Heavy Metals in Chinese Therapeutic Foods and Herbs

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Summary: In the present study 15 samples of Chinese therapeutic foods and herbs that are frequently consumed by people in both the East and West are analyzed, for the content of cadmium, mercury, lead, arsenic, cuprum and zinc, by atomic absorption spectrophotometer. The results showed that the highest mean value for Cd(0.49 $mg{\cdot}kg^{-1}$), Hg(0.34 $mg{\cdot}kg^{-1}$), Pb(9.01 $mg{\cdot}kg^{-1}$), As(3.26 $mg{\cdot}kg^{-1}$), Cu(33.56 $mg{\cdot}kg^{-1}$) and Zn(38.32 $mg{\cdot}kg^{-1}$) were found in *Radix Salviae Miltitorrhizae*, *Radix Puerariae*, *Radix Salviae Miltitorrhizae* , *Radix Sophorae Flavescentis* , *Fructus Crataegi* , *Herba andrographis* , respectively. The Cd levels of two samples and Cu levels of one sample were found to be higher than the recommended values of Food and Drug Administration (FDA) and World Health Organization (WHO), and may constitute a health hazard for consumers. Mercury, lead, and arsenic levels of one sample were higher than the recommended limit of China Pharmacopoeia, but it were below the WHO and FDA. All other heavy metals in other medicinal plants were found below the recommended tolerable limits.

Introduction

Herbal medicines have a history of recorded use in healing over thousands of years, and are still used widely in many cultures around the world. According to a World Health Organization (WHO) survey cited by Chan, about three-quarters of the world's population relies on on-conventional medicine in their primary healthcare, mainly of herbal origins [1]. Especially Chinese herbs, herbal extracts and formulized herbal compounds have been increasingly used throughout the world for the prevention and treatment of diseases. Chinese herbs are reputed to restore the body's proper balance to maintain optimum health. Some herbs have been used both as food supplements and as medicines [2]. The sale of such herbal medicines and products has increased dramatically in developed countries over the last decade. It should be noted that more than half of the medically important pharmaceutical drugs are either natural products or derivatives of natural products [3]

However, the safety of their use has recently been questioned due to the reports of illness and fatalities [4]. Poisonings associated with the presence of toxic metals in medicinal plants were reported in Asia, Europe and the United States [5-7].The major source of metal contamination is from their presence in the soil (including contamination of the plant material with soil, water or air [8]. High levels of toxic metals can occur in medicinal preparations when they are used as active ingredients, as in the case of Pb and Hg in some Chinese, Mexican and Indian medicines [9, 10] or when the plants are grown in polluted areas, such as near roadways or metal mining and smelting operations. In addition, high levels can be found when agricultural expedients are used, including cadmium containing fertilizers, organic mercury or lead based pesticides, and contaminated irrigation water [11]. Recently a study conducted on ayurvedic medicines by Harvard Medical School [12] also reported that some of these drugs had potentially harmful levels of lead, mercury and arsenic. Additional sources of heavy metal contamination are rainfall, traffic density, and 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 [13].

Heavy metals may cause serious health hazards such as renal failure, symptoms of chronic toxicity and liver damage [14]. 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 to the mark, to meet the criteria needed to support its use worldwide [15]. Thus, it becomes imperative that quality of these drugs must be maintained and it should be free from any type of contamination.

In this work, we analyzed 15 samples of the most commonly consumed therapeutic foods and herbs by the population of China, for the presence of cadmium, mercury, lead, arsenic, cuprum and zinc. This investigation is important to estimate the level of metal contamination in some herbal drugs to assure their quality.

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Results and discussion

Fifteen Chinese therapeutic foods and herbs analyzed for heavy metals, local name, parts used and medicinal properties are given in Table-1. A total of 6 metals (Cd, Pb, As, Hg, Zn and Cu) were determined in the Chinese therapeutic foods and herbs samples by AAS. Table-2 shows the mean concentration of trace metals in the medicinal herbal plants under study. The presence of certain heavy metals at trace levels in medicinal plants may be correlated with therapeutic properties against various health disorders. Whereas, increased levels of heavy metals have been known to cause various health disorders. So many countries and institutes have published some standards for heavy metals content of herbal medicines. The permissible limit for some heavy metals in different regulatory systems is shown in Table-3 [16].

Cadmium (Cd)

The Cd concentrations varied from <0.05 to 0.49 mg·kg^{-1}. Among 15 Chinese therapeutic foods and herbs, four herbal medicines (*Lotus Nelumbinis*, *Flos Carthami Tinctorii*, *Flos Lonicerae Japonicae*, *Aloe*) have not detected cadmium. Mean cadmium level of *Fructus lycii*, *Rhizoma Gastrodiae Elatae* and *Radix Salviae Miltitorrhizae* exceeded the WHO, FDA and Pharmacopoeia of China recommended limit of 0.3μg.g^{-1} other was well below the permissible limit. Higher level of cadmium poses serious toxicological impacts on human health and ingestion via food, especially plant-based foodstuffs. The kidney, especially the renal tract, is the critical organ of intoxication after exposure to cadmium. Excretion is slow, and renal accumulation of Cd may result in irreversible impairment in the re-absorption capacity of renal tubules [17].

Copper (Cu)

Cu concentration varied between 2.30 and 33.56 mg·kg^{-1}. Only *Radix Sophorae Flavescentis* has no Cu. *Fructus Crataegi* contains the highest level of Cu and *Flos Carthami Tinctorii* contains the lowest. The Cu level of *Fructus Crataegi*, *Flos Lonicerae Japonicae*, *Radix Rhizoma Polygoni Cuspidati* exceeded the permissible limits 20.0 mg.kg^{-1} of WHO and FDA. The results of the present investigation have shown that copper concentration is not a matter of concern from the toxicity point of view for the observed herbs.

Mercury (Hg)

There are 8 herbal medicines which were free from mercury contamination in this study. Hg concentration varied between 0.06 and 0.34 mg·kg^{-1}. The highest concentration of Hg was 0.34 mg.kg^{-1} in *RadixPuerariae*, which is lower than the limit of 1 mg.kg^{-1} recommended in drugs, in FDA and WHO. But the mercury level of *Radix Puerariae* and *Folium Ginkgo Biloba* exceeded the permissible limit of 0.2 mg·kg^{-1} in Pharmacopoeia of China. Mercury exposure for the general population occurs mainly from consumption of fish, as methyl mercury [18, 19] and possibly from dental amalgam fillings (WHO,2003), and it is unlikely that the exposure through medicinal herbs will affect human health.

Lead (Pb)

It is appeared from the observed data that the lead concentration varies between 0.09 and 9.01 mg·kg^{-1}, except for Rhizoma Gastrodiae Elatae and Herba andrographis, which were free from lead contamination. The values of Pb are below the prescribed limits i.e 10 mg·kg^{-1} set by the regulatory authorities WHO and FDA. Only one sample(*Radix Salviae Miltitorrhizae*) Pb level exceeded the limit of 5 mg·kg^{-1} by Pharmacopoeia of China. The Lead in herbal drugs was due mainly to aerial deposition or absorption by external parts. Exposure to Pb is of concern mainly because of its acute toxicity even at trace levels and numerous studies have revealed that it can adversely affect the central and peripheral nervous system, growth and development, cognitive development, renal system, blood circulation, mental retardation, reproductive health and even can cause death [20, 21].

Arsenic (As)

The arsenic was analyzed with AAS and result revealed that six of fifteen samples were contaminated by arsenic in the range 0.25–3.26 mg·kg^{-1}. Only two samples (*Lotus Nelumbinis* and *Radix Sophorae Flavescentis*) were found beyond the permissible limits of Pharmacopoeia of China (2.0 mg·kg^{-1}) , but all the concentration levels of As below the permissible limit i.e. 10 mg/kg of WHO/FDA.

Zinc (Zn)

The Zn concentration varied between 2.34 and 38.32 mg·kg^{-1} and most of the samples contain Zn concentration in the range of 15 to 30 mg·kg^{-1}. According to the WHO and FDA, permissible limits Zn in herbal medicines is 50 mg·kg^{-1}. The results revealed that zinc concentration in all the samples was within the permissible limit as prescribed by WHO and FDA. However, Zn metal is an essential element for humans, but it also dangerous beyond the permissible limits [22].

Table-1: 15 Chinese Therapeutic Foods and Herbs under investigation(name, parts and medicinal).

Sample	Latin Name	English Name	Chinese name	Part used	Medicinal properties
1	Radix glycyrrhizae	Liquoric root	Gancao	Root	treatment of sore throats, coughs and bronchial catarrh. allergic reactions, rheumatism and arthritis. prevent liver toxicity.
2	Fructus lycii	Barbary wolfberry fruit	Gouqi	Fruit	dyspeptic complaints and gallstones
3	Flos Trollii	Chinese Globeflower flower	Jinlianhua	Flower	toothache, minor infections of the mouth and skin. sore throats and coughs
4	Radix Puerariae	Kudzuvine root, pueraria	Ge Gen	Root	Vents muscles and papules, abates fever, generates body fluids, stop diarrhea, initial stage of measles,
5	Rhizoma Gastrodiae Elatae	Gastrodia tuber	Tian Ma	Stems	Convulsions, spasm, dizziness, headache, numbness of the limb.
6	Fructus Crataegi	Hawthorn fruit, crataegus	Shan Zha	Fruit	Indigestion, food accumulation, diarrhea and dysentery, postpartum pain in the abdomen.
7	Lotus Nelumbinis	Weeping forsythiae capsule	Lianqiao	Fruit	coronary heart disease ，nocturnal calf-muscle spasms, oedema.
8	Flos Carthami Tinctorii	Safflower flower, carthamus	Hong Hua	Flower	Painful menstruation, menstrual block, postpartum pain in the abdomen, abdominal masses, severe heart pain.
9	Folium Ginkgo Biloba	Gingko biloba leaf	Yin Xing Ye	Leaves ,seed	Chest fullness sensation and pain, chronic cough, dysentery and vaginal discharge.
10	Flos Lonicerae Japonicae	Honeysuckle flower, lonicera	Jin Yin Hua	Flower	Infections, dysentery
11	Radix Salviae Miltitorrhizae	Salvia root	Dan Shen	Root	Menstrual irregularities, painful menstruation, menstrual block,pain in the chest, palpitations and insomnia.
12	Radix Sophorae Flavescentis	Lightyellow sophora root	Ku Shen	Root	Jaundice, dysentery, vaginal discharge, itchy skin, scabies andtinea, eczema, hemorrhoids.
13	Radix Rhizoma Polygoni Cuspidati	Japanese knotweed	Hu Zhang	Root, leaves	Jaundice, vaginal discharge, pain in the sinews and bones,menstrual block, retention of lochia.
14	Herba andrographis	Common andrographis herb	Chuanxinlian	Leaves, stems	bacillary dysentery, bronchitis, carbuncles, colitis, coughs, fevers, malaria, mouth ulcers,
15	Aloe	Aloes	Luhui	leaves	constipation. seborrhoeic dermatitis, peptic ulcers, fungal infections, and for reduction of blood sugar levels

Table-2: Concentration of elements in Chinese Therapeutic Foods and Herbs.

Herbal drugs	Elements concentration ($mg \cdot kg^{-1}$) (mean±SD n=5)					
	Cd	Cu	Hg	Pb	As	Zn
Radix glycyrrhizae	0.22±0.02	8.34±0.34	0.06±0.01	0.72±0.04	—	21.34±1.45
Fructus lycii	0.31±0.04	6.25±0.48	0.11±0.01	0.69±0.04	—	20.24±1.02
Flos Trollii	0.05±0.01	9.12±1.21	—	0.56±0.03	—	6.89±0.81
Radix Puerariae	0.17±0.03	9.64±0.98	0.34±0.04	2.34±0.03	—	17.98±0.74
Rhizoma Gastrodiae Elatae	0.36±0.01	3.26±0.16	—	—	0.25±0.01	2.34±0.02
Fructus Crataegi	0.09±0.01	33.56±1.27	—	3.21±0.27	1.03±0.38	18.12±0.69
Lotus Nelumbinis	—	6.52±0.43	0.05±0.01	0.54±0.02	2.28±0.41	—
Flos Carthami Tinctorii	—	2.30±0.08	—	0.14±0.03	1.36±0.26	6.05±0.59
Folium Ginkgo Biloba	0.07±0.04	12.47±2.42	0.31±0.02	1.28±0.24	—	2.37±0.21
Flos Lonicerae Japonicae	—	22.34±2.15	0.06±0.02	2.39±0.36	—	15.01±0.75
Radix Salviae Miltitorrhizae	0.49±0.01	16.14±3.21	—	9.01±0.28	—	15.31±1.34
Radix Sophorae Flavescentis	0.17±0.04	—	0.12±0.03	1.16±0.13	3.26±0.24	32.64±2.12
Radix Rhizoma Polygoni Cuspidati	0.14±0.02	28.38±3.57	—	1.22±0.14	—	28.46±3.62
Herba andrographis	0.06±0.03	9.78±2.31	—	—	—	38.32±2.03
Aloe	—	13.26±3.19	—	0.09±0.01	0.78±0.04	24.38±2.58

Table-3: Permissible limit for some heavy metal in herbal drugs.

Test for heavy/ toxic metals	WHO	US Food and Drugs Administration(FDA)	Health Science Authority (HSA)singpore	Pharmacopoeia of china
cadmium	0.2ppm	0.3ppm	0.05ppm	0.3ppm
lead	10.0 ppm	10.0 ppm	20.0 ppm	5.0 ppm
arsenic	10 ppm	10 ppm	5 ppm	2.0 ppm
mercury	1.00 ppm	1.00 ppm	0.5 ppm	0.2 ppm
Cuprum	20.0 ppm	20.0 ppm	150.0 ppm	20.0 ppm
zinc	50 ppm	50 ppm	—	—

Experimental

Fifteen samples of powdered dry or dried extract of therapeutic foods and herbs were donated by local stores. Table-1 shows the plant Latin, English and Chinese alphabetical names and their application, the form of the sample analyzed. The samples (approximately 100 g) arrived in the laboratory in plastic bags and were kept at room temperature until analyzed.

All the materials and glassware were used of Analytical grade. Ultrapure water was used for all analytical work. All glassware was washed with 2% Extran solution, soaked in 3N HCl for 24 h, and rinsed with ultrapure water before use. Metal standard solution of Cd, Hg, Pb, Cu, As and Zn, containing 1000mg.L^{-1} of each metal were obtained from Merck (Darmstadt, Germany).

Heavy metals were analyzed in plant samples by atomic absorption spectrometry (AAS), The AAS analysis was performed on an AFS-230E (manufactured by Beijing Haiguang Instrument Company, China). The methods described by Gomez *et al* [23, 24] were used for the preparation of sample and analysis. In summary, the samples were ground and sieved to ensure a particle size <20um, the powdered samples were dried for 24hrs at a temperature of 104 □ and stored in polyethylene bags prior to analyses. About 2 g dried powder sample of each herb was digested in a Teflon vessel with 20ml of cone. HNO_3 near to dryness and then 10ml $HCLO_4$, HF was added to digest the silica salt until white fumes were observed, the residue was quantitatively transferred to 25 ml volumetric flask and made up to volume with ultrapure water. 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^{-1} on dry weight basis of sample.

Conclusions

The population growth in the world and the increasing interest in the industrialized nations have greatly expanded the demand for medicinal plants and their products. Approximately 80% of the world population uses the medicinal plants. Due to various factors such as soil, agro-climatic conditions, anthropogenic activities, presence and distribution of heavy metals is inevitable, therefore, their assimilation in herbs is very likely. So it is important to establish the identity, purity and quality assurance of herbal drugs for their acceptance by the consumer. The present study reveals the heavy metal profile in various herbal drugs prepared in China. Such studies are useful to estimate the level of metal contamination in herbal drugs to assure their safety and quality.

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