4.50 p/sample (2.25 p/L) in soft drinks, 10.5 p/sample (29.3 p/L) in fruit drinks, and n. d. in bottled teas (Table S10, Supplementary Material). The median value of MPs in fruit drinks was higher than that in soft drinks and bottled tea. Fruit drinks might be more vulnerable to MP contamination than other processed drinks because of the bigger pore sizes of filters during production. For instance, a separation procedure of pulp to produce fruit nectars containing fruit flesh may require multiple filtrations through pore sizes of 0.4–6 mm (Horvath et al., 1990). On the other hand, the water treatment for carbonated soft drink requires membrane filtrations, removing particles down to 0.1 nm (Tatlock et al., 2007). Besides, MPs originated from the food container's inner surfaces also cannot be excluded. There were no significant differences in abundances of MPs among the three beverage types ( $p > 0.05$ ). Shruti et al. (2020) reported a median MP value of 2 p/L in soft drinks, which is comparable with our results. The authors reported 1–6 p/L MPs in cold tea, which was higher than the range of n. d.–2.00 p/L in bottled tea in this study, which can be explained by the different methods used to identify and enumerate MPs.

Beer samples were divided into domestic and imported beer. They were further divided into container types (glass, aluminum cans, and PET bottles). Overall, the number of detected MPs ranged from n. d. (IB-7) to 46.0 (DB-10) p/L, with a median value of 9.00 p/L. They were 10.00 p/L for domestic beers and 7.00 p/L for imported beers (Fig. 2b). Regarding the contamination of beer from container materials, beer contained in glass bottles and cans had the lowest and highest median value of MPs, respectively (Tables S11 and S12; Supplementary Material). Two studies (Kosuth et al., 2018; Liebezeit and Liebezeit, 2014) reported the anthropogenic particles detected in beer (16–254 p/L and n. d.–14.3 p/L, respectively). However, the findings were not chemically verified, complicating the comparison. Two studies (Li et al., 2022; Shruti et al., 2020) reported visually detected MPs that were partially verified using micro-Raman spectroscopy. The MPs abundance of 20,000–80,000 p/L (Li et al., 2022) was significantly higher than the MPs abundance (n.d.–46.00 p/L) in this study. The values of Shruti et al. (2020) ranged from n. d. to 28 p/L, which were more comparable to the present results. Li et al. (2022) collectively referred to all polymeric particles ( $\leq 5$  mm) as MPs and chemically verified a partial number of particles, which could explain the significantly elevated number of MPs reported. To identify the source of contaminating MPs, the manufacturing processes of beers were investigated. Most of the commercial beers were filtered through a 0.2–30  $\mu$ m filter in the middle or final stage of the manufacturing process before packaging. The dominance of 45–99  $\mu$ m MP in beers suggested the source of MPs from the packaging processes after filtration.

### 3.3. Effect of washing on contamination of seaweed with microplastics

No change in the abundances of MPs during handling and cooking is possible for directly consumed foods, such as beverages and beers, and some directly added cooking ingredients (soy sauce and fish sauce). However, the actual intake of MPs through food, especially solid foods, can be affected by the addition or loss of MPs during preprocessing and cooking. In this study, dried seaweed and dried kelp were chosen to investigate the effects of washing before consumption on the contamination by MPs, because seaweed and kelp had the highest ARM reported in this study and are commonly washed before cooking or eating. The number of MPs in dried seaweed and kelp decreased after repeated washing in comparison to those after the first scenario, with significant reduction by washing once (Fig. 3a and b). MPs reduction by 70% and 84% in dried seaweed and dried kelp, respectively, were observed after washing twice with tap water, and an additional 7% and 6% reduction was expected when washing three times (Table S13; Supplementary Material). As reported in our method validation study (Kim et al., 2022),

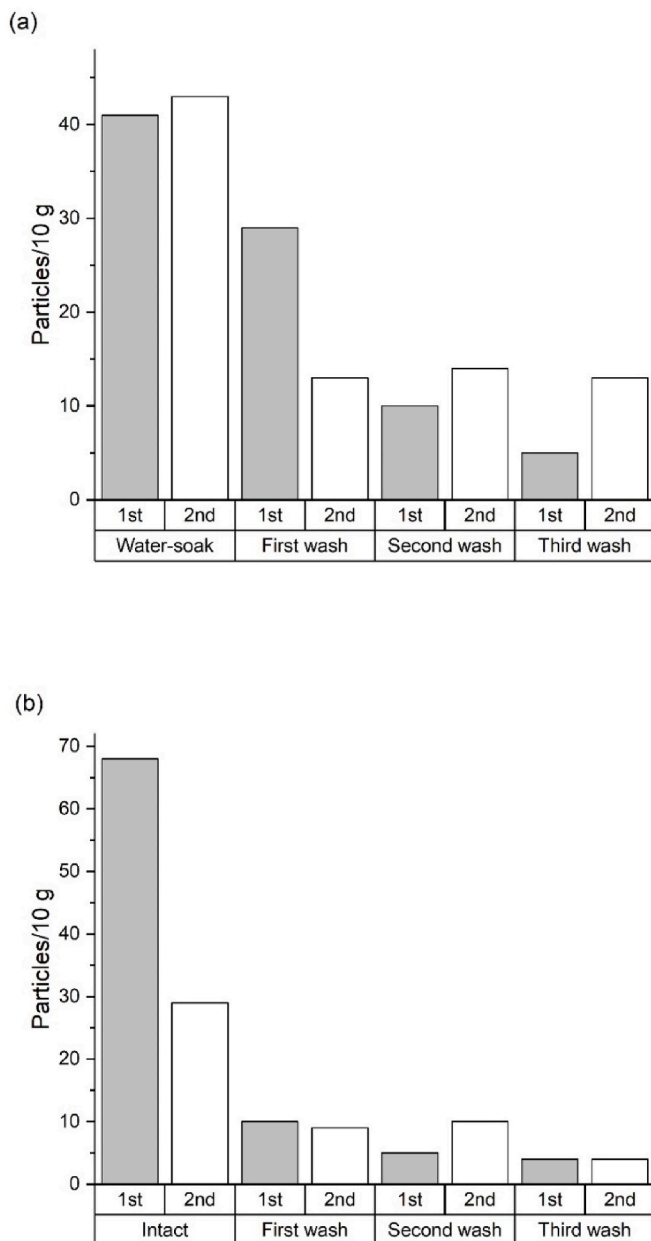


Fig. 3. Abundances of microplastic after four preparing scenarios in (a) dry seaweed and (b) dried kelp.

the number of MPs in salted pollock roe samples was also significantly decreased by surface washing with tap water. Because ARMs are high in solid foods that require preprocessing and cooking, such as seaweed and salted seafood, further research is needed to examine the actual intake of MPs through food with preprocessing and cooking.

### 3.4. Microplastic size and composition

Most of the MPs detected in all food types were PE, polypropylene (PP), and PET (Tables S3–S11, Supplementary Material). PE was dominant in all soy sauce, imported beer, and salted seafood products, as well as in most fish sauces and seaweed types (Fig. 4a). PP accounted for the highest proportion among the MP types in all honey and domestic beers, two salt types (SA, SC-1–3), and one seaweed type (SW). PET was most prominent in the two other salt types (SB and SC-4–5). Four beer products contained in PET bottles were dominant in PP and PE. The findings indicate that MPs in beer likely come from manufacturing

processes, rather than PET containers.

The majority of MPs detected in all food types were <300  $\mu\text{m}$  (70–100%) (Fig. 4b). Excluding two beer types (IB and DB), two seaweed types (SW and LV), one salted seafood type (PR), and one soy sauce type (FA), the sizes of MPs (<100  $\mu\text{m}$ ) of other products occupied the highest proportion (50–100%). The 45–99  $\mu\text{m}$  range of MPs size was the most dominant (38–72%) among the eight food types, except for sea salt (SA), which contained the highest proportion of 20–44  $\mu\text{m}$  MP (54%). Because sea salt also has the highest MPs abundance among salt types, the outweigh occupation of 20–44  $\mu\text{m}$  MP can be explained by the pore size of the filter paper during the manufacturing process.

### 3.5. Evaluation of human exposure

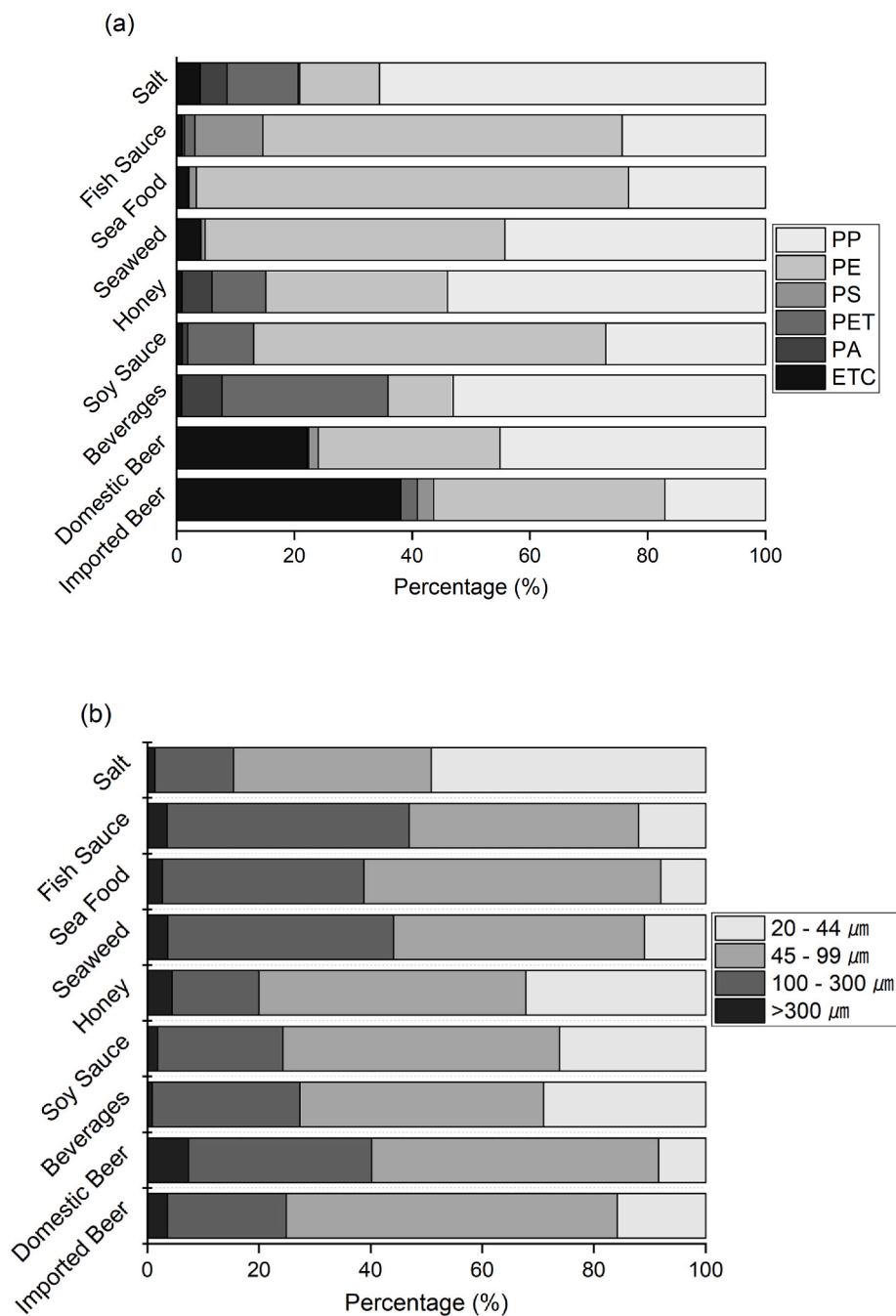
#### 3.5.1. Deterministic calculation

The deterministic calculation of human exposure to MPs revealed that ANMs due to the consumption of seaweed, salt, and fruit drinks were among the highest group at 24.5, 8.8, and 6.8 p/week, respectively. Honey (0.63 p/week), soy sauce (1.9 p/week), fish sauce (1.1 p/week), soft drinks (0.89 p/week), and bottled tea (0.037 p/week) had the lowest ANM values (Table 1). The detailed parameters used in the deterministic calculations are listed in Table S14 (Supplementary Material). Most food items in the smallest ANM group were liquid. Although these products had the highest consumption by mass among the food types in this study (Table 1), the results suggest that filtration can significantly alleviate MPs contamination in manufacturing processes. However, beer and fruit drinks had greater ANMs than other liquid foods, possibly because of the high consumption of beer in Korea and the complex filtration of fruit drinks. The ANMs of seafood, salt, honey, and beer reported by Senathirajah et al. (2021) were 6.5–313.2, 0.79–20.9, 1.1–2.1, and 3.4–16.7 p/week, respectively. These lower values are comparable with the average ANMs measured in this study, possibly because of the similar ANM calculation methods used in both studies.

The geometric mean of the MP size was used to minimize the influence of outliers in the dataset. The average value in all food types was 90.1  $\mu\text{m}$ , equating to an average AMM of 0.38  $\mu\text{g}$ /particle. MPs found in seaweed and salt had the highest and lowest AMM of 0.92 and 0.10  $\mu\text{g}$ /particle, respectively (Table 1). Senathirajah et al. (2021) reported AMM abundances of 13,000 and 480  $\mu\text{g}$ /particle in salt and beer, respectively, which were five and three orders of magnitude higher than the values reported in this study and unreasonably high (Pletz, 2022). Although the MPs were assumed to be spherical in both studies, the weight means of particle size used by Senathirajah et al. (2021) can be easily influenced by extreme data points, leading to exaggerated results. Consequently, ARMs of salt (0.88  $\mu\text{g}$ /week) and beer (2.6  $\mu\text{g}$ /week) in this study were significantly lower than those values of 140,000 and 4800  $\mu\text{g}$ /week calculated by Senathirajah et al. (2021). The detailed parameters used in the Monte Carlo calculation are listed in Table S14 (Supplementary Material). The ARMs of the other food items are listed in Table 1. The highest and lowest average MPs exposures were through intaking seaweed (22.4  $\mu\text{g}$ /week) and bottle tea (0.12  $\mu\text{g}$ /week), respectively (Table 1).

#### 3.5.2. Monte Carlo calculation

Exposures to MPs through seaweed and bottle tea had highest and lowest average rate of 84.8  $\mu\text{g}$ /week (39.3 p/week) and 0.050  $\mu\text{g}$ /week (0.074 p/week) (Table 2) (Fig. S3, Supplementary Material). The geometric mean, geometric standard deviation of MPs abundance/size, and repeated 100,000 runs of Monte Carlo simulations were believed to provide reasonable ranges considering uncertainties in both MP measurements and food consumption data. Utilizing the geometric means of particle size to reduce the influence of extreme data points is important for evaluating the toxicity of MPs because of their strong dependence on size distribution (Deng et al., 2017).



**Fig. 4.** (a) Microplastic material compositions and (b) size distribution in eight food types. (PP: polypropylene, PE: polyethylene, PS: polystyrene, PET: polyethylene terephthalate, PA: polyacrylate, ETC: other materials).

### 3.5.3. Aggregate human exposure via food consumption

The ARM (ANM) of fishes, shellfishes, and water calculated by the deterministic method were 90.2  $\mu\text{g}/\text{week}$  (146.5 p/week), 16.5  $\mu\text{g}/\text{week}$  (16.8 p/week), and 0.55  $\mu\text{g}/\text{week}$  (0.38 p/week), respectively (Table 2). The average and geometric mean of abundance of MPs in fishes, shellfishes, and water are shown in Table S15 (Supplementary Material). The deterministic aggregated ARM and aggregated ANM values were 139.9  $\mu\text{g}/\text{week}$  and 219.8 p/week, respectively. Those values were significantly smaller than aggregated ARM of 0.1–5.5 g/week reported by Senathirajah et al. (2021) including only four categories (shellfishes, salt, beer, and water), but rather close to the median intake of 4.1  $\mu\text{g}/\text{week}$  for adults (Mohamed Nor et al., 2021). Considering that spherical particles were assumed in this study whereas shape factors were considered in Mohamed Nor et al. (2021), differences between two

studies could be explained. The large difference in aggregated ARMs between this study and Senathirajah et al. (2021), although two studies used the same calculation method, can be explained by the different derivations of the representative particle size. In their study, the ARM of salt derived from a representative particle size of 2940  $\mu\text{m}$  accounted for more than 95% of the aggregated ARM. The size distribution of the salt used to estimate the representative particle size was obtained from two studies (Karami et al., 2017; Kim et al., 2018). The exclusion of MPs <math><100 \mu\text{m}</math> in these studies should have led to a higher estimation of the representative size, especially when weighted means are used. The much greater ANM reported by Senathirajah et al. (2021) compared to the value in this study (0.38 p/week) was due to the selection of data from the literature. The average number of MPs from different size ranges can result in an overestimation of the ANM. For example, Mintenig et al.

**Table 1**  
Deterministic calculation of mean number and mass consumed.

Food type	AMM (g/particle)	ANM (p/week)	ARM (g/week)
Salt	1.0E-07	8.8	8.8E-07
Soy sauce	2.3E-07	1.6	3.8E-07
Fish sauce	7.0E-07	1.1	7.6E-07
Salted seafood	5.8E-07	6.7	3.8E-06
Seaweed	9.2E-07	24.5	2.2E-05
Beer	5.1E-07	5.1	2.6E-06
Soda	2.8E-07	0.89	2.5E-07
Fruit drink	2.1E-07	6.8	1.4E-06
Bottle tea	5.4E-07	0.037	2.0E-08
Honey	1.9E-07	0.62	1.2E-07
Shellfishes	9.8E-07	16.8	1.6E-05
Fishes	6.2E-07	146.5	9.0E-05
Water	1.4E-06	0.38	5.5E-07
<b>Total</b>		219.8	1.4E-04

**Table 2**  
Monte Carlo simulation of mean number and mass consumed.

Food type	ANM (p/week/person)		ARM (g/week/person)	
	Mean	95%	Mean	95%
Salt	12.1	45.8	3.3E-06	1.2E-05
Soy sauce	128.0	173.0	5.5E-05	7.0E-05
Fish sauce	10.4	25.0	2.1E-05	3.4E-05
Salted seafood	7.6	26.4	9.6E-06	3.7E-05
Seaweed	39.3	117.0	8.5E-05	3.3E-04
Beer	4.3	7.6	8.1E-06	3.1E-05
Soda	0.89	2.6	7.9E-07	3.0E-06
Fruit drink	13.6	52.6	8.5E-06	2.8E-05
Bottle tea	0.074	0.074	5.0E-08	1.2E-07
Honey	0.65	1.9	5.8E-07	2.1E-06
Water	0.38	0.82	5.8E-07	1.4E-06
Fishes	145.0	534.0	9.1E-05	3.3E-04
Shellfishes	23.5	86.7	2.3E-05	8.4E-05
<b>Total</b>	385.8	1073.4	3.1E-04	9.6E-04

(2019) maximumly detected 0.7 p/L in the size range of 50–150  $\mu\text{m}$ , while Pivokonsky et al. (2018) detected up to 4102 p/L down to the size of 1  $\mu\text{m}$ . As a result, these variances overestimated the ARM in Senathirajah et al. (2021), causing difficulties in comparison. The comparisons suggested that the size range should be specified before selecting data to maintain the consistency of the correlation between MP abundance and the representative size of MPs.

The aggregated ARM and aggregated ANM calculated by Monte Carlo simulations are reported for the first time in this study. Aggregated RM (ANM) values of fishes, shellfishes, and water were 90.8  $\mu\text{g}/\text{week}$  (145.0 p/week), 22.5  $\mu\text{g}/\text{week}$  (23.5 p/week), and 0.58  $\mu\text{g}/\text{week}$  (0.38 p/week), respectively (Table 2). Consequently, the Monte Carlo calculated aggregated ARM and aggregated ANM were 305.6  $\mu\text{g}/\text{week}$  and 385.8 p/week, respectively, which were higher than those that were calculated by the deterministic method. In the case of the highest exposed group (95th percentile), Koreans can consume an MPs aggregate mass of 962.6  $\mu\text{g}/\text{week}$  and an aggregate abundance of 1073.4 p/week (Table 2).

The results suggest that data should be selected more carefully to ensure the quality of the estimations. In this study, the criteria were strictly set to investigate human exposure to 20–1000  $\mu\text{m}$  MP. The abundance and size distributions of MPs in fishes, shellfishes, and water were extracted from literature studies using spectroscopic methods, preferably FTIR, so that the size could be consistent within the 20–1000  $\mu\text{m}$  range. Geographical restrictions within Korea might limit the applicability of the dataset globally. Although MP pollution levels varied between diets of different regions of the world, the variations would be likely within an order of magnitude. Moreover, this study omitted the MPs and nanoplastics (NPs) having sizes smaller than 20  $\mu\text{m}$ . Despite of toxicological importance, experimental data on MPs/NPs <10  $\mu\text{m}$  in

foods are very scarce (Thiele et al., 2021). Nevertheless, human exposure to MPs (>20  $\mu\text{m}$ ) can be used as a surrogate to more toxicologically meaningful exposure to smaller particles considering the size distribution and total exposure in mass of MPs.

Although this study provides reasonable and up-to-date estimates of dietary exposure to MPs, further elaborations are needed. Although the total MPs ingested mass of 13 food items is reported for the first time in this study, additional data on highly consumed foods, such as milk, rice, bread, meat, and other foods, are required to extend future estimations. Because MPs can be added or destroyed during preparation and cooking processes before the final consumption, further research should be conducted on the impact of preparation and cooking processes. To validate the estimates based on the occurrence of MPs in major food contributors, it is also required to analyze MPs in human feces quantitatively. Further research is also needed to identify the MPs <20  $\mu\text{m}$  qualitatively and quantitatively in various food. Finally, the threshold of MPs ingested toxicity must be determined toward more reliable risk assessment.

#### 4. Conclusions

This study proposes pretreatment and analytical methods for eight types of food. The median MPs abundance in honey, salt, fish sauce, seaweed, and salted seafood were 0.18, 0.22, 0.60, 4.00, and 5.30 p/g, respectively. The median MPs abundances in beverages, beer, and soy sauce were 1.75, 9.00, and 30.00 p/L, respectively. PE, PP, and PET were the most dominant MPs detected in all food types. Most MPs detected were smaller than 300  $\mu\text{m}$  (70–100%). MPs can originate from external sources, such as the manufacturing process of salt and beverages, packaging, and the salt added to seafood, seaweed, and soy sauce, rather than the food itself. Human exposure assessment of MPs was conducted using deterministic calculations and Monte Carlo simulations. The highest and lowest MPs ingested masses were through consuming seaweed at 62.6  $\mu\text{g}/\text{week}$  and drinking bottled tea at 0.050  $\mu\text{g}/\text{week}$ . Additional data on fishes, shellfishes, and water were extracted from the literature to measure the total number of MPs (aggregated ANM) and mass ingested (aggregated ARM). By deterministic and Monte Carlo calculations, Koreans could potentially ingest  $1.4 \times 10^{-4}$  and  $3.1 \times 10^{-4}$  g MP per week, respectively. The larger value in the earlier estimation was likely from biases choosing and extracting literature data on abundance and size distribution of MPs. Therefore, standardizations of calculations/parameters and geographical boundaries are required to ensure the quality of measurements. This study extends the understanding of the occurrence of MPs in foods and provides a more complete estimate of the amount of MPs that may be ingested by Koreans for the further development of human health risk assessments, management, and policy.

#### Author statement

Dat T. Pham: Conceptualization, methodology, investigation, validation, visualization, and writing—original draft preparation. Jinwoo Kim: conceptualization, methodology, and investigation. Sang-Hwa Lee: Methodology and investigation. Juyang Kim: Methodology and investigation. Dowoon Kim: Methodology and investigation. Soonki Hong: supervision and funding acquisition. Jaehak Jung: supervision, funding acquisition, writing—reviewing and editing. Jung-Hwan Kwon: Conceptualization, supervision, funding acquisition, writing—reviewing and editing.

#### 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.envpol.2023.121153>.

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