

Investigation of Heavy Metal Migration from Food Contact Materials used for Food Delivery Using an Inductively Coupled Plasma–Mass Spectrometer

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ABSTRACT - The surge in food delivery systems during the coronavirus 2019 pandemic necessitated this study of heavy metal migration from food contact materials (FCMs). A total of 104 samples of FCMs, comprising 51 polypropylene (PP), 21 polyethylene (PE), and 32 polystyrene (PS) samples of six different types of FCMs (containers, covers, table utensils, cups, pouches, and wrappers) used for food delivery distributed in Korea, were collected and investigated for migration of three heavy metals (Pb, Cd, and As) using inductively coupled plasma–mass spectrometry (ICP-MS) to determine whether they complied with Korea’s Standards and Specifications for Utensils, Containers, and Packages. Acetic acid (4%, v/v) was used as the food simulant, and tests were performed at 100°C (in harsh conditions) for 30 min. Linearity of Pb, Cd, and As showed acceptable results with a coefficient of determination (R^2) value of 0.9999. Limit of detection (LOD) and limit of quantification (LOQ) of Pb, Cd, and As were 0.001, 0.001, and 0.001 $\mu\text{g/L}$ and 0.002, 0.003, and 0.003 $\mu\text{g/L}$, respectively. Accuracy and precision results complied with the criteria presented in the European Commission Joint Research Centre guidelines. The average concentration of Pb, Cd, and As migration detected in a total of 104 samples was 0.009–0.260 $\mu\text{g/L}$, which was very low compared with the migration specification set in the Standards and Specifications for Utensils, Containers, and Packages. The maximum level of Pb corresponded to 0.23% of the migration limit. There were no samples exceeding the limit. Thus, this study confirmed that the heavy metal contents of FCMs used for delivery food distributed in Korea were safely managed. The data from this study represent an invaluable source for science-based safety management of hazardous heavy metals migrating from FCMs used in the food delivery industry.

Key words: Food contact materials, Polypropylene, Polyethylene, Polystyrene, Heavy metals migration

Recently, food delivery systems have surged owing to the increased number of single-person households and the coronavirus 2019 pandemic. The type of food items delivered has expanded not only to meals but also to desserts, such as ice cream and coffee. The surge in food delivery, which increased by 75% in 2021, also means increased use of food delivery containers¹⁾.

In this study, take-out containers, as well as containers used for food delivery, were included as delivery containers. The types of delivery containers are diverse, including lunch boxes, salad containers, rice containers, soup/sauce containers,

side dish containers, and dessert containers. Materials mainly used for food delivery containers are polypropylene (PP), polyethylene (PE), and polystyrene (PS). These three materials were tested for migration of three heavy metals, including Pb, Cd, and As. PP is a synthetic resin containing 50% or more propylene units in the basic polymer²⁾. PP is glossy with a high thermal resistance up to between 121 and 165°C, moisture resistance, and transparency. It is widely used as a food delivery container, particularly for hot water, as a steamer, porridge container, airtight container, and microwave-safe container, due to its high heat resistance³⁾. PE is a synthetic resin containing 50% or more ethylene units in the basic polymer²⁾. It has a lower heat resistance temperature than PP. Among the many types of PE, the three most common PE grades are low-density polyethylene (LDPE), high-density polyethylene (HDPE), and linear low-density polyethylene (LLDPE). HDPE has high tensile strength, a high melting point, and good rigidity but has the disadvantage of low impact resistance. It is opaque and lacks flexibility, but it is mainly used in high-temperature heated

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food plastic containers due to its high heat resistance temperature^{3,4}). To improve these disadvantages, LDPE began to be developed. Although LDPE has poor heat resistance, it is used for frozen food packaging due to its high cold resistance, and because it is transparent and flexible, it is widely used for fruit packaging film and paper cup inner coating. PS is a synthetic resin containing 50% or more styrene or α -methylstyrene units in the basic polymer⁵). Compared to PP and PE, it is not microwave-safe due to its low heat resistance temperature, but it is used for various purposes due to excellent moldability. It is divided into general-purpose polystyrene (GPPS), high-impact polystyrene (HIPS), and expanded polystyrene (EPS). GPPS is colorless, transparent, hard, and lightweight, so it is mainly used in disposable cups, food storage containers, and jelly containers. However, it has a disadvantage in that it is vulnerable to impact. HIPS is added with rubber elastomer. HIPS is hazy milky white and is used in containers, such as trays of frozen food, ice cream, and yogurt. EPS is weak against impact but has excellent insulation. It is mainly used in instant food containers or cup-type food containers, such as cup noodles³).

Heavy metals refer to metal elements with a specific gravity of 4 or more. Pb, Cd, and As, which were investigated in this study, are relatively toxic to the human body, so they need to be managed. Heavy metal contamination in foods sometimes occurs as a result of leaching from food contact materials (FCMs) which become unintentionally contaminated during manufacture or because heavy metals are added to improve marketability and processability. Pb has good color composition and is easy to process due to its low melting point, corrosion resistance, flexibility, and softness, so it is widely used in tableware, plates, containers, paints and pigments, and cosmetics⁶). Cd has been used in the form of sulfide or selenide as yellow, red, and orange pigments in plastics and various types of paints. Cd stearate has been used as a stabilizer for plastics in the past, and Cd can be detected in glaze and enamel ceramics because of its use as a pigment in certain enamels^{7,8}). As is mainly used to increase corrosion resistance, for the preservation of glass, fiber, paper, and wood, and in antibacterial synthetic resins⁹). The International Agency for Research on Cancer (IARC) classified inorganic Pb as Group 2A (human carcinogen) and organic Pb compounds as Group 3¹⁰), and Cd and inorganic As as Group 1 (carcinogenic substances)^{11,12}). In addition, the Joint Food and Agricultural Organization of the United Nations (FAO)/World Health Organization (WHO) Expert Committee on Food Additives (JECFA) set the provisional tolerable weekly intake of As and Pb at 15 and 25 $\mu\text{g}/\text{kg}$ body weight/week^{13,14}), respectively, but both were withdrawn. In the case of Cd, considering that it has a long biological half-life, the provisional tolerable monthly intake was set at 25 $\mu\text{g}/\text{kg}$ body weight/month in

2010 by the JECFA¹⁵), and subsequently by the Korea Food and Drug Administration in 2011¹⁶).

In Korea, the migration specifications for FCMs are set and managed in the Standards and Specifications for Utensils, Containers, and Packages (Notification 2021-76). The specific migration limit of 1 mg/L is set for Pb from PP, PE, and PS. FCMs used for delivery food are mainly inexpensive and disposable synthetic resins. Human exposure to FCMs can occur at any age, regardless of health status or disease, so special attention is needed. To date, several theses¹⁷⁻¹⁹) have monitored heavy metal migration in FCMs. However, research about heavy metal migration from FCMs used for delivery food is relatively insufficient.

This study was designed to contribute to filling this knowledge gap. Over 100 samples of FCMs (PP, PE, and PS) used for delivery food distributed in Korea were collected and examined for the migration of Pb, Cd, and As by inductively coupled plasma–mass spectrometry (ICP-MS).

Materials and methods

Sample collections

A total of 104 samples of FCMs (PP, PE, and PS) used for delivery food distributed in Korea were purchased through supermarkets, wholesale and retail stores, and online markets. Samples analyzed in this study were mostly domestic products, besides some imported products from China and Malaysia. By type, containers ($n = 61$) were the most common, followed by 15 covers, 13 table utensils (forks, spoons, and knives), 7 cups, 3 pouches, and 5 wrappers. By material, PP was the most common ($n = 51$), followed by 32 PS and 21 PE.

Standards and reagents

The Multi-Element Calibration Standard (10 mg/L each of Pb, Cd, and As) supplied by Perkin Elmer (Waltham, MA, USA) was used for the analysis of Pb, Cd, and As. Acetic acid (ACS reagent grade 99.0%) and nitric acid (69-71%) were from Kanto Chemical Co. (Tokyo, Japan). Distilled water was purified to a resistivity of 18.2 $\text{M}\Omega\text{-cm}$ by a Milli-Q ultrapure water purification system (Millipore Co., Bedford, MA, USA).

Analytical instrument

In this study, the ICP-MS instrument used to analyze Pb, Cd, and As was the NexION 2000B model manufactured by Perkin Elmer. The instrument analytical conditions were as follows: concentric glass nebulizer, glass cyclonic spray chamber, RF power of 1600 W, nebulizer gas flow of 1.0 L/min, auxiliary gas flow of 1.2 L/min, plasma gas flow of 18

L/min, and reaction gas (Ar) flow of 0.45 mL/min. Ions monitored (m/z) were Pb (208), Cd (111), and As (75).

Preparation of standard solutions

To prepare the standard stock solution, 1 mL of Pb, Cd, and As standard (10 mg/L; Multi-Element Calibration Standard) was placed into a 100 mL volumetric flask and was diluted to 100 mL with 4% (v/v) acetic acid. To prepare the standard solution, the standard stock solution was diluted by weight with 4% acetic acid (v/v) to obtain concentrations of Pb, Cd, and As in the range of 0.05-15.0 $\mu\text{g/L}$.

Sample preparation

Test solutions of all samples were prepared according to the Standards and Specifications for Utensils, Containers, and Packages²⁰. Acetic acid (4%, v/v) was used as the food simulant, and if the sample could be filled with liquid, sample is fully filled with food simulant, which is heated at 100°C, and covered with watch-glass and maintained at 100°C. If the sample could not be filled with liquid, dip the sample in the food simulant, which is heated at 100°C at the rate of 2 mL/cm² surface area (calculated by combining surface area of both sides) and then covered with watch-glass. The tests of all samples were performed at 100°C (in harsh conditions) for 30 min.

Method validation

Linearity

A calibration curve was prepared from the analysis results obtained by injecting standard solutions of Pb, Cd, and As prepared at concentrations of 0.05, 0.1, 0.5, 1.0, 5.0, 10.0, and 15.0 $\mu\text{g/L}$ by diluting the standard stock solution with 4% acetic acid into the ICP-MS instrument. The linearity of the calibration curve was confirmed by determining the coefficient of determination (R^2).

Limit of detection (LOD) and limit of quantification (LOQ)

LOD and LOQ were measured by referring to the European Commission Joint Research Centre (JRC) guidelines for performance standards and verification procedures of the analysis method used to manage FCMs²¹. LOD was obtained from the equation of X_{BL} (average concentration of blank

sample)+ $3 \cdot X_{SD}$ (standard deviation of the blank sample) by analyzing 4% acetic acid more than 10 times. LOQ was obtained from the equation of X_{BL} (average concentration of blank sample) + $10 \cdot X_{SD}$ (standard deviation of the blank sample).

Accuracy and precision

Accuracy (recovery) was repeatedly analyzed three times after adding a low, medium, and high concentration standard solution to the sample using 4% acetic acid used as the food simulant. For precision verification, intra-day precision was expressed as the relative standard deviation (RSD%), which is the standard deviation of recovery obtained by repeated analysis three times a day divided by the average value, and inter-day precision was expressed as RSD%, which is the standard deviation of recovery obtained by repeated analysis for 3 days divided by the average value. The suitability for recovery and precision was verified by referring to the JRC guidelines for performance standards and verification procedures of the analysis method used to manage FCMs²¹.

Results and discussion

Method validation

Linearity, LOD, and LOQ

Pb, Cd, and As calibration curves were prepared at concentrations of 0.05, 0.1, 0.5, 1.0, 5.0, 10.0, and 15.0 $\mu\text{g/L}$, respectively. The coefficient of determination (R^2) value showed excellent linearity of 0.9999 or more over the tested concentration range of the Pb, Cd, and As calibration curves (Table 1).

The LOD was 0.001 $\mu\text{g/L}$ for each of the three heavy metals, and the LOQs were 0.002, 0.003, and 0.003 $\mu\text{g/L}$ for Pb, Cd, and As, respectively. Low values for LOD and LOQ were confirmed to be suitable for monitoring Pb, Cd, and As migrated from PP, PE, and PS FCMs used for delivery food (Table 1).

Accuracy and precision

Accuracy and precision results are shown in Table 2. The accuracy (recovery) of the established analysis method for Pb, Cd, and As in PP was 95.71-101.27%, 98.37-100.70%, and 98.49-102.05%, respectively. For precision verification,

Table 1. The results of linearity, slope, intercept, LOD, and LOQ

Substance	Linearity (R^2)	Slope	Intercept	LOD ($\mu\text{g/L}$)	LOQ ($\mu\text{g/L}$)
Pb	0.9999	84615	1726	0.001	0.002
Cd	0.9999	12346	-279	0.001	0.003
As	0.9999	43536	-527	0.001	0.003

Table 2. Accuracy and precision results of heavy metals analysis in PP, PE, and PS by ICP-MS

Materials	Substance	Spiked level ($\mu\text{g/L}$)	Accuracy ¹⁾ (%)	Precision (RSD%)	
				Intra-day	Inter-day
PP	Pb	0.5	101.27±0.79	0.10	0.87
		5.0	96.16±1.21	0.08	1.97
		10.0	95.71±1.21	0.68	1.60
	Cd	0.5	100.70±0.36	0.22	0.37
		5.0	98.37±2.35	0.28	2.63
		10.0	99.45±2.07	0.40	2.14
	As	0.5	102.05±0.53	0.42	0.43
		5.0	98.49±0.44	0.25	0.37
		10.0	99.26±0.43	0.44	0.30
PE	Pb	0.5	101.69±1.12	0.75	0.71
		5.0	93.18±2.30	0.50	2.61
		10.0	94.26±1.28	0.18	1.84
	Cd	0.5	98.66±0.61	0.45	0.75
		5.0	98.80±0.69	0.60	0.54
		10.0	101.32±0.69	0.49	0.63
	As	0.5	100.53±1.69	0.59	2.25
		5.0	100.89±1.79	0.66	2.72
		10.0	101.28±1.27	0.15	1.98
PS	Pb	0.5	106.42±1.32	1.47	0.78
		5.0	102.03±1.47	0.85	1.21
		10.0	96.43±1.49	0.33	1.54
	Cd	0.5	99.69±1.34	1.53	1.47
		5.0	102.17±1.49	1.44	1.77
		10.0	100.78±0.35	0.27	0.42
	As	0.5	103.34±1.41	1.08	1.53
		5.0	107.72±1.45	0.81	1.64
		10.0	103.97±1.63	0.23	2.42

Mean±SD (n=3)

intra-day precision was 0.08-0.68% for Pb, 0.22-0.40% for Cd, and 0.25-0.44% for As, and the corresponding inter-day precision values were 0.87-1.97%, 0.37-2.63%, and 0.30-0.43%, respectively.

Accuracy for Pb, Cd, and As in PE was 93.18-101.69%, 98.66-101.32%, and 100.53-101.28%, respectively. For precision verification, intra-day precision was 0.18-0.75% for Pb, 0.45-0.60% for Cd, and 0.15-0.66% for As, and the corresponding inter-day precision values were 0.71-2.61%, 0.54-0.75%, and 1.98-2.72%, respectively.

Accuracy for Pb, Cd, and As in PS was 96.43-106.42%, 99.69-102.17%, and 103.34-107.72%, respectively. For precision verification, intra-day precision was 0.33-1.47% for Pb, 0.27-1.53% for Cd, and 0.23-1.08% for As, and the

corresponding inter-day precision values were 0.78-1.54%, 0.42-1.77%, and 1.53-2.42%, respectively.

As a result of the validation, the accuracy (recovery) was 93.18-107.72%, satisfying the criteria (average recovery 40-120% at concentrations less than 10 $\mu\text{g/L}$) specified in the JRC guideline. The precision (RSD%) was 0.08-2.72%, satisfying the criteria (0.1 $\mu\text{g/L}$: 63.2%, 1 $\mu\text{g/L}$: 44.8%, 10 $\mu\text{g/L}$: 31.7%) calculated by the Horwitz equation presented in the JRC guideline²¹⁾.

Migration monitoring

Pb migration

The Pb migration results are shown in Fig. 1 and Table 3.

Table 3. Pb, Cd and As concentrations in total samples analyzed by ICP-MS

Materials	Type	Tested No.	Detected No.			Concentration ($\mu\text{g/L}$)			
			Pb	Cd	As	Pb	Cd	As	
PP	Container	36	32	20	22	Mean \pm SD ¹⁾	0.101 \pm 0.102	0.006 \pm 0.011	0.007 \pm 0.010
						Mean \pm SD ²⁾	0.114 \pm 0.101	0.011 \pm 0.013	0.011 \pm 0.010
						Min-Max	N.D. ³⁾ -0.419	N.D.-0.061	N.D.-0.052
	Cover	10	10	6	10	Mean \pm SD ¹⁾	0.282 \pm 0.348	0.003 \pm 0.003	0.013 \pm 0.014
						Mean \pm SD ²⁾	0.282 \pm 0.348	0.005 \pm 0.001	0.013 \pm 0.014
						Min-Max	N.D.-0.999	N.D.-0.006	N.D.-0.049
	Utensil	3	3	2	1	Mean \pm SD ¹⁾	0.874 \pm 1.228	0.017 \pm 0.024	0.001 \pm 0.002
						Mean \pm SD ²⁾	0.874 \pm 1.228	0.025 \pm 0.028	0.004 \pm 0.001
						Min-Max	0.059-2.286	N.D.-0.045	N.D.-0.004
	Wrapper	2	2	0	0	Mean \pm SD ¹⁾	0.007 \pm 0.001	N.D.	N.D.
						Mean \pm SD ²⁾	0.007 \pm 0.001	N.D.	N.D.
						Min-Max	0.006-0.008	N.D.	N.D.
	Total	51	47	28	33	Mean \pm SD ¹⁾	0.178 \pm 0.355	0.006 \pm 0.011	0.007 \pm 0.011
						Mean \pm SD ²⁾	0.194 \pm 0.366	0.010 \pm 0.013	0.012 \pm 0.011
						Min-Max	N.D.-2.286	N.D.-0.061	N.D.-0.052
PE	Container	8	8	3	4	Mean \pm SD ¹⁾	0.114 \pm 0.134	0.005 \pm 0.007	0.003 \pm 0.003
						Mean \pm SD ²⁾	0.114 \pm 0.134	0.013 \pm 0.004	0.006 \pm 0.001
						Min-Max	0.012-0.430	N.D.-0.016	N.D.-0.007
	Cup	7	6	4	2	Mean \pm SD ¹⁾	0.209 \pm 0.191	0.006 \pm 0.006	0.002 \pm 0.003
						Mean \pm SD ²⁾	0.244 \pm 0.183	0.011 \pm 0.003	0.006 \pm 0.003
						Min-Max	N.D.-0.412	N.D.-0.013	N.D.-0.008
	Pouch	3	3	2	0	Mean \pm SD ¹⁾	0.059 \pm 0.022	0.003 \pm 0.003	N.D.
						Mean \pm SD ²⁾	0.059 \pm 0.022	0.005 \pm 0.001	N.D.
						Min-Max	0.040-0.082	N.D.-0.005	N.D.
	Wrapper	3	3	2	3	Mean \pm SD ¹⁾	0.243 \pm 0.127	0.006 \pm 0.005	0.034 \pm 0.015
						Mean \pm SD ²⁾	0.243 \pm 0.127	0.009 \pm 0.003	0.034 \pm 0.015
						Min-Max	0.128-0.380	N.D.-0.011	0.024-0.051
	Total	21	20	11	9	Mean \pm SD ¹⁾	0.156 \pm 0.152	0.005 \pm 0.006	0.007 \pm 0.013
						Mean \pm SD ²⁾	0.164 \pm 0.152	0.010 \pm 0.004	0.015 \pm 0.016
						Min-Max	N.D.-0.430	N.D.-0.016	N.D.-0.051
PS	Container	17	16	8	15	Mean \pm SD ¹⁾	0.100 \pm 0.109	0.004 \pm 0.006	0.010 \pm 0.008
						Mean \pm SD ²⁾	0.107 \pm 0.109	0.009 \pm 0.006	0.011 \pm 0.008
						Min-Max	N.D.-0.374	N.D.-0.021	N.D.-0.033
	Cover	5	5	3	5	Mean \pm SD ¹⁾	0.246 \pm 0.219	0.006 \pm 0.007	0.016 \pm 0.011
						Mean \pm SD ²⁾	0.246 \pm 0.219	0.010 \pm 0.007	0.016 \pm 0.011
						Min-Max	0.063-0.596	N.D.-0.018	0.004-0.033
	Utensil	10	10	5	10	Mean \pm SD ¹⁾	0.512 \pm 0.592	0.004 \pm 0.006	0.025 \pm 0.018
						Mean \pm SD ²⁾	0.512 \pm 0.592	0.009 \pm 0.004	0.025 \pm 0.018
						Min-Max	0.093-1.683	N.D.-0.015	0.004-0.056
	Total	32	31	16	30	Mean \pm SD ¹⁾	0.252 \pm 0.385	0.004 \pm 0.006	0.015 \pm 0.014
						Mean \pm SD ²⁾	0.260 \pm 0.389	0.009 \pm 0.005	0.017 \pm 0.014
						Min-Max	N.D.-1.683	N.D.-0.021	N.D.-0.056
	Total	104	98	55	72	Mean \pm SD ¹⁾	0.197 \pm 0.334	0.005 \pm 0.009	0.010 \pm 0.013
						Mean \pm SD ²⁾	0.209 \pm 0.341	0.010 \pm 0.010	0.014 \pm 0.013
						Min-Max	N.D.-2.286	N.D.-0.061	N.D.-0.056

¹⁾ Average and standard deviation of all samples²⁾ Average and standard deviation of detected samples³⁾ Not detected (below LOQ)

Pb was detected in 98 of the 104 samples. On average, the detected Pb migration was $0.209 \pm 0.341 \mu\text{g/L}$, and the Pb migration of all samples was $0.197 \pm 0.334 \mu\text{g/L}$ on average. By material, the average concentrations of Pb migration detected in PP, PE, and PS samples were 0.194 ± 0.366 , 0.164 ± 0.152 , and $0.260 \pm 0.389 \mu\text{g/L}$, respectively. The average concentration of Pb migration was the highest in PS FCMs. Among the types of FCMs, a spoon within the PP samples presented the maximum Pb migration, which was $2.286 \mu\text{g/L}$.

Cd migration

The Cd migration results are shown in Fig. 2 and Table 3. Cd was detected in 55 of the 104 samples. On average, the detected Cd migration was $0.010 \pm 0.010 \mu\text{g/L}$, and the Cd migration of all samples was $0.005 \pm 0.009 \mu\text{g/L}$ on average. By material, the average concentrations of Cd migration detected in PP, PE, and PS samples were similar, showing 0.010 ± 0.013 , 0.010 ± 0.004 , and $0.009 \pm 0.005 \mu\text{g/L}$, respectively. A container within the PP samples presented the maximum Cd migration, which was $0.061 \mu\text{g/L}$. There

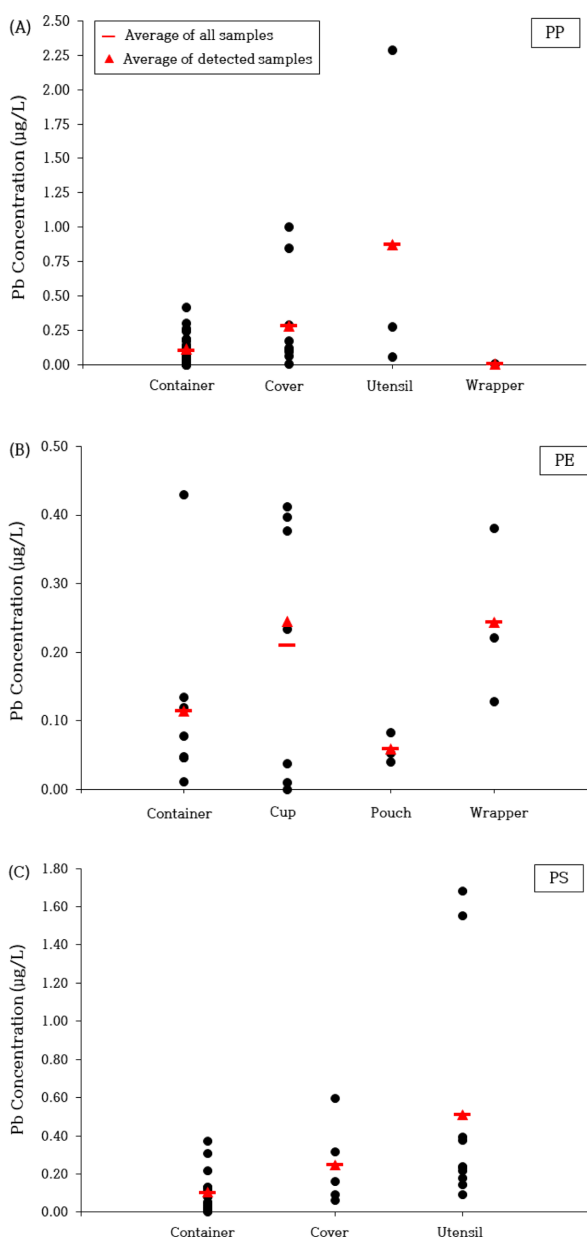


Fig. 1. Scatter plots of migration of Pb in PP, PE, and PS samples. (A) PP, (B) PE, and (C) PS.

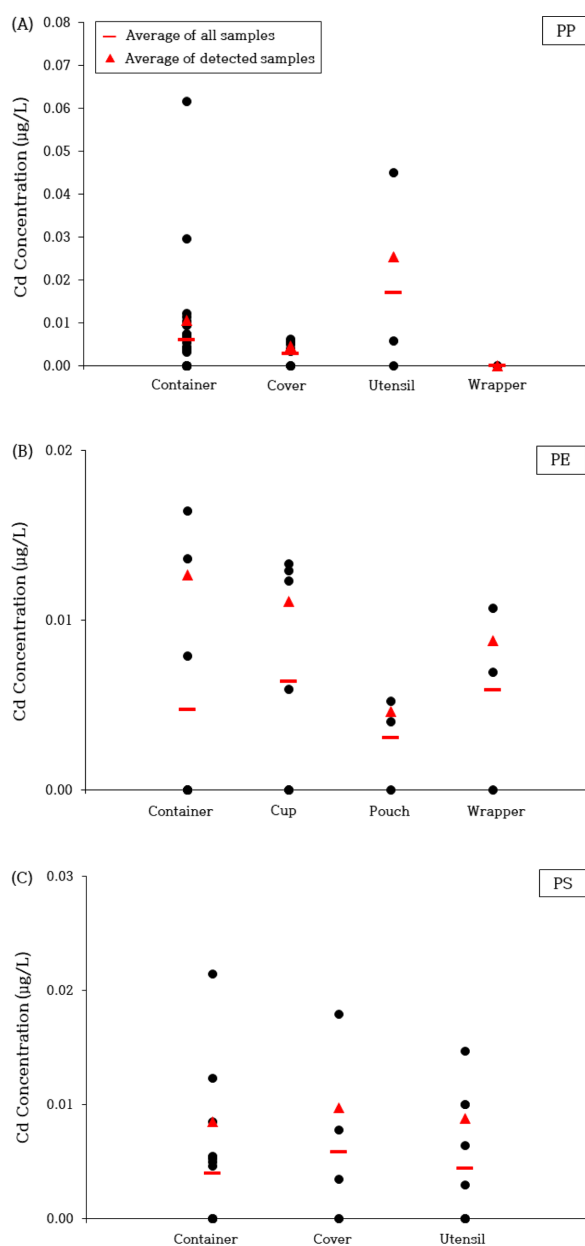


Fig. 2. Scatter plots of migration of Cd in PP, PE, and PS samples. (A) PP, (B) PE, and (C) PS.

were 49 samples in which Cd was measured below the LOQ, and Cd migration among the total samples was very low.

As migration

The As migration results are shown in Fig. 3 and Table 3. As was detected in 72 of the 104 samples, On average, the detected arsenic migration was $0.014 \pm 0.013 \mu\text{g/L}$, and arsenic migration of all samples was $0.010 \pm 0.013 \mu\text{g/L}$ on average. By material, the average concentrations of As

migration detected in PP, PE, and PS samples were similar, presenting 0.012 ± 0.011 , 0.015 ± 0.016 , and $0.017 \pm 0.014 \mu\text{g/L}$, respectively. A fork among the utensils within the PS samples recorded the maximum As migration, which was $0.056 \mu\text{g/L}$. There were 32 samples in which As was measured below the LOQ, and As migration among the total samples was very low.

Distribution of heavy metal concentrations in FCMs

The frequency histogram of the heavy metals (Pb, Cd, and As) in PP, PE, and PS FCMs used for delivery food is shown in Fig. 4. Samples with heavy metal concentrations measured below the LOQ (N.D.) had the highest distribution of Cd and As, and samples with heavy metal concentrations in the range of LOQ-0.1 $\mu\text{g/L}$ had the highest distribution of Pb.

Although Pb showed the highest migration among the three heavy metals analyzed, the amount detected was very low compared to the migration specification of $1000 \mu\text{g/L}$ set in the Standards and Specifications for Utensils, Containers, and Packages in Korea, and no samples exceeded the standard. As a result of this study, it was confirmed that the heavy metal contents of FCMs used for delivery food distributed in Korea were safely managed. According to the previous studies on the heavy metal contents of FCMs, in a paper published in 2018¹⁸⁾, a total of 200 samples of FCMs (Polycarbonate) were tested at both 70°C and 100°C for migration of heavy metals, the migration of lead, cadmium, and arsenic performed at 100°C was $0.0003\text{-}1.19 \mu\text{g/L}$ on average, and the maximum level of Pb corresponded to 0.5% of the migration limit. Additionally, in a paper published in 2018¹⁹⁾, a total of 211 samples of FCMs (Polylactide) were tested at both 70°C and 100°C for migration of heavy metals, and the migration of lead, cadmium, and arsenic performed at 100°C was $0.06\text{-}1.57 \mu\text{g/L}$ on average, and the maximum level of Pb corresponded to 1.0% of the migration limit. In this study, the migration of lead, cadmium, and arsenic was $0.009\text{-}0.260 \mu\text{g/L}$ on average, and the maximum level of Pb corresponded to 0.23% of the migration limit. As a result, the migration of heavy metals in this study was found to be lower than in previous studies. The results from this study represent a valuable source for science-based safety management of hazardous heavy metals migrating from FCMs into delivery food by the food industry.

국문요약

국내 유통되고 있는 배달식품용 폴리프로필렌, 폴리에틸렌 및 폴리스티렌 기구류가 안전하게 관리되고 있는지 파악하기 위해 배달식품용 기구류를 수거하여 중금속 이행

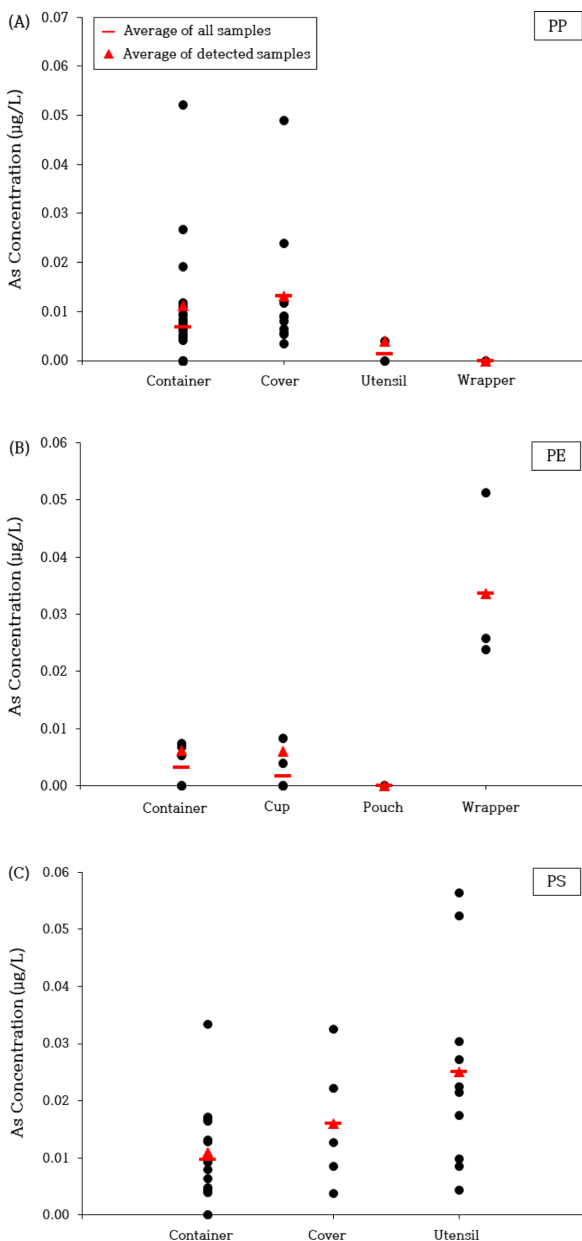


Fig. 3. Scatter plots of migration of As in PP, PE, and PS samples. (A) PP, (B) PE, and (C) PS.

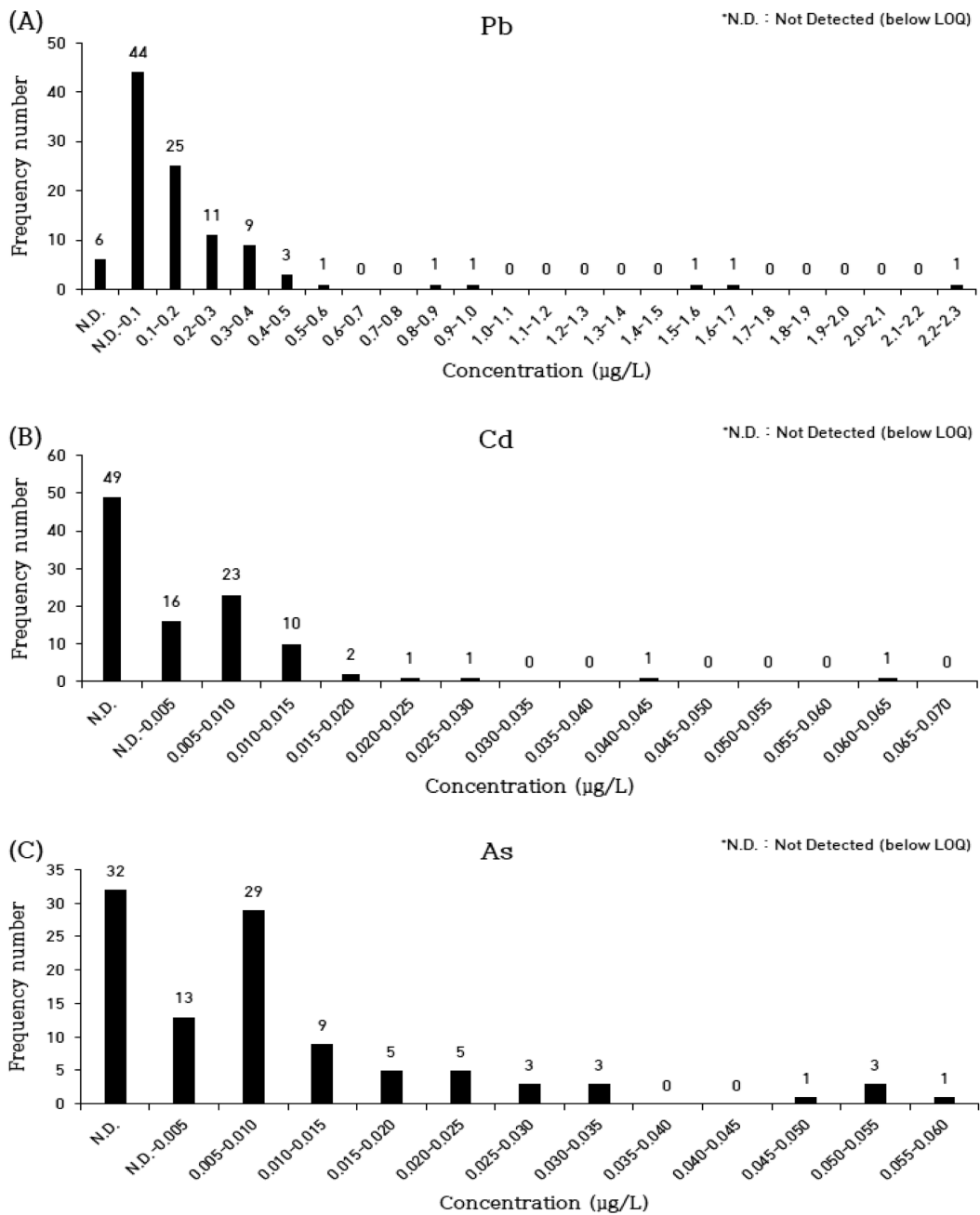


Fig. 4. Frequency distribution histogram of migration in total samples. (A) Pb, (B) Cd, and (C) As.

량(납, 카드뮴 및 비소)을 조사하였다. 모니터링에 사용된 시료는 용기, 뚜껑, 컵, 테이블용 기구, 파우치, 포장재로 총 6개의 품목으로 구성되어 있으며, 폴리프로필렌 51건, 폴리에틸렌 21건, 폴리스티렌 32건으로 총 104건이었다. 「식품용 기구 및 용기·포장 공진」의 시험법에 따라 식품모사용매로는 4% acetic acid를 사용하였고, 총 104건의 시료를 가혹조건인 100°C로 통일하여 용출한 것을 시험용액으로 하여 ICP-MS를 이용하여 납, 카드뮴 및 비소를 분석하였다. 납, 카드뮴 및 비소의 직선성은 결정계수(R^2) 값이 0.9999 이상의 우수한 직선성을 보였으며, 납, 카드뮴

및 비소의 검출한계는 각각 0.001 µg/L, 0.001 µg/L 및 0.001 µg/L이었고, 정량한계는 각각 0.002 µg/L, 0.003 µg/L 및 0.003 µg/L로 나타났다. 또한, 정확성 및 정밀성은 JRC 가이드라인에서 제시하고 있는 기준을 만족하였다. 중금속 이행량 결과를 살펴보면, 총 104건의 시료에서 검출된 납, 카드뮴 및 비소의 이행량은 평균 0.009-0.260 µg/L로 「식품용 기구 및 용기·포장 공진」에서 설정된 용출규격인 1,000 µg/L 대비 매우 낮은 수준이었고, 기준·규격을 초과하는 시료는 없었다. 이를 통하여 현재 유통되고 있는 배달식품용 폴리프로필렌, 폴리에틸렌 및 폴리스티렌 기구

류의 중금속 함량이 안전한 수준으로 관리되고 있음을 확인하였다. 본 연구 결과는 배달식품용 기구류의 중금속 이행량 관리에 대한 과학적 기초자료로 활용될 수 있을 것으로 사료된다.

Conflict of interests

The authors declare no potential conflict of interest.

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References

1. Ministry of Environment. 2021. A study on the standardization and improvement of delivery container reduction, Ministry of Environment, Sejong. Korea, pp. 9-12.
2. Ministry of food and drug safety. 2022. Standards and Specifications for Utensils, Containers and Packages III. Specification for Individual Materials 1-1 Olefin-based Resins e. polyethylene (PE) and f. polypropylene (PP).
3. Ministry of food and drug safety. 2013. About Materials of Plastic, The monthly packaging world, Serial No. 244, Korea Packaging Association, Seoul, Korea, pp. 118-123.
4. Cho, S.J., Kim, A.K., Kwak, J.E., Kim, J.Y., Kim, S.J., Kum, J.Y., Kim, I.Y., Kim, J.H., Chae, Y.Z., Monitoring of 1-hexene and 1-octene in Hygienic Polyethylene-based Packaging, *J. Food Hyg. Saf.*, **26**, 383-387 (2011).
5. Ministry of food and drug safety. 2022. Standards and Specifications for Utensils, Containers and Packages III. Specification for Individual Materials 1-3 Styrene-based Resins e. polystyrene (PS).
6. Ministry of food and drug safety, Risk management team. 2007. What is lead in food? Risk profile in food 1-2, Cheongju, Korea, pp. 3-5.
7. Ministry of food and drug safety, Risk management team. 2007. What is cadmium in food? Risk profile in food 1-14, Cheongju, Korea, pp. 3-8.
8. Mason, R. W., Jones, S. D. J., (1990). In JECFA Monographs 089, Retrieved from <http://inchem.org/documents/pims/chemical/cadmium.htm>
9. The Committee of Experts on Packaging Materials for Food and Pharmaceutical Products. 2013. Metals and alloys used in food contact materials and articles, First Edition. European Directorate for the Quality of Medicines & HealthCare (EDQM). Council of Europe, Strasbourg, France, pp. 119-123.
10. IARC Working Group on the Evaluation of Carcinogenic Risks to Human. 2006. Inorganic and organic lead compounds, Vol 87C, IARC, Lyon, France, pp. 9-378.
11. IARC. 2012. Arsenic, Metals, Fibres and Dusts, IARC monographs on the evaluation of carcinogenic risks to humans, Vol 100C, IARC, Lyon, France, pp. 121-145.
12. IARC. 2012. Arsenic, Metals, Fibres and Dusts. IARC monographs on the evaluation of carcinogenic risks to humans, Vol 100C, IARC, Lyon, France, pp. 41-93.
13. WHO. (2000). Evaluation of certain food additives and contaminants (Fifty-third report of the Joint FAO/WHO Expert Committee on Food Additives), World Health Organization, Accessed http://apps.who.int/iris/bitstream/10665/42378/1/WHO_TRS_896.pdf
14. JECFA. (1998). Arsenic, In JECFA Monographs 658. (WHO Food Additives Series 24), World Health Organization, Geneva, Switzerland, Accessed <http://www.inchem.org/documents/jecfa/jecmono/v024je08.htm>
15. JECFA. 2010. Summary and conclusions of seventy-third meeting (JECFA/73/SC), Geneva, Switzerland, pp. 12.
16. National Institute of Food and Drug Safety Evaluation. (2022, January 27). Human exposure safety criteria. Retrieved from https://www.nifds.go.kr/wpge/m_280/cont_03/cont_03_08_05_03_02.do
17. Choi, J.C., Park, S.J., Goh, H., Lee, J.Y., Eom, M.O., Kim, M.H., A Study on Migration of Heavy Metals from Kitchen Utensils Including, Glassware, Ceramics, Enamel, Earthenware and Plastics, *J. Food Hyg. Saf.*, **29**, 334-339 (2014).
18. Park, S.J., Park, S.R., Kim, M., Choi, J.C., A Study on the Migration of Heavy Metals from Polycarbonate Food Contact Materials Using an Inductively Coupled Plasma Mass Spectrometry (ICP-MS), *J. Korea Soc. Packag. Sci. & Tech.*, **24**, 107-112 (2018).
19. Kim, H., Park, S.Y., Jo, Y.E., Park, Y., Park, S.J., Kim, M., Monitoring of Heavy Metals Migrated from Polylactide (PLA) Food Contact Materials in Korea, *J. Food Hyg. Saf.*, **33**, 102-109 (2018).
20. Ministry of food and drug safety. 2022. Standards and Specifications for Utensils, Containers and Packages IV. Test Method 2-6 Preparation of migration test solution for each material.
21. Bratinova, S., Raffael, B., Simoneau, C. 2009. Guidelines for performance criteria and validation procedures of analytical methods used in controls of food contact materials, JRC Scientific and Technical Reports, EUR 24105 EN-1st edition, European Commission, pp.32-43.