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TRADITIONAL AYURVEDIC TABLETS MAY CONTAIN HEAVY METALS

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ABSTRACT

Plan: *Several authors have reported the presence of alarmingly high levels of heavy metals like lead, arsenic, cadmium and mercury in ayurvedic medicines. Ayurveda employs several classes of medicines, among which gulika (tablets) are an important one. However, it is not known whether traditional ayurvedic pills are free from heavy metals. The present study was therefore, undertaken to study the heavy metal content of some representative ayurvedic tablets manufactured using herbs.*

Methodology: *Samples of ayurvedic tablets were obtained from the market and the content of lead, arsenic, cadmium and mercury was determined using ICP-MS.*

Outcome: *Among the 16 Ayurvedic tablets analyzed, 4 failed for lead, 2 for arsenic and 6 for mercury. All the tested samples had cadmium below the limit set by Government of India. A review of the available reports and the present study support the contention that tablets are the worst affected ayurvedic dosage form. Urgent measures are to be taken to control this problem.*

1. INTRODUCTION

Ayurveda has been practiced in India since ancient times. According to the modest estimates made by European scholars, it has a track record of 20 centuries [1]. However, protagonists of the system claim that it is more ancient[2]. Following the renewed worldwide interest in traditional medicine spurred by the Alma Ata Declaration of 1978[3], greater attention is now being paid to the quality control of ayurvedic medicines[4]. In this context the content of heavy metals in these herbal products assumes great importance. The presence of heavy metals in ayurvedic medicines was first reported by Saper et al (2004)[5]. This was followed by several reports of heavy metal contamination in ayurvedic medicines[6-10].

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Ayurveda employs several classes of medicines, among which *gulika* (tablets) are an important one. Nearly thirty *gulika* are used in contemporary ayurvedic practice[11]. However, it is not known whether traditional ayurvedic tablets are free from heavy metals. The present study was therefore, undertaken to study the heavy metal content of some representative ayurvedic tablets manufactured using herbs.

2. MATERIALS AND METHODS

This work was carried out when the authors were employed at Confederation for Ayurveda Renaissance Keralam Ltd, KINFRA Small Industries Park, Nalukettu Road, KINFRA Park P.O. - 680 309, Koratty, Thrissur District, Kerala, India (2012-2015). Samples of traditional ayurvedic tablets were procured from various parts of the southern Indian province of Kerala, where Ayurveda is very popular.

2.1. Standards and reagents

Multielement standards for ICP-MS were prepared from stock solutions of Pb, As, Cd and Hg at 100 ppm concentrations obtained from Merck KGaA, Frankfurt, Germany. 1 ppm stock solution was prepared from the 100 ppm standard. This solution was further diluted to 5 ppb, 50 ppb, 100 ppb, 200 ppb and 250 ppb. Ultrapure nitric acid and hydrochloric acid were procured from Panreac Química S.A.U., Barcelona, Spain. HPLC water (Merck Specialities Pvt Ltd., Mumbai, India) was used for washing laboratory plastic ware and for preparing sample and standard solutions.

2.2. Microwave-assisted digestion

250-500 mg of a sample of ayurvedic tablet was put into a PFA tube. 5 ml of ultrapure nitric acid and 0.5 ml of ultrapure hydrochloric acid were added. The tube was capped and placed in microwave-accelerated reaction system (CEM Corporation, Matthews, North Carolina, U.S.A.) for 1 hour. The instrument was programmed to reach 180^0 C in 15 minutes and stay on hold for 35 minutes. The PFA tube was taken out of the digester and allowed to cool down to room temperature. The sample was quantitatively transferred to 50 ml plastic tube and made up to 50 ml, using HPLC water. The sample was saved in the refrigerator until taken up for analysis.

2.3. Heavy metal analysis

Pb, As, Cd and Hg in the digested samples were analyzed using an Agilent 7700X inductively-coupled mass spectrometer (ICP-MS) (Agilent Technologies, U.S.A.). The operating conditions of the instrument were as shown in Table 1. The ICP-MS installed in a temperature-controlled room (17-20 ^{0} C) was allowed a period of 3 hours to stabilize before analyses was carried out.

Before attempting to analyze the heavy metal content of the samples, some of the parameters like linearity, range, accuracy, precision, limit of detection and limit of quantification were validated. The data are presented in Table 2.

Table 1: Operating conditions of Agilent 7700X ICP-MS

Parameter	*Value*
Plasma gas flow rate	15 L/min
Auxiliary gas flow rate	1 L/min
Carrier gas flow rate	1 L/min
Makeup gas flow rate	1 L/min
Collision gas flow rate	4.3 mL/min
RF power	1500 W
Nebuliser	Micromist
Torch injector internal diameter	2.5 mm
Sample depth	10 mm
Interface cone	Ni sampler cone, Ni skimmer
CeO+/Ce+	0.50 %

Table 2: Validation parameters for determination of Pb, As, Cd and Hg

Element	*Range (ppm)*	*Linear equation*	*Correlation coefficient*	*Precision (%RSD)*	*LOD (ppb)*	*LOQ (ppb)*
Pb	0.008-50	y= 44763.7306x + 7758.90	0.9985	0.0415	8.9100	29.7003
As	0.007-25	y= 2381.7508 x + 77.78	0.9992	0.0533	7.9867	26.6225
Cd	0.009-50	y= 9690.4582 x + 30.00	0.9990	0.0459	9.3077	31.0258
Hg	0.009-25	y= 9893.0718 x + 3758.43	0.9988	0.0929	9.4733	31.5777

2.4. Detection limits

The minimum detection and instrument detection limits were calculated for Pb, As, Cd and Hg as three times the standard deviation of the concentrations of the blanks and sum of the minimum detection limit and element concentration of the blank respectively[12].

3. RESULTS AND DISCUSSION

Among the 16 Ayurvedic tablets analyzed, 4 failed for lead, 2 for arsenic and 6 for mercury. All the tested samples had cadmium below the limit set by Government of India, according to which, lead, arsenic, cadmium and mercury in any Ayurveda, Siddha or Unani medicine should not exceed 10, 3, 0.3 and 1 ppm respectively (Table 3)[13].

Of the 14 Ayurvedic products reported by Saper et al (2004)[5] to contain high levels of heavy metals, 8 belong to the class of tablets. It was earlier reported that *Kāṅkāyan Baṭi* and *Candraprabhā Vaṭi* which are prepared exclusively with herbal ingredients also contain high levels of heavy metals [6, 10]. This indicates that tablets are the highly affected dosage form. Sebastian et al (2015)[14] carried out an extensive study of the heavy metal content of traditional ayurvedic medicines. The content of lead, arsenic, cadmium and mercury in 126 Ayurvedic medicines manufactured by 32 companies was analyzed using inductively coupled plasma mass spectrometry (ICP-MS).

Table 3: Content of heavy metals in traditional ayurvedic tablets

Sl. No.	Name of formulation	Content of heavy metals (ppm)			
		Lead	Arsenic	Cadmium	Mercury
1	*Ajālyādi Gulika*	**28.771**	ND	ND	ND
2	*Candraprabhā Vaṭika*	**12.17**	2.26	0.055	0.235
3	*Dhānvantaram Gulika*	6.41	1.27	0.08	ND
4	*Dhanwantharī Gulika*	**14.80**	0.77	BDL	**13.91***
5	*Hiṅgvādi Gulika*	2.55	1.57	0.09	ND
6	*Kaiśōra Guggulu* (1)	**25.67**	0.77	BDL	BDL
7	*Kaiśōra Guggulu* (2)	1.62	0.23	ND	ND
8	*Kāñcanāra Guggulu*	1.609	BDL	BDL	ND
9	*Nirguṇḍyādi Gulika*	7.18	2.45	0.073	**258.51**
10	*Sinhanād Guggulu*	5.293	**16.762**	BDL	**45.218**
11	*Sukhabhēdi Gulika*	3.7	1.41	BDL	**779.97**
12	*Tālīsapatrādi Vaṭakam*	0.186	0.108	BDL	ND
13	*Vilvādi Gulika*	2.01	**4.09**	BDL	**85.37**
14	*Yōgarāja Guggulu* (1)	9.03	0.49	0.069	**1.45**
15	*Yōgarāja Guggulu* (2)	7.41	0.896	0.059	ND
16	*Yōgarāja Guggulu* (3)	2.04	0.24	BDL	ND

*Figures in bold font indicate element exceeding limit

All the samples of *kvātha* (decoctions), *cūrṇa* (powders), *taila* (medicated oils), *ghṛta* (medicated clarified butter), *lēhyam* (electuaries), *āsava* and *ariṣṭa* (fermented products), *drāvakam* (distillates) and *lēpa* (pastes) analyzed in the study conformed to the limits for heavy metals set by Government of India. However, unlike the results reported by Sebastian et al (2015)[14] many of the traditional ayurvedic tablets investigated in the present study contain high levels of heavy metals. Heavy metal content of Ayurvedic minerallo-metalic tablets is already reported[5, 10]. It is noteworthy that in several of these tablets, mercury is present in alarming amounts like 31,000 ppm or 70,000 ppm[10].

One major source of heavy metals in ayurvedic tablets is the herbal raw materials themselves. There are several reports of heavy metal contamination of medicinal herbs. Dghaim et al (2015)[15] reported high levels of cadmium in chamomile, basil, sage, oregano and thyme collected from the United Arab Emirates. High content of cadmium have been reported in Egyptian and Iranian herbs also[16, 17]. The heavy metal content of ayurvedic herbs traded in India also needs to be determined.

Another possible source of heavy metal contamination may be the traditional method of preparing the tablets. Powders of herbs are mixed with various fluids like water, decoctions or juices of herbs, lemon juice, juice of pomegranate flowers or fruits, latex of *Jatropha curcas*, cow milk, breast milk, cow urine, goat urine, castor oil, honey, clarified butter or sesame oil. The mass is ground on granite stone slabs using granite pestles. The grinding is carried out continuously for long periods lasting even seven to eighteen days[18].

Thereafter, the ground mass is rolled into tablets of various sizes. Granite is basically an igneous rock and contains mercury. Its mercury content is nearly twice that of crustal material[19]. Granite also contains in addition to other heavy metals, 0.2-13.8 ppm of arsenic, 0.003-0.18 ppm of cadmium and 6-30 ppm of lead[20, 21]. It is possible that during the continuous grinding process, these heavy metals may be leaching out into the herbal mass being ground. It needs to be ascertained whether such a grinding method influences the heavy metal content of the ayurvedic tablets.

Heavy metal contamination of herbal medicines has been reported from other countries as well. Mousavi et al (2014)[22] quantitatively determined concentrations of lead and cadmium in eleven herbal medicines sampled from pharmacies in Tehran, Iran, using atomic absorption spectrophotometry. Several of them contained these heavy metals above the specified levels. Obi et al (2006) [23] estimated the content of heavy metals including mercury in ready-to-use herbal products from Nigerian market. Results showed that 100% of the samples contained high levels of the heavy metals.

4. CONCLUSION

Heavy metals are those metals which possess a specific density of more than 5 g/cm^3 and adversely affect the environment and living organisms [24]. Lead, cadmium, mercury and arsenic are the heavy metals that pose a major threat to humans. Though the adverse health effects of heavy metals have been known for long, exposure to them continues especially in less developed countries[25]. The present study shows that some Ayurvedic tablets also can be a source of heavy metal poisoning. The magnitude of this problem can be gauged from the fact that ayurvedic tablets can be bought and consumed in India without a medical prescription. As the present investigation is limited by the small size of the studied samples, Bureau of Indian Standards (BIS) and the Department of AYUSH (Government of India) should exigently institute a detailed investigation into the presence of heavy metals in ayurvedic tablets. This report is intended to spur that action.

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Conflict of Interest

CARe Keralam Ltd was established for the advancement of Ayurveda. As this report is directly related to improving ayurvedic pharmaceutics, the authors do not have any conflict of interest.

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