Journal of Food Composition and Analysis 114 (2022) 104856

Contents lists available at ScienceDirect

Journal of Food Composition and Analysis

journal homepage: www.elsevier.com/locate/jfca

Development and validation of analytical methods for detecting and identifying microplastics in salts, soy sauce, and salted pollock roe

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ARTICLE INFO

Keywords:
Microplastics
FT-IR
Food analysis
Method validation
Food composition

ABSTRACT

One of the main routes of human exposure to microplastics is food consumption; therefore, the quantitative analysis of microplastics in foods is important. Researchers have reported the occurrence of microplastics in different ways, necessitating the validation and standardization of analytical methods. In this study, we optimized pretreatment methods for three representative foods (i.e., salts, soy sauce, and salted pollock roe) and verified analytical methods, including material identification using Fourier-transform infrared spectroscopy, in four independent laboratories based on blind tests. We prepared custom-made cylindrical reference materials, visually identifiable under a microscope, using five plastic materials (polyethylene, polypropylene, polyethylene terephthalate, polystyrene, and polyamide-6). Each food sample was spiked with a randomized number of reference materials and sent to testing laboratories for analysis without notifying them the material type and number of reference materials. The average recovery rates of the reference materials for salt, soy sauce, and salted pollock roe were 73.2%, 76.9%, and 86.2%, respectively. Two-way analysis of variance of the experimental results demonstrated that the recoveries of the reference materials did not depend on any of the five plastic materials or the participating laboratories, indicating that the proposed methods are capable of reliably determining microplastics greater than 100 μm in selected foods.

1. Introduction

Microplastics are ubiquitous in diverse environments, including seawater (Savoca et al., 2019; Sun et al., 2018), marine sediments (Claessens et al., 2011; Reed et al., 2018), soils (Liu et al., 2018; Scheurer and Bigalke, 2018; Zhou et al., 2020), and air (Amato-Lourenço et al., 2020; Prata, 2018). Owing to the increasing accumulation of microplastics in the environment, it is suspected that they exist in various foods as well. Microplastic ingestion via food consumption has become one of the most significant contemporary environmental problems (Addo Ntim et al., 2018). In the last decade, several studies have demonstrated the existence of microplastics in sea salts and seafoods (Akhbarizadeh et al., 2020; Cho et al., 2019; Daniel et al., 2020; Devriese et al., 2015; Fadare et al., 2021; Gundogdu, 2018; Hermabessiere et al., 2019; Hossain et al., 2020; Iniguez et al., 2017; Karami et al., 2018; Lee et al., 2019; Li et al., 2015; Li et al., 2016; Piyawardhana et al., 2021; Sathish et al., 2020; Sparks et al., 2021; Van Cauwenberghe and Janssen, 2014; Wootton et al., 2021; Yang et al., 2015). Recently, microplastics have been observed in diverse foods, including beer (Diaz-Basantes et al., 2020; Kosuth et al., 2018; Liebezeit & Liebezeit, 2014), milk (Diaz-Basantes et al., 2020; Kutralam-Muniasamy et al., 2020), beverages (Shruti et al., 2020), and honey (Diaz-Basantes et al., 2020; Liebezeit & Liebezeit, 2013; Liebezeit & Liebezeit, 2015).

The isolation and identification of microplastics, especially in foods,

Abbreviations: ANOVA, analysis of variance; EDX, energy dispersive X-ray spectroscopy; FT-IR, Fourier transform infrared spectroscopy; HDPE, high density polyethylene; PA-6, polyamide-6; PET, polyethylene terephthalate; PP, polypropylene; PS, polystyrene; SEM, scanning electron microscopy.

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https://doi.org/10.1016/j.jfca.2022.104856
Received 8 February 2022; Received in revised form 9 August 2022; Accepted 22 August 2022
Available online 23 August 2022

are very challenging because microplastics are embedded in complex food matrices containing several natural and anthropogenic polymeric materials (Kwon et al., 2020). Depending on the nature and characteristics of the surrounding matrix, various pretreatment methods for isolating microplastics from a given matrix have been applied (Sridhar et al., 2022). In addition, various identification methods were attempted, including optical stereomicroscopy, Fourier-transform infrared spectroscopy (FT-IR), Raman spectroscopy, and scanning electron microscopy (SEM) with energy-dispersive X-ray spectroscopy (EDX). For microplastics in seawater and sediment, an experimental protocol was drafted by the National Oceanic and Atmospheric Administration (NOAA) (Masura et al., 2015), and each step of the protocol was validated for marine sediment samples (Vermeiren et al., 2020). However, isolating and detecting microplastics from foods and other environmental matrices may require additional steps. Further validation of the experimental protocols for these matrices is required. The results reported by individual researchers often span a wide range and sometimes include possible false-positive detections. Differences in size cutoff, instruments used for identifying microplastics, and related analytical protocols among researchers make it difficult to assess the occurrence of microplastics in various foods and their corresponding human exposure (Koelmans et al., 2019). Thus, the validation of methods applied for quantifying microplastics in foods is an urgent requirement.

The competence and recovery of analytical methods for microplastics can be validated using spiked reference materials in test matrices. Reference materials have been applied to various matrices, such as water (Müller et al., 2020), sediment (Karlsson et al., 2017), biota (Karlsson et al., 2017), and soil (Hurley et al., 2018; Perez et al., 2022). Spectroscopic identification methods such as FT-IR have been validated using reference materials in most studies. However, a limited number of studies have evaluated the robustness of thermal analysis using reference materials (Hermabessiere et al., 2018; Ishimura et al., 2021; Matsueda et al., 2021). The reference materials utilized in earlier studies can be classified into four types based on their shape: fragments, beads, pellets/granules, and fibers. Fragments are relatively easy to produce in large quantities compared to other shapes because they can be prepared by physically grinding plastics. However, they are often indistinguishable from microplastic fragments that already exist in the matrix. Beads are easy to distinguish and can be produced in a sub-micrometer size. However, depending on the type of filter used, in the case of flat and small pore-sized filters such as silicone and Anodisc™ filters, reference material beads often slide on the filters and are difficult to locate under a microscope. Industrial pellets (>1 mm) are also used as the reference materials (e.g., Nuelle et al., 2014). Although they are easily identifiable, they may not represent the extraction recovery of relatively smaller microplastics (<1 mm) that could be lost during the pretreatment steps. Fiber reference materials are often easier to identify. However, there is a risk of underestimation because of the potential intertwining between fibers (Pirc et al., 2016).

In this study, we refined and validated analytical methods for isolating and identifying microplastics in three food matrices (i.e., salts, soy sauce, and salted pollock roe) in five independent laboratories. After a thorough review of the existing methods, the most appropriate pretreatment methods were chosen. The reference materials of five different plastic materials (high-density polyethylene (HDPE), polypropylene (PP), polystyrene (PS), polyethylene terephthalate (PET), and polyamide-6 (PA-6)) randomly spiked into sample matrices were analyzed to obtain recovery. The robustness of the methods was evaluated based on extraction recovery using an analysis of variance (ANOVA).

2. Materials and methods

2.1. Materials and chemicals

Salts, soy sauce, and salted pollock roe were purchased from an online market, as they are the major brands with a high market share in Korea. They were bought as packages to ensure their origin from the same batch. Salts were sealed in a PE bag, soy sauce was stored in a glass bottle with a PP cap, and the salted pollock roe was wrapped in polyethylene (PE) film in a PET box. Concentrated hydrogen peroxide (H_2O_2, 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 ($FeSO_4 0.7 H_2O$) and glucose were purchased from Wako Chemicals (Osaka, Japan) and Duksan Chemicals (Seoul, Republic of Korea), respectively. Stainless steel filters with pore sizes of 5 (KF-STC2505) and 20 μm (KF-STC2520 and KF-STC4720) were kindly provided by the Korea Institute of Analytical Science and Technology (Seoul, Republic of Korea). Glass microfiber filters (GF/D) were purchased from Whatman (Buckinghamshire, UK). Commercial pellets of HDPE, PP, PS (LG Chem Ltd., Seoul, Republic of Korea), PET (Lotte Chemical, Seoul, Republic of Korea), and PA-6 (Hyosung TNC Co., Seoul, Republic of Korea) were purchased and transformed into fiber form using an LME mini extruder (Dynisco, Westwood, MA, USA). The produced bundles of fibers were wrapped in parafilm and cut into cylinders with a height of approximately 100 μm using a Microm HM400 microtome (Microm International, Walldorf, Germany). Images of the reference materials were captured using a BX 51 microscope coupled with a DP20 camera (Olympus, Tokyo, Japan) (Fig. 1). The diameter and length of the cylindrical reference materials were measured based on image analysis of microscope images using IMAGEJ software 1.53a (National Institutes of Health, Bethesda, MD, USA). Table S1 (Supplementary Material) summarizes the characteristics of the reference materials used in this study.

2.2. Optimization of digestion methods

Among many digestion methods attempted for biota and organic-rich media, the methods suggested in ISO/TR 21960:2020 (International Standard Organization (ISO), 2020) and the references cited therein were modified to choose six candidate pretreatment methods, as presented in Table 1. To compare digestion efficiency, the remaining total carbon in the solution was measured using a TOC-V carbon analyzer coupled with an SSM-5000A solid sample combustion unit (Shimadzu, Kyoto, Japan) at 900 °C for 8 min as an indicator, assuming a negligible contribution of inorganic carbon.

One hundred grams of salt was dissolved in 400 mL of pretreatment solution (Table 1). Soy sauce (100 mL) was first filtered through a 20 μm (25 mm diameter) stainless steel filter and mixed with 200 mL of pretreatment solution, followed by 10 min of sonication. The filters were retrieved and reused during the subsequent filtration. Salted pollock roe (10 g) was added to 200 mL of pretreatment solution.

After the digestion time determined in preliminary experiments, the samples were filtered through a 5 μm (25 mm diameter) stainless steel filter. Salted pollock roe samples with low digestion efficiency were filtered with a 20 μm (47 mm diameter) stainless steel filter because of the high loading of remaining solids. The optimal volume ratios of the pretreatment solution to the samples were determined through trial and error in a series of preliminary experiments. The filters were dried for 24 h in a desiccator and analyzed using a TOC analyzer. The total carbon content of the sample filter was subtracted from that of the blank filters in triplicate. A standard curve was drawn with glucose over the range of 0.5–10 mg of carbon. Samples of salted pollock roe that contained more than 10 mg of carbon after digestion were further homogenized in a mortar and 10 mg of solids was used for the analysis of the total carbon in the remaining solids. The total carbon content of the sample was calculated based on the total solid mass on the filter.

For preventing microplastic contamination, we excluded all plastic tools and used metal scalpel and glass materials during the sampling and post-sampling processes. Except for field collection and sampling, all sample preparation, pretreatment, and filtration steps in the laboratory were performed inside of a laminar flow box (SINAN Science Industry,

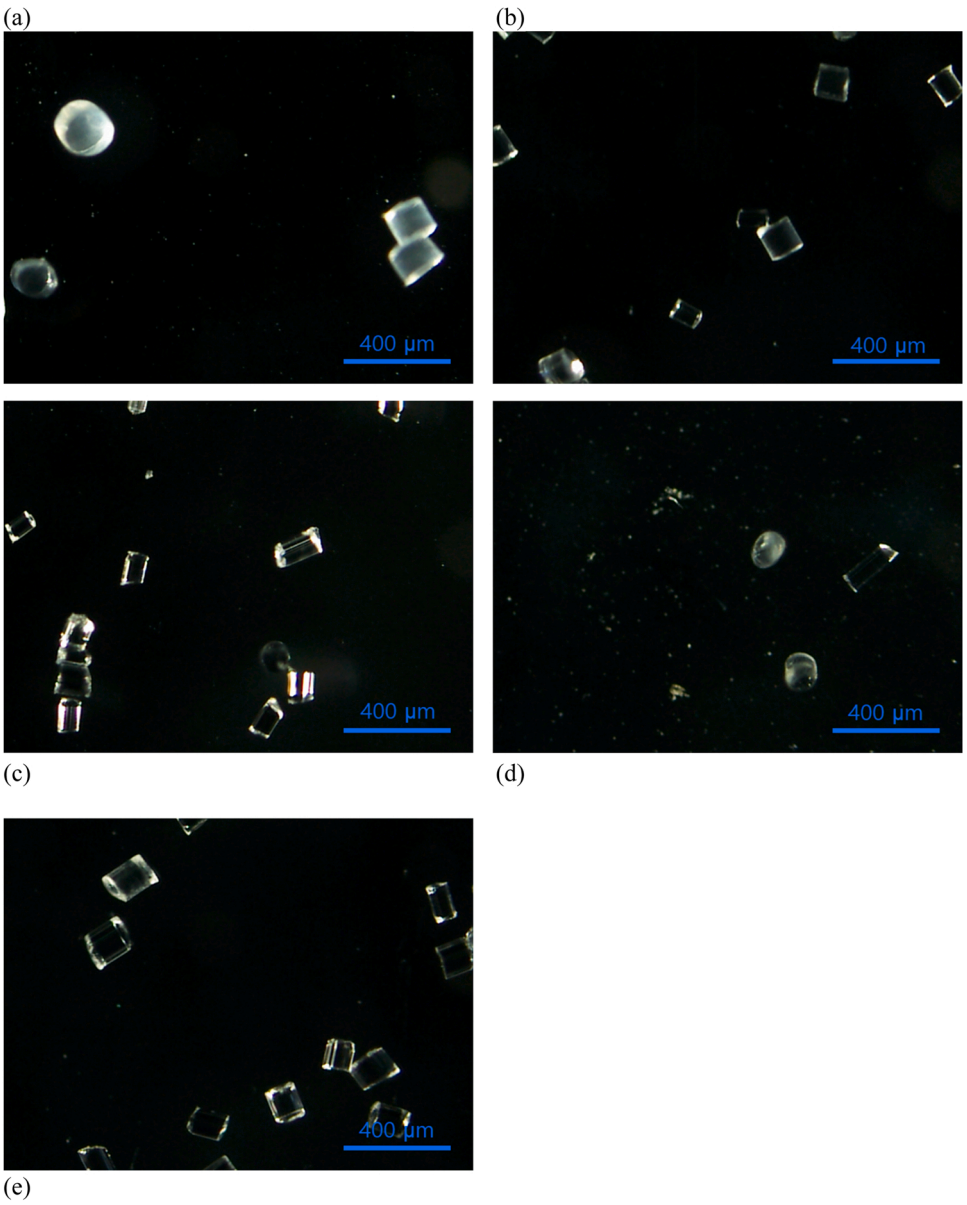

Fig. 1. Microscopic (4 ×) images of cylindrical reference materials of (a) polyethylene (PE), (b) polypropylene (PP), (c) polyethylene terephthalate (PET), (d) polystyrene (PS), (e) polyamide 6 (PA6) prepared in this study.

Table 1
Summary of pretreatment methods applied to salts, soy source, and salted pollock roe for total carbon analysis.

Method	Pretreatment solution	Reference
1	15% (w/w) H_2O_2, 7 d	Avio et al. (2017)
2	$FeSO_4$0.7 H_2O 6.67 mg/mL + 30% H_2O_2, 1 h	Tagg et al. (2017)
3	KOH 10% (w/v), 7 d	Cho et al. (2019)
4	KOH 30% (w/v), 7 d	This study
5	30% (w/v) KOH: 35% (v/v) H_2O_2 (3:1) 5 d in KOH solution, followed by another 5 d after adding H_2O_2 solution	This study
6	Distilled water, 7 d	Renzi & Blašković (2018)

HSCV-1300) to prevent contamination by indoor airborne microplastics. All solutions were previously filtered using a metal filter (5 μm) before use. Filtered water and ethanol were used to clean all glassware prior to laboratory experiments. All samples were covered with aluminum foil when they were moved outside the laminar flow hood. To minimize any contamination of the samples, the use of plastic material was avoided whenever possible, and nitrile gloves and cotton coats were used during all processing steps.

2.3. Method validation

We tested the robustness of the methods based on the extraction recovery of reference materials. We conducted inter-laboratory validation in four laboratories as pre-validation (Laboratories A, B, C, and D) and validation (Laboratories A, B, C, and E). Each sample was spiked with a randomly generated number of microplastics (0–10 reference materials in the pre-validation and 5–10 reference materials in the validation). Each reference material was picked using a tweezer and transferred to a sample with great care. Samples were prepared in a laminar flow box, and the labware used during the experiment was washed with distilled water and then rinsed with ethanol. A glass bottle was used as the container in the pre-validation, whereas a beaker with a flat wall was used in the validation. While the samples were transported, the liquid samples were kept frozen using dry ice in a Styrofoam box.

The sample containers were covered with aluminum foil and sealed with parafilm. Samples were delivered to testing laboratories via postal service in the pre-validation stage, whereas they were delivered in person in the validation stage to avoid any potential losses or contamination of microplastics during transportation.

All samples were analyzed using the same optimized methods. After the samples were filtered, the side walls of the beaker and glass funnel were rinsed with at least 300 mL of distilled water and ethanol, and the rinsate was filtered through the same filter. Rinsing with ethanol dissolved the viscous saponified gel formed by the reaction between lipids in salted pollock roe and alkali solution and reduced the color staining from soy sauce (Dawson et al., 2020; Shruti et al., 2020). Distilled water and solutions used in the experiment were filtered through a Whatman GF-D glass fiber filter before use. The stainless steel filters used in the experiment were sonicated in distilled water, methanol, and hexane and dried. After filtration, the samples were stored in glass petri dishes.

To prevent potential contamination of microplastics, we excluded all the plastic tools and used metal scalpels and glass materials during the sampling and post-sampling processes prior to FT-IR analysis. Except for the field collection and sampling, the entire sample preparation, pretreatment, and filtration steps in the laboratory were performed in a laminar flow box. All the solutions were filtered using a metal filter (5 μm) before use. Filtered water and ethanol were used to clean all glassware prior to the laboratory experiments. All samples were covered with aluminum foil when they were moved outside the laminar flow hood. To minimize any contamination of the samples, the use of plastic material was avoided whenever possible, and nitrile gloves and cotton coats were used during all processing steps. At least one negative blank (400–1000 mL deionized and filtered water, depending on the laboratory) was included in a batch and data were reported only when no microplastics greater than 20 μm were observed.

Laboratories A, C, and D conducted analysis using Nicolet iN10 FT-IR instruments (ThermoFisher Scientific, Cleveland, OH). Laboratory B used a LUMOS 2 FT-IR instrument (Bruker, Fremont, CA), and laboratory E used a Hyperion 3000 FT-IR instrument (Bruker, Fremont, CA). The samples were analyzed using the library offered by the manufacturers, with additional spectrum data of pre-shared reference materials measured in each laboratory. Laboratories A, B, C, and D using FT-IR instruments that support automatic mapping mode, scanned the entire area of the filter and compared the similarities with the spectral data of each material. Laboratory E captured video images of the whole filter using a stereomicroscope and characterized microparticles with a cylindrical shape. The detailed settings of the analysis followed the standard protocol of each laboratory owing to differences in equipment and are summarized in Table S2, Supplementary Material.

2.4. Statistical analysis

The participating laboratories reported the number and material types of reference material in each sample. As the number of spiked reference materials were different in the samples, the weighted mean ($\bar{x}$) and standard deviation (s) of the recovery rate were calculated using the sample matrix and material type as follows:

$$\bar{x} = \frac{\sum_{i=1}^{N} w_i x_i}{\sum_{i=1}^{N} w_i} \quad (1)$$

$$s = \sqrt{\frac{\sum_{i=1}^{N} w_i (x_i - \bar{x})^2}{\frac{(M-1)}{M}\sum_{i=1}^{N} w_i}} \quad (2)$$

where N represents the number of samples; M is the number of non-zero weights; w_i is the weight of sample i, which is the number of spiked reference materials; x_i is the recovery rate of sample i (%); $\bar{x}$ and s are the weighted mean and standard deviation of the recovery rate (%), respectively.

Using the weighted mean of recovery (%) of each material type ($N = 3$) as input, we conducted a 2-way ANOVA (Pozo et al., 2019) with Tukey's post hoc test to compare the differences caused by the type of reference materials and analyzed with a Greenhouse–Geisser correction to meet the assumption of sphericity of the data based on the assumption of normality, independence, and equality of the variance. Statistical significance was set at $p < 0.05$.

2.5. Characterization of the size of microplastics other than reference materials

Laboratories A, B, and C also identified microplastics present in food samples other than the spiked reference materials. For comparison of particle size, rhombus-shaped microplastics were assumed, and diagonal height and width were determined to estimate the projected area.

$$\text{Area} = \frac{\text{Height} \times \text{Width}}{2} \quad (3)$$

The characteristic length of each particle was calculated as the diameter of a circle with the same projected area. The size classification was then divided into five groups: < 20 μm, 20–45 μm, 45–100 μm, 100–300 μm, and > 300 μm.

$$\text{Characteristic length} = 2\sqrt{\frac{\text{Area}}{\pi}} \quad (4)$$

3. Results and discussion

3.1. Optimization of pretreatment methods

The remaining carbon content after the pretreatment of the salt samples was below the detection limit, except for those using the KOH solution (methods 3, 4, and 5) (Fig. S1a, Supplementary Material). The formation of insoluble carbonate salts at high pH can explain the increase in total carbon in the filtrate, indicating that alkali treatment is not desirable for salts. We chose method 1 using H_2O_2 because any organic materials potentially present in the salt could be removed and microplastics may be colored after Fenton's reagent treatment.

The total carbon content of the soy sauce samples after treatment with H_2O_2 (method 1) and Fenton's reagent (method 2) was below the detection limit whereas those after other treatments were slightly above the detection limit (Fig. S1b, Supplementary Material). For the salt samples, we chose method 1 using H_2O_2 to prevent the coloring of microplastics after Fenton's reagent treatment.

Unlike salt and soy sauce samples, the removal of total carbon was enhanced by alkali treatments (methods 3, 4, and 5) for salted pollock roe samples (Fig. S1c, Supplementary Material), whereas oxidative methods demonstrated poor efficiency. Treatment with 30% KOH (Method 4) and 30% KOH with H_2O_2 (Method 5) resulted in a total carbon content below the detection limit. Considering the removal of total carbon and potential roles of the oxidizing agent for removing organic particles, method 5 using both KOH and H_2O_2 solution was chosen, although it took slightly more time. The detailed procedures of the three selected methods are schematically presented in Fig. 2 and are described below.

Salt: 100 g of salt was measured in a 600 mL beaker with a scale where the reference materials were spiked. Salt with reference materials was dissolved in distilled water to obtain a total solution volume of 200 mL, and later 200 mL of 30% H_2O_2 was added. The solution was agitated at 300 rpm for 7 d at room temperature. The treated solution was filtered through a 20 μm (25 mm diameter) stainless steel filter. The sidewalls of the beaker and filter holder were washed with a sufficient amount of distilled water. The filter was retrieved from the holder assembly and dried in a desiccator for 1 d, and the particles on the filters were analyzed using FT-IR.

Soy sauce: 100 mL of soy sauce was measured using a volumetric

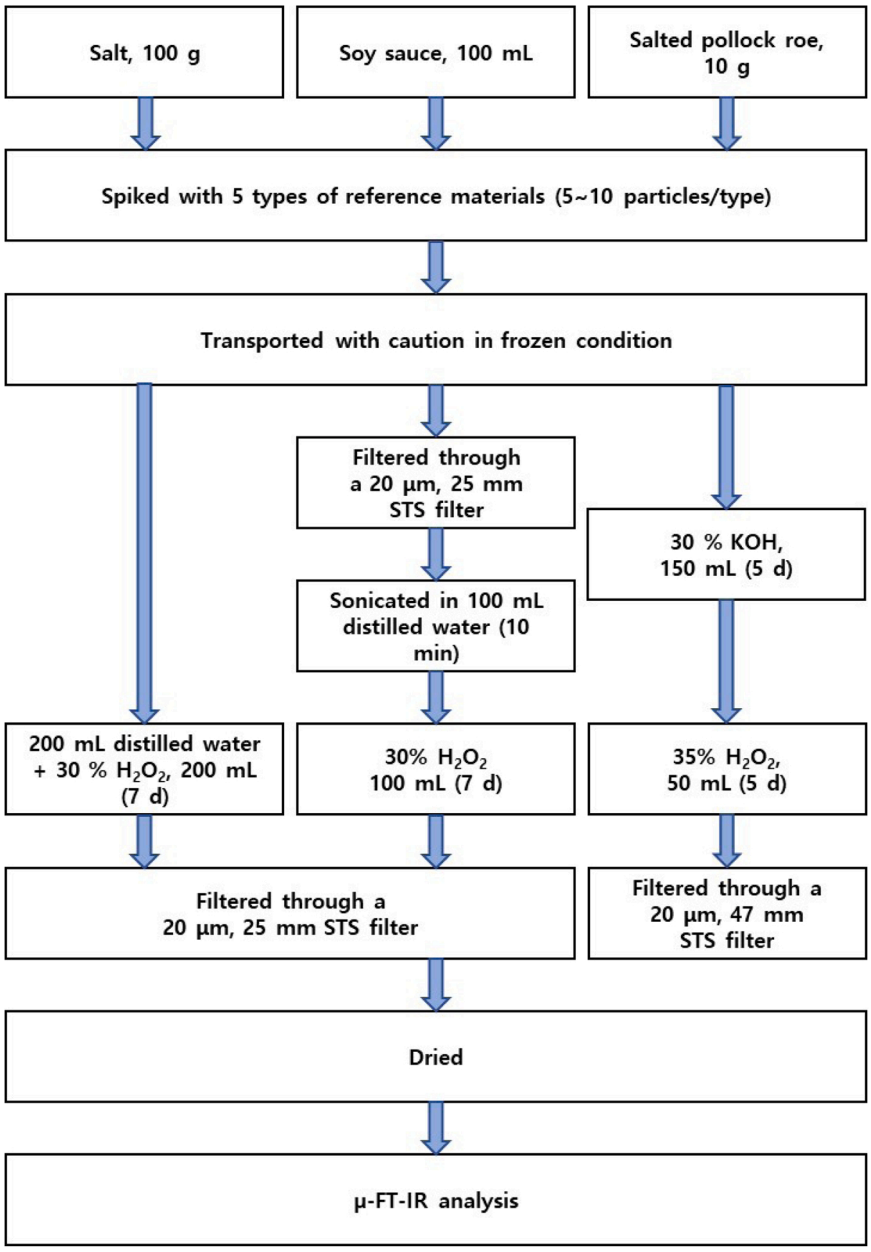

Fig. 2. Flow chart of the microplastic pretreatment method validation in three food matrixes: salt, soy sauce, salted pollock roe.

flask and poured into a 400 mL beaker, and the reference materials were spiked. Soy sauce samples with reference materials were filtered through a 20 µm (25 mm diameter) stainless steel filter. After filtration, the filter was sonicated in 100 mL distilled water for 10 min. Then, 100 mL of 30% H_2O_2 was added, and the solution was left at room temperature for 7 d. After the solution became clear, it was filtered through a filter used for the primary filtration. The filter was retrieved from the holder assembly and dried in a desiccator for 1 d. The particles on the filter were analyzed using FT-IR.

Salted pollock roe: Ten grams of salted pollock roe was measured and placed in a 1 L beaker, where the reference materials were spiked. After adding 150 mL of 30% KOH, the solution was agitated at 300 rpm for 5 d at room temperature. After alkali digestion, 50 mL of H_2O_2 was added slowly to prevent vigorous reactions, and the solution was agitated at 300 rpm for another 5 d at room temperature. The solution was then further filtered through a 20 µm (47 mm diameter) stainless steel filter. The filter was retrieved from the holder assembly and dried in a desiccator for 1 d. The particles on the filter were analyzed using FT-IR.

3.2. Method validation

The reference materials recovered from the samples did not exhibit any signs of deterioration or change in color in any of the samples. They appeared transparent and maintained their shape initially prepared by visual inspection. The numbers of spiked and recovered reference materials in the pre-validation and validation periods are summarized in Table 2 and S3 (Supplementary Material). The average recovery rates of reference materials in the validation were 76.7% (HDPE), 83.5% (PP), 71.0% (PET), 84.0% (PS), and 74.2%, (PA-6). In some cases, experimenters reported a greater number of reference materials than spiked. Two possible reasons include errors in spectral determination or confusion with microplastics that already exist in the test matrix.

Evidently, we noticed a significant improvement in the recovery rate when comparing the results of the pre-validation and validation stages. As presented in Fig. 3, the standard deviations of the recovery rates were dramatically reduced, although the average values did not change significantly. There are several reasons for this improvement. The

Table 2
Number of reference materials spiked and recovered in the validation test.

Sample type	Laboratory	Replicant	Recovery (Recovered reference material/Spiked reference material)					
			HDPE	PP	PET	PS	PA-6	Total
Salt	A	1	4/5	6/8	7/8	3/10	5/7	25/38
		2	4/5	6/5	5/5	6/9	6/5	27/29
		3	10/10	10/6	8/10	4/5	6/8	38/39
	B	1	10/10	9/10	6/7	6/6	5/6	36/39
		2	7/6	6/5	5/7	6/9	7/8	31/35
		3	4/8	5/8	5/7	5/9	4/7	23/39
	C	1	3/8	6/6	7/8	6/9	8/10	30/41
		2	0/9	3/10	3/9	1/10	0/5	7/43
		3	9/9	7/8	7/8	7/8	7/7	37/40
	E	1	4/5	6/9	11/10	5/9	0/5	26/38
		2	6/7	5/8	9/9	8/8	7/8	35/40
		3	4/6	3/6	4/5	4/8	3/5	18/30
Soy sauce	A	1	5/9	12/10	9/9	7/8	2/5	35/41
		2	10/10	9/9	8/8	10/9	7/9	42/43
		3	6/7	6/6	7/7	5/5	2/9	26/34
	B	1	8/10	8/9	6/7	4/8	5/8	31/42
		2	3/6	4/10	2/5	7/9	4/8	20/38
		3	4/6	8/10	7/7	6/7	6/5	31/35
	C	1	8/10	3/5	2/8	5/7	4/10	22/40
		2	7/8	7/6	7/6	7/10	7/5	35/35
		3	5/9	6/9	8/9	7/9	7/9	33/45
	E	1	7/7	6/7	8/10	7/8	3/7	31/39
		2	6/7	8/9	2/5	5/8	6/8	27/37
		3	10/10	5/6	5/8	9/10	8/10	37/44
Salted pollock roe	A	1	7/10	5/9	0/8	7/8	4/10	23/45
		2	8/9	8/8	10/8	9/8	4/5	39/38
		3	5/5	6/5	4/5	8/10	1/10	24/35
	B	1	8/6	0/6	4/6	5/7	6/5	23/30
		2	8/8	5/10	3/9	8/8	6/9	30/44
		3	9/8	8/9	0/8	8/8	9/8	34/41
	C	1	6/5	8/10	4/8	11/9	5/5	34/37
		2	8/8	7/5	8/9	1/1	6/5	36/34
		3	7/5	7/6	6/7	6/5	9/9	35/32
	E	1	8/9	6/6	4/9	6/5	10/9	34/38
		2	10/10	7/6	4/10	12/7	7/8	40/41
		3	5/6	8/9	1/7	16/9	10/7	40/38

increase in the number of spiked reference materials in the samples could have lowered the weighted standard deviation by decreasing the contribution of missing or false-positive detection of reference materials. Improved sample control and delivery may have improved the potential loss of the spiked reference materials. Glass bottles with necks were used in the pre-validation, whereas beakers with flat walls were used in the validation. The absence of bottlenecks may have facilitated the transfer of microplastics from the container. More careful delivery of samples to testing laboratories in the validation could further prevent the potential leakage of liquid or contamination of microplastics. The process of identifying reference materials by the analyst participating in the experiment could also be improved.

Except for the salted pollock roe, the recovery rate was not significantly affected by material type or different laboratories according to two-way ANOVA at $p < 0.05$. The ANOVA result of the recovery rate in salt was $F(3, 12) = 2.03$ and $p = 0.1632$ for laboratories and $F(4, 12) = 1.75$ and $p = 0.2033$ for material types; for soy sauce, it was $F(3, 12) = 1.14$ and $p = 0.3740$ for laboratories and $F(4, 12) = 1.28$ and $p = 0.3300$ for material types; in salted pollock roe, it was $F(3, 12) = 2.03$ and $p = 0.1635$ for laboratories and $F(4, 12) = 3.53$ and $p = 0.0398$ for material type. A statistically significant correlation was observed between the type of reference material and the recovery rate in the salted pollock roe samples. A significantly lower recovery was observed for PET from salted pollock roe. Although further confirmation is required, the lower recovery may be caused by salted pollock roe containing more organic substances, which require a strong alkali solution to decompose them, unlike the two other matrices in this study. Other studies using density separation have also reported that PET exhibits a lower recovery rate than PE, PP, or PS (Hurley et al., 2018; Quinn et al., 2017; Vermeiren et al., 2020). Several studies have claimed that the higher density of PET compared to other thermoplastics is responsible for its lower recovery rate (Quinn et al., 2017). Although the density separation step was not included in this study, the density of the final solution after adding KOH (1.24 g/cm^3) was greater than that of PET. The floating PET in this pretreatment solution may be more difficult to transfer during handling. It is also generally accepted that smaller microplastic particles are more difficult to isolate (Quinn et al., 2017); as such, the slightly smaller size of PET reference materials with regard to other reference materials may explain our results further.

3.3. Occurrence of microplastics in salts, soy sauce, and salted pollock roe

During the method validation, the average number of particles detected in the three replicates excluding reference materials was 169 ± 29, 332 ± 207, and 77.3 ± 37.7 items/100 g salt in laboratories A, B, and C, respectively. In soy sauce samples, they were 32.3 ± 16.1, 78.7 ± 21.2, and 12.3 ± 2.9 items/100 mL in laboratories A, B, and C, respectively. In salted pollock roe, 25.3 ± 11.8, 24.7 ± 11.7, and 39.0 ± 10.4 items/10 g were detected in laboratories A, B, and C, respectively.

Lee et al. (2021) summarized the range of microplastics in sea salts (0–39800 (189 ± 639; median = 7.8) items/100 g) reported in literature, including sea salts from Korea (Kim et al., 2018). The observed level in this study is close to the mean value but greater than the median (Lee et al., 2021) and the range reported by Kim et al. (2018) (9.8–23.2 items/100 g). Studies based on visual inspection tended to report more microplastics than studies based on FT-IR or Raman spectroscopy (Lee et al., 2021). Thus, more microplastics were detected in this study than in previous studies using FT-IR spectroscopy. However, microplastics larger than 20 μm were identified in this study, whereas the majority of

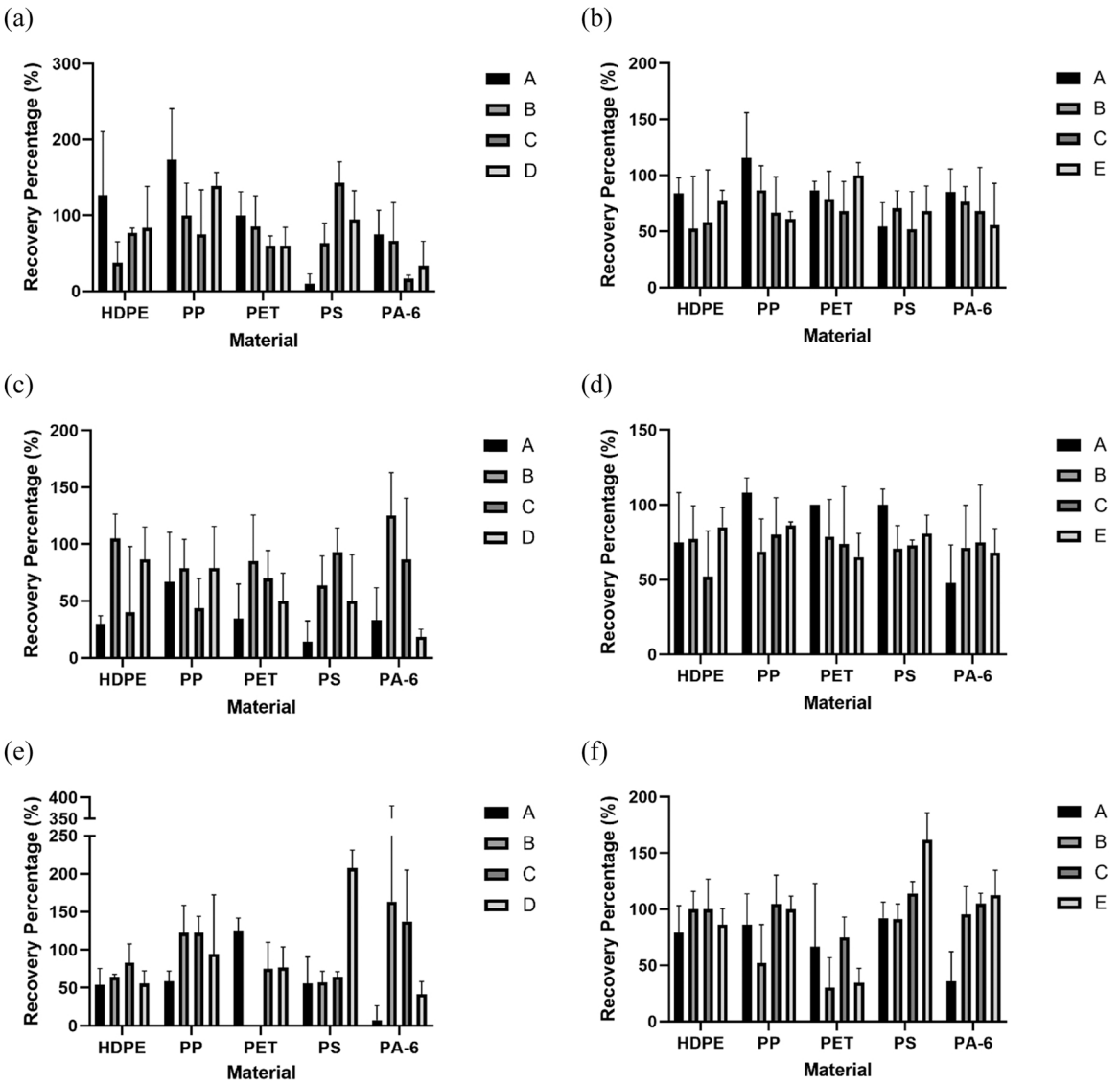

Fig. 3. Percent recoveries of reference materials in the pre-validation test from (a) salt, (c) soy sauce, and (f) salted pollock roe samples, and validation test from (b) salt, (d) soy sauce, and (e) salted pollock roe samples. Bars indicate mean recovery rates and error bars describe weighted standard deviations. **Abbreviations:** HDPE, high-density polyethylene; PA6, polyamide-6; PET, polyethylene terephthalate; PP, polypropylene; PS, polystyrene.

previous studies used larger size cut-offs (Karami et al., 2017; Kim et al., 2018; Lee et al., 2019; Yang et al., 2015). Thus, it is desirable to compare different studies based on the mass concentrations of microplastics.

As this is the first study to report microplastics in soy sauce and pollock roe, it was impossible to compare our results with those in the literature. Larger microplastics (>100 μm) were rarely detected in internal soft tissues of fish (Daniel et al., 2020; Park et al., 2020). The reported high levels of microplastics in salted pollock roe are likely from salts, although further investigation is required. Laboratory C compared the levels of microplastics in salted pollock roe samples after washing with flowing tap water and physically removing external membranes. These treatments dramatically reduced the concentration of microplastics in the salted pollock roe samples (Fig. 4) supporting the hypothesis although only one sample was analyzed after each treatment.

In the validation, the size range of non-reference material microplastics was defined, and their shapes were divided into fibers and fragments based on an aspect ratio cutoff of 3. Laboraboties A and C reported significantly fewer fibers than laboratory B (Fig. S2, Supplementary Material). The most dominant size range reported in laboratories A and C was 45–100 μm (laboratory A: 49%, laboratory C: 56%). Moreover, laboratory B reported that 66% of the identified microplastic particles were between a size range of 20 and 45 μm (Fig. S3,

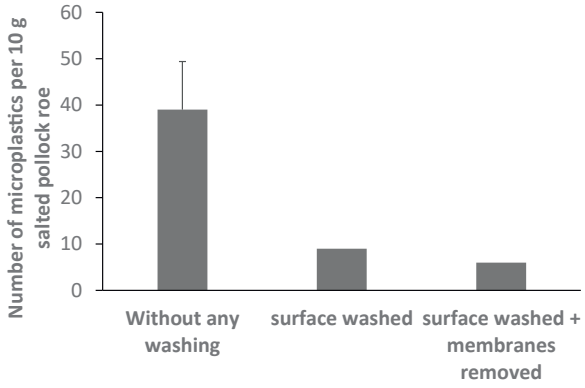

Fig. 4. Changes in the number of microplastics isolated and detected in 10-g salted pollock roe samples after surface washing and physical removal of membranes.

Supplementary Material). The difference among laboratories could be owing to the difference in the model of the FT-IR instruments used for identification. Laboratories A and C used the iN10 model (ThermoFisher Scientific), whereas laboratory B used the LUMOS 2 model (Bruker). However, it is unclear whether the differences originated from variation in samples during preparation or the size measurement algorithm of the software used in the instruments. Further studies are required to compare the performance of commercial instruments with standardized samples.

4. Conclusions

Various pretreatment methods that can be used to isolate microplastics from foods have been evaluated and optimized for salt, soy sauce, and salted pollock roe. There should not be a universal pretreatment method but rather a specific method should be chosen considering the nature of the organic matter interfering with the analysis. The selected pretreatment methods for FT-IR identification of microplastics were found to be highly robust based on the 70–80% recoveries of randomly spiked reference materials of five different microplastic materials in five independent laboratories. Although the isolation and identification methods for microplastics were found to be robust based on the blind recoveries of diverse microplastic reference materials, it is still uncertain whether the selected methods are sufficient for smaller microplastics because the size of the reference materials used in this study was approximately 100 μm. As potential adverse effects of microplastics are expected for smaller particles, the microplastics evaluated in this study would have limited toxicological implications, and further validation is necessary for smaller microplastics.

Funding

This research was supported by a grant (20162MFDS029) from the Ministry of Food and Drug Safety of Korea in 2021.

CRediT authorship contribution statement

Jinwoo Kim: Conceptualization, Methodology, Data curation, Formal analysis, Writing – original draft. **Dat Thanh Pham:** Investigation, Data curation. **Hee-Jin Park:** Investigation, Data curation. **Min-Young Chae:** Data curation. **Sang-Hwa Lee:** Methodology, Investigation, Data curation. **Soonki Hong:** Data curation, Funding acquisition. **Ju-Yang Kim:** Methodology, Investigation, Data curation. **Jaehak Jung:** Methodology, Writing – review & editing, Funding acquisition. **Byung-Tae Lee:** Data curation. **Jung-Hwan Kwon:** Conceptualization, Supervision, Writing – review & editing, Funding acquisition.

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.

Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jfca.2022.104856.

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