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Quality assessment of trace Cd and Pb contaminants in Thai herbal medicines using ultrasound-assisted digestion prior to flame atomic absorption spectrometry

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ABSTRACT

A simple, efficient, and reliable ultrasound-assisted digestion (UAD) procedure was used for sample preparation prior to quantitative determination of trace Cd and Pb contaminants in herbal medicines using flame atomic absorption spectrometry. The parameters influencing UAD such as the solvent system, sample mass, presonication time, sonication time, and digestion temperature were evaluated. The efficiency of the proposed UAD procedure was evaluated by comparing with conventional acid digestion (CAD) procedure. Under the optimum conditions, linear calibration graphs in a range of 2–250 μg/L for Cd, and 50–1000 μg/L for Pb were obtained with detection limits of 0.56 μg/L and 10.7 μg/L for Cd and Pb, respectively. The limit of quantification for Cd and Pb were 1.87 μg/L and 40.3 μg/L, respectively. The repeatability for analysis of 10 μg/L for Cd and 100 μg/L for Pb was 2.3% and 2.6%, respectively. The accuracy of the proposed method was evaluated by rice flour certified reference materials. The proposed method was successfully applied for analysis of trace Cd and Pb in samples of various types of medicinal plant and traditional medicine consumed in Thailand. Most herbal medicine samples were not contaminated with Cd or Pb. The contaminant levels for both metals were still lower than the maximum permissible levels of elements in medicinal plant materials and finished herbal products sets by the Ministry of Public Health of Thailand. The exception was the high level of Cd contamination found in two samples of processed medicinal plants.

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1. Introduction

Herbal medicines such as herbal materials (raw or processed medicinal plants, e.g., powder and slice) and traditional herbal products (decoctions, tablets, pills, or capsules containing crude herbal materials or crude herbal extracts) [1], are traditionally used in developing countries. Nowadays, interest in natural therapies has also become popular, resulting in the rapidly increasing worldwide consumption of these herbal medicines. Therefore, a critical evaluation of their safety and quality is important. The World Health Organization [2] has established standards for the quality control of medicinal plants including the classification, botanical identification, determination of active principles, and identification of contaminants.

One of the most frequent contaminants likely to be found in herbal materials or herbal products is heavy metals [1,3]. Ingestion of heavy metals through medicines and foods can cause accumulation in organisms, producing serious health hazards such as injury to the kidneys, symptoms of chronic toxicity, renal failure, and liver damage [4]. There are three key mechanisms that have been proposed to explain heavy metal contamination of medicinal plant-based products: (1) contamination during cultivation (e.g., from contaminated soil or atmosphere); (2) inadvertent cross-contamination during processing; and (3) purposeful introduction of heavy metals for alleged medicinal purposes [1,5]. The World Health Organization sets the maximum permissible levels of heavy metals in medicinal herbs for As, Cd, Cu, Hg, Pb, and Zn at 10 mg/kg, 0.3 mg/kg, 20 mg/kg, 1 mg/kg, 10 mg/kg, and 50 mg/kg, respectively [6]. However, in Thailand, the Ministry of Public Health sets the maximum permissible levels of heavy metals in medicinal plant materials and finished herbal products for only As, Cd, and Pb at 4 mg/kg, 0.3 mg/kg, and 10 mg/kg, respectively [7].

Several analytical methods have been reported for trace heavy metals analysis in medicinal plants and its products such as flame atomic absorption spectrometry (FAAS) [8–13], graphite furnace atomic absorption spectrometry [14–18], inductively coupled plasma-optical emission spectrometry [17,19–22], inductively coupled plasma-mass spectrometry [7,23], stripping voltammetry [4], and solid contact ion-selective electrode [24]. Nevertheless, conventional acid digestion of medicinal plant and herbal medicine samples was performed in most of the published studies with tedious preparation steps, long digestion time, and high chemical reagent consumption.

Ultrasound-assisted sample pretreatment approaches (e.g., digestion, dissolution, and extraction) of solid samples have been proved in the context of green analytical chemistry with clean protocol, safety, short operation time (<1 hour), and moderate volume/concentration consumption of solvents and energy [25]. Under ultrasonic irradiation, the dissolution of a solid sample in a liquid phase can be enhanced by mechanical effects from the acoustic cavitation phenomenon, and chemical effects from the formation of free radicals and various other species [26]. Ultrasound-assisted digestion (UAD) protocols have been applied for digestion and determination of heavy metals in various environmental and biological samples [27,28], meat and mussel samples [29,30], multivitamin tablet samples [31], and biodiesel samples [32].

In this study, a simple ultrasound-assisted treatment using an ultrasonic bath was applied for heavy metals determination in herbal medicine samples such as medicinal plants and traditional medicines. The development of a highly efficient analytical approach with short analysis time and less chemical consumption is required for analysis of real samples. The concentrations of Cd and Pb found in the medicinal plant and traditional medicine provided safety information for the database of Thai herbal medicine and for human dietary intake.

2. Materials and methods

2.1. Chemicals and reagents

All chemicals used were of analytical reagent grade unless otherwise stated. Deionized water from a Simplicity 185 (Millipore, Billerica, MA, USA) with resistivity of 18.2 MΩ cm was used throughout the experiments. Metal standards of Cd and Pb (1000 mg/L for AAS, Merck, Darmstadt, Germany) were used for all the experiments. Working standard solutions of Cd and Pb with different concentrations were prepared by appropriately diluting the stock solution. Nitric acid (HNO_3, 65%, extra pure grade) and hydrogen peroxide (H_2O_2, 35%, extra pure grade; QRëC, Auckland, New Zealand) were used for the digestion of the samples.

Accuracy was evaluated using certified reference material (CRM): (1) TRM-F-4001 (elements in glutinous rice powder; Cd 0.69 ± 0.06 mg/kg, Cu 1.5 ± 0.1 mg/kg, Mn 7.8 ± 1.0 mg/kg, and Zn 21.2 ± 1.0 mg/kg) obtained from the National Institute of Metrology, Ministry of Science and Technology, Thailand; and (2) IRMM-804 (rice flour; As 0.049 ± 0.004 mg/kg, Cd 1.61 ± 0.07 mg/kg, Cu 2.74 ± 0.24 mg/kg, Mn 34.2 ± 2.3 mg/kg, Pb 0.42 ± 0.07 mg/kg, and Zn 23.1 ± 1.9 mg/kg) obtained from the Institute for Reference Materials and Measurements, European Commission Joint Research Centre, Geel, Belgium. The materials were dried in an oven at 60°C for 4 hours and stored in a desiccator at room temperature for about 10 days until it reached a constant mass. All glassware and plastic materials used were treated for 24 hours in 10% volume/volume nitric acid and rinsed with deionized water.

2.2. Instrumentation

The FAAS measurements were performed with an Agilent 280FS AA atomic absorption spectrometer (Agilent Technologies, Santa Clara, CA, USA) equipped with a deuterium background corrector. A high intensity UltrAA coded multi-element (Ag/Cd/Pb/Zn) hollow cathode lamp (Agilent Technologies) was used as the radiation source. The detection wavelength for Cd and Pb was 228.8 nm and 217.0 nm, respectively. All instrumental conditions were followed according to the manufacturer's recommendation [33] using air/acetylene flame. UAD was carried out with a high-power ultrasonic cleaning unit (Sonorex digitec DT 255/H; Bandelin electric GmbH & Co. KG, Berlin, Germany) with technical specifications: timer 0–30 minutes, 230 V, 50/60 Hz, 35 kHz, and built-in heater 20–80°C.

2.3. Sample collection

Twenty-three samples of herbal medicines in the form of powder, capsule, and tablet were selected and bought directly from herbal drug stores in Maha Sarakham, Ubon Ratchathani, and Lop Buri Provinces, Thailand between March 2016 and May 2016. The selected herbal medicine samples including 10 of processed medicinal plants (P1–P10) in powdered form. The other 13 traditional medicine samples (M1–M13) were in the form of capsule and tablet. All samples were from domestic cultivated plants and produced in Thailand. After delivery to the laboratory, coarse particles like powders, capsules, or tablets were ground to fine particles using mortar and pestle. The fine powdered form samples were dried in an oven at 60°C for 4 hours and stored in a desiccator at room temperature for about 10 days until it reached a constant mass. The dried samples were then individually packaged in clean polyethylene bags and stored in a desiccator. Each sample was analyzed in triplicate.

2.4. Procedures

2.4.1. UAD

For UAD optimization, different solvent systems [concentrated (conc.) HNO_3, a mixture of conc. HNO_3:H_2O_2, and H_2O_2], sample mass (0.1 g, 0.3 g, and 0.5 g for 3 mL of solvent), sonication time (0 minutes, 5 minutes, 10 minutes, 20 minutes, 30 minutes, and 40 minutes) and digestion temperature (30°C, 50°C, 60°C, 70°C, and 80°C) were tested. To evaluate the efficiency of the process, the results obtained with the UAD procedure were compared with those from a conventional acid digestion (CAD) procedure.

Approximately 0.1 g of all herbal medicine samples were accurately weighed into glass test tubes with a cap (50 mL capacity), and 2.0 mL of 2:1 acid–oxidant mixture (conc. HNO_3:H_2O_2) was added. The glass tube cap was tightly closed. All tubes were allowed to stand for 10 minutes at room temperature, and marked as presonication time. The two-step sequential UAD was then carried out. The tubes were immersed in the ultrasonic water bath and subjected to ultrasonic energy at 35 kHz for 10 minutes, termed as sonication time. The temperature range of the ultrasonic bath was set at 60°C using a built-in heater. After 10 minutes of the first digestion step, 1.0 mL of 2:1 acid–oxidant mixture (conc. HNO_3:H_2O_2) was added and the tubes were digested under the same condition for a further 10 minutes. After sonication, the sample digestion was made up to 10 mL in volumetric flasks with deionized water and then was filtered through filter paper. The final volume was stored in polyethylene bottles at 4°C for analysis. Blanks were also treated in the same manner without samples for each experiment.

2.4.2. CAD

Acid digestion of all samples was prepared by following the wet digestion of plant analysis reference procedures [34]. Approximately 0.5 g of each sample was accurately weighed into a beaker, 10 mL of conc. HNO_3 was then added, and the beaker was covered with a watch glass. The beaker was allowed to stand overnight, and the contents were heated on a hot plate for 1 hour. The digestion solution was then cooled to room temperature, followed by the addition of 1 mL H_2O_2 (35%) and heated on a hot plate for 20 minutes. A further 1 mL H_2O_2 (35%) was added, and heating was continued until the color of the digestion solution became transparent. After digestion, the digestion solution was filtered through filter paper. The final volume was made up to 25 mL in volumetric flasks with deionized water, and stored in polyethylene bottles at 4°C for analysis. Blanks were also treated in the same manner without samples for each experiment.

3. Results and discussion

3.1. Optimization of UAD

The medicinal plant sample (P1) having matched matrices to real samples was used for optimization. The digestion efficiency or percentage recovery of the results provided by UAD variable parameters was evaluated considering the optimum conditions. The parameters influencing the UAD efficiency were optimized within the variation as listed in Table 1. All the results were compared with those obtained by applying the conventional acid digestion.

The effect of solvent systems such as acid (conc. HNO_3), oxidant (H_2O_2), and a acid–oxidant mixture (HNO_3:H_2O_2; 2:1) was investigated in a univariate mode by fixing the other variables at 0.1 g sample, solvent volume 3 mL, presonication time 30 minutes, and temperature 70°C. Significantly higher recoveries of Cd(II) and Pb(II) were obtained from the acid–oxidant mixture (sonication time 30 minutes) than from acid alone (sonication time 50 minutes). Also, we found that H_2O_2 combined with HNO_3 provided a clear solution at short digestion time compared to acid alone. These results indicated that the presence of oxidant can improve the efficiency of the digestion of metal ions from the medicinal plant sample, and this result was consistent with published literature on UAD of biological samples [27].

The mass sample was evaluated in a univariate mode by fixing the other variables at HNO_3:H_2O_2 (2:1) volume 3 mL, presonication time 30 minutes, sonication time 30 minutes, and temperature 70°C. At 30 minutes sonication time, incomplete digestion samples (visual aspect) were obtained when >0.1 g of the sample was used. Therefore, a sample mass of 0.1 g was suitable for UAD under this condition.

The effect of presonication time, the time for treatment of the medicinal plant samples with acid–oxidant mixtures before subjection to the ultrasonic bath for 30 minutes, was evaluated for different time intervals (0–30 minutes). The

Table 1 – Optimum conditions for the ultrasound-assisted digestion of medicinal plant samples.

Digestion parameters	Variation	Optimized value
Solvent system	HNO_3	Series of
	HNO_3:H_2O_2 (2:1)	HNO_3:H_2O_2 (2:1) 2 mL
	H_2O_2	HNO_3:H_2O_2 (2:1) 1 mL
Sample mass	0.1–0.5 g	0.1 g
Presonication tim	0–30 min	10 min
Sonication time	0–40 min	20 min
Digestion temperature	30–80°C	60°C

optimum time was selected at 10 minutes (>85% recovery for Cd and >75% recovery for Pb) because there was no significant effect on the recoveries of both metals studied after 10 minutes.

The influence of sonication time on the digestion efficiency was investigated in a univariate mode at their optimal values. The percentage recoveries obtained from different sonication time of both metals did not differ (Figure 1). However, incomplete digestion samples (visual aspect) were observed and the percentage recoveries of both metals from UAD (<95% for Cd and <77% for Pb) were still lower than CAD (100.5 ± 2.1% for Cd and 92.7 ± 3.0% for Pb). Then, the sequential UAD procedure was tested by adding a series of acid–oxidant mixtures. Three milliliters of HNO_3:H_2O_2 (2:1) was divided into 2 mL and 1 mL for two steps of sonication (10 minutes each). After sonication for 20 minutes, the clear solutions of digest samples were obtained. This sequential UAD procedure provided higher percentage recoveries for Cd (99.1 ± 2.0%) and Pb (91.0 ± 2.4%) than the ordinary method.

The effect of digestion temperature was then evaluated from 30°C to 80°C. The results shown in Figure 2 indicate that the UAD efficiency increased with temperature from 30°C to 50°C, and then reached equilibrium. However, incomplete digestion samples (visual aspect) were observed at 50°C. Therefore, the optimum temperature was selected at 60°C with percentage recoveries of 101.0 ± 2.1% for Cd and 92.6 ± 2.8% for Pb because there was no significant effect on the recoveries of both metals studied after 60°C.

The optimized values for UAD procedure are presented in Table 1. In addition, comparison of the digestion efficiency of both UAD and CAD procedures are summarized in Table 2. According to *t* test at 95% confidence limit, the results obtained from both procedures were in agreement ($t_{critical} = 2.180$, $t_{calculate} = 1.177$ and −0.031 for Cd and Pb, respectively). Satisfactory recoveries between both procedures were obtained at 101.4% and 98.7% for Cd and Pb, respectively. These results indicated that the digestion efficiency of the proposed UAD procedure is comparable to that of the CAD procedure. Moreover, the relative standard deviations (SDs) of the metal concentration obtained after digestion by UAD tended to be lower than for CAD. The proposed UAD procedure provides some advantages over the CAD procedure such as ease of operation, short operation time, low chemical consumption, high precision, and high sample throughput of more than a dozen samples that can be treated simultaneously using only a simple ultrasonic bath with a built-in temperature control.

3.2. Analytical features of the proposed system

The analytical characteristics of the proposed method were investigated. Using the optimum conditions as described above, the standard calibration in the range of 2–250 μg/L for Cd and 50–1000 μg/L for Pb were constructed by plotting the absorbance against concentrations. Under the selected conditions, a linear calibration graph was obtained for Cd and Pb, with the calibration equations $y = (6.1 \times 10^{-4} \pm 7.2 \times 10^{-6})x + (2.8 \times 10^{-4} \pm 2.6 \times 10^{-5})$, $R^2 = 0.9999$ for Cd, and $y = (5.2 \times 10^{-2} \pm 4.3 \times 10^{-3})x - (1.1 \times 10^{-4} \pm 3.7 \times 10^{-5})$, $R^2 = 0.9994$ for Pb, respectively. The limit of detection (LOD) (3σ/s) and limit of quantification (LOQ) (10σ/s) [where σ is SD of digestion blank ($n = 11$) and s is the slope of calibration curve] were obtained at LOD 0.56 μg/L, LOQ 1.87 μg/L for Cd, and LOD 10.7 μg/L, LOQ 40.3 μg/L for Pb, respectively. The relative SDs for 11 replicate determinations of 10 μg/L for Cd and 100 μg/L for Pb were 2.3% and 2.6%, respectively. The reproducibility for seven determinations of 10 μg/L for Cd and 100 μg/L Pb was 3.5% and 3.9%, respectively.

Accuracy of the proposed UAD procedure was evaluated by CRM of glutinous rice powder and rice flour. The CRMs were analyzed for six measurements spread over 2 weeks. The results of the standard reference materials were in good agreement with the certified values as presented in Table 3. For analysis of Cd, using the comparison method of the European Reference Materials [35], the differences between the certified and measured values were compared with its uncertainty. Because $\Delta_m \leq U_\Delta$ there was no significant difference between the measurement result and the certified value for both of CRMs at the 95% confidence level. The spiked CRMs with 200 μg/L Pb were used for accuracy investigation due to the low level of Pb content in IRMM-804 (lower than LOD of Pb) and noncertified value presented in TRM-F-4001. The recoveries for both spiked CRMs were obtained in the acceptable range at 93–95%. These results indicate that the proposed UAD procedure can be applied for analysis of Cd and Pb in real herbal medicine samples with high efficiency.

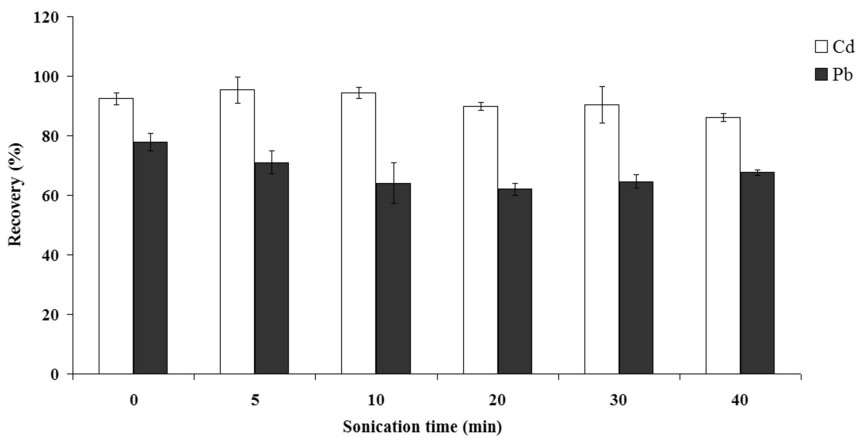

Figure 1 – Effect of sonication time on the ultrasound-assisted digestion efficiency.

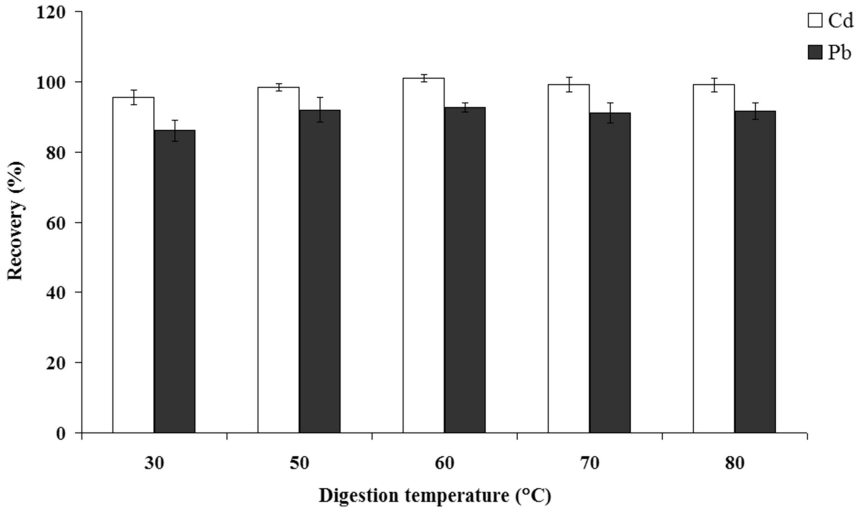

Figure 2 – Effect of digestion temperature on the ultrasound-assisted digestion efficiency.

Table 2 – Digestion efficiency of proposed UAD procedure at optimum conditions.

Metal	Spiked medicinal plant sample (50 μg/L for Cd and 200 μg/L for Pb)		
	CAD ($n = 7$)	UAD ($n = 7$)	% Recovery[a]
Cd	49.1 ± 1.4 (2.9% RSD)	49.8 ± 1.0 (2.0% RSD)	101.4
Pb	185.6 ± 7.8 (4.2% RSD)	183.1 ± 5.3 (2.9% RSD)	98.7

CAD = conventional acid digestion; RSD = relative standard deviation; UAD = ultrasound-assisted digestion.
[a] Recovery (%) = (Metal found with UAD/metal found with CAD) × 100.

3.3. *Analysis of herbal medicine samples*

The proposed UAD procedure was used for FAAS determination of Cd and Pb in medicinal plant and traditional medicine samples. The analysis results of Cd and Pb in all samples are presented in Table 4. To perform the recovery study, all samples were spiked with Cd and Pb at 50 μg/L and 200 μg/L, respectively. Satisfactory results for the concentration levels studied were obtained for Cd and Pb, with percentage recoveries of 90–101% and 92–109%, respectively.

As presented in Table 4, most of all studied samples were not contaminated with Cd and Pb. Concentrations of Cd and Pb found in 19 and 14 herbal medicine samples were below the detection limit (LOD 0.0056 mg/kg for Cd and 1.07 mg/kg for Pb) of the proposed method. The maximum permitted level (MPL) of Cd and Pb in medicinal plant materials and finished herbal products are 0.3 mg/kg and 10 mg/kg, respectively [7]. The amounts of Cd found in two samples of medicinal plants (P7 and P8) were higher than the MPL level. However, concentration of Pb found in the samples of medicinal plant (P8) and herbal medicine (M13) were within the MPL level. The amount of Cd and Pb found in medicinal plant samples (P1–P10) tended to be higher than the amount of Cd and Pb found in herbal medicine samples (M1–M13). All traditional medicine samples (M1–M13) were certified by the Food and Drug Administration, Ministry of Public Health of Thailand, whereas the medicinal plant samples were bought directly as medicinal plant materials without the certification from any administrative agency.

4. Conclusions

A high efficiency sample preparation procedure based on UAD was successfully applied for the acid digestion of the herbal medicine samples. Determination of Cd and Pb was performed by FAAS with sensitive, precise, and accurate results. The recommended method offered fast, convenient, high sample throughput, and low chemical consumption for digestion of herbal medicine samples, compared to the conventional wet acid digestion. The amount of Cd and Pb

Table 3 – Accuracy of the proposed UAD procedure at optimum conditions.

CRM	Amount of Cd (mg/kg)				Spiked CRM with Pb		
	Certified value	UAD ($n = 6$)	Δ_m[a]	U_Δ[b]	Added (μg/L)	Found (μg/L)	Recovery[c] (%)
TRM-F-4001	0.69 ± 0.06	0.66 ± 0.06	0.030	0.077	200	189.2 ± 9.2	94.6 ± 4.6
IRMM-804	1.61 ± 0.07	1.58 ± 0.03	0.030	0.074	200	185.2 ± 2.4	92.6 ± 1.2

CRM = certified reference material; UAD = ultrasound-assisted digestion.
[a] Absolute difference between mean measured value and certified value.
[b] Expanded uncertainty of difference between measured value and certified value.
[c] Recovery (%) = (Metal found/metal added) × 100.

Table 4 – Analysis of Cd and Pb in herbal medicine samples by UAD-FAAS method.

Type of sample[a]	Form of medicine	Common name/ commercial name	Scientific name/ingredient	Cd		Pb	
				Amount (mg/kg)	Recovery[b] (%)	Amount (mg/kg)	Recovery[b] (%)
P1	Powder	Sappan wood	*Caesalpinia sappan* Linn.	ND[c]	100.6 ± 1.0	ND	92.8 ± 2.9
P2	Powder	Turmeric	*Curcuma longa* Linn.	ND	96.8 ± 1.4	ND	—
P3	Powder	Mangosteen (peel)	*Garcinia mangostana* Linn.	ND	—	ND	107.2 ± 2.6
P4	Powder	Soap pod (leaf)	*Acacia concinna* (Wild.) DC.	ND	90.4 ± 4.0	ND	—
P5	Powder	Cassia tree or Thai copper pod (leaf)	*Senna siamea* (Lam.)	ND	—	ND	100.9 ± 2.9
P6	Powder	Horseradish tree (leaf)	*Moringa oleifera* Lam.	ND	93.6 ± 1.4	<LOQ[d] 1.29 ± 0.34	—
P7	Powder	Stevia/Ya wan	*Stevia rebaudiana* Bertoni.	0.68 ± 0.06	—	<LOQ 3.17 ± 0.60	101.8 ± 4.5
P8	Powder	Jiaogulan	*Gynostemma pentaphyllum* (Thunb.) Makino	1.57 ± 0.02	94.8 ± 3.5	5.45 ± 1.10	—
P9	Powder	Indian snake grass/Fa thalai chon	*Andrographis paniculata* (Burm. f.) Wall. ex Nees	ND	—	<LOQ 2.08 ± 0.45	97.5 ± 3.9
P10	Powder	Sacred lotus (stamen)	*Nelumbo nucifera* Gaertn.	<LOQ 0.06 ± 0.01	96.1 ± 1.8	ND	—
M1	Capsule	Bora phet phung chang	*Stephania pierrei* Diels. 350 mg Others 150 mg	ND	—	ND	103.7 ± 2.5
M2	Capsule	Kwao khruea khao	*Pueraria mirifica* Airy Shaw & Suvat. 95 mg *Terminalia chebula* Retz. 135 mg Others 270 mg	ND	93.3 ± 1.8	ND	—
M3	Capsule	Garlic	*Allium sativum* Linn. 300 mg Others 200 mg	ND	—	<LOQ 2.37 ± 0.84	101.4 ± 4.1
M4	Capsule	Phlu khao	*Houttuynia cordata* Thunb. 300 mg Others 200 mg	ND	90.2 ± 0.7	<LOQ 1.47 ± 0.56	—
M5	Capsule	Umbrella tree/Hanuman prasan kai	*Schefflera leucantha* Vig. 300 mg Others 200 mg	ND	—	<LOQ 2.38 ± 0.73	108.7 ± 3.3
M6	Capsule	Sea holly/Ngueak pla mo	*Acanthus ebracteatus* Vahl. 300 mg Others 200 mg	ND	95.4 ± 16	ND	—
M7	Capsule	East Indian Senna	*Cassia angustifolia* Vahl. 268 mg Others 232 mg	ND	—	ND	108.7 ± 2.1
M8	Capsule	Cat's whiskers	*Orthosiphon aristatus* Miq. 350 mg Others 150 mg	ND	94.3 ± 1.3	ND	—
M9	Capsule	Yah pak king	*Murdannia loriformis* 200 mg Others 200 mg	ND	—	ND	96.7 ± 3.9

(continued on next page)

Table 4 – (continued)

Type of sample[a]	Form of medicine	Common name/ commercial name	Scientific name/ingredient	Cd		Pb	
				Amount (mg/kg)	Recovery[b] (%)	Amount (mg/kg)	Recovery[b] (%)
M10	Tablet	Java ginger/Wan chak motluk	*Curcuma zanthorrhiza* Roxb. and others	ND	98.5 ± 2.8	ND	—
M11	Tablet	Prabchompootaweep	*Leonurus sibiricus* L., *Piper nigrum* L., *Acanthus ebracteatus* Vahl. and others	ND	—	ND	105.9 ± 6.9
M12	Tablet	Ceylon calumba root/Ham	*Coscinium fenestratum* (Gaertn.) Colebr. 500 mg	ND	98.6 ± 4.3	<LOQ 3.41 ± 0.29	—
M13	Capsule	Indian snake grass/Fa thalai chon	*Andrographis paniculata* (Burm. f.) Wall. ex Nees 470 mg Others 30 mg	<LOQ 0.07 ± 0.01	—	4.43 ± 0.77	92.2 ± 3.8

FAAS = flame atomic absorption spectrometry; LOQ = limit of quantification; ND = not detected; UAD = ultrasound-assisted digestion.

[a] P1–P10, medicinal plant; M1–M13, traditional medicine.

[b] Recovery (%) = (Metal found/metal added) × 100.

[c] Not detected or below detection limit of Cd 0.56 μg/L; 0.056 mg/kg and Pb 10.7 μg/L; 1.07 mg/kg.

[d] Below limit of quantification of Cd 1.87 μg/L; 0.187 mg/kg and Pb 40.3 μg/L; 4.03 mg/kg.

contaminants in the medicinal plants and traditional medicines investigated were found at different levels. Only two of medicinal plant samples [*Stevia rebaudiana* Bertoni and *Gynostemma pentaphyllum* (Thunb.) Makino] were contaminated with Cd at a higher level than the MPL. This study provides significant data on the safety and quality of herbal medicine consumed in Thailand. In addition, the proposed method has potential as a good alternative for analysis of Cd and Pb contaminants in various biological samples.

Conflict of interest

All contributing authors declare no conflicts of interest.

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