

Heavy Metals Content in Traditional Medicines Sold in the Local Market of Sarawak and Online Shopping Platform in Malaysia

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Abstract: Traditional medicine has been widely used all around the world, since, ancient times. This medicine might be accidentally contaminated with heavy metals during the preparation or intentionally added into the medicine as it is believed to cure diseases. Exposures to heavy metals can greatly affect important organs like the brain, kidney, heart and lungs. Heavy metals (Pb, Ni, Cr, As and Cu) were analyzed in twenty nine samples ($n = 29$) of traditional medicines mainly Malay traditional medicine, Indonesian traditional medicine, Chinese traditional medicine and Ayurveda traditional medicine that were purchased from the local market and online shopping platform in Malaysia. Wet digestion method was used prior to the analysis of metal using flame atomic absorption spectroscopy (Shimadzu AA-7000). The result showed the highest concentrations of Pb, Ni and Cr in tablet/capsule form of Ayurvedic sample at $(38.54 \pm 4.03 \text{ mg/kg})$, $(26.59 \pm 3.43 \text{ mg/kg})$ and $(371.06 \pm 3.10 \text{ mg/kg})$, respectively. In green tea samples, the digested tea leaves showed high Pb $(10.44 \pm 0.0001 \text{ mg/kg})$ and Cu $(17.22 \pm 0.0000 \text{ mg/kg})$ but low Cd $(0.04 \pm 0.0001 \text{ mg/kg})$ content. Meanwhile, spices showed moderate concentrations of As, Cr and Ni with (0.446 ± 0.014) , (0.689 ± 0.003) and (0.476 ± 0.007) , respectively. Some of these samples showed metal concentrations above the standard limit set by World Health Organization (WHO), Ministry of Health Malaysia (MOH) and ASEAN. Traditional medicine products should be monitored, especially, the unregistered products that are readily sold in the local market and online stores. It is important for the public to be aware of the consequences from consuming these medicines.

Key words: Heavy metals, Malay traditional medicines, Chinese traditional medicines, Ayurveda traditional medicines, Indonesian traditional medicines, wet digestion, atomic absorption spectroscopy

INTRODUCTION

Traditional medicine has become one of the earliest treatments ever known to history and this can be proved by the fossil record found during the primal years where the Palaeolithic's human is likely to use plant-derived medicine as ointments (Fabricant and Farnsworth, 2001). The community in Malaysia believes that traditional remedies can cure diseases ranging from mild to severe. These traditional medicines can be grouped into four main types which are Malay Traditional Medicine (MTM), Chinese Traditional Medicine (CTM), Indonesian Traditional Medicine (ITM) and Ayurvedic Traditional Medicine (ATM).

Loads of plant-based medicines were used by traditional medicine practitioners (Newman and Cragg, 2007). Spices are also used as an alternative medicine as it possesses antioxidant and antimicrobial activity (Arora and Kaur, 1999; Hinneburg *et al.*, 2006; Samotya and Urbanowicz, 2005; Krejpcio *et al.*, 2007).

Countries such as Malaysia, Indonesia and India are famous for being the producer of various spices. Meanwhile green tea which originated from CTM possessed active constituent of catechin which is beneficial to human life (Ferrara *et al.*, 2001). Based on green tea article (2015), 3-5 cups/day (1200 mL) of green tea provide at least 250 mg/day of catechins.

Traditional medicines may contain heavy metals that were intentionally added in the drugs formulation or contaminated the herbal plant itself (Martin and Griswold, 2009). Heavy metals contamination in traditional medicine occurred as herbal plants are sensitive to its surrounding. They tend to accumulate heavy metals in high quantities via. root uptake, foliar adsorption and leaves. Thus, it changes the overall elemental composition of the plants (Zollman and Vickers, 1998). Nnorom (2014) also stated that the usage of herbal and traditional medicine raises some issues about the safety whether herbal medicine is safe to be consumed.

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It is reported that the herbal products consisting a significant amount of heavy metals such as lead (Pb), Cadmium (Cd), Chromium (Cr), Arsenic (As) and Nickel (Ni) (Saeed *et al.*, 2011). CTM was reported to contain some poisonous herbs with a significant quantity of As and Pb (Espinoza *et al.*, 1995). In 2014, National Survey on the Use of Medicine (NSUM) stated that they are 17.8% out of 3500 correspondents from the thirteen states of Malaysia preferred consuming traditional medicines (Azmi *et al.*, 2016).

The aim of this study is to determine the presence of heavy metals (Pb, Ni, Cr, As and Cu) in MTM, ITM, ATM and CTM products that were readily sold from the local market and online stores in Malaysia.

MATERIALS AND METHODS

Sampling and classification: The 29 samples of traditional medicines in the form of tablet, capsule and powder, spices and green teas were purchased from the local market and drugstores in Sarawak and also through online stores. The selection of the samples is based on the popularity of the products among consumers, highest sales and recommendation by the traditional medicine practitioners. All of the samples were stored at dry place in its original packaging. If the samples are being drawn from its original packaging, it must be stored in a cleaned container to avoid any contamination (Wee, 2016). The classification of samples is presented in Table 1.

Experimental procedures: Tablet, capsule and powder: about 1 g of sample was weighed, ground and digested using wet digestion method in mixture of HNO₃: HCl (1:3) in 50 mL and boiled in a water bath with the temperature 95°C for about 4-5 h or until the solution has finally dissolved.

Spices 1 g of dry-grounded sample was weighed and digested in 4 mL of H₂SO₄ and 7 mL of 30% H₂O₂. The mixture was heated to 100°C and gradually increased to 250°C and let it cool for 30 min before re-added 2 mL of 30% H₂O₂ and reheated. The process was repeated until a clear solution was obtained (Jones and Case, 1990).

Green tea prior to sample digestion, the tea bag was oven-dried to a constant weight at 60°C. Then the sample was crushed using a sterile mortar and pestle at room temperature. About 10 mL concentrated HNO₃ was added to 1 g of green tea sample and allowed to stand overnight at room temperature. The sample was heated on a hot plate until the solution becomes clear and semi dried. Based on Ambadekar *et al.* (2012) if brown colour appears, acid with the same volume will be added and reheat. The temperature is raised gradually until clear, transparent or slightly yellow solution obtained.

All the digested solution was filtered using Whatman filter paper No. 42 and transferred into 100 mL volumetric flask. Ultrapure water is added up to the calibration mark. Clear solution will be used for the analysis of heavy metals using FAAS Shimadzu AA-7000. Blank was digested using the same method.

Table 1: Type and uses of each traditional medicine samples

MTM and ITM			CTM			ATM		
Code	Form	Uses	Code	Form	Uses	Code	Form	Uses
KB	Powder	Increases sexual energy, vitality and stamina	HG	Capsule	Relieves muscle joint ache and strengthen bone	ASW	Capsule	Improve stamina and helps calming down nerves
MX	Tablet	Reduce pain during menstruation	CK	Tablet	Treat diarrhea and stomach ache	SP	Powder	Treating flu, head ache and migraine
MK	Capsule	Strengthening uterus after childbirth	NC	Tablet	Regulate menstrual cycle and improve appetizer	VV	Powder	Prevent tooth decay and relieve tooth pain
MS	Capsule	Reduce fat and burn excess fat	FP	Powder	Relieve fever and cool down body	DT	Tablet	Reduce weight and helps to balance digestion
BK	Powder	Example: ghout, diabetes and asthma	ST	Tablet	Example: sore throat, stomatitis ulcerative and dermatitis	Black pepper	Berry	Digestive disorder and nerves
SM	Powder	Example: aid in indigestion and urination	GP	Tablet	Example: muscle aches, waist, shoulder, back and joints pain	Cardamom	Capsule	Diarrhea and stabilize pregnancy
SA	Powder	Reduce body weight, fat burner and aid in indigestion	TBH	Green tea bag	Digestion, promote urination and bowel movement	Ginger	Rhizome	Digestive and respiratory disorder
PX	Powder	Reduce body weight and promoting urination	TDC					
			TDH					
			TTL					
			TYH					
			TSK					
			Cinnamon	Bark	Cold, nausea, diarrhea			
			Cassia	Bark	For eye problems and improve bowel movement			

Table 2: Heavy metal concentrations (mean ± SD) in tablet, capsule and powdered form of traditional medicine (mg/kg)

Types/ Sample codes	Form	Mean concentration (mg/kg)		
		Cr	Ni	Pb
CTM				
HG	Capsule	70.47±13.29	7.45±2.17	5.13±0.82
CK	Tablet	40.67±8.79	15.36±4.16	9.28±1.01
NC	Tablet	40.72±6.64	1.84±0.76	11.10±0.58
FP	Powder	45.42±2.21	5.87±4.00	22.01±0.27
ST	Tablet	12.67±0.88	12.24±0.8	32.40±3.05
GP	Tablet	21.39± 0.55	12.31±1.48	34.59±4.34
ITM				
KB	Powder	70.06±0.29	17.44±3.66	9.99±0.01
MX	Tablet	29.33±0.59	14.68±1.22	22.76±2.14
MK	Capsule	25.77±0.83	15.71±0.74	19.49±1.31
MS	Capsule	62.64±4.43	21.94±0.71	37.99±3.25
SA	Powder	18.19±0.64	11.10±0.89	10.38±1.47
PX	Powder	18.60±0.99	12.60±0.52	6.38±1.41
ATM				
ASW	Capsule	142.72±59.0	26.59±3.43	38.54±4.03
SP	Powder	371.06±3.10	4.05±0.66	4.94±1.67
VV	Powder	70.27±24.84	2.49±0.16	25.91±0.59
DT	Tablet	103.20±3.11	n/a	34.95±0.58
MTM				
BK	Powder	6.8±0.84	8.21±0.83	18.35±2.04
SM	Powder	10.41±0.72	9.71±1.15	25.91±3.4

*(n/a = not applicable)

Table 3: Heavy metal concentrations (mean±SD) in spices (mg/kg)

Types/ Sample codes	Form	Mean concentration (mg/kg)		
		Cr	Ni	As
CTM				
Cinnamon	Bark	0.689±0.003	0.476±0.007	0.446±0.014
Cassia	Bark	0.684±0.000	0.453±0.001	0.415±0.000
ATM				
Black pepper	Berry	0.686±0.001	0.464±0.004	0.427±0.009
Cardamom	Capsule	0.686±0.002	0.459±0.001	0.424±0.002
Ginger	Rhizome	0.683±0.000	0.448±0.000	0.409±0.001

Table 4: Heavy metal concentrations (mean ± SD) in green tea (mg/kg)

Types/ Sample codes	Form	Mean concentration (mg/kg)	
		Cu	Pb
CTM			
TBH	Tea bag	2.14±0.0002	12.00±0.0001
TDC		3.02±0.0001	13.36±0.0006
TDH		9.81±0.0002	17.22±0.0000
TTL		6.92±0.00003	10.20±0.0006
TYH		7.93±0.0001	8.29±0.0005
TSK		10.44±0.0001	4.53±0.0000

RESULTS AND DISCUSSION

The mean concentrations of heavy metal (Pb, Ni, Cr, As and Cu) in 29 samples of MTM, ITM, ATM and CTM were tabulated in Table 2-4 based on the form of the medicine sample.

Chinese Traditional Medicine (CTM): CTM showed the lowest concentrations of Ni and Cr analysed in the capsule, tablet and powdered form of traditional medicine. High concentration of Pb found in sample NC (11.10± 0.58), FP (22.01±0.27), ST (32.40±3.05) and GP

(34.59± 4.34) which exceeded the standard limit set by WHO (2007) and ASEAN both at 10 mg/kg. Only samples NC and FP showed low Ni value with 1.84±0.76 and 5.87±4.00 mg/kg, respectively which is below than the regulatory limits set by USP at 5.0 mg/kg. Cr recorded the highest HG (70.47±13.29 mg/kg) followed by FP at 45.42±2.21 mg/kg. In spices, low Cr, Ni and As recorded ranging from 0.415±0.000 to 0.689±0.003 mg/kg. Nordin and Selamat (2013) also found that the amount of as in most of the local spices in Malaysia has the similar range of value. In this study, as concentration in all spices samples were lower than all permissible limits set by WHO and FAO (0.5 mg/kg) and MOH (5.0 mg/kg). In green tea samples, Pb was detected the highest in TSK (9.81±0.0002 mg/kg) and the lowest lead was detected in TBH (2.14±0.0002 mg/kg). All samples have exceeded the permissible limit for Pb from MOH (Malaysia) and MOH (Chinese) which is 2 mg/kg and 5 mg/kg, respectively. Cu was found the highest in TDH (17.22±0.00 mg/kg) and the lowest in TSK (4.53±0.00 mg/kg). All green tea samples showed Cu concentration below than the established permissible limit set by MOH (Chinese) and EU from 30-150 mg/kg.

Ayurveda Traditional Medicine (ATM): ATM showed ASW sample has the highest concentration of all heavy metals analysed with Cr at 142.72±59.0 mg/kg, Ni at 26.59±3.43 mg/kg and Pb at 38.54±4.03 mg/kg which exceeded the permissible limit in traditional medicine provided by WHO (2007) and USP. Only SP sample showed both Ni and Pb value below than the standard permissible limit but the concentration for Cr in this sample recorded the highest with 371.06 ±3.10 mg/kg. The contamination of chromium might occur during the preparation of the medicine where the chromium in the air may pollute the medicine (ATDSR., 2000). High Cr content in SP because it is made up from turmeric which is a type of rhizome that is grown under the soil. Huffman and Allaway (1973) stated that a high concentration of chromium is the highest in the root which is about 98% and only 0.1% in seed. The accumulation of chromium in the plants root is because both of the Cr (III) and Cr (IV) are being trapped inside the vacuoles of the root cells (Shanker *et al.*, 2005). Both of the Cr (III) and Cr (IV) will cross the endodermis layer but only Cr (IV) cannot cross this layer, hence, Cr (IV) will be reduce into Cr (III) and diffuse into the endodermis layer and later will be transported to the vascular tissue (Shanker *et al.*, 2005).

None of the capsule, tablet and powdered form of ATM followed the regulation and the permissible limit provided by WHO (2007) and these medicine should not

be traded in the market. Based on the previous research done by Hina *et al.* (2011), the level of Pb in the analysed herbal drugs ranged from 3.26 to 30.46 mg/kg and 71.6% of the analysed samples exceeded WHO permissible limit. There is less significant difference with the obtained result and the research done by Hina *et al.* (2011) as the maximum lead found in this study did not deviate too much from the previous study.

ASW sample contains the root extract of *Withania somnifera* or also known as Indian ginseng. The possible explanation behind high Pb content is due to the uptake of Pb through the root's plant. The roots of the plant are able to take up a significant amount of Pb from the soil that is mainly originated from the automobiles, factories and pesticides (Sharma and Dubey, 2005). The Pb available in the soil will bind on the root surface which later accumulates in the root apoplast which is the space outside the plasma membrane (Sharma and Dubey, 2005). The second highest concentration of Pb in ATM is in DT sample (34.95±0.58 mg/kg). This might be due to the ruptured endodermis layer of the root. Sharma and Dubey (2005) stated that when the plants has already up taken a high amount of Pb, it will caused cell injury and thus disturb the barrier provided by the endodermis layer and able to diffuse into the leaves, fruits and stem of the plant. In India, majority of the children consume traditional medicine like ASW that is added in milk for treating emaciation problems and it is one of the best medication thus it is essential to ensure that the medicine is free from heavy metals (Sharma, 1995). As we all know, the effect of lead poisoning is more dangerous in children. According to Canfield *et al.* (2003), children that consume a high concentration of lead which is >10 µg/dL are likely to have lower Intelligence Quotient (IQ) test or intelligence functions.

In spices, ATM showed low Cr, Ni and As with the value ranged at 0.409±0.001 to 0.686±0.001mg/kg which is below that the permissible limit set by WHO.

Indonesian Traditional Medicine (ITM): Heavy metals (Ni, Pb and Cr) in all ITM samples have exceeded the permissible limit except Pb concentration in KB and PX with 9.99±0.01 and 6.38±1.41 mg/kg, respectively. Cr showed the highest amount of the heavy metals analysed in KB (70.06±0.29 mg/kg) followed by MS (62.64 ±4.43 mg/kg). Meanwhile, Ni concentration was found the lowest in SA at 11.10±0.89 mg/kg but this amount of nickel still beyond the permissible limit provided by USP at 5.0 mg/kg and (WHO., 2007) at 1.5 mg/kg.

Malay Traditional Medicine (MTM): BK and SM samples showed Cr content with 6.8±0.84 mg/kg and

10.41±0.72 mg/kg, respectively which also exceeded the permissible limit for chromium set by WHO (2007) which is 2.0 mg/kg. The presence of chromium might be due to the contamination during the processing of the products and agriculture soil. Both samples in MTM showed high Pb content and have exceeded the permissible limit by WHO (10.0 mg/kg) and ASEAN (10.0 mg/kg).

Trend of heavy metals in BK sample is Pb>Ni>Cr whereas SM was observed with Pb>Cr>Ni. High Pb content in this sample is probably due to environment factors which caused the contamination of such products.

CONCLUSION

In this study, the concentrations of heavy metals which are Pb, Ni, Cr, As and Cu were analysed in twenty nine samples of traditional medicines mainly from Malay traditional medicine, Indonesian traditional medicine, Chinese traditional medicine and Ayurveda traditional medicine that were purchased from the local market and online shopping platform in Malaysia. Wet digestion method was used prior to the analysis of metal using flame atomic absorption spectroscopy (Shimadzu AA-7000). The result of the analysis showed that these heavy metals were present in these four types of traditional medicine. Most of the samples analysed exceeded the standard limit regulated by World Health Organisation (WHO). Even though traditional medicine may sound safe, some of the traditional medicine products may cause adverse health effects and it is important to have knowledge about the medicine before consuming it.

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