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REGULAR ARTICLE

HEAVY METALS CONTAMINATION IN HERBAL PLANTS FROM SOME GHANAIAN MARKETS

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ABSTRACT

A study was conducted to investigate the magnitude of heavy metals (arsenic [As], copper [Cu], cadmium [Cd] and mercury [Hg]) contamination that may be present in some Ghanaian medicinal herbs/plants available in local markets and also to compare the levels with recommended levels by the International Organization. A total of 267 samples of herbal plants representing 18 different plants collected from several markets in Ghana were tested for heavy metals contamination. Atomic Absorption Spectrophotometry was used for the analyses, and content of metals per sample was expressed as percent µg/g. The study showed differences in metal concentrations according to the parts analysed (leaf, fruit, root bark and crown). The obtained results which showed the predominance of Cd in almost all the analysed parts of the samples followed by Zn, Cu, As and Hg. However, Hg was the least predominant metal detected in the analyzed samples. All the monitored metals in the herbal plants were within the safe limit approved by Codex Alimentarius Commission and FAO/WHO limit for spices. The findings generally suggest that consumers of these herbal products would not be exposed to any risk associated with the intake of herbal plant products for the management of diseases.

Keywords: Herbal plants, contamination, heavy metals, maximum residue limit, Ghana

INTRODUCTION

The widespread of contamination with heavy metals in the last decade has raised public and scientific interest due to their dangerous effects on human health **(Gilbert, 1984).** This has lead researchers all over the world to study the pollution with heavy metals in the air, water, and foods to avoid their harmful effects and to determine their permissibility for human consumption. The accumulation of heavy metals can have middle-term and long term health risks, and strict periodical surveillance of these contaminants is therefore advisable.

Herbal plants are dried parts of plants widely used as raw materials for pharmaceutical preparations (Galenic products) and as a supplement for dietetic products and especially for 'self-medication' in the general population. Some are often used as diet components often to improve color, aroma, palatability and acceptability of food **(Abou-Arab and Abou Donia, 2001)**.

Most of these herbs consist of rhizomes, barks, leaves, fruits, seeds, and other parts of the plant. Most of these are fragrance, aromatic and pungent. The bulk of the dried material of most herbs contains carbohydrates and organic compounds having diverse functional groups. Some of these herbs have been reported to contain significant quantities of some trace metals **(Al-Eed *et al.*, 1997)**.

Environmental pollution is the main cause of heavy metal contamination in food chain. These metals may reach and contaminant plants, vegetables, fruits and canned foods through air, water, and soil during cultivation **(Hussain *et al.*, 1995)** and also during industrial processing and packaging **(Tsoumbaris and Tsoukali-Papadopoulou, 1994)**.

These trace metals in herbal plants play vital role as structural and functional components of metal proteins and enzymes in living cell. The addition of herbs that may contaminate with trace and heavy metals to food as a habit may result in accumulation of these metals in human organs. Subjecting to trace and heavy metals above the permissible limit affect the human health and may result in illness to human fetus, abortion and preterm labor and mental retardation to children. Adults also may experience high blood pressure, fatigue and kidney and brain troubles **(Hifsa *et al*., 2009)**.

Ghana, like most African countries has a high diversity of plants used as herbal medicine to help meet some of their primary health care needs. In Africa, up to 80 % of the population in rural areas depends on traditional medicine (TM), while in India the corresponding figure is 65 % according to World Health Organization (WHO) estimates **(WHO, 2003)**. Several herbs and spices are either produced on small farmlands or naturally grow in different regions.

There is little information available about the safety of herbal plants and their products in respect to heavy metals contamination. In Ghana, the use of herbal medicines has increased markedly in rural areas because of its potency and it is therefore consumed more than other beverages. Due to the use of enormous amount of herbal medicine, it is important to know the toxic metal contents in this product.

The objective of this work is to investigate the magnitude of heavy metals (arsenic [As], copper [Cu], cadmium [Cd] and mercury [Hg]) contamination in some Ghanaian medicinal herbs/plants available in local markets and also to compare the levels with recommended levels by the International Organization.

MATERIAL AND METHODS

Sample preparation and treatment

A total of 267 samples of fresh Ghanaian medicinal herbs/plants were purchased in open market and herbal dealers in Greater Accra region of Ghana. The samples (Table 1) were transferred into a crucible and oven dried at 105 ^{o}C for 24 h. The dried samples were poured in a mixer grinder taking care not to overheat the sample. Few drops of concentrated nitric acid were added to the solid as an ashing aid. Dry-ashing process was carried out in a muffle furnance by stepwise increase of the temperature up to 550 ^{o}C and then left to ash at this temperature for 4 h. The ash was left to cool and then decompose using concentrated nitric acid (10 ml). The ash suspension was filtered into a 25 ml volumetric flask using Whatman filter paper No. 41 and the solution was completed to the mark using deionized water.

Table 1 Medicinal plant samples collected from several selected markets in Ghana

Common name/Local name	Scientific name	No. of collected items
Leaves		
Bitter leaf (Awonwono)	*Vernonia amygdalina*	9
Fever plant	*Ocinum veride*	7
Awonta	*Diadiulome*	8
Moringa	*Moringa oleifera*	11
Goat weed	*Ageratum conyzoids*	6
Duawusu	*Cassia occidentalis*	12
Aubergine du diable	*Solanum torvum*	10
Velvet tamarind	*Dialium guineense*	11
Prekese	*Tetrapleura tetrafera*	13
Bomma gu woakyi	*Phyllanthus nuriri*	8
Crab's eye	*Abrus pricatorius*	9
Horn-beam-leaved	*Sida acuta*	16
Neem	*Azadiracta indica*	13
Dry zone mahogany	*Khaya senegalensis*	9
Fruit		
Duawusu	*Cassia occidentalis*	12
Aubergine du diable	*Solanum torvum*	9
Prekese	*Tetrapleura tetrafera*	13
Baobab	*Adansonia digitata*	8
Root		
Fever plant(Nunum)	*Ocinum veride*	10
Duawusu	*Cassia occidentalis*	8
Aubergine du diable	*Solanum torvum*	11
Bark		
Bitter leaf(Awonwono)	*Vernonia amygdalina*	6
Baobab	*Adansonia digitata*	9
Neem	*Azadiracta indica*	11
Dry zone mahogany	*Khaya senegalensis*	8
Crown		
Sweet broom weed(Odwankyene)	*Scoparia dulcis*	12
Nsirinsiri	*Diodia rubiscosa*	8

Atomic Absorption Spectrophotometer determination

Analysis of heavy metals of interest was performed using a Varian model AA 240 FS Atomic Absorption Spectrophotometer. Measurements were made using a hollow cathode lamp for arsenic [As], copper [Cu], cadmium [Cd] and mercury [Hg]) at wavelengths of 193.7, 324.7, 228.8, and 253.7 nm respectively. The slit width was adjusted for all metals at 0.5 nm. The calibration curves were prepared from standards by dissolving appropriate amounts of the metal salts in purified nitric acid, diluting with deionised water and storing as stock solutions in a quatz flask. Fresh working solutions were obtained by serial dilution of stock solutions.

Quality assurance

Appropriate quality assurance procedures and precautions were carried out to ensure reliability of the results. The recovery and reproducibility of the method was carried out by spiking and homogenizing several already analyzed samples with varied amounts of standards solutions of the metals and processed as previously described. Reagent blank determinations were used to correct the instrument readings and the limit of detection (LOD) of the analytical method for each metal was calculated as double the standard deviation of a series of measurement of a solution, the concentration of which is distinctly detectable above, but close to blank absorbance measurement **(US EPA, 1983)**. The limit of quantification (LOQ) was determined by preparing two solutions of each sample and three separate readings were made for each solution according to **International Accreditation Criteria for Laboratories Performing Food Chemistry Testing Method (1999)**. Table 3 shows the LODs and LOQs obtained for each element by AAS (Table 2).

Table 2 Average recovery, Limit of detection (LOD) and quantitation (LOQ) of elements assayed by AAS

Metal	Average Recovery±SD	LOD	LOQ
Arsenic (As)	93.5±6.3 %	0.001	0.003
Cadmium (Cd)	89.6±10.2 %	0.002	0.007
Copper (Cu)	94.2±5.8%	0.001	0.003
Zinc (Zn)	91.6±8.9%	0.002	0.007
Mercury (Hg)	94.7±4.8%	0.002	0.007

SD- Standard deviation; LOD- Limit of detection; LOQ- Limit of quantitation

RESULTS AND DISCUSSION

A total of 267 samples of medicinal plant were analyzed for heavy metals. The identities of heavy metals found in the leaf group samples of medicinal plants are given in Table 3-7. Examination of the data show that, in some analyzed samples, heavy metals of some of the tested samples were below the detection limits. The other samples contained variously different levels of the heavy metals. Among the various metals in the leaf samples, Zn and Cd are the predominant metals followed by Cu, Hg and As. As was detected only in *Ageratum conyzoids* whiles Hg was detected in *Vernonia amygdalina* and *Cassia occidentalis* (Table 3).

Table 3 The distribution levels (µg/g + SD) of heavy metals detected in some Ghanaian medicinal plant sample (Leave group)

Leave group	Concentration and range of metals				
	As	Cd	Cu	Hg	Zn
Vernonia amygdalina	<LOD	0.02±0.01	0.24±0.08	0.02±0.01	2.21±1.40
Ocinum veride	<LOD	0.08±0.04	0.11±0.09	<LOD	1.02±0.09
Diadiulome	<LOD	0.06±0.02	0.13±0.07	<LOD	1.32±0.08
Moringa oleifera	<LOD	0.03±0.01	<LOD	<LOD	1.31±0.07
Ageratum conyzoids	0.12±0.07	0.03±0.01	0.23±0.11	<LOD	2.01±0.06
Cassia occidentalis	<LOD	0.05±0.02	0.12±0.08	0.01±0.00	3.11±0.19
Solanum torvum	<LOD	0.02±0.01	0.38±0.13	<LOD	2.31±1.40

The detected levels of metals varied greatly. For instance, the maximum concentration value of Zn concentration (3.11µg/g) was highest in *Cassia occidentalis*. These values were lower than those reported by **Sharma *et al.*, (2006)** and **Islam *et al.*, (2007)**. In the leafy group concentration of Cd, Cu, and Zn were lower during the present study as compared to those obtained by **Sridanhara Chary *et al.*, (2008)** and those reported by **Nkansah and Amoako (2010)** as well as **Singh *et al.*, (2010).** In all the leafy samples with the highest metal concentrations are *Cassia occidentalis* followed by *Solanum torvum, Vernonia amygdalina, Ageratum conyzoids, Diadiulome, Moringa oleifera* and *Ocinum veride.*

Considering the fruit group of the herbal samples (*Cassia occidentalis, Solanum torvum, Tetrapleura tetrafera* and *Adansonia digitata*), Zn concentration was highest in *Solanum torvum* (2.08 µg/g). The observed figure was lower than the one recorded in lady's finger by **Singh *et al*., (2010),** however, within the range of values of 1.3-3.7 µg/g obtained by **Sridanhara Chary *et al*., (2008).** The minimum value of As (0.03 µg/g) was recorded in *Cassia occidentalis* and was below the detection limit in the other tested three samples, notably *Solanum torvum, Tetrapleura tetrafera* and *Adansonia digitata*. The predominant metals detected were Cu, Zn and Cr. The levels of these metals were far lower than those reported by **Nkansah and Amoako (2010)**. However, Hg was below detection limit in all the analyzed fruit group herbal samples (Table 4). In total, *Solanum torvum* was the fruit group with the highest concentration of heavy metal content followed by *Cassia occidentalis, Adansonia digitata* and *Tetrapleura tetrafera.*

Table 4 The distribution levels (µg/g + SD) of heavy metals detected in some Ghanaian medicinal plant samples (Fruit group)

Fruit group	**Concentration and range of metals**				
	As	**Cd**	**Cu**	**Hg**	**Zn**
Cassia occidentalis	0.03±0.01	0.18±0.08	0.14±0.06	<LOD	1.42±0.12
Solanum torvum	<LOD	0.22±0.11	0.33±0.13	<LOD	2.08±1.21
Tetrapleura tetrafera	<LOD	0.55±0.37	0.17±0.09	<LOD	0.62±0.08
Adansonia digitata	<LOD	0.42±0.24	0.18±0.08	<LOD	1.04±0.14

Among the root group, As and Hg were below the detection limit in all the analysed samples (Table 5). Zn was however present in only one sample (*Cassia occidentalis*) whiles Cd and Zn was the predominant metals. The highest concentration value of 0.44 µg/g was achieved by Cd in *Ocinum veride* whiles minimum concentration value of Zn (0.08 µg/g) was observed in the same *Ocinum veride.* However, *Ocinum veride* is a root group sample with the highest concentration of heavy metal content followed by *Solanum torvum* and *Cassia occidentalis* in that order.

Table 5 The distribution levels (µg/g + SD) of heavy metals detected in some Ghanaian medicinal plant samples (Root group)

Root group	**Concentration and range of metals**				
	As	**Cd**	**Cu**	**Hg**	**Zn**
Ocinum veride	<LOD	0.44±0.26	<LOD	<LOD	0.08±0.00
Cassia occidentalis	<LOD	0.14±0.08	0.23±0.15	<LOD	0.10±0.04
Solanum torvum	<LOD	0.44±0.22	<LOD	<LOD	0.06±0.02

The distribution levels of heavy metals detected in the bark of Ghanaian medicinal plants are presented in Table 6. The most predominant metal was Cd, followed by Zn and As, then to Cu which was present in only one sample (*Vernonia amygdalina*). Hg was below the detection limit in all the analysed samples. The maximum concentration of Zn (0.57 µg/g) was recorded in *Azadiracta indica* whiles minimum value of 0.01 µg/g of As was recorded in *Vernonia amygdalina.* The order of bark group samples with the highest concentration of heavy metals is *Vernonia amygdalina, Azadiracta indica, Adansonia digitata* and *Khaya senegalensis.*

Table 6 The distribution levels (µg/g + SD) of heavy metals detected in some Ghanaian medicinal plant samples (Bark group)

Bark group	**Concentration and range of metals**				
	As	**Cd**	**Cu**	**Hg**	**Zn**
Vernonia amygdalina	0.01±0.00	0.51±0.27	0.08±0.02	<LOD	0.04±0.02
Adansonia digitata	<LOD	0.45±0.23	<LOD	<LOD	<LOD
Azadiracta indica	0.04±0.02	0.57±0.31	<LOD	<LOD	0.02±0.01
Khaya senegalensis	<LOD	0.33±0.09	<LOD	<LOD	<LOD

Among the crown group, Cd was the highest with a concentration of 0.52 µg/g whiles Zn achieved a minimum value (0.03 µg/g) in *Diodia rubiscosa*. As and Hg were below detection limit in the two analysed herbal samples (Table 7). Meanwhile *Scoparia dulcis* samples recorded the highest concentration of heavy metals.

Table 7 The distribution levels (µg/g + SD) of heavy metals detected in some Ghanaian medicinal plant samples (Crown group)

Crown group	Concentration and range of metals				
	As	Cd	Cu	Hg	Zn
Scoparia dulcis	<LOD	0.38±0.01	0.51±0.02	<LOD	0.06±0.02
Diodia rubiscosa	<LOD	0.52±0.01	0.09±0.1	<LOD	0.03±0.01

With respect to all the heavy metals, Cd was predominant in all the analyzed samples of herbal medicine followed by Zn, Cu, As and Hg. The overall results indicated clearly that heavy metals are present in Ghanaian medicinal plants and that the contents of these metals were within permissible limits of **Codex Alimentarius Commission (1991)** and **FAO/WHO (1999)** safe limit for spices [Table 8]. Therefore, herbal formulations of these plant species can also be beneficial sources of appropriate and essential trace elements, though care must be taken to avoid As and Hg toxicity, especially in higher doses. Generally most of the herbal plants available on the market are safe for human consumption as far as trace metal levels are concerned.

Table 8 Safe values for As, Cd, Cu, Zn and Hg in some medicinal plants by Codex Alimentarius Commission (1991).

Element	Maximum allowable limits of elements in fruits and vegetables (mg/kg dry weight)
As	0.2
Cd	0.2
Cu	40
Zn	60
Hg	10

10* mg/kg represents FAO/WHO limits for Hg in spices

CONCLUSION

Heavy metals are environmental hazardous and many developed and developing countries have been continuing to monitor the trends of its concentrations in food, spices, herbal medicines and other biota. But maximum residue limit (MRL) for heavy metals in food and other herbal products are currently not in force in most countries. This study shows that there has been some heavy metals contamination in herbal medicines. The analyzed plant species contained safe levels of the heavy metals analyzed and hence may have no adverse effects normally associated with heavy metal toxicity on people who patronize these products for their health needs. It indicates that the present monitoring program that has been conducted only for heavy metals in herbal plants is insufficient. Although heavy metals in herbal plants do not pose any immediate risk to human health so far, a yearly monitoring program for heavy metals in food and other herbal products is a necessity.

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