

**Research Article** 

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# Reduction of Toxic Heavy Metals in Traditional Asian Herbs By Decoction Preparation

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#### Abstract

Heavy metal contents in traditional Asian herbs need to be monitored closely because of potential health risks of heavy metals at high concentrations. Total amount of toxic heavy metals (lead, copper, cadmium, chromium, mercury and arsenic) in each herb listed in the recipe of Ssanghwatang (one of the most popular herbal drinks in Korea) was 22.6-42.1 mg/kg as determined by Inductively Coupled Plasma - Atomic Emission Spectrometry (ICP-AES). These levels were notable because they were comparable to or above the guideline (30 mg/kg) set by Korean Food and Drug Administration (KFDA). However, herbal tea prepared by decoction preparation (a process of extracting medicinal components from the herbs by boiling them in water) contained only 6.35-12.2 % of the original toxic heavy metals in the herbs. Instead the remainder of toxic heavy metals was found in the herbal residue and the hempen cloth filter. The result suggests that drinking herbal tea is a much safer way of taking beneficial nutrients from the herbs as compared with consuming the whole herbs.

**Keywords:** Herbal tea; Ssanghwatang; Heavy metals; KFDA guideline; Decoction: ICP-AES

## Introduction

The world market size of traditional Asian herbs amount to 200-250 billion US \$ per year, among which 30% is consumed in North America [1,2]. In spite of the medical efficacy of herbal treatment a wider use of herbs has been limited due to public concerns on contamination by heavy metals. Herbs, like other plants, absorb and accumulate heavy metals from soil during growth. Therefore, herbs may contain too high level of heavy metals if they are grown on soil contaminated by heavy metals coming from mining or manufacturing industry, the use of synthetic products containing heavy metals (pesticides, insecticides, paints, and batteries, etc.), and the land application of industrial wastes or domestic sludge [3,4]. In agricultural area pesticides and fertilizers are known to be the main sources of heavy metal pollution [5].

Heavy metals are defined as metals with a density higher 5 g/cm<sup>3</sup>. However, based on the solubility at physiological conditions the number of heavy metals available for living cells and of importance for organism and ecosystem are limited to 17 [6,7]. Among these some heavy metals such as iron (Fe), molybdenum (Mo), and manganese (Mn) are important as micronutrients, but nickel (Ni), copper (Cu), vanadium (V), cobalt (Co), tungsten (W), and chromium (Cr) are toxic at high concentrations. Arsenic (As), mercury (Hg), cadmium (Cd), and lead (Pb) have no known function as nutrients and seem to be toxic to plants and micro-organisms [8,9]. These heavy metals are transported from soil to plants through plant cell wall and membranes, and plants are able to accumulate heavy metals to certain level in their tissues [10,11]. These toxic heavy metals will eventually be transferred to human body through food web [12-14].

When the concentration of heavy metals in human body reaches a certain high level, it will cause various acute and chronic disorders (Table 1) [15]. Chronic exposure to heavy metals was linked to the development of various diseases [16-18]. To resolve health related concerns, we must monitor the level of heavy metals in the herbs before the herbs are applied for treatment. The management and regulation of herb distribution systems have been established and are currently in effect in East Asian countries including South Korea, and the level of heavy metals and pesticide residues are controlled for both domestic and imported traditional Asian herbs. Nonetheless, when the media reports on heavy metal contamination of medicinal herbs, the public becomes anxious and hesitant to select herbal treatment for their illnesses.

In this paper we determined the levels of heavy metals (lead, copper, cadmium, chromium, mercury and arsenic) in nine herbs used for the preparation of Ssanghwatang. Compared with the guideline of Korean Food and Drug Administration [19] the results were noteworthy. When we applied decoction preparation (a process of boiling herbs in water in order to extract medicinal components from the herbs), the level of heavy metals in the herbal decoction (tea) was lowered far below the KFDA guideline. This was because most of the metals in the herbs were left in the residue and hempen cloth filter, but not extracted during decoction process.

# Materials and Methods

### Sources of traditional Asian herbs

Ssanghwatang is one of the most popular traditional herbal drinks available in Korea. In the name "Ssang" means a couple, "hwa" means a harmony, and "tang" means decoction, and it is said to be originally prepared by a court lady for a king after she slept with him at night. As the name stands for, it is known to be effective for both men and

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Heavy Metal	Acute	Chronic	Toxic Concentration		
Lead	Nausea, vomiting, encephalopathy (headache, seizures, ataxia, obtundation)	Encephalopathy, anemia, abdominal pain, ne- phropathy, foot-drop/ wrist-drop	Pediatric: symptoms or [Pb] ≥45 µ/dL (blood); Adult: symptoms or [Pb] ≥70 µ/dL		
Copper	Blue vomitus, GI irritation/ hemorrhage, hemolysis, MODS (ingested); MFF (inhaled)	Vineyard sprayer's lung (inhaled); Wilson disease (hepatic and basal ganglia degeneration)	Normal excretion: 25 μg/24 hours (urine)		
Cadmium	Pneumonitis (oxide fumes)	Proteinuria, lung cancer, osteomalacia	Proteinuria and/or ≥15 µg/g creatinine		
Chromium	GI hemorrhage, hemolysis, acute renal failure (Cr <sup>6+</sup> ingestion)	Pulmonary fibrosis, lung cancer (inhalation)	No clear reference standard		
Mercury	Elemental (inhaled): fever, vomiting, diarrhea, ALI; Inorganic salts (ingestion): caustic gastroenteritis	Nausea, metallic taste, gingivo-stomatitis, tremor, neurasthenia, nephrotic syndrome; hypersensitivity (Pink disease)	Background exposure "normal" limits: 10 µg/L (whole blood); 20 µg/L (24 hours urine		
Arsenic	Nausea, vomiting, "rice-water" diarrhea, encepha- lopathy, MODS, LoQTS, painful neuropathy	Diabetes, hypopigmentation/ hyperkeratosis, cancer: lung, bladder, skin, encephalopathy	24 hours urine: ≥50 µg/L urine, or 100 µg/g creatinine		

MODS, multi-organ dysfunction syndrome; MFF, metal fume fever; GI, gastrointestinal; LoQTS, long QT syndrome and a rare inborn heart condition; ALI, acute lung injury.

Table 1: Typical presentation of poisoning by six heavy metals.

Heavy metals	Wave length (nm)	Estimated detection limit (µg/L)	Alternate wave length (nm)	Calibration concentra- tion (mg/L)	Upper Limit concentra- tion (mg/L)	
Lead (Pb <sup>2+</sup> )	220.35	40	217	10.0	100	
Copper (Cu <sup>2+</sup> )	324.75	6	219.96	1.0	50	
Cadmium (Cd <sup>2+</sup> )	226.50	4	214.44	2.0	50	
Chromium (Cr6+)	267.72	7	206.15	5.0	50	
Mercury (Hg <sup>2+</sup> )	253.70	2	184.91	1.0	50	
Arsenic (As <sup>2+</sup> )	193.70	50	189.04	10.0	100	

Table 2: Operating conditions of inductively coupled plasma - atomic emission spectrometer (ICP-AES).

women in relieving physical fatigue, boosting vigor and energy after a sexual intercourse, and controlling cold sweat.

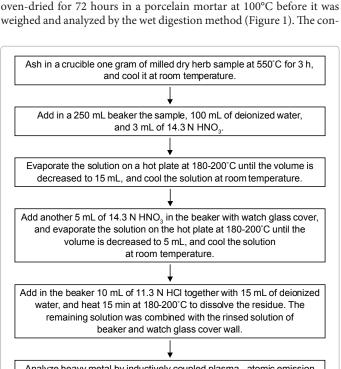
Traditional Asian herbs listed in the recipe of Ssangwhatang [20] were obtained from Oriental Medical Center of Kyung Hee University (Seoul, Republic of Korea), and identified by traditional Asian herbal medicinal experts. The herbs were originated from different locations in East Asia: *Paeoniae Radix Alba* (Dae-Jeon-Si, Republic of Korea), *Zizyphi Fructus* (Yeong-Cheon-Si, Republic of Korea), *Cnidii Rhizoma* (Yeong-Deok-Gun, Republic of Korea), *Rehmanniae Radix Preparata* (China), *Angelicae Gigantis Radix* (Cheong-Yang-Gun, Republic of Korea), *Astragali Radix* (Je-Cheon-Si, Republic of Korea), *Glycyrrhizae Radix* (China), *Cinnamomi Cortex Spissus* (China) and *Zingiberis Rhizoma* Crudus (China).

### Heavy metal analysis

Decoction is a process of boiling herbs in water, and extracting medicinal components from the herbs. When herbs are processed by decoction, we obtain the herbal decoction (tea) together with herbal residue and used hempen cloth filter. Therefore, heavy metals originally present in the herbs will be distributed in the herbal decoction, residue, and hempen cloth filter. The contents of lead (Pb<sup>2+</sup>), copper (Cu<sup>2+</sup>), cadmium (Cd<sup>2+</sup>), chromium (Cr<sup>6+</sup>), mercury (Hg<sup>2+</sup>) and arsenic (As<sup>2+</sup>) in herbal decoction, herbal residue, and hempen cloth filter were determined by inductively coupled plasma - atomic emission spectrometer (ICP-AES) (Shimazu Co., Japan). The decoction experiments were performed in triplicate, and the same sample from each experiment was analyzed twice. The detailed procedure followed literature information [21,22]. Operating conditions of the ICP-AES are shown in Table 2. In the analysis standard solution of heavy metal (1000 mg/L, Showa Co., Japan) was used with appropriated dilution.

### Preparation of herbal decoction

One gram of oven-dried herb was wrapped in one gram of an ovendried hempen cloth filter, and was boiled for 3 hours in a flask with



300 mL of deionized water. The herbal residue and hempen cloth filter

were oven-dried for 24 hours in a porcelain mortar at 100°C before dry weight analysis. The dried herbal residue and hempen cloth filter were

milled separately in a pestle and subsequently digested by acids (HNO<sub>3</sub>

and HCl) in a flask according to the wet digestion method (U.S. E.P.A.

Standard Method 3030 F) [23-26] (Figure 1). The herbal decoction was

Analyze heavy metal by inductively coupled plasma - atomic emission spectrometry (ICP-AES).

Figure 1: Procedure of wet digestion and heavy metal analysis (U.S. E.P.A. Standard Method 3030 F and ICP-AES).

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Heavy metal	Sample	Rehman- niae Radix Preparata	Zizyphi Fructus	Cnidii Rhi- zoma	Cinnamomi Cortex Spis- sus	Astragali Radix	Paeoniae Radix Alba	Glycyrrhizae Radix	Angelicae Gi- gantis Radix	Zingiberis Rhizoma Crudus
Lead (Pb <sup>2+</sup> ) (mg/kg) ±error	Herbal decoc- tion	0.16±0.03	0.30±0.09	0.24±0.06	1.15±0.14	0.69±0.05	0.93±0.03	0.61±0.10	0.11±0.01	0.12±0.03
	Herbal resi- due	3.13±0.14	2.96±0.44	1.42±0.20	2.19±0.09	3.64±0.34	6.53±0.74	7.40±0.56	4.19±0.53	5.96±0.36
	Hempen cloth filter	0.59±0.12	0.42±0.07	0.13±0.22	0.16±0.04	0.23±0.03	0.43±0.01	0.68±0.04	0.11±0.02	1.38±0.17
	Total level	3.88±0.23	3.68±0.42	1.79±0.28	3.49±0.18	4.56±0.33	7.89±0.72	8.69±0.53	4.41±0.52	7.46±0.20
	% in decoc- tion	4.12	8.15	13.4	33.0	15.1	11.8	7.02	2.49	1.61
Copper (Cu <sup>2+</sup> ) (mg/kg) ±error	Herbal decoc- tion	0.23±0.03	0.34±0.10	0.56±0.32	0.51±0.09	0.87±0.07	0.50±0.09	0.59±0.04	0.65±0.06	0.43±0.06
	Herbal resi- due	2.18±0.17	7.27±0.86	6.93±0.59	7.11±1.02	4.11±1.06	7.43±0.27	3.91±0.14	8.24±0.76	2.45±0.13
	Hempen cloth filter	0.58±0.08	0.36±0.14	0.71±0.03	1.42±0.03	1.72±0.39	0.55±0.07	0.71±0.05	0.54±0.06	0.59±0.04
	Total level	2.99±0.10	7.97±0.84	8.20±0.61	9.04±1.09	6.70±1.22	8.48±0.41	5.21±0.22	9.43±0.77	3.47±0.10
	% in decoc- tion	7.69	4.27	6.80	5.64	13.0	5.90	11.3	6.89	12.4
Cadmium (Cd <sup>2+</sup> ) (mg/kg) ±error	Herbal decoc- tion	0.07±0.02	0.05±0.02	0.09±0.02	0.08±0.01	0.08±0.01	0.05±0.01	0.04±0.01	0.10±0.03	0.09±0.03
	Herbal resi- due	1.25±0.12	0.28±0.09	0.55±0.08	0.68±0.18	0.39±0.07	0.84±0.12	0.75±0.04	2.11±0.05	4.64±0.37
	Hempen cloth filter	0.11±0.05	0.11±0.04	0.06±0.01	0.10±0.04	0.25±0.10	0.20±0.11	0.14±0.02	0.07±0.01	0.22±0.04
enor	Total level	1.43±0.10	0.44±0.05	0.70±0.11	0.87±0.20	0.72±0.16	1.09±0.23	0.93±0.04	2.28±0.03	4.95±0.43
	% in decoc- tion	4.90	11.4	12.9	9.20	11.1	4.59	4.30	4.39	1.82
	Herbal decoc- tion	1.09±0.02	1.83±0.17	1.16±0.11	0.94±0.02	0.85±0.04	1.05±0.05	0.51±0.01	0.70±0.05	1.65±0.23
Chromium	Herbal resi- due	6.48±0.20	4.46±0.31	6.10±0.51	5.64±0.31	5.93±0.26	4.00±0.76	6.69±0.29	7.74±0.18	12.7±1.58
(Cr <sup>6+</sup> ) (mg/kg) ±error	Hempen cloth filter	0.45±0.10	0.91±0.37	0.71±0.31	1.48±0.07	1.67±0.22	1.73±0.32	2.19±0.25	1.84±0.32	1.94±0.06
	Total level	8.02±0.12	7.20±0.67	7.97±0.38	8.06±0.27	8.45±0.13	6.78±0.54	9.39±0.13	10.3±0.12	16.3±1.43
	% in decoc- tion	13.6	25.4	14.6	11.7	10.1	15.5	5.43	6.80	10.1
Mercury	Herbal decoc- tion	0.00±0.01	0.14±0.02	0.20±0.03	0.00±0.00	0.17±0.01	0.09±0.01	0.00±0.00	0.21±0.02	0.12±0.03
	Herbal resi- due	1.61±0.07	2.33±0.43	3.21±0.31	1.45±0.17	3.16±0.52	2.27±0.13	3.72±0.15	4.60±0.53	2.11±0.08
Hg²+) mg/kg) :error	Hempen cloth filter	1.49±0.07	0.11±0.02	0.10±0.04	0.16±0.03	0.67±0.05	0.02±0.01	1.34±0.28	0.01±0.01	0.10±0.01
enoi	Total level	3.10±0.14	2.58±0.41	3.51±0.28	1.61±0.20	4.00±0.50	2.38±0.12	5.06±0.13	4.82±0.54	2.33±0.09
	% in decoc- tion	0.00	5.43	5.70	0.00	4.25	3.78	0.00	4.36	5.15
	Herbal decoc- tion	0.42±0.02	0.51±0.03	0.58±0.05	0.34±0.02	0.59±0.01	0.59±0.05	0.38±0.12	0.67±0.07	1.40±0.13
vrsenic	Herbal resi- due	1.29±0.03	3.03±0.07	3.21±0.22	5.45±0.16	3.56±0.11	2.56±0.31	1.86±0.03	6.05±0.06	4.82±0.12
(As <sup>2+</sup> ) (mg/kg) ±error	Hempen cloth filter	1.45±0.21	0.62±0.22	1.39±0.15	1.32±0.16	1.89±0.11	0.87±0.05	0.18±0.03	0.49±0.05	1.44±0.14
	Total level	3.16±0.18	4.16±0.21	5.18±0.29	7.11±0.25	6.04±0.19	4.02±0.37	2.41±0.12	7.21±0.05	7.66±0.29
	% in decoc- tion	13.3	12.3	11.2	4.78	9.77	14.7	15.4	9.29	18.3
Total heavy metals in herbal decoction in mg/kg (%)		1.97 (8.73)	3.17 (12.18)	2.83 (10.35)	3.02 (10.01)	3.25 (10.67)	3.21 (10.48)	2.12 (6.69)	2.44 (6.35)	3.81 (9.04)
Total heavy metals in herbal residue in mg/kg		15.94 (70.59)	20.33 (78.10)	21.42 (78.32)	22.52 (74.62)	20.79 (68.23)	23.63 (77.12)	24.33 (76.77)	32.93 (85.69)	32.68 (77.51)
Total heavy metals in hempen cloth filter in mg/kg		4.67 (20.68)	2.53 (9.72)	3.1 (11.33)	4.64 (15.37)	6.43 (21.10)	3.80 (12.40)	5.24 (16.54)	3.06 (7.96)	5.67 (13.46)
Sum of heavy metals in de- coction, residue and hempen cloth filter in mg/kg		22.58 (100)	26.03 (100)	27.35 (100)	30.18 (100)	30.47 (100)	30.64 (100)	31.69 (100)	38.43 (100)	42.16 (100)

Table 3: Distribution of heavy metals in the herbal decoction (tea), the herbal residue, and the hempen cloth filter.

tents of heavy metals in herbal decoction, herbal residue, and hempen cloth filter were determined by ICP-AES.

#### Heavy metal contents in the unused hempen cloth filter

Because hempen cloth is also from a plant source, we need to determine the initial level of heavy metals in the unused hempen cloth filter. We calculated a net gain of heavy metals in hempen cloth filter by subtracting the initial level from the final level of used hempen cloth filter. The unused hempen cloth filter was oven-dried for 24 hours in a porcelain mortar at 100°C. One gram of oven-dried hempen cloth was milled by a pestle, and followed procedures in Figure 1. The grey white ash was obtained at the completion of the ashing procedure. The ICP-AES was used to analyze the concentration of the heavy metals in the clear digested solution. The initial levels of heavy metals in one gram of dry unused hempen cloth filter were 0.41 mg/kg (lead), 1.03 mg/kg (copper), 0.13 mg/kg (cadmium), 1.74 mg/kg (chromium), 1.38 mg/kg (mercury) and 1.50 mg/kg (arsenic), respectively.

#### Calculation of heavy metal percentage in herbal decoction

Percentage of heavy metal in the herbal decoction was obtained as follows;

Percentage of heavy in the herbal decoction =  $\frac{M(B)}{M(A)} \times 100$  (1) Where,

M (A): The sum of heavy metals in herbal decoction, herbal residue, and a gain of heavy metals in hempen cloth filter

M (B): Mass of heavy metals in herbal decoction

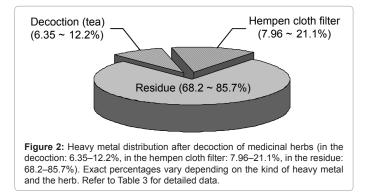
## **Results and Discussion**

### Total heavy metal levels for each traditional Asian herb

Total amount of toxic heavy metals (Pb<sup>2+</sup>, Cu<sup>2+</sup>, Cr<sup>6+</sup>, Hg<sup>2+</sup> and As<sup>2+</sup>) in traditional Asian herbs listed in the recipe of Ssanghwatang varied from 22.6 to 42.1 mg/kg, and the value for each herb was as follows (in mg/kg): *Rehmanniae Radix Preparata* (22.6), *Zizyphi Fructus* (26.0), *Cnidii Rhizoma* (27.4), *Cinnamomi Cortex Spissus* (30.2), *Astragali Radix* (30.5), *Paeoniae Radix Alba* (30.6), *Glycyrrhizae Radix* (31.7), *Angelicae Gigantis Radix* (38.4), and *Zingiberis Rhizoma Crudus* (42.2) (Table 3). This level of heavy metals was close to or above the guideline (30 mg/kg) set by Korean Food and Drug Administration. Especially, the heavy metal contents of *Angelicae Gigantis Radix* (38.4 mg/kg) and *Zingiberis Rhizoma Crudus* (42.2 mg/kg) were 1.28 and 1.40 times higher than the guideline of KFDA.

### Individual heavy metal level in each traditional Asian herb

Heavy metal contents for each herb are presented in Table 3. Lead



(Pb<sup>2+</sup>) content in the herbs ranged from 1.79 (*Cnidii Rhizoma*) to 8.69 mg/kg (*Glycyrrhizae Radix*). Copper (Cu<sup>2+</sup>) content was from 2.99 (*Rehmanniae Radix Preparata*) to 9.43 mg/kg (*Angelicae Gigantis Radix*). Cadmium (Cd<sup>2+</sup>) content varied from 0.44 (*Zizyphi Fructus*) to 4.95 mg/kg (*Zingiberis Rhizoma Crudus*). Chromium (Cr<sup>6+</sup>) content ranged from 6.78 (*Paeoniae Radix Alba*) to 16.3 mg/kg (*Zingiberis Rhizoma Crudus*). The level of mercury (Hg<sup>2+</sup>) was between 1.61 (*Cinnamomi Cortex Spissus*) and 5.06 mg/kg (*Glycyrrhizae Radix*). Arsenic (As<sup>2+</sup>) content ranged from 2.41 (*Glycyrrhizae Radix*) to 7.66 mg/kg (*Zingiberis Rhizoma Crudus*).

In the case of *Zingiberis Rhizoma Crudus* the levels of cadmium (4.95 mg/kg) and chromium (16.3 mg/kg) were noticeably higher compared with other herbs. This result concurs with a literature report that cadmium and chromium were easily taken up by root of plants and transported to different plant parts [5]. Total heavy metal level was also highest for *Zingiberis Rhizoma Crudus* probably because this root type of herb was directly influenced by soil pollution [3].

#### Heavy metal levels in each herbal decoction

Herbal decoction (tea) can be prepared from traditional Asian herbs by decoction process: wrapping herbs in hempen cloth and steeping them for a few hours in boiling water [20, 27-29]. Unlike the unprocessed herb the level of heavy metals in the herbal decoction was much less than the guidelines set by KFDA. The percentage (contents) of heavy metals (Pb2+, Cu2+, Cd2+, Cr6+, Hg2+ and As2+) in the herbal decoction (tea) as defined by equation (1) was 8.73% (1.97 mg/ kg) of the total contents for Rehmanniae Radix Preparata. And the percentage (contents) for other herbs were 12.2% (3.17 mg/kg) for Zizyphi Fructus, 10.4% (2.83 mg/kg) for Cnidii Rhizoma, 10.0% (3.02 mg/kg) for Cinnamomi Cortex Spissus, 10.7% (3.25 mg/kg) for Astragali Radix, 10.5% (3.21 mg/kg) for Paeoniae Radix Alba, 6.69% (2.12 mg/kg) for Glycyrrhizae Radix, 6.35% (2.44 mg/kg) for Angelicae Gigantis Radix, and 9.04% (3.81 mg/kg) for Zingiberis Rhizoma Crudus (Table 3). Our results indicate that decoction preparation leaves most of (87.8-93.7%) of heavy metals in the residue and hempen cloth, and the problem of heavy metals in traditional Asian herbs can be solved efficiently by applying decoction preparation. Also our results suggest that in order to prepare powder form of herbal medicine we better use decoction process followed by drying rather than washing procedure. In our previous work washing procedure removed 17.8-36.7% of heavy metals from the herbs [30], which was much lower than the decoction process (87.8-93.7%). Decoction process may increase the cost of powder preparation due to a large evaporation load during drying process. However, preparation of healthier herbal medicine (tea) with less heavy metal content is definitely a more important issue to be addressed.

# Conclusion

Total amount of toxic heavy metals (Pb<sup>2+</sup>, Cu<sup>2+</sup>, Cd<sup>2+</sup>, Cr<sup>6+</sup>, Hg<sup>2+</sup> and As<sup>2+</sup>) in each traditional Asian herbs listed in the recipe of Ssanghwatang are notable because they are comparable to or above the guideline (30 mg/kg) set by Korean Food and Drug Administration (KFDA). However, after decoction process most of the heavy metals were present in the herbal residue and used hempen cloth filter, and the percentage of heavy metals in the herbal decoction (tea) was only 6.35-12.2% of the total amount, which was far below the guideline set by KFDA. This result suggests that public concerns on the level of heavy metals in traditional Asian herbs can be resolved efficiently by applying decoction process (a process of extracting medicinal components from the herbs by boiling them in water).

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