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Original Research Article

Determination of Arsenic Content of Available Traditional Medicines in Malaysia using Hydride Generation Atomic Absorption Spectrometry

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Abstract

Purpose: *To determine the content of arsenic (As) in some locally available traditional medicines in the East Coast region of Malaysia.*
Methods: *The determination of As was conducted using hydride generation-atomic absorption spectrometry (HG-AAS). Sample preparation entailed mineral acid digestion using hydrochloric acid and nitric acid mixture in a ratio of 1:3. Sixty samples were collected from different locations including shops and open markets in East Coast region of Malaysia, namely, Pahang, Terengganu and Kelantan states. Most of these preparations were not registered with Malaysian drug authority.*
Results: *Out of sixty traditional medicine samples, twenty six contained As in a concentration range of 0.2150 - 1.3254 ppm. As for the rest, they were below the limit of quantification (LOQ).*
Conclusion: *Traditional medicine samples available in the east coast region of Malaysia contain levels of arsenic that can adversely affect health upon consumption.*

Keywords: *Traditional medicine, Arsenic, Hydride Generation –Atomic Absorption Spectrometer HG-AAS.*

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INTRODUCTION

Traditional medicines (TM) had been used long before the recorded history [1]. For last few decades, enormous growth in popularity of traditional healing modalities was observed in developing and developed countries [2,3]. Safety and efficacy has become a concern worldwide due to this popularity as toxicity from TM has often been reported. One of the major causes of toxicity is the presence of toxic heavy metals in such products. There is a high possibility that some of the herbal products may contain toxic heavy metals [4,5]. Heavy metal toxicity can cause damage to vital body organs such as brain, kidneys and liver [6,7].

In Malaysia a large segment of the population rely on TM. One of the studies reported that 8% of TM samples contained Pb in a concentration range of 10.64 - 20.72 µg/g which exceeds the permissible limit of lead imposed by Drug Control Authority (DCA) in Malaysia [8]. Another study revealed that 26 products contained 0.53 – 2.35 µg/g of mercury and therefore, did not fulfill the requirements of the regulatory authorities [9].

Arsenic (As) is one of the potential toxic heavy metals which may be present in some herbal products as it can penetrate the plant via contaminated soil and water. Both trivalent and pentavalent forms of As are rapidly absorbed from the gastrointestinal tract [10]. Arsenic (As) also has a direct toxic effect on cellular respiration in mitochondria [11].

The rationale for this study was to identify (if any) trace amount of arsenic in locally available traditional medicines in East Coast region of Malaysia in order to determine if they are safe for human consumption.

EXPERIMENTAL

In this study we analyzed sixty samples for the content of arsenic (As) using HG-AAS. All samples were purchased from various places such as shops and markets from three major states in the East Coast Region of Malaysia, namely, Pahang, Terengganu and Kelantan. We have followed the non-probability purposive sampling protocol. All samples were mostly of herbal origin and in various dosage forms, including tablets, pills, capsule and powder. The quantity of samples, according to dosage form type, ranged from 2 (for tablets) to 30 (capsules). The study samples were being used nedicinal purposes, and not as supplements or energy enhancing agents; they were mainly unregistered with the Malaysian Drug Control Authority (DCA).

Chemical and reagents

All chemicals and reagents used in this study were of analytical grade and trace metal grade. Concentrated hydrochloric acid (HCl) and nitric acid (HNO_3) were obtained from Fisher. Arsenic standard solution (1000 ppm) was obtained from Perkin Elmer. Sodium-borohydride, sodium hydroxide, ascorbic acid and potassium iodide (KI) were obtained from Merck**.**

Treatment of glassware

All glassware were soaked in 5% nitric acid for 2 h and then washed with deionized water prior to use. The quartz cell was soaked with a mixture of two concentrated mineral acids (hydrochloric acid 37% and nitric acid 65%) in 1:3 ratio for 2 h, rinsed with deionized water and dried using lab-dryer FDD-720 prior to use.

Standards and reagent preparation

Working standards were prepared by serial volume/volume (v/v) dilution of the stock standard solution (1000 ppm). Freshly prepared standards were used for calibration purposes.

Freshly prepared reductant solution, sodium borohydride (0.2% $NaBH_4$ + 0.05% NaOH) was dissolved in 1 L of deionized water and a carrier solution 10% HCl was also prepared prior to the analysis. Calibration for As was obtained in the standard concentration range 1 - 10 ppb. This calibration range exhibited good linearity with a coefficient of determination (R^2) > 0.990.

Sample preparation

Traditional medicine samples were obtained from different sources like shops and markets from different places of east coast region of Malaysia that comprises three states namely Pahang, Terengganu and Kelantan. Samples were digested using a mixture of two concentrated mineral acids, (Nitric acid 65% , Hydrochloric acid 37%) in 1:3 (v/v) ratio. 0.5 g of each sample was weighed and placed in 100 ml PTFE beaker after which 9 ml of freshly prepared acid mixture was added. Then the mixture was boiled gently over a water bath (95°C) for 4 - 5 h (or until the sample had completely dissolved). It was allowed to cool at room temperature and filtered with Whatman 42 grade filter paper. A sufficient amount of de-ionized water was added to make the final volume up to 50 ml [12]. For arsenic analysis, standards and samples were pre-reduced from arsenate pentavalent (V) to arsenite trivalent (III) state. This was achieved by adding a reducing solution containing 5% w/v KI, 5% w/v ascorbic acid and 10% HCl, and the treated samples and standards were allowed to stand at room temperature for approximately 40 min prior to analysis.

Total arsenic (As) determination was carried out using Perkin Elmer Hydride Generation Atomic Absorption Spectrometer (HG-AAS) (A Analyst 800), controlled by Win Lab 32 software and equipped with arsenic (As) electrode-less discharge lamp (EDL) which provides higher energy than the corresponding hollow cathode lamp (HCL) and results in improved sensitivity and detection limit. The technique involves the reaction of acidified aqueous samples with a reducing agent like sodium borohydride. The sodium borohydride/acid reduction generates volatile hydrides which are transported to a quartz cell by means of an argon carrier gas. In the heated quartz cell, the hydrides are converted to gaseous metal atoms. The spectrophotometer was operated at 193.7 nm with a slit width of 0.7 nm. The carrier gas flow

was optimized to 80 mL/min prior to calibration in order to achieve the highest sensitivity.

Statistical analysis

The results for each determination were reported as mean ± standard deviation (SD, n = 3) for each sample using Microsoft Office Excel 2007 software for descriptive analysis.

RESULTS

In this study, sixty samples were collected for the determination of a trace amount of arsenic. The Limit of Quantification (LOQ) for As was 1.1752 ppb. Out of sixty TM samples, twenty six were found to contain As in a concentration range of 0.2150-1.3254 ppm as listed in Table 1. The remaining samples were below the LOQ.

Table 1: Mean concentration of arsenic (As) in some traditional medicines

Sample ID	Mean content of As (±SD, n = 3, ppm)
TM – 4	0.4562(±0.0066)
TM – 7	0.2970(±0.0074)
TM – 10	0.5044(±0.0070)
TM – 12	0.2386(±0.0030)
TM – 13	0.7894(±0.0060)
TM – 14	0.3220(±0.0170)
TM – 18	0.3628(±0.0048)
TM – 20	0.3346(±0.0040)
TM – 21	0.3728(±0.0058)
TM – 24	0.4546(±0.0068)
TM – 25	0.3498(±0.0080)
TM – 26	0.3958(±0.0074)
TM – 28	0.3554(±0.0064)
TM – 31	0.9736(±0.006)
TM – 34	0.8294(± 0.0180)
TM – 35	0.2544(0.0040)
TM – 41	1.2100(±0.0330)
TM – 42	0.2678(±0.0036)
TM – 43	1.1792(±0.0674)
TM – 45	0.9250(±0.0060)
TM – 46	1.3254(±0.0306)
TM – 47	1.1670(±0.0208)
TM – 52	0.2826(± 0.0020)
TM – 53	0.2742(± 0.0048)
TM – 55	0.3660(± 0.009)
TM – 57	0.2150(±0.0360)

TM codes: *Capsule = 18, 20, 24, 25, 34, 35, 42, 52, 53, 55, 57; round pill = 10, 12, 13, 28, 31, 41, 43, 45, 46, 47; powder = 4, 7, 14, 21, 26,*

DISCUSSION

Arsenic (As) is generally found at trace concentration levels in ground water, soil, and air, mostly due to industrial activities such as mining, combustion of fossil fuels or from agrochemical waste [13]. A study conducted in Sungai Lembing, an old tin mining town located 34 km northwest of Kuantan in Pahang (Malaysia), showed the presence of arsenic (As) concentrations in significant values in soil samples [14]. The uptake and accumulation by plants is considerably affected by As concentration in the soil and increases with increase in As levels [15].

The results of the present study show that about 43% of the total number of the samples contained As but did not exceed the permissible limit of 5 ppm stipulated by the National Pharmaceutical Control Bureau (NPCB) of Malaysia. The highest concentration 1.32 ± 0.03 µg/g was detected in TM 46 which was an unregistered product, and was purchased from an open market in Kuantan.

Though the results state that As concentrations were below the permissible limit for several of the preparations evaluated, it is not enough to conclude that these medicines are absolutely safe in terms of As contamination because As, like other heavy metals, can bioaccumulate and subsequently disrupt the functions of vital organs in the human body such as brain, kidneys and liver [7].

CONCLUSION

The samples tested contain arsenic at varying concentrations. Among them, none exceeds the permissible level stipulated by NPCB. However, caution should be exercised in declaring these products as they may cause adverse health effects in consumers, especially if consumed in amount that exceeds permissible levels.

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