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Metals in Herbal Drugs from Himalayan Region

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Abstract Various herbal products from Himalayan region may provide a huge source of supply in the domestic and international markets. In this study, the heavy metal load in various herbal drugs of the region was investigated. The studied toxic elements were present in the herbal drugs (0.2–8.34 mg/kg As, 0.11–0.48 mg/kg Cd, 2.5–6.0 mg/kg Pb). Zinc was found in the range 7–32 mg/kg and all the samples were free from mercury contamination.

Keywords Heavy metal · Toxic · Zinc · AAS · Herbal drugs

The herbal drugs are getting importance throughout the world and consumed by masses during the last few decades as evidenced by rapidly growing global and national markets of herbal drugs. Now people rely more on herbal drugs than the synthetic drugs due to their high prices and harmful side effects (Tang et al. 2007). Herbal drugs have much positive points over the allopathic system of medicine. This trend is growing, not only in developing countries but also in developed countries. According to World Bank, trade in herbal industry reaches about US $ 62 billion, with annual increase of 5%–15% (Patwardhan et al. 2003). However there is an inherent health risk associated with many of these medicines due to presence of some toxicants including heavy metals. The major source of metal contamination is agricultural expedients, such as cadmium containing dung, organic mercury fungicides, and the insecticide lead arsenate (Gosslim et al. 1984).

Recently a study conducted on ayurvedic medicines by Harvard Medical School (Saper et al. 2004) also reported that some of these drugs had potentially harmful levels of lead, mercury and arsenic. Lead is widely distributed in spices and herbal plants (Divrikli et al. 2003; De Leonardis et al. 2000). Cadmium in foods is mostly derived from various sources of environmental contamination (Adriano 2001). The source of contamination of arsenic (As) is mainly due to use of fertilizers and pesticides, or due to other pollutions the soil As level could be elevated (Chen et al. 2006; Sierra-Alvarez et al. 2006). Additional sources of heavy metal contamination are rainfall, traffic density, use of oil or fossil fuels for heating. Contamination may also occur during processing in industries and leaching of metals from metallic containers during storage (Alabduláaly and Mujahid 2009).

One of the major causes of adverse health effects is directly linked to presence of heavy metals in herbal medicines**.** Heavy metals may cause serious health hazards such as renal failure, symptoms of chronic toxicity and liver damage (Andrew et al. 2003 Shaw et al. 1997). Cadmium is a non-essential element in foods and natural waters, and it accumulates principally in the kidneys and liver. Cd causes osteomalacia and pyeloephritis. Amongest known toxic heavy metals, lead (Pb) in any form seems to be a ubiquitous environmental poison to any form of life (D'Souza et al. 2002). Pb may cause renal tumors and other carcinomas. However, Zn is less toxic than former ones. Due to these reasons, increasing popularity of herbal medicines has also brought concern and fears because the quality, safety and batch to batch consistency of herbal formulations are not up

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to the mark, to meet the criteria needed to support its use worldwide (Liang et al. 2004). Thus, it becomes imperative that quality of these drugs must be maintained and it should be free from any type of contamination.

In India, Himalayan region may provide a huge source of supply for herbal medicines in the domestic and international markets. The monitoring of toxic component in vegetable drugs, their mixtures, preparations and chemical drugs are controlled by the same drug law. The objective of the present research is to study of heavy metal profile to assess the safety and quality of ayurvedic herbal medicine products of Himalayan region commercially available in the markets. Therefore, such study is important to estimate the level of metal contamination in some herbal drugs to assure their quality.

Materials and Methods

Atomic absorption spectrophotometer grade standard stock solutions (1000 mg/L) supplied by Merck, Germany, were used to prepare calibration curve for each metal. Distilled-deionized (Millipore, Direct Q_3) water was used for all analytical works. The chemical use for digestion and analysis such as nitric acid (HNO_3), perchloric acid ($HClO_4$) from Merck, Mumbai, India and HCl from Ranbaxy, India. The filter paper obtained from Qualigens (0.45 μm), 615A, Germany. The galsswares were soaked in 2N HCl for 24 h and rinsed with deionized-distilled water before use. The sodium borohydride (4.0%) and 5 M HCl was prepared for determination of arsenic (As) and mercury (Hg) by Hydride Vapour Generator (HVG). Shimadzu model AA 6300 Atomic Absorption Spectrophotometer (AAS) (Tokyo Japan) coupled with deuterium corrector and Hydride Vapour Generator (HVG) with appropriate hallow cathode lamps (HAMAMATSUPHOTONICS K.K. JAPAN) as radiation source was used for measurement of heavy metals from samples. The 14 herbal drug samples were collected from the local market. The plant name was described in Table 1 from which herbal drugs manufacture.

The fine powdered samples (1.0 g each in triplicate) were digested in glass vessel containing 15 mL of HNO_3 at

Table 1 Plant used in herbal drugs

Sample no.	Scientific name of plants
1	*Withania somnifera, Centella asiatica, Sida cardifolia, Spilanthes acmella*
	Orchis latifolia, Tribulus terrestris, Astercantha longifolia, Celastrus, panniculatus
	Saussurea lappa, Cissus quadrangularis, Mucuna pruriens, Colchicum luteum
	Muria puama
2	*Silybum marianum, Boerhaavia diffusa, Picrorhiza kurroa, Eclipta elba,*
	Phyllanthus amarus, Tephrosia purpurea, Anchographis paniculata
	Swertia chirayita
3	*Gymnema sylvestre, Pterocarpus marsupium, Ocimum basilicum, Momordia charantia, Azadirachta indica, Vinca rosea, Salacia chinensis, Aegel marmelos*
	Trigonella foenum graecum, Syzygium cumini
4	*Saraca indica, Centella asiatica, Bamboomanna, Aloe vera, Symplocos racemosa*
5	Trifla
6	*Rauwalfia serpentina, Convolvulus pluricaulis, Terminalia arjuna*, Rosa powder
	Inula racemosa, Onosma bracteatum, Boerhaavia diffusa
7	*Withania somnifera, Centella asiatica, Gingko biloba, Bacopa monnieri, Ashatvarga, Pueraria tuberosa, Asparagus somnifera*
8	*Cyperus rotundus, Coelus forskohlii, Rubia cardiofolia, Tinospora cordiofolia*
	Trifla, *Azadirachta indica*
9	*Commiphora mukul*
10	*Dioscorea deltodia, Terminalia arjuna, Asparagus racemosa, Saraca indica*
	Cissus quadrangularis
11	*Dioscorea deltodia, Ocimum basilicum, Tribulus terrestris, Saxifraga ligulate*
	Crataeva religiosa, Achyranthes aspera, Rubia cordifolia, Mimosa pudica
	Dolichos biflorus, Cyperus retundus
12	*Withania somnifera*
13	*Curcuma longa, Zingiber officinale, Cochicum luteum, Withania somnifera*
	Pine apple, *Boswellia serrata*
14	*Convolvulus pluricaulis, Terminalia arjuna, Cyperus retundus*

140°C for 2 h. The solution was further oxidized with perchloric acid (3 mL) at 240°C until clear solution obtained and finally made up the volume to 100 mL using distilled-deionized water. Stored the solution in acid washed container. The analysis of each metal was carried out in triplicate for precision of results. The metal concentrations were calculated from each replicate absorbance value, which was then used to calculate an average metal concentration. Metal concentrations were expressed in mg/kg on dry weight basis of sample.

Results and Discussion

The heavy metal content in the spiked and non spiked samples was determined by above described method. Values were compared to calculate percent recovery. It is evident from Table 2 that recoveries for all the metals except lead were within the range 82%–100% which is considered satisfactory (ICH Guideline, 1994, http://www.ich.org). The detection limits of Hg and As using HVG were 0.020 and 0.005 μg/mL and Pb, Cd and Zn using flame method were 1 μg/mL, 0.2 and 0.1 μg/mL.

Concentration of heavy metal in herbal drug sample investigated are presented in Table 3. Concentrations of cadmium were low in nearly all analyzed samples, ranging from 0.11 to 0.48 mg/kg. Cadmium was well below the permissible limit (1.5 mg/kg) of the Prevention of Food Adulteration Acts & Rules (PFA) in almost all the samples. Only two sample exceeded permissible limit (0.3 mg/kg) of World Health Organization (WHO)/Food & Drug Administration (FDA). The cadmium concentration in

Table 2 Method recoveries

Element	Base value (mg/Kg)	Quantity added (mg/Kg)	Quantity found (mg/Kg)[a]	Recovery (%)[b]
Cd	0.38	10	8.55 ± 1.1	82
Pb	–	10	5.95 ± 0.94	59.5
As	–	0.010	0.009 ± 0.0005	90
Hg	–	0.020	0.019 ± 0.004	95
Zn	31.84	10	41.88 ± 1.34	100

Recovery test

[a] Mean ± SD (n = 3)

[b] 100* [(Found-base)/added]

Table 3 The level of metals concentration in herbal drugs

Herbal Drugs	Elements concentration (mg/Kg)[a]				
	Cd	Pb	As	Hg	Zn
Sample 1	BDL	BDL	BDL	BDL	16 ± 1.8
Sample 2	BDL	BDL	BDL	BDL	15 ± 2.0
Sample 3	BDL	BDL	BDL	BDL	27 ± 4.5
Sample 4	BDL	BDL	BDL	BDL	31.5 ± 3.4
Sample 5	BDL	BDL	BDL	BDL	6.73 ± 0.31
Sample 6	BDL	BDL	1.45 ± 0.3	BDL	7 ± 0.66
Sample 7	0.48 ± 0.11	6 ± 0.66	8.34 ± 0.54	BDL	22 ± 1.0
Sample 8	0.11 ± 0.01	4 ± 0.7	BDL	BDL	32 ± 2
Sample 9	0.38 ± 0.11	2.5 ± 0.58	BDL	BDL	21 ± 0.17
Sample 10	BDL	BDL	BDL	BDL	24 ± 0.30
Sample 11	BDL	4 ± 0.80	BDL	BDL	29 ± 3.0
Sample 12	0.16 ± 0.04	BDL	0.31 ± 0.07	BDL	16 ± 0.70
Sample 13	0.11	BDL	0.75 ± 0.1	BDL	6.45 ± 1.55
Sample 14	0.29 ± 0.08	BDL	0.72 ± 0.07	BDL	14 ± 1.66
Regulatory Limits					
PFA	1.5	10	5.0	1.0	50
WHO/FDA	0.3	10	10	1.0	50

[a] The value represents the mean ± SD. (n = 3)

BDL: Below detection limit

herbs was related to pH value and Zn concentration of soil. Low pH and Zn concentration in soil enhanced the transfer of Cd from soil to herbs (Wu et al. 2008).

The lead concentration in 4 samples was in the range of 2.5–6.0 mg/kg. This may be the good indicator of Pb contamination level contributed by the vehicle exhaust as the raw material samples were taken from the road sides. The values of Pb are below the prescribed limits i.e. 10 mg/kg set by the regulatory authorities (WHO 2003). The arsenic was analyzed on the HVG coupled with AAS and result revealed that five of fourteen samples were contaminated by arsenic in the range 0.31–8.34 mg/kg. Only one sample (8.34 mg/kg) was found beyond the permissible limits of PFA and rest below the permissible limit i.e. 10 mg/kg of WHO/FDA.

With regard to Zinc content was found in the range of 6.45–32 mg/kg in all samples. According to the PFA, WHO/FDA, permissible limits Zn in herbal drugs is 50 mg/kg. The results revealed that zinc concentration (6.45–32 mg/kg) in all the samples was within the permissible limit as prescribed by PFA and WHO/FDA. However, Zn metal is essential micronutrient, but it also dangerous beyond the permissible limits (Somers 1974; Schumacher et al. 1991). The mercury was analyzed on the HVG-AAS and the result showed that all samples were free from mercury contamination. The similar result was reported in the literature that the herbal samples from Western Himalayan region are free from Hg contamination (Vartika et al. 2008).

In conclusion, the recent increasing demand of herbal drugs and possible access to such drugs by common people it is important to establish the identity, purity and quality assurance of herbal drugs for their acceptance by the consumer. Due to various factors such as soil, agroclimatic conditions, anthropogenic activities, presence and distribution of heavy metals is inevitable, therefore, their assimilation in herbs is very likely. The present study reveals the heavy metal profile in various herbal drugs prepared in the Himalayan region. Such studies are useful to estimate the level of metal contamination in herbal drugs to assure their quality.

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