Lee *et al. Appl Biol Chem* (2019) 62:64
https://doi.org/10.1186/s13765-019-0470-0

KSABC
The Korean Society for Applied Biological Chemistry

ARTICLE

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Effects of food processing methods on migration of heavy metals to food

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Abstract

Heavy metals including Lead (Pb), Cadmium (Cd), Arsenic (As) and Aluminium (Al) were analysed in oilseeds, noodles, tea leaves and their processed or cooked products to study the effects of food processing methods on migration of heavy metals. The heavy metals were determined with ICP-MS and ICP-OES following microwave-assisted acid digestion. Heavy metals in oilseeds, noodles and teas were reduced by extracting oils, boiling noodles, and infusing teas. And the transfer of heavy metals into boiling water and infusion tea was increased as the boiling and infusion time is increased. Heavy metals in foods are water soluble and heavy metals in foods would be decreased when foods are processed or cooked with water. Furthermore, it is needed to determine the migration rates in other cooked foods and assess the risk of heavy metals with concentrations calculated by the migration rates.

Keywords: Heavy metal, Migration, Food processing, Oilseed, Noodle, Tea

Introduction

As societies are industrialized, a variety of contaminants cause serious environmental problems. Heavy metals are one of the main contaminants and they are ubiquitous in environment [1]. Heavy metals naturally exist on the earth's crust and they are significantly contaminated to environment by mining and processing metal ore. Human activity such as pesticide and herbicide applications also occur heavy metals in environment [2]. Heavy metals are accumulated in plants from environment such as soils and air, and they are transferred to animals through the food chains. People intake heavy metals by consuming foods which are contaminated with heavy metals [3].

Heavy metals can impact on human health. The U.S. Environmental protection Agency (EPA) has listed 20 hazardous substances in 2001, and As, Pb, Mercury (Hg) and Cd were ranked in the top of the list [4]. Pb is regarded as potential carcinogen and causes a number of serious health problems. Headache, irritability and various symptoms relating to the nervous system can be caused by short-term exposure to Pb, and memory deterioration, prolonged reaction time and reduced ability to understand are some of symptoms of long-term exposure to Pb [5]. As is also considered as carcinogen being related to the lung, kidney, bladder and skin cancer. Hypertension and cardiovascular diseases are strongly related to arsenic exposure [6]. Cd is one of the hazard heavy metals. Cd is first reported as a reason of itai-itai disease from Japan in 1950s. Long-term exposure to high level of Cd causes skeletal damage and it has been classified as a human carcinogen by the IARC [7]. Aluminium (Al) is considered as a strong neurotoxicant, and irritation can be caused by inhalation or injection exposure to Al in animal experiments. Modest evidences for reproductive and bone toxicity exist following oral and injection exposure, respectively [8]. Several studies have shown that Al in drinking water and Alzheimer's disease have significant relations, even though the relation is still on debate [9].

A number of studies have determined heavy metals in different food categories. EFSA published the scientific opinions about the contaminations of Pb, Cd, Al and As in food [10–12]. Especially, oilseeds of plants easily uptake heavy metals from soil and accumulate them, even though different plants have different distributional

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character of heavy metals to organs such as roots, stems, leaves, fruit shell and seeds. Chizzola [13] ascertained that seeds of sunflowers contained high level of Cd, while some studies show that roots and leaves accumulate more heavy metals than seeds [14, 15]. However, edible oils do not contain as high level of heavy metals as seeds [16]. Tea also have heavy metals in their leaves. However, levels of heavy metals in tea infusion were not as high as those in tea leaves [17]. Heavy metal contents in food would be changed during cooking or producing processes. Therefore, in this study, we analyzed the levels of Pb, Cd, As and Al in foods and their cooked and processed foods to determine migration and residual rates of Pb, Cd, As and Al in food according to cooking and processing methods on different conditions. We chose oilseeds, noodles and teas as representative foods to compare the differences of extracting heavy metals with oil and water. There are some research to compare the heavy metal contents in oilseeds and their oils. But they did not consider different extracting methods such as pressing extraction and solvent extraction and different kinds of oilseeds [16]. Teas also have been studied to compare heavy metal contents in teas and their infusions. However, they analyzed only Pb and Cd, and they did not compare the levels of heavy metals according to infusion time [17, 18]. And there are no studies to compare heavy metal contents before and after boiling noodles. We extracted oils from seeds, boiled noodles and infused teas by different methods. And the migration rates occurred based on water solubility of heavy metals were estimated by extracting them with oil and water. Especially, we consider the two cases in which we intake water or remove water after extracting heavy metals with water by analyzing noodles and teas.

Materials and methods

Chemicals and materials

The standards of Pb, Cd, As and Al were purchased from Merck (Darmstadt, Germany) and nitric acid and hydrogen peroxide were of electronic grade from Wako (Osaka, Japan). *n*-hexane from Merck (Darmstadt, Germany) were prepared. The standards of Pb, Cd and As of 1000 mg/kg were diluted to working solutions of 100 μg/kg with HNO_3 (3%). The standard of Al was prepared at the level of 1000 mg/kg, and it was diluted to working solution of 100 mg/kg with HNO_3 (3%). Water purified by a Milli-Q System (Millipore, Bedford, MA, USA) was used and edible oil, peach leaves, spinach leaves and fortified ginseng for certified reference material (CRM) were purchased from FAPAS (TET009RM, York, UK), NIST (1547, Gaithersburg, MD), NIST (1570a, Gaithersburg, MD) and KRISS (Daejun, Korea). Edible oil contained Pb of 0.332 mg/kg, peach leaves contained Pb of 0.869 mg/kg, Cd of 0.0261 mg/kg and Al of 248.0 mg/kg, spinach leaves contained Cd of 2.876 mg/kg, As of 0.068 and Al of 310.0 mg/kg and fortified ginseng contained Pb of 10.749 mg/kg and Cd of 1.178 mg/kg. Argon gas was of spectral purity of 99.9998%.

Samples

Sesame seeds, perilla seeds and flaxseeds were chosen for comparing the levels of heavy metals in seeds and oils and 3 samples of each seed were purchased at Korean offline markets. Sesame oil and perilla oil are the most consumed oils in Korea according to the Korea National Health and Nutrition Examination survey (KNHANES) [19] and flaxseed oil can be highly contaminated by Cd [20]. Sesame and perilla were originated from Korea and flaxseeds were imported from Canada. And flour noodle and sweet potato glass noodle were selected for comparing the heavy metals contents before and after boiling. Flour noodles and glass noodles are highly consumed in Korea by KNHANES [19]. Flour noodle was made from Korean wheat flour, and glass noodle is made from sweet potato starch in Korea, which is widely used in Asian foods. 3 flour noodles and 3 glass noodle were purchased at Korean offline markets. For infusing teas, black, green and Solomon's seal tea were selected. Black, green and Solomon's seal tea would be contaminated with heavy metals because black and green tea are made of the leaves of the plant *Camellia sinensis* and solomon's seal tea is made of the roots of plant, *Polygonatum odoratum var. pluriflorum* Ohwi. Leaves and roots accumulated heavy metals. Black teas were made in Sri Lanka and green and Solomon's teas were made in Korea. 3 black teas and 3 green teas and 3 Solomon's teas were purchased at Korean offline markets.

Cooking and processing methods

Oil extraction

The most widely used methods for extractions of edible oils are mechanical pressing, solvent and supercritical-fluid extraction [21]. The mechanical pressing technique has been the most used method with relatively low costs. However, its efficiency is very low comparing to other two methods [22]. Solvent extraction has been used recently in the last century. It has advantage of extraction efficiency (> 99 wt%), while its quality is reduced because of the solvent recovery process [22, 23]. Supercritical-fluid extraction is way to extract oil with supercritical fluids which have gas-like diffusivities but liquid-like densities. Carbon dioxide (CO_2) is the most frequently used for extracting edible oils [24]. We had oils extracted by these three methods in oil-extracting companies.

Pressing extraction

Compression oil extractor, P-16(S) (Poong-jin, Busan, Korea) was used for extracting oil. Flaxseeds of 4840 g, sesame of 5000 g and perilla of 5000 g were fried before pressing at 160 °C for 10 min, 180 °C for 20 min, and 160 °C for 10 min. And then, fried seeds were pressed by 200×10^5 Pa at 70 °C for 15 min. Flaxseed oil of 1670 g, sesame oil of 1530 g and perilla oil of 1528 g were extracted from seeds.

Solvent extraction

n-Hexane was used for solvent extraction with solvent extractor, E-816 HE (Buchi Labortechnik AG, Flawil, Switzerland). Seeds were extracted for 100 min at 165 °C followed by being evaporated for 20 min at 150 °C. It was dried for 10 min at 150 °C. Flaxseeds of 20.32 g, sesame of 11.81 g and perilla of 11.78 g were extracted to oil of 5.72 g, 3.42 g and 3.14 g, respectively.

Supercritical CO_2 extraction

Supercritical extraction was performed with a supercritical-fluid extractor, L-1000 Bar-Pilot Plant (Natex, Ternitz, Austria) equipped with Mass flow meter, independently controlled heating circuits for the extractor and the separators, CO_2 recycling system including unit and computerized control system for 240 min. Extractor and separator run at 400×10^5 Pa at 50 °C and 40×10^5 Pa at 40 °C and CO_2 run at a flow rate of 60 mL/min. The extracts were collected in a vessel attached to a refrigerated bath at −2 °C. Flaxseed oils of 95.7 g, sesame oils of 74.3 g and perilla oils of 75.3 g were extracted from flaxseeds of 250 g, sesame of 200 g and perilla of 200 g, respectively.

Noodle boiling

The noodles of 50 g were boiled in water of 1 L at 100 °C for 3, 5, and 10 min. Noodles were stirred with chopsticks during boiling, and water was removed from noodles after boiling. Figure 1 shows the boiling procedure.

Tea infusion

The teas of 1.2 g were put in tea bags which were made from polyethylene and polypropylene, and tea bags were infused in water of 150 mL at 98 °C for 2, 10, and 30 min. Tea bags were separated from the tea infusion and water was removed from tea bags after infusing. Figure 1 shows the infusing procedure.

Microwave-assisted acid digestion

Following homogenization of samples, 0.20 g of samples were weighed in polytetrafluoroethylene (PTFE) flasks of 20 mL, and the flasks were kept for pre-digestion at 80 °C for 15 min and at 140 °C for 25 min with HNO3 (60%) of 10 mL and H2O2 (30%) of 1 mL. And then, the flasks were placed in a microwave oven, Easy Control-280 (Milestone, Sorisole, Italy) for digestion by increasing temperature from 0 to 80 °C for 5 min, and then digestion temperature were decreased to 50 °C for 3 min. The flasks were finally digested at 180 °C for 20 min following being heated to 180 °C for 15 min. After cooling the flasks, distilled water was added to wash the residue in the flask up to 25 g.

Determination of heavy metals with ICP-MS and ICP-OES

Pb, Cd and As were determined with inductively-coupled plasma (ICP) with mass spectrometry (MS), Dionex ICS-5000+SP (Thermo Fisher, Waltham, MA, USA). It was operated with RF power of 1.3 kW, argon gas flow rates of 15 mL/min for plasma, 0.9 L/min for auxiliary and 1–1.1 L/min for nebulizer. The quadrupole mass analyser ionized Pb, Cd and As in the single ion monitoring mode to analytical masses of m/z 208, m/z 111 and m/z 75, respectively.

A inductively-coupled plasma (ICP) optical emission spectrometer (OES), Optima 8300 (Perkinelmer, Waltham, MA, USA) was used for determining Al. It was equipped with Mirawire nebulizer, BPC3-50R6 (Elemental Scientific, Omaha, NE), cychronic spray chamber and quarts torch, and it was operated with RF power of 1.50 kW, argon gas flow rates of 10.0 L/min for coolant, 0.2 L/min for auxiliary and 0.7 mL/min for nebulizer and wavelength of 396.153 nm. Samples were taken at the flow rate of 1.0 mL/min.

Method validation

The analytical method was verified for quality control with performance parameters: Limit of Detection (LOD), Limit of Quantification (LOQ), linearity of calibration curves, and accuracy including recovery and precision. LOD and LOQ were calculated by multiplying standard deviation by 3 and 10, and a standard deviation was derived from measurements of 10 blank samples. Calibration curves for heavy metals were obtained by computing regression equations with plots pointed by peak areas against concentrations in a range of 0.01 to 20 μg/kg for Pb, Cd and As and 0.1 to 30 mg/kg at different 3 days. The linearity of calibration curve was evaluated by the coefficient of correlation of regression equation. The accuracy was measured by analyzing CRM of edible oil, Ginseng powder and peach leaves three times a day. The recoveries were calculated by comparing the concentrations of CRMs and analyzed concentrations. The repeatability was obtained by calculating standard deviations of results of inter-day tests.

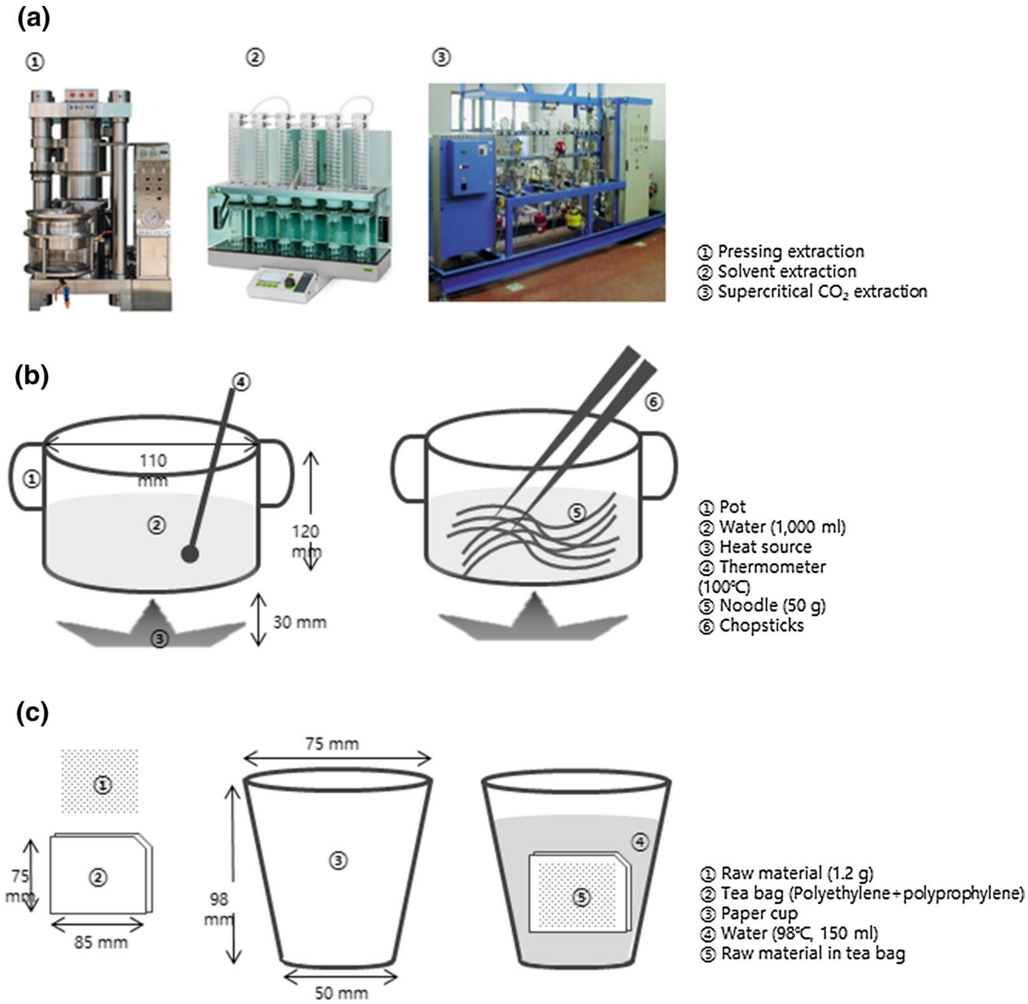

Fig. 1 Designs of cooking methods to study the effects on transfer of heavy metals: **a** oil extraction, **b** noodle boiling and **c** tea infusion

Every sample batch was analyzed with double distilled deionized water for checking the instruments' conditions and CRM was included in sample batch once a month. Instruments were calibrated and maintained every 3 months by instrument maintenance teams.

Statistical analysis

Data was calculated as mean with standard deviation of triplicate measurements. To assess significant differences in levels of heavy metals in the different procedures, the student's t-tests were conducted by using Microsoft Office Excel 2007 (Microsoft Corporation, Redmond, WA, USA). Migration rates and residual rates were calculated by the amount of heavy metals because the concentrations of heavy metals were changed by the amount of food and water and they did not show the exact the migration and residual rates of heavy metals. Extracting, boiling and infusion were carried out three times for each food.

Results and discussion

Method validation

Table 1 shows the performance parameters for verifying the analytical methods. The LODs for Pb, Cd, As and Al were ranged from 0.010 to 0.859 μg/kg and the LOQs were from 0.035 to 2.863 μg/kg. The linear regression analysis ensured that the calibration curves had good linear relationships between peak areas and concentrations with regression coefficients of 0.999. The relative recoveries of Pb, Cd, As and Al from CRM were from 71.5 to 107.5% and relative standard deviations for repeatability were 0.84 to 18.55%. values were

Table 1 Linear equation, limits of detection and quantitation (LOD and LOQ), relative recovery, and precision obtained for heavy metals

Heavy metals	Linear equation	Regression coefficient	Matrix	LOD (μg/kg)	LOQ (μg/kg)	Relative recovery (%)	RSD^r (%)
Pb	y = 1.0032x − 0.0465	0.999	Ginseng powder	0.859	2.863	89.919	2.963
			Edible oil	0.011	0.036	78.071	1.385
			Peach leaves	0.122	0.405	81.452	7.723
Cd	y = 1.0008x − 0.0121	0.999	Spinach leaves	0.123	0.410	71.495	1.995
			Ginseng powder	0.023	0.076	77.300	0.838
			Peach leaves	0.012	0.040	83.250	18.552
As	y = 0.9988x + 0.017	0.999	Spinach leaves	0.010	0.035	77.644	6.580
Al	y = 51,332.1x + 4246.1	0.999	Spinach leaves	0.047	0.158	101.306	5.016
			Peach leaves	0.016	0.054	107.450	2.027

RSD^r repeatability in single-laboratory

acceptable according to criteria recommended by the AOAC [25].

Change of heavy metal contents by extracting oils

The mean amounts of Pb in seed were 0.758 ± 0.152 μg to 936.000 ± 97.083 μg. And contents of Cd were from 0.133 ± 0.038 to 2610.251 ± 429.354 μg. Seeds were contaminated from 0.612 ± 0.086 to 678.788 ± 33.686 μg by As and 0.498 ± 0.003 to 1023.546 ± 187.529 mg by Al (Table 2). Seeds contain wide levels of heavy metals, and Al was highly detected than other heavy metals such as Pb, Cd and As. Different kinds of seeds contain different levels of heavy metals. This is because different plants have different proper to uptake heavy metals [2]. However, the effect of soil contamination could not be estimated without information about the area where the plants grew up. Flaxseeds contain higher level of Cd comparing to Pb and As while sesame contains higher level of As and Perillas contain higher level of Pb. Flaxseeds have some Cd binding proteins and contains high level of Cd [26], and sesame is a strong accumulator of arsenic [27].

Heavy metal contents in extracted oils were much less than those in seeds. The mean amounts of Pb were from 0.019 ± 0.016 to 36.010 ± 53.832 μg, and contents of Cd were from ND to 70.030 ± 103.731 μg. And oils were contaminated from 0.011 ± 0.013 to 15.185 ± 8.418 μg by As and 0.029 ± 0.008 to 29.814 ± 3.463 mg by Al (Table 2). Levels of Pb and As in this study were similar with other studies, in which Pb of 0.056 to 0.072 μg and As of 0.060 to 0.079 μg in sesame oil of 4 g were determined in China [28] and Pb of 0.192 (LB) and 0.288 (UB) μg in vegetable fats and oils of 4 g were detected in EU [29]. Vegetable fats and oils of 4 g were containing As of 0.025 (LB) and 0.135 (UB) μg in EU [11] and Cd of ND to 0.416 μg in fats of vegetable and animal of 4 g [12].

All heavy metals were significantly decreased ($p < 0.05$) in oils by extracting them from seeds (Fig. 2). Pb, Cd, As and Al in seeds were transferred to oils by 1.1 to 32.5%, 0 to 27.3%, 0.9 to 20.0% and 1.7 to 23.9%, respectively. Heavy metals were transferred from seeds to oils less than 10% by pressing and solvent extractions other than Cd in Perilla oils. This would be because heavy metals combine to some minerals and dissolves easily in water. They are up-taken by plants through water. Therefore, heavy metals in seeds are very water-soluble [30]. The transfer rates of heavy metals were up to 30% in supercritical-fluid extraction. Supercritical-fluid extraction extracts more heavy metals rather than other extraction processes. This would be because supercritical-fluid extraction is more effective in extracting components of seeds rather than solvent and pressing extractions. Omega-6-fatty acid and mega-3-fatty acid were more extracted by supercritical-fluid extraction rather than by other extraction processes [21, 31]. Even though supercritical-fluids procedure is one of the most efficient extraction processes in food products [15], it needs to be improved on selectivity not to extract hazardous components such as heavy metals.

Change of heavy metal contents by boiling noodles

Flour noodles of 50 g were containing Pb of 0.820 ± 0.399 μg, Cd of 0.714 ± 0.042 μg, As of 0.055 ± 0.023 μg and Al of 0.768 ± 0.032 mg. And glass noodles of 50 g were contaminated by Pb of 0.634 ± 0.150 μg, As of 0.086 ± 0.022 μg and Al of 0.981 ± 0.005 mg. Cd were not detected in glass noodles (Table 2). Noodles including glass noodles were made from grains or vegetables. Grains and vegetables also uptake heavy metals from environment and accumulate them [2]. Heavy metals would be transferred to noodles from raw materials. Especially, Al is allowed to add to noodles as a leavening agent, acidic sodium aluminum

Table 2 Contents of heavy metals in oilseeds and their extracted oils, noodles and their boiled noodles and teas and their infusions

Method	Food	Status/time	Heavy metal			
			Pb (μg)	Cd (μg)	As (μg)	Al (mg)
Oil extraction						
Pressing	Flaxseed	Seed	373.624 ± 142.869[a]	2610.251 ± 429.354[a]	557.181 ± 259.443[a]	118.702 ± 0.705[a]
		Oil	25.652 ± 36.963[b]	70.030 ± 103.731[b]	3.102 ± 1.036[b]	29.8144 ± 3.463[b]
	Sesame	Seed	321.154 ± 64.165[a]	448.447 ± 44.324[a]	678.788 ± 33.686[a]	849.674 ± 116.487[a]
		Oil	36.010 ± 53.832[b]	0.000 ± 0.000[b]	15.185 ± 8.418[b]	15.974 ± 14.484[b]
	Perilla	Seed	936.000 ± 97.083[a]	56.446 ± 16.043[a]	259.843 ± 36.355[a]	1023.546 ± 187.529[a]
		Oil	35.519 ± 34.428[b]	8.642 ± 14.705[b]	14.109 ± 3.926[b]	25.546 ± 10.514[b]
Solvent	Flaxseed	Seed	1.569 ± 0.600[a]	10.961 ± 1.803[a]	2.340 ± 1.089[a]	0.498 ± 0.003[a]
		Oil	0.168 ± 0.063[b]	1.059 ± 1.314[b]	0.148 ± 0.126[b]	0.029 ± 0.008[b]
	Sesame	Seed	0.758 ± 0.152[a]	1.059 ± 0.105[a]	1.603 ± 0.080[a]	2.007 ± 0.275[a]
		Oil	0.025 ± 0.009[b]	0.000 ± 0.000[b]	0.037 ± 0.052[b]	0.047 ± 0.034[b]
	Perilla	Seed	2.206 ± 0.229[a]	0.133 ± 0.038[a]	0.612 ± 0.086[a]	2.412 ± 0.442[a]
		Oil	0.019 ± 0.016[b]	0.000 ± 0.000[b]	0.011 ± 0.013[b]	0.041 ± 0.033[b]
Supercritical-fluid	Flaxseed	Seed	19.299 ± 7.380[a]	134.827 ± 22.177[a]	28.780 ± 13.401[a]	6.131 ± 0.036[a]
		Oil	6.249 ± 3.989[b]	8.300 ± 3.321[b]	5.716 ± 4.432[b]	0.175 ± 0.113[b]
	Sesame	Seed	12.846 ± 2.567[a]	17.938 ± 1.773[a]	27.152 ± 1.347[a]	33.987 ± 4.659[a]
		Oil	0.140 ± 0.221[b]	0.025 ± 0.031[b]	0.430 ± 0.086[b]	1.232 ± 0.728[b]
	Perilla	Seed	37.440 ± 3.883[a]	2.259 ± 0.642[a]	10.394 ± 1.454[a]	40.942 ± 7.501[a]
		Oil	4.183 ± 3.778[b]	0.574 ± 0.993[b]	0.805 ± 0.330[b]	3.031 ± 1.668[b]
Boiling noodle	Flour noodles	Control	0.820 ± 0.399	0.714 ± 0.042	0.055 ± 0.023	0.768 ± 0.032
		3 min	0.354 ± 0.250[a]	0.173 ± 0.122[a]	0.000 ± 0.000[a]	0.257 ± 0.008[a]
		5 min	0.298 ± 0.190[a]	0.103 ± 0.076[a]	0.002 ± 0.005[a]	0.217 ± 0.017[b]
		10 min	0.251 ± 0.126[a]	0.049 ± 0.009[a]	0.011 ± 0.018[a]	0.203 ± 0.002[b]
	Glass noodles	Control	0.634 ± 0.150	0.000 ± 0.000	0.086 ± 0.022	0.981 ± 0.005
		3 min	0.359 ± 0.132[a]	0.000 ± 0.001	0.002 ± 0.004[a]	0.556 ± 0.004[a]
		5 min	0.289 ± 0.177[a]	0.000 ± 0.000	0.000 ± 0.000[a]	0.476 ± 0.004[b]
		10 min	0.191 ± 0.036[a]	0.000 ± 0.000	0.000 ± 0.000[a]	0.347 ± 0.002[c]
Infusing tea	Green tea	Control	0.219 ± 0.008	0.017 ± 0.002	0.073 ± 0.015	4.694 ± 0.119
		2 min	0.047 ± 0.002[a]	0.003 ± 0.001[a]	0.003 ± 0.005[a]	1.441 ± 0.119[a]
		10 min	0.056 ± 0.001[b]	0.003 ± 0.003[a]	0.006 ± 0.002[b]	1.957 ± 0.147[b]
		30 min	0.065 ± 0.002[c]	0.005 ± 0.003[a]	0.007 ± 0.005[b]	2.462 ± 0.098[c]
	Black tea	Control	0.657 ± 0.061	0.011 ± 0.001	0.049 ± 0.005	1.039 ± 0.039
		2 min	0.330 ± 0.081[a]	0.003 ± 0.003[a]	0.023 ± 0.009[a]	0.663 ± 0.128[a]
		10 min	0.429 ± 0.138[a]	0.006 ± 0.006[b]	0.038 ± 0.017[a]	0.786 ± 0.126[a]
		30 min	0.558 ± 0.194[b]	0.010 ± 0.009[b]	0.049 ± 0.019[b]	0.864 ± 0.054[b]
	Solomon's seal tea	Control	0.474 ± 0.163	0.054 ± 0.008	0.053 ± 0.009	0.428 ± 0.026
		2 min	0.086 ± 0.017[a]	0.004 ± 0.004[a]	0.002 ± 0.003[a]	0.214 ± 0.022[a]
		10 min	0.175 ± 0.121[b]	0.014 ± 0.014[b]	0.009 ± 0.003[b]	0.277 ± 0.041[b]
		30 min	0.298 ± 0.118[c]	0.020 ± 0.004[c]	0.015 ± 0.003[c]	0.374 ± 0.041[c]

Values with different letters in each column are significantly different with p-value (< 0.05)

phosphate. Therefore, noodles can contain high level of Al. Levels of Pb, Cd and As in noodles were less than those in the previous studies. Pb and Cd were found in noodles of 40 g in the range of 46.8 to 66.8 μg and 21.2 to 32.8 μg, respectively [32]. They were also similar with the levels in raw materials such as flour and sweet potatoes. Pb of 3.26 (LB) and 4.00 (UB) μg in starchy roots of 40 g and 0.884 (LB) and 1.580 (UB) μg in cereal products of 40 g were detected in EU [29]. Cereal products excluding rice based products of 40 g were containing As of

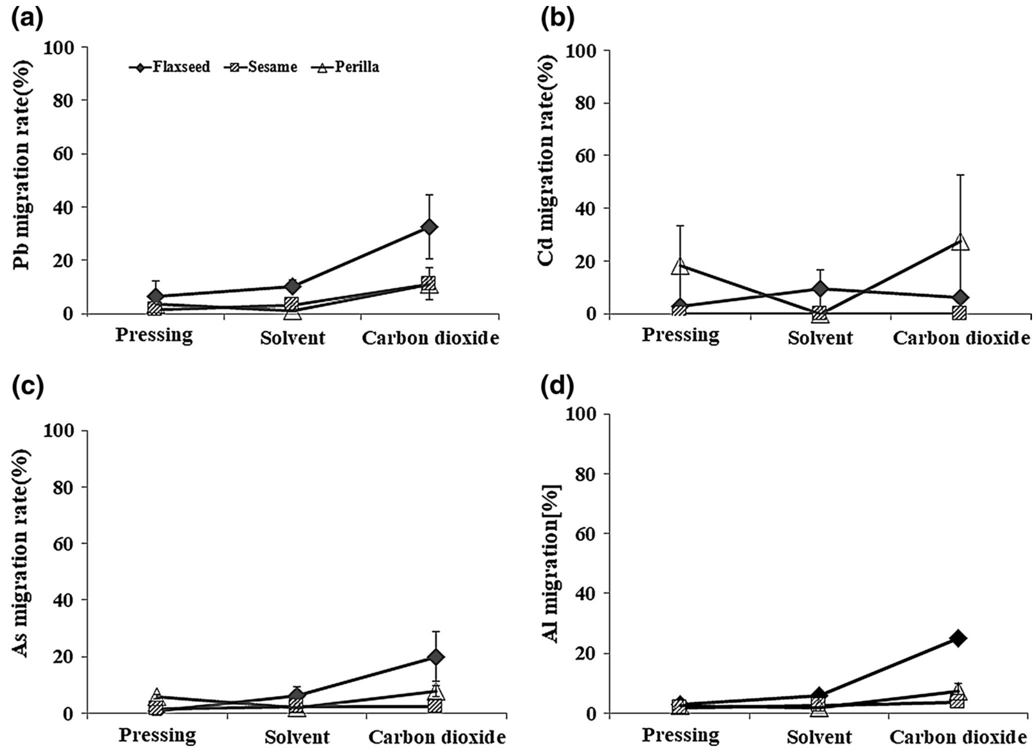

Fig. 2 Migration rates of heavy metals, **a** lead, **b** Cadmium, **c** Arsenic and **d** Aluminium during extracting edible oils from different seeds by different processing methods

0.428 (LB) and 1.188 (UB) μg and starchy roots or potatoes of 40 g were contaminated by As of 0.124 (LB) and 0.568 (UB) μg in EU [11] and wheat and flour contains Cd of 1.200 μg and other starchy roots excluding potatoes contain Cd of 0.632 μg in EU [12]. Stahl showed that the levels of Al in food were ND to 29.480 mg and flour was contaminated with Al in the range from 0.040 to 0.760 mg [33].

Heavy metals were significantly decreased ($p < 0.05$) during boiling noodles for 3 min (Fig. 3). Pb, Cd, and Al in flour noodles were remained by 43.8%, 21.4% and 33.4%, and Pb and Al in glass noodles were decreased by 53.8% and 56.7% during boiling noodles for 3 min. The levels of heavy metals in noodles were not significantly reduced from 3 to 10 min other than Al. This is because heavy metals were transferred to water during boiling noodles and most heavy metals were quickly dissolving in water during boiling. We did not study the effects of the amount of heavy metals, noodles and water. Further studies are needed to figure out factors which effect on the solubility of heavy metals. Al content was significantly decreased as boiling time increased. It would be because Al contents was much higher than other heavy metal contents. It would take more time for Al to dissolve in water rather than Pb, Cd and As.

The risk of heavy metals in noodles would be reduced when noodles are consumed after boiling them and removing the water in which noodles cooked. It would be much realistic risk assessment of heavy metals in noodles when considering the migration rates of heavy metals. In previous study, the risk of Al in instant noodles were of concern with Percentile 95% of target hazard quotient (THQ) of 1.789 for adult [34]. However, the risk of Al in instant noodles would be safe with THQ of 0.597 less than 1 when the residual rate of Al of 33.4% in flour noodle was considered.

Change of heavy metal contents by infusing teas

Black teas of 1.2 g were containing Pb of 0.657 ± 0.061 μg, Cd of 0.011 ± 0.001 μg, As of 0.005 ± 0.005 μg and Al of 1.039 ± 0.039 mg. And green teas of 1.2 g were contaminated by Pb of 0.219 ± 0.008 μg, Cd of 0.017 ± 0.002 μg, As of 0.073 ± 0.015 μg and Al of 4.694 ± 0.119 mg. Solomon's seal teas of 1.2 g showed the contaminations of Pb in 0.474 ± 0.163 μg, Cd in 0.054 ± 0.008 μg, 0.053 ± 0.009 μg and 0.428 ± 0.025 mg. (Table 2). Teas were also contaminated with Al at most, and Pb contents were higher than Cd and As contents. The levels of Pb, Cd and As in teas were similar with the previous studies. Al contents are higher than other heavy metals followed

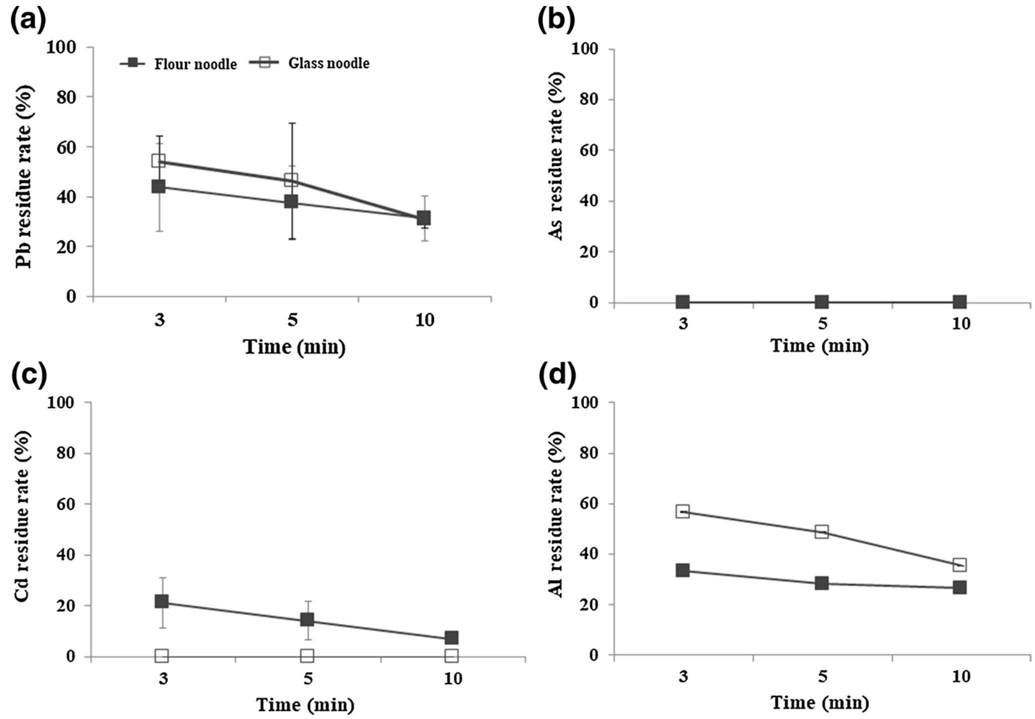

Fig. 3 Migration rates of heavy metals, **a** lead, **b** Cadmium, **c** Arsenic and **d** Aluminium during boiling noodles at different time

by Pb. Karak and Bhagat were reviewed the heavy metal contents in teas. Black teas of 1 g were contaminated by Pb in a range between ND to 240.1 μg, Cd of from ND to 1.92 μg, As of from ND to 4.3 μg and Al of from 0.168 to 2.716 mg [35]. Green teas of 1 g were contaminated by Pb from 0.11 to 3.92 μg, Cd from 0.013 to 0.114 μg, As from ND to 1.66 μg, and Al from 0.211 to 4.074 mg. However, there are no studies to determine heavy metals in Solomon's seal teas.

Heavy metals were significantly decreased in tea infusion after infusing tea for 2 min. Pb was transferred to tea infusion by 50.2% in black tea, 21.3% in green tea and 18.2% in Solomon's seal tea. Cd of 33.3% in black tea, 14.3% in green tea and 6.7% in Solomon's seal tea was transferred to tea infusion. In case of As, the migration rates were 46.3% in black tea, 4.9% in green tea and 4.5% in Solomon's seal tea. Al were transferred to infusion by 63.9% in black tea, 30.7% in green tea and 49.9% in Solomon's seal tea. The migration of heavy metals into tea infusion is metal dependent and heavy metals contents were increased as the infusion time increased from 2 to 10 and 30 min (Fig. 4). Pb and Cd were highly transferred to tea infusion with longer infusion time in other study [18]. Especially, black tea showed the highest migration rates and green tea had the lowest migration rates. Black tea were made from green tea by oxidation which is called fermentation procedure. Heavy metals bio-concentrate to metal chelates in plans and metal chelates would be changed by oxidation in black tea. Therefore, heavy metals in black tea would be easily migrated to infusion [36].

When assessing the risk of heavy metals in teas with analysing them, it would be much realistic to consider the migration rates of heavy metals in teas. Nkansah assessed the risk of heavy metal in black tea including As [37]. The average carcinogenic risk of As through drinking black tea was 1.48×10^{-4} and it was greater than the acceptable limits of 10^{-6}. However, this research was analysing black tea leaves and did not consider migration rate of As during infusing black tea. Therefore, the carcinogenic risk of As would be decreased to 6.83×10^{-5} when the migration rate of As in black tea of 46.2% evaluated in this study were calculated. The risk of As of Puerh tea was assessed in Puer, China. The 95th percentile carcinogenic risk were 1.07×10^{-4} and 1.19×10^{-4} for male, and they were of concern exceeding the acceptable level of 10^{-4} [38]. However, the carcinogenic risks would be reduced to 0 or 4.94×10^{-5} for all and 0 or 5.50×10^{-5} for male with migration rate of As in green tea or black tea. Therefore, the 95th percentile carcinogenic risk would not be concern.

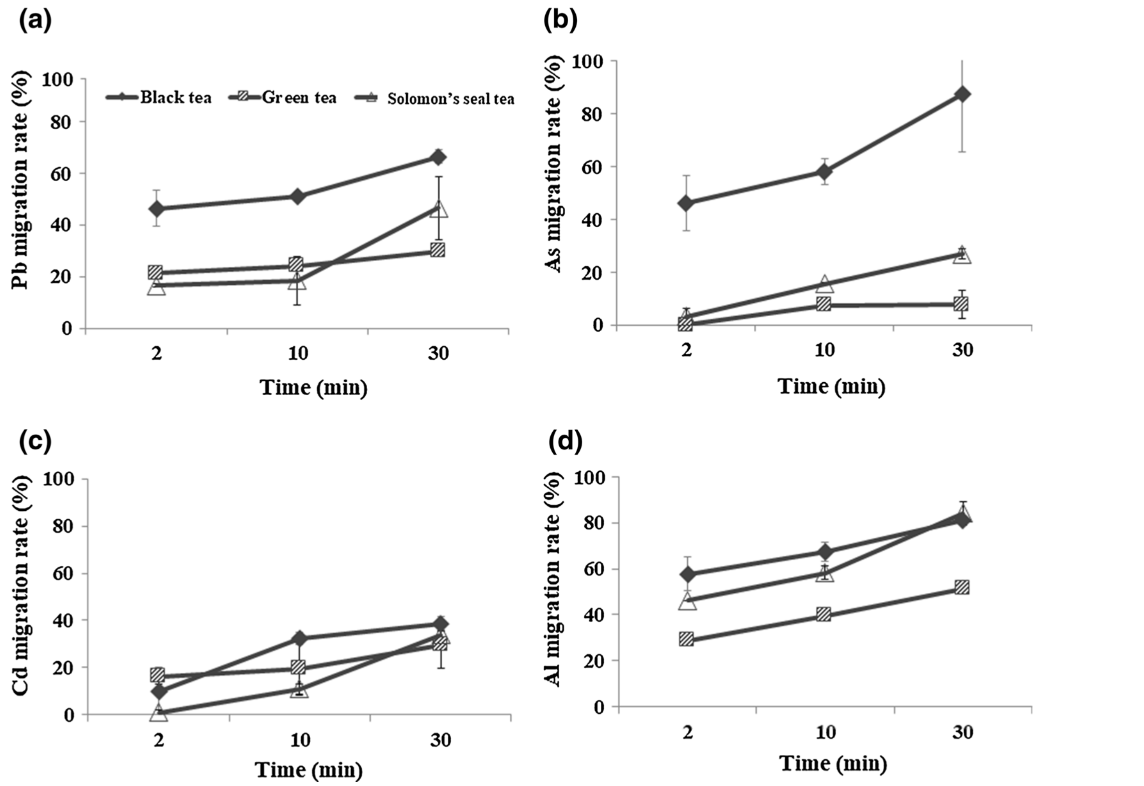

Fig. 4 Migration rates of heavy metals, **a** lead, **b** Cadmium, **c** Arsenic and **d** Aluminium during infusing teas at different time

Acknowledgements

This research was supported by a grant (18161MFDS020) from the Ministry of Food and Drug Safety in 2018.

Authors' contributions

JGL validated analytical method and analysed samples and JYH and HEL statistically analysed data. THK interpreted effects of food processing on contents of PAHs. JDC operated the analytical instruments and analyse data. GJG organized this study and manuscript. All authors read and approved the final manuscript.

Funding

This research was supported by a grant (18161MFDS020) from Ministry of Food and Drug.

Availability of data and materials

All data analysed during this study are included in this published article.

Competing interests

The authors declare that they have no competing interests.

Received: 8 September 2019 Accepted: 16 October 2019
Published online: 04 November 2019

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