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ENVIRONMENTAL CHEMISTRY, POLLUTION & WASTE MANAGEMENT | RESEARCH ARTICLE

Heavy metal contents of some medicinal herbs from Kumasi, Ghana

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Abstract: This research was conducted to assess the safety of herbs available in the Kumasi central market with regards to heavy metal contamination. A total of 15 medicinal herbs were randomly collected. The levels of iron (Fe), cadmium (Cd), lead (Pb) and zinc (Zn) were determined by atomic absorption spectrometry. The levels of the metals were in the range of 2.51–7.06 mg/kg for Fe, 0.44–0.89 mg/kg for Pb, 0.21–1.07 mg/kg for Zn and 0.11–0.53 mg/kg for Cd. This shows that most of the metal concentration levels in the herbs were within the WHO maximum permissible limits (WHO MPL) of 15 mg/kg for Fe, 100 mg/kg for Zn and 10 mg/kg for Pb. Four samples, however, had Cd levels above the WHO MPL of 0.3 mg/kg. There was a positive correlation between Cd and Zn. However, there was no correlation between the remaining metals. Considering the levels of heavy metals, the medicinal herb samples investigated are considered unsafe for human consumption.

Subjects: Analytical Chemistry; Environmental Chemistry; Environmental Sciences

Keywords: heavy metals; herbs; medicinal plants; contamination

1. Introduction

Herbal medicines usually refers to plant-derived substances that occur in nature and are utilized with little or no industrial processing for treatment of illnesses (Tilburt & Kaptchuk, 2008). Various plant parts are used in the formulation of herbal medicines. These include, but not limited to, leaves, roots, barks, fruits and seeds. Because of their natural origin, herbal remedies are perceived by people who patronize them to be safer than western pharmaceutical products. The World Health Organization (WHO) reckons that over 80% of the population in Africa and other developing

ABOUT THE AUTHORS

Marian Asantewah Nkansah, James Hawkins Ephraim and Samuel Takyi Hayford belong to the same work group at the Kwame Nkrumah University of Science and Technology. Our research interest spans varied areas of environmental chemistry; including pollution studies and remediation. This current work is part of a nation-wide survey of dust from streets, classrooms and other public places.

Lawrence Sheringham Borquaye is a lecturer at the Kwame Nkrumah University of Science and Technology with research interest in environmental chemistry in natural products, molecular biology and chemical biology.

PUBLIC INTEREST STATEMENT

Majority of the populace in Ghana depend on herbs and herbal formulations for their healthcare needs. There is the general perception that medications from natural or herbal sources are safe relative to orthodox medicine which is sometimes from artificial products. However due to the mode of cultivation, harvest, transportation and preparations of these herbal products, there is the tendency of anthropogenic contamination. This research therefore sought to look at metal content (both essential and toxic) of some common herbs in Ghana.

countries depend on herbal remedies for their healthcare needs (WHO, 2002). For most people in Africa, the high costs of western pharmaceuticals put modern health care services out of their reach and therefore heavily rely on herbal medicine and medicinal plants to meet their primary health care needs. In addition, western pharmaceuticals are most of the time inaccessible to most people in Africa and so herbal medicines have become one of the major options for treating various diseases (Debas, Laxminarayan, & Straus, 2006).

The environmental impact of toxic metals pollution and the accompanying health effects remain a great area of concern. Although trace amounts of some heavy metals are beneficial to human health, the presence beyond certain thresholds tends to be injurious, causing acute or chronic poisoning (Qelik & Oehlenschlager, 2007). Plants have the capacity to accumulate heavy metals in concentrations much higher than metal concentrations in the environment. This ability to bioaccumulate and hence bioconcentrate in plants increase their potential harmful effects (Rao & KumarMeena, 2011; Smical, Hotea, Oros, Juhasz, & Pop, 2008). When consumed beyond a certain threshold, lead (Pb) can increase blood pressure accompanied by debilitating effects to key organs such as the kidney and the brain. Cadmium (Cd) poisoning is associated with a number of respiratory disorders, renal failures and cardiovascular problems. Even though an essential mineral, an overdose of zinc (Zn) can cause fever, nausea and general weakness. Though iron deficiency causes anaemia, too much iron is particularly dangerous in children and could cause gastrointestinal and skin problems (Baby et al., 2010).

In Ghana, herbal medicines have been in use for a long period of time and are used by people of all walks of life. To recognize the important role of herbal medicines in Ghana's health care system, the Ghana Health Service have gradually integrated the use of herbal medicines into mainstream health delivery (Traditional & Alternative Medicine Directorate, 2009). Medicinal plants used in the preparation of herbal medicines are cultivated in various parts of the country. With the increase in mining activities in most parts of the country, medicinal plants are at a higher risk of being contaminated with heavy metals. In addition, medicinal plants can be contaminated with these heavy metals via irrigation with contaminated water, pesticides and fertilizers.

With the proliferation of herbal medicines on the Ghanaian market from both local and international sources, there is an urgent need to assess the extent of exposure to heavy metals as a result of the usage of these herbal medicines. To achieve this goal, the levels of Pb, Cd, Fe and Zn in 15 plants used in the preparation of different herbal medicines in Ghana were determined. These levels were then compared with WHO maximum permissible limits (WHO, 2007a, 2007b) to determine toxicity from herbal medicines due to their consumption.

2. Materials and methods

2.1. Sample collection and preparation

A total of 75 (five sub-samples each of 15 medicinal herb types) fresh commonly used medicinal herbs were obtained from the Kejetia market (also known as the central market) in Kumasi, Ghana. Selection of the herbs used was based on the popularity of the herbs among the general public. Samples were placed in polythene bags and brought to the laboratory. Plant identification was done at the Department of Herbal Medicine, College of Pharmacy and Pharmaceutical Science, Kwame Nkrumah University of Science and Technology (KNUST), Kumasi. The medicinal herbs and parts used for this study are summarized in Table 1. Samples were washed and air-dried for one week at room temperature in dust free environment, then ground with a porcelain mortar and pestle into fine particles. For each medical herb type, 1 g each of the five sub-samples were weighed to obtain 15 composites. The powdered composite samples were placed in plastic containers and stored in a cool dry cupboard prior to analysis.

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Table 1. Medicinal herbs used in this study

Scientific name	Local name	Part of plant used	Therapeutic indication
Vernonia amygdalina	Awonwone	Leaves	Hepatitis B and C
Ocimum viride	Eme	Leaves	Wound dressing, conjunctivitis
Paullinia pinnata	Tuoantini	Leaves	Malaria, skin infection, wound dressing
Azadirachta indica	Neem	Leaves	Fever, malaria, skin infections
Ocimum gratissimum	Nunum leaves	Leaves	Cough, asthma, tuberculosis and pulmonary infections
Momordica charantia	Nyanya	Leaves	Ulcer, piles, colitis, gastritis haemorrhoids
Psidium guajava	Gauva	Leaves	Germs protection and for wounds wash
Alcohornea cordifolia	Gyamma	Leaves	Diarrhoea, amoebic dysentery, bronchitis
Cympopogon citratus	Lemon grass	Leaves	Cold, headache pain, high blood pressure
Taraxacum officinale	Dandelion	Leaves	Liver, skin disease, gall bladder disease
Carica papaya	Pawpaw	Leaves	Malaria, stomach pain, genitourinary ailment
Gossypium herbaceum	Cotton leaf	Leaves	Fever, headache, menstrual disorder
Chromolena odorata	Achempong	Leaves	Wounds healings
Cassia occidentalis	Mofra borodia	Leaves	Heart disease and whooping cough
Garcinia kola	Twepia seed	Seeds	Throat infections, liver disorder

2.2. Sample digestion and analysis

For each medicinal herb, one gram of pre-weighed sample was placed in a 100 mL beaker. Acid mixture of 4 mL of H_2SO_4, 2 mL of $HClO_4$ and 2 mL of HNO_3 (a ratio 2:1:1) were added sequentially. The mixture was heated to 250°C and left at this temperature until a clear solution was obtained. The clear solution was then filtered into a 100 mL volumetric flask using a 0.45 pm pore size membrane filter paper (Whitman filter paper No. 41). The filtrate was topped to the mark with distilled water. Digested samples were transferred into plastic bottles and stored at 4°C prior to analysis.

2.3. Preparation of working solutions

Stock solutions of the metals were prepared from high purity standard solutions of the metals and calibration standards were obtained from the stocks.

2.3.1. Preparation of Fe solutions

A 1,000 ppm iron solution was prepared by dissolving 1 g of iron wire in 50 ml (1 + 1) HCl in a beaker. The resultant solution was transferred into a 1 L volumetric flask and made to the mark with 1% (v/v) HCl.

2.3.2. Preparation of Zn solutions

In order to obtain 1,000 mg/l of zinc solution, 1 g of zinc metal was dissolved in 50 ml (1 + 1) HCl in a beaker and diluted to 1 litre mark with 1% (v/v) HCl.

2.3.3. Preparation of Cd solutions

In order to obtain 1,000 mg/l of cadmium solution, 1 g of cadmium metal was dissolved in 50 ml (1 + 1) HCl in a beaker. The resultant solution was transferred into a 1 L volumetric flask and made to the mark with 1% (v/v) HCl.

2.3.4. Preparation of Pb solutions

A 1000 ppm lead solution was prepared by dissolving 1.598 g of lead nitrate, Pb(NO3)2, in 50 ml solution of 1% HCl in a beaker. The resultant solution was transferred into a 1 L volumetric flask and made to the mark with distilled water.

Table 2. Method validation data

Analyte	Matrix	LOD mg/kg	LOQ mg/kg	% Recovery	SD	RSD %	R^2 of calibration curve
Fe	Herbs	0.01	0.5	100.01	0.02	0.43	0.999
Zn	Herbs	0.01	0.5	98.5	0.024	4.5	0.998
Cd	Herbs	0.005	0.01	98	0.0027	1.35	0.999
Pb	Herbs	0.005	0.01	99.05	0.017	2.4	1.000

Notes: LOD: Limit of detection; LOQ: limit of quantification.

2.4. Method validation

Working standard solutions of concentration 1,000 mg/L were prepared for each of the metals under study using pure standards from Merck, Germany. In order to monitor consistency in the performance of the instrument, standards and blanks were run at regular intervals to ensure reproducibility and all sample analysis were performed in triplicates. Recovery analysis was done by spiking samples with standards of known concentrations. The percent recovery was determined according to Equation (1):

$$\%\,\text{Recovery} = \frac{\text{conc. of spiked sample} - \text{conc. unspiked sample}}{\text{conc. of known spike added}} \times 100\% \tag{1}$$

Information on method validation and quality assurance has been presented in Table 2.

2.5. Instrumentation

Standards and samples were analysed with a VARIAN SPECTRA AA220 Zeeman Atomic Absorption Spectrometer (AAS) (Varian Canada Inc) with a graphite furnace and a D2 lamp for background correction. Fe, Zn, Cd and Pb were determined with their specific hollow cathode lamps at wavelengths of 248.3, 213.0, 228.0 and 283.0 nm, respectively. Integration time was set at 5 s at 5.0 mA lamp current.

2.6. Statistical analysis

Results of the research were analysed for statistical significance using IBM statistical package software 20. Values in the text are shown in tabulated form as mean ± SD with ND as not detectable.

3. Results and discussion

A lot of research has been done on levels of heavy metal contamination of functional foods, medicinal herbs and herbal preparations in various regions of Ghana (Annan, Dickson, Nooni, & Amponsah, 2013; Nkansah & Amoako, 2010; Sarpong & Boateng, 2013). Due to the high risk associated with human consumption of contaminated foods and herbs, it is imperative that an effective continuous monitoring programme is developed. The plant samples used in this study are sold on the market by herbalists who give specific instructions to their customers on preparation and administration. The herbs investigated in this study are presented in Table 1. Information on individual metal concentrations and mean values have been presented in Tables 3.

3.1. Concentration of metals

Lead is one of the most toxic heavy metals which have no nutritive value (Chionyedua, Onwordi, Agbo, & Ogunwande, 2015; Ullah et al., 2012). Progressive exposure to lead results in a decrease in the performance of the nervous system and affects renal clearance (Salawu et al., 2009). Inorganic lead is also a carcinogen and may cause miscarriage in pregnant women. In general, the levels of lead recorded were very low. The mean concentration of Pb in all the medicinal herb samples analysed was 0.67 ± 0.03 mg/kg and the levels ranged from 0.44 ± 0.01 to 0.89 ± 0.01 mg/kg (Table 3). The highest recorded concentration of lead was 0.89 ± 0.01 mg/kg in *T. officinale* (Dandelion), with the lowest recorded in *G. kola* (Twepia seed). The WHO MPL for lead in medicinal plants is 10 mg/kg (WHO 2007a,

Table 3. Levels of heavy metals in medicinal herb samples (*n* = 75)

Name of plant	Abbreviation	Mean ± SD concentration (mg/kg)			
		Fe	Zn	Cd	Pb
Vernonia amygdalina	VA	5.31 ± 0.06	0.21 ± 0.02	0.12 ± 0.01	0.88 ± 0.02
Ocimum viride	OV	4.95 ± 0.02	0.33 ± 0.02	0.14 ± 0.01	0.83 ± 0.02
Paullinia pinnata	PP	3.86 ± 0.02	0.54 ± 0.03	0.52 ± 0.01	0.63 ± 0.04
Azadirachta indica	AI	5.11 ± 0.02	0.37 ± 0.04	ND	0.59 ± 0.01
Ocimum gratissimum	OG	7.06 ± 0.01	0.55 ± 0.04	0.43 ± 0.01	0.74 ± 0.01
Momordica charantia	MC	7.06 ± 0.01	0.35 ± 0.01	0.24 ± 0.01	0.57 ± 0.04
Psiduim guajava	PG	6.22 ± 0.02	0.52 ± 0.05	ND	0.67 ± 0.02
Alcohornea cordifolia	AC	2.95 ± 0.04	0.33 ± 0.01	ND	0.55 ± 0.01
Cymbopog oncitratus	CON	2.51 ± 0.03	0.86 ± 0.03	0.42 ± 0.01	0.66 ± 0.01
Teraxacum officinale	TO	4.83 ± 0.03	0.21 ± 0.03	0.11 ± 0.03	0.89 ± 0.01
Carica papaya	CP	4.88 ± 0.02	0.97 ± 0.01	0.53 ± 0.03	0.85 ± 0.02
Gossypium herbaceum	GH	4.84 ± 0.03	0.37 ± 0.02	0.12 ± 0.02	0.75 ± 0.03
Chromolena odorata	COD	3.53 ± 0.01	0.54 ± 0.03	0.13 ± 0.02	0.77 ± 0.01
Cassia occidentis	COC	4.32 ± 0.02	0.77 ± 0.03	ND	0.62 ± 0.01
Garcinia kola	GK	2.51 ± 0.01	1.07 ± 0.02	0.20 ± 0.01	0.44 ± 0.01
Mean		4.66	0.53	0.02	0.67
Range		2.51–7.06	0.21–1.07	ND–0.53	0.44–0.89
WHO MPL (mg/kg) (WHO, 2007a, 2007b)		15.0	100.0	0.3	10.0

Notes: SD–Standard deviation; ND–not detected.

2007b). All 15 medicinal plant samples analysed were within this permissible limit. Sarpong and Boateng (2013) reported similar results from different samples of unregistered herbal preparations marketed in and around Kumasi, with the levels of Pb below the WHO MPL. Because of its ability to bio-accumulate in biological tissues, patients who use medicinal herbs with even low concentrations of Pb over a long period of time might be at risk of chronic Pb toxicity and should be monitored for any signs of lead poisoning. Plants are particularly efficient at absorbing Pb from the soil and are reported to retain up to 7%. Soil naturally contains up to 50 mg/kg of Pb, although this is significantly higher in rocky (igneous) soils (Sarpong & Boateng, 2013). The medicinal plants used in this work are usually transported to the markets from farms in the forest zones. Annan et al. (2013), attributed the observed levels of Pb beyond WHO MPL to vehicular deposition of the mineral into the soil. The low concentrations observed might be due to the low intake of Pb by these medicinal herbs (Annan et al., 2013).

Another toxic metal analysed was Cd. The WHO MPL of Cd in medicinal herbs is 0.3 mg/kg (WHO 2007a, 2007b). On average, the Cd concentration in samples analysed was 0.20 mg/kg (Table 3). *C. papaya* had the highest concentration of 0.5 mg/kg while it was undetected in four samples. Cadmium may be taken up and accumulated in the leafy tissues of plants. Subsequent ingestion of those plants by humans and animals may then result in accumulation in the liver and kidneys, which can result in kidney damage (Palizban, Badii, Asghari, & Nafchi, 2015). Cadmium adsorbs strongly to organic matter and soil. As such, it is usually unavailable to roots of medicinal plants. Another source of Cd contamination of plants comes from the use of phosphate fertilizers, which contains up to 100 mg/kg of Cd (Sarpong & Boateng, 2013). Four of the samples (*P. pinnata, O. gratissimum, C. citratus* and *C. papaya)* recorded mean concentrations far greater than that permitted by the WHO. In two other samples (*G. kola* and *M. charantia)*, the concentrations were closer to the permissible limit. High Cd levels have been reported by other researchers in some of the samples they worked on (Annan et al., 2013; Sarpong & Boateng, 2013). Similarly high cadmium levels have also been observed for medicinal plants from Iran, Egypt as well as Dubai (Abou-Arab & Abou Donia, 2000;

Dghaim, Al Khatib, Rasool, & Ali Khan, 2015; Ziarati, 2012). The findings of this work and those of previous work suggest Cd contamination might be a general problem that needs to be critically investigated.

Zinc is an essential element; it plays an important role in growth and has a recognized action in more than 300 enzymes by participating in their structure or their catalytic and regulatory action. Zinc deficiency causes growth retardation and hypogonadism. Zinc deficiency may also affect the bone metabolism and gonadal function (Nishi, 1996). The average zinc concentration of the 15 medicinal plant samples analysed was 0.53 ± 0.02 mg/kg (Table 3). The highest concentration was recorded in *G. kola* (1.07 ± 0.02 mg/kg) followed by *C. papaya* (0.97 ± 0.01 mg/kg). Concentrations of Zn recorded were much lower than WHO MPL. These values are relatively low compared to other studies. For instance, a study on the levels of Zn in some medicinal plants from India gave concentrations in the range 21.3–48.5 μg/g (Rathore & Mohit, 2013). A study in Pakistan also gave Zn levels slightly above that of this study in the range of below detection −502 μg/g (Ansari et al., 2004).

Iron is an essential element for human growth and development and an essential component of haemoglobin. It facilitates the oxidation of carbohydrates, proteins and fats to control body weight, which is a very important factor is diabetes management. Iron is necessary for the formation of haemoglobin and also plays an important role in oxygen and electron transfer in the human body. Low iron content causes gastrointestinal infection, nose bleeding and myocardial infection (Maiga, Diallo, Bye, & Paulsen, 2005). Iron's mean concentration was 4.66 ± 0.02 mg/kg (Table 3), the highest of all metals analysed. The concentrations ranged from 2.51 ± 0.03 to 7.06 ± 0.03 mg/kg. The highest concentration was recorded in both *O. gratissimum* as 7.06 ± 0.01 mg/kg and *M. charantia* with C. *citrates* and *G. Kola* having the lowest. In comparison with WHO MPL of 15 mg/kg, all the herbs had Fe content below this limit (WHO 2007a, 2007b). These levels are relatively low compared to iron in most medicinal plants for the treatment of anaemia. A study conducted in Poland on selected medical herbs known for the treatment of anaemia have iron concentrations within the range 6.67–223 μg/g (Konieczyński, Wesołowski, & Rafalski, 2007).

3.2. Correlation between metals

There are significant correlations between the heavy metals in the herbal plant samples under study (Table 4). Fe shows a weak negative correlation with Cd. This is because Cd and Co have been shown to inhibit Fe absorption (Hamilton & Valberg, 1974). This trend has been observed in children with anaemia (iron deficiency) who happen to have high cadmium levels in their blood. High urine levels of cadmium have also been observed with pregnant women with low iron status. There has been an established inverse relationship between cadmium and iron levels in a wide range of people (Silver, Lozoff, & Meeker, 2013). The mechanism of absorption of Cd in roots is similar for all divalent irons including Zn. Cd is a chemical analogue of Zn and plants may not be able to differentiate between the two ions thus indicating the strong positive correlation between these two metals. Since the bivariate Cd and Zn are analogous, they are positively correlated (Yashim, Kehinde Israel, & Hannatu, 2014). Pb is weakly correlated with Fe. This observation is also supported by literature reports of Pb and Fe concentrations being positively correlated in leaf tissue, also due to their shared bivalent nature. This could also be due to emissions from exhaust of vehicles and other anthropogenic activities close to the source of these herbs (Singh, 2005).

Table 4. Pearson correlation coefficients of available heavy metal concentration in herbal plant samples (N = 15), p = 0.05

Parameters	Fe	Zn	Cd	Pb
Fe	1			
Zn	−0.433	1		
Cd	−0.005	0.451	1	
Pb	0.306	−0.370	0.135	1

4. Conclusion

From the study, the medicinal herbs investigated had levels of Pb below detection. However, Cd levels in some of the medicinal herbs were above the WHO permissible limit of 0.3 mg/kg. The high Cd content observed in some samples has the potential of posing health risks to consumers of these herbs. Essential metals Fe and Zn were present in all samples with levels within the WHO permissible limits. Correlation analysis between the metals indicated that there was a positive correlation of 0.451 between cadmium (Cd) and zinc (Zn) but the other metals were weakly or negatively correlated with each other. There is therefore the need of constant monitoring of local medicinal herbs and their formulations by the development of routine programmes in order to safeguard the safety of the population, many of who patronize them.

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