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Heavy Metal Accumulation in Some Herbal Drugs

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Abstract

Accumulation of heavy metals, namely Pb, Cd, Cu, and Zn, was estimated in some of the important herbal drugs of India, viz., *Alpinia galanga, Artemisia parviflora, Butea monosperma, Coleus forskohlii, Curcuma amada, Euphorbia prostrata, Leucas aspera, Malaxis acuminata* and *Pueraria tuberosa.* A total of 34 samples, genuine as well as market samples of aforesaid plant species, were procured for estimation of heavy metals. The concentration of Pb and Cd was found beyond the WHO permissible limits in most of the samples studied. It is also notable that difference in heavy metal concentration in a particular plant species collected from different regions is related to the site from where the samples were collected. For example, Junagadh sample of *Artemisia parviflora* leaves accumulated more Pb and Zn as compared to the Tarikhet sample. This may be due to higher industrial activity at Junagadh district of Gujrat State. Similarly, Lucknow samples of *Alpinia galanga* and *Curcuma amada* showed maximum concentrations of Pb, Cd, Cu and Zn, as compared to the other regional samples of the same species, and the air pollution due to automobiles is quite high in this city.

Keywords: Heavy metals, herbal drugs, market samples.

Introduction

In the present day scenario, the importance of herbal drugs is increasing due to their lesser side effects and acceptability to the majority of the population of third world countries. Thus, there is an urgent need to establish the identity, purity and quality assurance of herbal drugs in order to have full efficacy and safety of the herbal products. The presence and wide distribution of heavy metals in soil, due to geoclimatic conditions and environmental pollution is inevitable, therefore, their assimilation in plants is obvious. It has been reported that traces of cadmium and lead can be detected in all plants and food stuffs (Piscator, 1985; Sherlock et al., 1983), and have a toxicological significance too, whereas some metals like copper and zinc, etc., are essential micronutrients for plants, but these metals can also be dangerous beyond the permissible limits (Somers, 1974; Schumacher et al., 1991). The World Health Organization in a number of resolutions has also emphasized the need to ensure the quality control of plant products by using modern techniques and applying suitable standards (WHO, 1992). Heavy metals are considered harmful for human beings. For instance, Cd causes osteomalacia and pyelonephritis. Likewise, Pb may cause renal tumors and other carcinomas. However, Cu and Zn are less toxic than former ones.

Unfortunately, very little work is on record regarding heavy metal accumulation in herbal drugs or medicinal plants. For example, Wong et al. (1993) estimated the concentrations of nine metals (Cd, Co, Cu, Fe, Mn, Ni, Pb, Zn and Hg) in 42 Chinese herbal medicinal plants. Similarly, Kwapulinski et al. (1996) and Chizzola and Franz (1996) determined the heavy metal concentrations of various medicinal plant species growing in Poland and Austria, respectively. However, in India, no such study is reported in herbal drugs. Parmar et al. (1993) showed that medicinal values of *Agave americana*, *Sambucus nigra* and *Silybum marianum* used in Homeopathic systems may be due to the presence of Ca, Cr, Cu, Fe, Mg, K and Zn. Thus, the present study has great potential and significance in herbal drug research.

In the present communication, estimation of Pb, Cd, Cu and Zn accumulation in some herbal drugs was undertaken.

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The species selected for study are *Alpinia galanga* Willd. ("Kulanjan"), *Artemisia parviflora* Roxb. ("Masipachchai"), *Butea monosperma* Kuntze ("Palash"), *Coleus forskohlii* Briq. ("Gandira"), *Curcuma amada* Roxb. ("Amra Haridra"), *Euphorbia prostrata* W. Ait. ("Dudhika"), *Leucas aspera* Link ("Thumbai"), *Malaxis acuminata* D. Don ("Jeevak") and *Pueraria tuberosa* DC. ("Vidarikand"), due to their importance in indigenous systems of medicine (Table 1). These drugs are also included in Ayurvedic Pharmacopoeia of India.

Materials and methods

The plant parts which have medicinal value were collected from different parts of India and an attempt was also made to procure market samples sold under the same vernacular names for estimation of heavy metals (Table 1). A total of 34 samples belonging to 9 different plant species were analyzed. Plants were washed in fresh running water to eliminate dust, dirt and possible parasites, and then they were washed again with deionized water (Zurera et al., 1987). Each completely dried sample (1 g) was digested in concentrated nitric acid and perchloric acid (3 : 1) until a clear solution was obtained. After adequate cooling, solutions were reconstituted to the desired volume, i.e., 25 ml, with deionized water, and stored in test tubes. All necessary precautions were adopted to avoid possible contamination of the samples. The prepared samples were analyzed on an atomic absorption spectrophotometer (Perkin Elmer 5000). Hollow cathode lamps were employed for detection of lead, cadmium, copper and zinc. The instrument was caliberated with standard solutions using the concentration mode.

Results and discussion

Heavy metals as environmental contaminants is not a recent phenomenon. They are ubiquitous in trace concentrations in soil and vegetation. In fact, many are required by plants and animals as micronutrients. The man made sources of metal contamination are mainly associated with certain industrial activities, agriculture practices, automobile emission, coal fired generating plants, municipal incinerators, etc. In the

Table 1. Procurement of material and medicinal properties.

Plant species	Part used	Genuine (Collected)	Commercial	Medicinal properties
Alpinia galanga Willd. "Kulanjan" (Zingiberaceae)	Rhizome	Lucknow	Bombay Delhi Jammu	Stomachic, carminative, cardiodepressant, used in rheumatism and catarrhal affections.
Artemisia parviflora Roxb.* "Masipachchai" (Asteraceae)	Leaves	Junagadh Tarikhet	–	Diuretic and antiviral
Butea monosperma Kuntze "Palash" (Fabaceae)	Seeds	Allahabad Ranikhet	Aligarh Bombay Pune	Aperient and rubefacient.
Coleus forskohlii Briq.* "Gandira" (Lamiaceae)	Roots	Agrakhal Tarikhet Vijaywada	–	Cardioactive, hypotensive, used in constipation.
Curcuma amada Roxb. "Amra Hardra" (Zingiberaceae)	Rhizome	Lucknow Tirunelveli	Delhi Nagpur	Stomachic, carminative used in bruises and sprains.
Euphorbia prostrata W. Ait. "Dudhika" (Euphorbiaceae)	Whole plant	Lucknow Tarikhet Tirunelveli	Patiala	Stimulant, astringent, anthelmintic and laxative.
Leucas aspera Link* "Thumbai" (Lamiaceae)	Whole plant	Banglore Bhubaneswar Calcutta Thiruvananthapuram Tirunelveli	–	Antipyretic, stimulant, expectorant, aperient, diaphoretic and used in chronic rheumatism.
Malaxis acuminata D. Don "Jeevak" (Orchidaceae)	Tubers	Ranikhet Tarikhet	Aligarh Bombay	Aphrodisiac, febrifuge and have a cooling effect.
Pueraria tuberosa DC. "Vidarikand" (Fabaceae)	Tubers	Dehra Dun Mandi	Delhi	Aphrodisiac, tonic, galactogogue, diuretic and cures leprosy.

* Crude drugs were not available in the markets of India surveyed.

present communication, heavy metal concentrations were estimated in collected as well as in commercial samples of some important herbal drugs of India. Table 1 lists the place of procurement as well as medicinal properties of herbal drugs, which are included in the indigenous Pharmacopoeia. Tables 2 and 3 show the mean concentration values of various metals, like Pb, Cd, Cu and Zn, in herbal drugs.

From the present study, it was revealed that all the metals accumulated to a greater or lesser extent by all the nine plant species studied. The minimum Pb concentration was found in *Alpinia galanga* (Bombay market), and the maximum in *Artemisia parviflora* (Junagadh), which interestingly also accumulated maximum concentrations of Zn and Cd. The minimum concentration of Cu and Zn was found in Delhi market sample of *Pueraria tuberosa*. Maximum Cu was present in the *Curcuma amada* Lucknow sample.

According to the WHO, permissible limits for medicinal plants, based on ADI (Acceptable Daily Intake) for Pb and Cd are 10 ppm and 0.3 ppm, respectively. As Cu and Zn are considered micronutrients, WHO limits for these metals have not yet been established. However, Bowen (1966) and Allaway (1968) studied the range of different metals in uncontaminated agricultural crop plants and found that Cu ranges from 4 to 15 and Zn from 15 to 200 ppm (dry weight). After comparing these limits with our results, it was found that most of the plant samples have heavy metal accumulation beyond these ranges. However, the Zn concentration in all the samples studied was found to be within the range. Out of 34 samples studied, only eight samples, namely, *Coleus forskohlii* (Vijaywada), *Malaxis acuminata* (Tarikhet and Ranikhet), *Alpinia galanga* (Bombay), *Curcuma amada* (Nagpur, Delhi and Tirunelveli), *Butea monosperma* (Allahabad) accumulated Pb within the permissible limit, i.e., less than 10 ppm. Similarly, the concentration of Cd in the majority of the samples, namely, *Artemisia parviflora* (Tarikhet and Junagadh), *Coleus forskohlii* (Vijaywada), *Curcuma amada* (Lucknow and Tirunelveli), *Pueraria tuberosa* (Mandi and Dehra Dun), and all samples of *Leucas aspera*, was beyond the permissible limit, i.e., 0.3 ppm. Cu accumulation in *Coleus forskohlii* (Agrakhal), *Curcuma amada* (Lucknow and Nagpur), *Leucas aspera* (Banglore and Thiruvananthapuram), *Artemisia parviflora* (Junagadh and Tarikhet), *Euphorbia prostrata* (Lucknow and Tarikhet) and all samples of *Butea monosperma* exceeded the prescribed range, i.e., 15 ppm.

Difference in heavy metal concentration in plants from different regions are related to the site from where the samples are collected. It is quite evident from Tables 2 and 3 that accumulation of various heavy metals vary in the same species of plant collected from different places of India. For instance, the leaves of *A. parviflora* collected from the Junagadh district of Gujrat state accumulated Pb and Zn, 99.33 and 144.28 ppm, respectively, as compared to the Tarikhet sample, which has Pb, 13.75 and Zn, 72.55 ppm. This may be due to higher industrial activity at Junagadh. Similarly, the Lucknow sample of *Alpinia galanga* and *Curcuma amada* has maximum concentrations of Pb, Cd, Cu and Zn in comparison with three other samples of the same species. Lucknow samples of these species were collected from National Botanical Research Institute, which is situated in

Table 2. Heavy metal (Pb, Cd, Cu, Zn) concentrations [ppm (dw)] in underground parts of some herbal drugs.

Herbal Drug	Place	Pb	Cd	Cu	Zn
Alpinia galanga	Bombay*	2.92 ± 1.82	0.08 ± 0.117	4.33 ± 0.235	28.66 ± 0.656
	Delhi*	14.0 ± 1.42	0.08 ± 0.117	5.66 ± 0.311	33.00 ± 0.353
	Jammu*	26.66 ± 2.18	0.08 ± 0.117	5.00 ± 0.204	35.25 ± 0.408
	Lucknow	43.25 ± 2.5	0.166 ± 0.117	9.00 ± 0.735	39.78 ± 0.967
Coleus forskohlii	Agrakhal	46.16 ± 5.92	0.33 ± 0.117	17.16 ± 0.656	80.64 ± 0.344
	Tarikhet	39.75 ± 8.72	0.33 ± 0.117	13.66 ± 0.235	67.22 ± 2.41
	Vijaywada	7.58 ± 0.92	0.50 ± 0.0	15.33 ± 0.656	27.41 ± 1.12
Curcuma amada	Delhi*	5.08 ± 1.35	0.25 ± 0.204	15.08 ± 0.424	59.66 ± 1.74
	Lucknow	19.5 ± 3.0	0.92 ± 0.117	28.58 ± 1.55	71.50 ± 2.22
	Nagpur*	7.83 ± 0.513	0.25 ± 0.204	18.25 ± 1.33	65.15 ± 2.12
	Tirunelveli	9.0 ± 1.59	0.42 ± 0.117	15.16 ± 1.23	24.08 ± 0.471
Malaxis acuminata	Aligarh*	95.58 ± 9.20	0.16 ± 0.117	10.5 ± 1.33	30.75 ± 3.08
	Bombay*	20.33 ± 2.16	0.08 ± 0.117	7.41 ± 0.117	25.42 ± 0.589
	Ranikhet	3.66 ± 0.84	0.16 ± 0.117	7.66 ± 0.589	18.42 ± 0.849
	Tarikhet	4.75 ± 0.61	0.08 ± 0.117	7.33 ± 0.471	23.75 ± 0.707
Pueraria tuberosa	Dehra Dun	21.75 ± 1.42	1.16 ± 0.117	6.5 ± 0.204	21.66 ± 0.424
	Delhi*	11.08 ± 1.24	0.33 ± 0.311	2.0 ± 0.0	8.75 ± 0.204
	Mandi	20.33 ± 0.82	1.33 ± 0.117	5.41 ± 0.117	30.17 ± 1.89

Values are arithmetic mean ± SD of 3 determinations in each case.
* Market samples.

Table 3. Heavy metal (Pb, Cd, Cu, Zn) concentrations [ppm (dw)] in aerial parts of some herbal drugs.

Herbal Drug	Place	Pb	Cd	Cu	Zn
Artemisia parviflora	Junagadh	99.33 ± 7.56	1.41 ± 0.117	17.91 ± 0.513	144.28 ± 1.62
	Tarikhet	13.75 ± 1.22	1.08 ± 0.117	22.16 ± 1.16	72.55 ± 5.38
Butea monosperma	Allahabad	8.08 ± 1.71	0.33 ± 0.117	24.66 ± 0.772	58.54 ± 1.49
	Aligarh*	10.83 ± 0.716	0.33 ± 0.117	27.91 ± 0.942	56.36 ± 2.26
	Bombay*	10.66 ± 1.49	0.16 ± 0.117	28.50 ± 0.816	56.99 ± 1.93
	Pune*	14.66 ± 2.97	0.16 ± 0.117	23.83 ± 1.44	52.16 ± 1.65
	Ranikhet	27.03 ± 1.36	0.25 ± 0.0	21.58 ± 1.47	54.66 ± 3.52
Euphorbia prostrata	Lucknow	14.50 ± 1.96	0.66 ± 0.235	16.08 ± 0.311	74.95 ± 5.34
	Patiala*	17.75 ± 2.01	0.25 ± 0.204	12.25 ± 0.54	45.82 ± 2.95
	Tarikhet	16.75 ± 0.612	0.33 ± 0.235	17.16 ± 1.64	65.78 ± 0.315
	Tirunelveli	13.0 ± 1.87	0.66 ± 0.117	12.58 ± 1.73	34.50 ± 1.59
Leucas aspera	Banglore	22.0 ± 0.35	0.83 ± 0.117	17.58 ± 0.623	48.03 ± 0.659
	Bhubaneswar	21.04 ± 3.36	0.66 ± 0.117	11.58 ± 0.235	67.85 ± 2.62
	Calcutta	16.00 ± 2.40	0.75 ± 0.0	8.8 ± 1.23	52.11 ± 0.846
	Thiruvananthapuram	52.83 ± 5.51	0.66 ± 0.117	18.16 ± 0.772	47.89 ± 3.34
	Tirunelveli	27.16 ± 0.23	0.66 ± 0.117	13.33 ± 0.513	50.58 ± 1.53

Values are arithmetic mean ± SD of 3 determinations in each case.
* Market samples.

the heart of the city with higher vehicular pollution, and this may be the chief reason of higher concentration of aforesaid metals. It is interesting to note that some samples procured from Bombay and Delhi markets have lesser accumulation of heavy metals inspite of these cities being marked as highly polluted. However, it may be possible that these samples were collected from less polluted areas and were sent to the markets of these cities.

From the present study, it was revealed that the detection of heavy metal accumulation is highly relevant for the assessment of herbal drugs quality. This study has provided a status of heavy metal concentration in various herbal drugs sold in Indian markets. Thus, it is concluded that there should be periodical assessment of heavy metal concentration in all herbal drugs, genuine as well as market samples, in order to have quality assurance and safer use of herbal products.

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