Global Advanced Research Journal of Educational Research and Review (ISSN: 2315-5132) Vol. 2(4) pp. 098-104, April, 2013
Available online http://garj.org/garjerr/index.htm

Full Length Research Paper

Hazardous metals concentrations in traditionally used unregistered herbal drugs sold at six selected Suburbs of Kumasi, Ashanti Region of Ghana

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Accepted 25 April, 2013

The emerging global significance and the possible faster rate of increase in interest of herbal drugs for treating various illnesses cannot be underestimated. This coupled with the associated health risk posed by these drugs due to hazardous metal contamination of various herbs used in the preparation of these drugs gives credence to concerns raised by health conscious people. In this study the Atomic Absorption Spectroscopic analysis was conducted on some selected traditionally used unregistered herbal drugs sold in Kumasi, Ghana to assess the concentrations of four hazardous metals lead (Pb), arsenic (As), cadmium (Cd) and zinc (Zn). Results indicated that with the exception of Cd which generally occurred in higher concentrations (ranging from 0.000 to 2.560 mg/L) than the WHO maximum permissible limits (MPL) of 0.3 mg/L, the others, with WHO MPL of 100 mg/L for Zn, 10 mg/L for Pb and 1.0 mg/L for As were generally lower; As (0.000 mg/L to 1.012 mg/L), Pb (0.001 mg/L to 1.510 mg/L) and Zn (ranging from 0.004 mg/L to 0.830 mg/L) were lower. This, not withstanding poses growth impairment threat. This situation calls for a more effective enforcement measures of the foods and drugs law in the bid to address the problem of consumption of unsafe herbal products.

Keywords: Hazardous metals, concentration, herbal drugs

INTRODUCTION

Herbal drugs play a very important role in the health care delivery systems of many countries. The World Health Organization (WHO) estimates that eighty percent (80%) of the world's population depends on non-conventional medicines mainly of herbal origin for their primary health care needs (Chan, 2003). Herbal medicine enjoys a wider acceptability among people probably because it blends readily and is deeply rooted into the socio-cultural life of the people. It's relatively low cost when compared to allopathic drugs, coupled with the usual difficulty in accessing health care facilities could be the driving forces behind the heavy reliance on herbal drugs by many people for treatment of their sicknesses. Also herbal medicine was the only form of health care available before the advent of modern medicine. Presently, even people close to hospitals consult traditional healers employing herbal drugs as a first choice (Maclean, 1971). Drew and Myers (1997) noted that there has been an increasing growth in popularity of herbal drugs usage in many countries. A reflection of this in local context such as in Ghana is the upsurge in the advertisement and sale of herbal drugs in both electronic and print media. The advertisers claim and stress that herbal drugs are 'natural', safe with no toxic effects. These claims have

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gained acceptance by a portion of the populace.

The Food and Drugs Board (FDB) of Ghana is mandated by law to ascertain the safety and wholesomeness of all food and drug products and duly register them before they are freely sold on the Ghanaian markets by manufacturers and vendors. While some manufacturers and vendors of herbal drugs have complied with the law others have not, causing the markets to be flooded with unregistered herbal drugs.

Unregistered herbal drugs may likely be contaminated with many various substances including banned pesticides, microbes, unknown concentrations of hazardous metals and chemical toxins. Hazardous metals contamination of herbal drugs could result from employing medicinal plants collected from sites where pesticides, herbicides and other harmful agrochemicals were applied (Chan, 2003) as the raw material. The medicinal plants absorb these harmful substances from the environment and store them in their tissues. Other sources of herbal drugs contaminations are water contaminated with hazardous metals used in preparation of the drugs, and the use of utensils containing hazardous metals such as lead, copper and aluminum during manufacture: holding, cooking and storage of the herbal drugs.

According to Gaur and Adholeya (2004), some hazardous metals are micronutrients necessary for plant growth while others such as cadmium (Cd), lead (Pb), and mercury (Hg) have unknown biological functions. Copper (Cu), selenium (Se) and zinc (Zn) are essential to maintain the metabolism processes of the human body (Collin, 2003). Zinc is an important cofactor for several enzymatic reactions in the human body and vitamin B-12 an important nutrient for formation of red blood cells to prevent pernicious anaemia (a very harmful anaemia type), and also for the formation of nucleus has a cobalt (Co) atom as its core. Chromium (Cr), manganese (Mn) and molybdenum (Mo) are required by the body in small amounts for bone and tendon formation, enzyme formation and regulation of sugar use respectively.

Goyer (1996) observed that hazardous metals at concentrations above the WHO MPL may be detrimental to the health of consumers as they adversely affect growth and development, cause cancer, organ and nervous system damage and in extreme cases death. The trivalent methylated arsenic is more efficient at causing DNA breakdown (Tangahu *et al*., 2011). Hazardous metals can also increase the acidity of blood. Markowitz *et al*., (1994) reported that the body draws calcium from the bones to help restore proper blood pH. Several cases of adverse effects of herbal drugs have been reported in many countries which are allegedly caused by taking herbal products containing hazardous metals. The adverse effects include vomiting, stomach upsets, feverishness and sometimes death as a result of intake of herbal drugs containing excessive amounts of hazardous metals (Wong *et al*., 1993).

In Ghana, useful medicinal plants for managing a wide spectrum of diseases such as cold, gastrointestinal troubles, hypertension, cancer, viral infection and other parasitic diseases (Ayitey-Smith, 1989) are employed to prepare herbal drugs sold in various dosage forms such as tablets, capsules, decoctions and creams. The herbal drugs are sold at public places such as lorry terminals, schools, markets, and even at premises of some hospitals. It is reported that the health of about 65% - 70% Ghanaians are managed by traditional/herbal practitioners (The Lancet, 2003). Although may be hazardous, the body requires some of the metals for the proper functioning and control of the various biological processes. The levels of hazardous metals in unregistered herbal drugs are unknown and may pose serious health risks to consumers (Xudong *et. al* 2011). Excessive amounts are toxic to consumers and amounts below the WHO MPL for hazardous metals adversely affect the body. These compounds exert cumulative effect and toxicity even in low quantities on humans (Ozkutlu *et al*, 2006). Therefore, this study, which sought to ascertain the concentrations of hazardous metals (As, Cd, Pb and Zn) in traditionally prepared unregistered herbal drugs sold in Kumasi, a Ghanaian city characterized by brisk socio-economic activities due to its vantage location, would be helpful in ascertaining the risk or otherwise posed by such drugs to users for further mitigation measures to be taken.

MATERIALS AND METHODS

Study Area

Kumasi ($6^{o}35'$N - $40'$N, $1^{o}30'$W) is the capital town of Ashanti Region of Ghana and is located 250-300m above sea level. It is the second most populous city after the nation's capital, Accra. The Ghana Districts (2012) reported that the population of Kumasi is two million, twenty-two thousand nine hundred and nineteen (2,022,919) and covers an area of two hundred and fifty-four square kilometers (254 km^2). Kumasi is relatively at the central location of Ghana and serves as the trading center for all people from the ten regions of the country. The products traded include medicinal plants, foodstuffs and vehicular parts.

Collection of unregistered traditionally used herbal drugs selected for the study

Unregistered traditionally used herbal drug samples (decoctions) claimed to cure various illnesses sold on the market in various suburbs of Kumasi were purchased from vendors. The samples, suburbs, claimed indications, organoleptic properties determined and recorded (Table 1). The samples were kept in polystyrene bottles. Each

Table 1 Selected herbal drugs from Kwadaso, Asafo, Bantama, Kentinkrono, Krobo Odomase and Mbrom

Name of Herbal Drugs	Code	Claimed Indications	Organoleptic Colour	Taste	Odour
Agacious Herbal Mix	K_1	Cough & sore throat	Red	Sweet	Pleasant
Ebenezer Miracle Herbal	K_2	Menstrual pains	Brown	Bitter	Unpleasant
Aduana Blood Tonic	K_3	Anaemia & menstrual pains	Red	Sweet	Pleasant
K2K Ulcerplex	K_4	Peptic ulcer	Red	Bitter	Pleasant
Godbless Herbal	K_5	Malaria	Colourless	Bitter	Unpleasant
Maadwoa Typhoid Herbal	K_6	Typhoid fever	Brown	Sour	Pleasant
Piesie Typhoid Herbal	K_7	Typhoid fever	Red	Sweet	Unpleasant
Supreme Typ. Mix	K_8	Typhoid fever	Wine	Sour	Pleasant
Aduroye Herbal Mix	K_9	Piles & rheumatism	Wine	Bitter	Pleasant
Power Herbal	K_{10}	Sexual weakness	Brown	Sweet	Pleasant
Tatahwe	K_{11}	Piles	Reddish	Sour	Pleasant
Dua ma Aduro	A_1	Diabetes & menstrual pains	Red	Sour	Pleasant
Osaa Nerbal	A_2	Malaria & jaundice	Brown	Sour	Unpleasant
Broni Bitters	A_3	Piles & sexual weakness	Red	Bitter	Pleasant
Sunkwa Bitters	A_4	Typhoid fever	Red	Bitter	Pleasant
Aduropa Mixture	A_5	Fever	Colourless	Sweet	Pleasant
Okunin Mixture	A_6	Piles	Brown	Bitter	Unpleasant
Besohwe Mixture	A_7	Janundice & malaria	Orange	Sour	Pleasant
Abusua Pile Bitters	A_8	Piles & sexual weakness	Brown	Bitter	Pleasant
Nkwanahia	A_9	Anaemia	Red	Sweet	Pleasant
Barima Mixture	A_{10}	Sexual weakness	Brown	Bitter	Pleasant
Enopa Mixture	A_{11}	Menstrual pains	Red	Sour	Pleasant
Suronipa Herbal Mixture	B_1	Ulcer	Brown	Sweet	Pleasant
Susunipaho	B_2	Piles	Brown	Bitter	Pleasant
Kasanipaho Herbal	B_3	Malaria & jaundice	Red	Sour	Pleasant
Sompa Pile Mixture	B_4	Piles	Orange	Sweet	Pleasant
Nkwahia	B_5	Diabetes	Brown	Sour	Pleasant
Okyeame	B_6	Asthma	Red	Bitter	Pleasant
Anuanom Mixture	B_7	Hypertension	Brown	Sweet	Pleasant
Kanawu Mixture	B_8	Menstrual pains	Orange	Sour	Pleasant
Kroye Mixtures	B_9	Sexual weakness	Red	Bitter	Unpleasant
Adomnyame	B_{10}	Diabetes	Brown	Sour	Pleasant
Adeepena	Ke_1	Ulcer	Red	Sweet	Pleasant
Kasamafo Mixture	Ke_2	Migraine	Brown	Sour	Unpleasant
Okunafoagya	Ke_3	Stomachache	Orange	Sour	Pleasant
Odima Herbal Mixture	Ke_4	Sexual weakness	Red	Bitter	Unpleasant
Odofo Mixture	Ke_5	Measles	Brown	Sweet	Pleasant
Mmoborohunu Mixture	Ke_6	Diarrhoea	Brown	Sour	Pleasant
Adasamma Bitters	Ke_7	Gonorrhea	Red	Bitter	Pleasant
Susudua Herbal Mixture	Ke_8	Hypertension	Brown	Sour	Pleasant
Osuani Mixture	Ke_9	Migraine	Red	Bitter	Pleasant
Yadeeya Mixture	Ke_{10}	Sexual weakness	Orange	Bitter	Pleasant
Nsuro Herbal Mixture	KO_1	Diabetes	Red	Sour	Pleasant
Obrapa Herbal	KO_2	Cough	Brown	Bitter	Pleasant
Obaatanpa Mixture	KO_3	Measles	Red	Sour	Unpleasant
Odofo Mixture	KO_4	Sexual weakness	Wine	Sweet	Pleasant
Awieye	KO_5	Hypertention & stroke	Red	Sweet	Pleasant
Gyewani Mixture	KO_6	Impotence	Brown	Sour	Pleasant
Surowodifo Mixture	KO_7	Gonorrhoea	Wine	Sweet	Pleasant
Atanfoaffire	KO_8	Stroke	Red	Sweet	Pleasant
Asuo Mixture	KO_9	Waist pains	Brown	Bitter	Pleasant
Asagyefo	KO_{10}	Waist pains	Wine	Sour	Unpleasant
Nananom Mixture	Mb_1	Toothache	Red	Bitter	Pleasant
Obideaba Bitters	Mb_2	Malaria & jaundice	Red	Sour	Pleasant
Asetenapa Bitters	Mb_3	Impotence	Wine	Bitter	Pleasant
Sarfo Piles Mixture	Mb_4	Waist pains	Colourless	Bitter	Pleasant
Oboafo Bitters	Mb_5	Rheumatism	Brown	Bitter	Pleasant
Oseefo	Mb_6	Skin rashes	Brown	Sweet	Unpleasant
Osorommoa Piles Mixture	Mb_7	Piles	Red	Bitter	Pleasant
Osiakwan	Mb_8	Fibroid	Brown	Sour	Pleasant
Owonta	Mb_9	Stomachache	Brown	Bitter	Pleasant
Odiawaresem	Mb_{10}	Sexual weakness	Red	Bitter	Pleasant

K = KWADASO, A = ASAFO, B = BANTAMA, Ke = KENTINKRONO, KO = KROBO ODUMASE, Mb = MBROM

sample was mixed thoroughly using a multi-mixing shaker to obtain a homogenous sample and stored in a refrigerator.

Digestion and analysis of selected unregistered traditionally used herbal drug samples

The samples (decoctions) were sent to the Council for Scientific and Industrial Research –Soil Research Institute (CSIR-SRI) at Kwadaso, Kumasi for digestion and analysis. Di-acid method of digestion was adopted from the protocol of Perkin Elmer manual for Atomic Absorption Spectrometry. Twenty milliliters (20ml) of each sample was measured and twenty milliliters each of concentrated nitric acid (HNO_3) and perchloric acid ($HClO_4$) in the ratio 9:4 added to each in a 200ml digestion tube. Each mixture was then heated on a block digester until the brown fumes of HNO_3 ceased and the mixture became clear. The digestion was stopped and distilled water added to each sample solutions to obtain a total volume of 20ml.The final solutions were filtered through 0.45µm pore size membrane filter papers (Whatman filter paper No. 41) to obtain a particle-free solutions. Each analyte was poured into a beaker and the capillary dipped into it and aspirated on the VARIAN SPECTRA AA220 Zeeman Atomic Absorption Spectrometer (AAS) (Varian Canada Inc). Determination

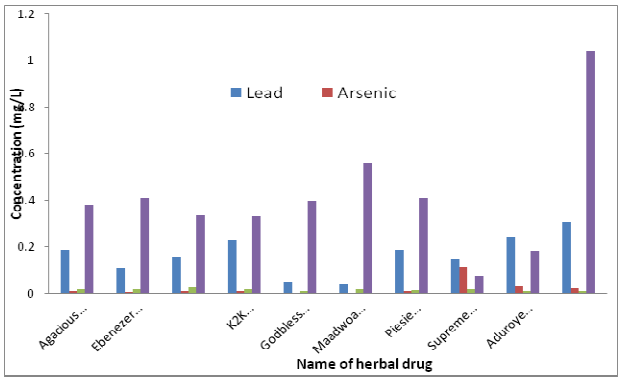

Figure 1 Mean concentration in mg/L of hazardous metals in unregistered traditionally used herbal drugs from Kwadaso, a suburb of Kumasi

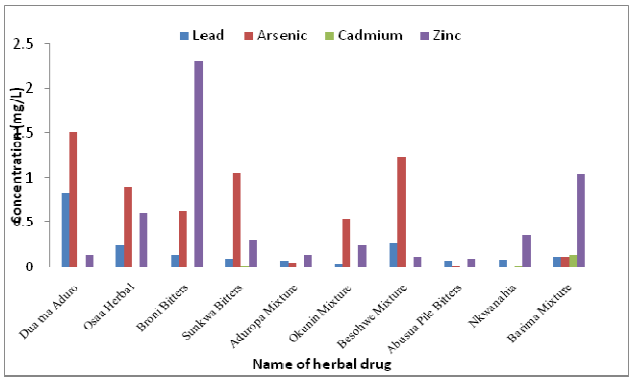

Figure 2 Mean concentration in mg/L of hazardous metals in unregistered traditionally used herbal drugs from Asafo, a suburb of Kumasi.

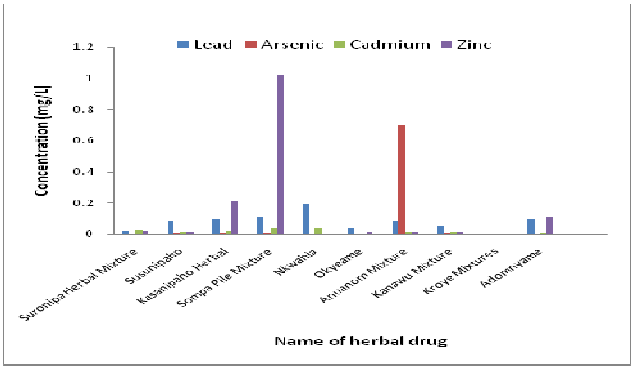

Figure 3 Mean concentration in mg/L of hazardous metals in unregistered traditionally used herbal drugs from Bantama, a suburb of Kumasi.

of the various metals (triplicates) in each sample was then performed and the mean values of samples recorded.

RESULTS AND DISCUSSIONS

Herbal drugs samples were given a range of names dominated by those that express ability to set at liberty from illness and disgrace, love, God's grace and so on. They were of various colours majority of which were brown followed by red probably due to the dry state in which most of these raw materials occurred. They exhibited sweet, sour and bitter taste sensations and were of either pleasant or unpleasant odour. The illnesses claimed to cure included both internal and external illnesses (Table 1). Uptake from the soil and atmospheric deposition are the two main entry routes of

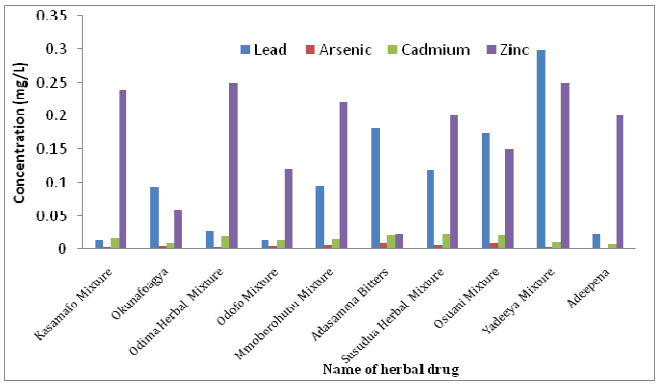

Figure 4 Mean concentration in mg/L of hazardous metals in unregistered traditionally used herbal drugs from Kentinkrono, a suburb of Kumasi.

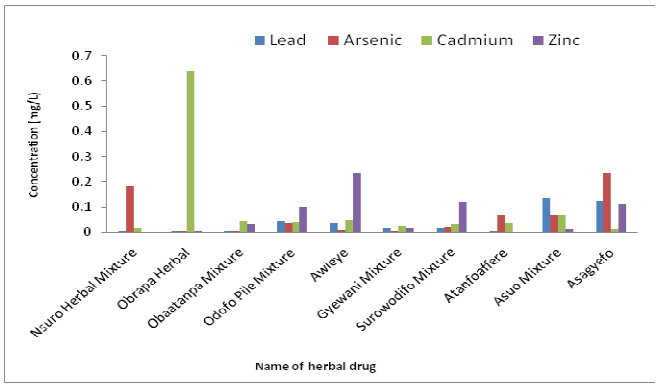

Figure 5 Mean concentration in mg/L of hazardous metals in unregistered traditionally used herbal drugs from Krobo Odumase, a suburb of Kumasi

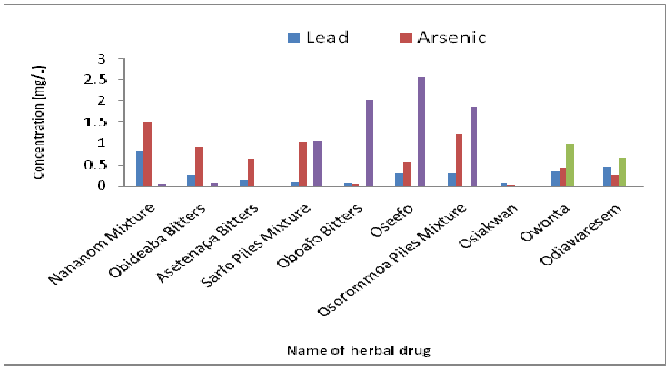

Figure 6 Mean concentration in mg/L of hazardous metals in unregistered traditionally used herbal drugs from Mbrom, a suburb of Kumasi.

hazardous metal in plants.

The mean levels in mg/L of zinc were generally higher than those of the other hazardous metals (Pb, As, and Cd) studied in unregistered traditionally used herbal drugs in the six suburbs of Kumasi (Figures 1 to 6).

Arsenic

There was a relatively wider range of As concentration beginning from 0.001 mg/L as recorded by Godbless Herbal (Figure 1) to 1.510 mg/L by Nananom Mixture (Figure 6) and Dua Ma Aduro (Figure 2) in the studied samples.

Concentrations of hazardous metals such as As recorded in other herbal drugs e. g. Sunkwa Bitters (1.050mg/L) and 1.230mg/L for Besohwe Mixture (Figure 2) and 0.900 mg/L by Obideaba Bitters and Osaa Herbal were also remarkable relative to the 0.002 mg/L by Maadwoa Typhoid Herbal, Suronipa Herbal Mixture,

Nkwahia, Kroye Mixtures and Adomnyame (Figure 3).

According to the World Health Organization (WHO), the safe concentration of As in herbal drug is 1.0 mg/L and the International Agency for Research on Cancer (IARC) has indicated the existence of evidence to conclude that As and As compounds can cause cancer in humans.

Anthropogenic contamination may explain As build-up in herbal drugs. Environmental contamination of As may occur because it is a constituent of agricultural pesticides. The amounts of As found in plants vary depending on such factors as the level of local contamination. The background levels of As in soil and sediment increase if there are natural and/or man-made sources of As contamination present.

The herbs for these drugs were harvested from areas of or close to agricultural activities where evidence of heavy use of pesticides exists. Mishandling, including improper disposal of the containers of these arsenic-containing pesticides could have contributed to the current levels of arsenic in these herbal drugs. Arsenic is found in the natural environment; abundant in the earth's crust and in small quantities in rock, soil, water and air. In weathered rock or soil, it can be picked up and moved by the wind and water. Many As compounds bind to soil and only move short distances when water percolates down through the soil. Plants on land can accumulate As compounds via uptake from soil and/or deposition from air onto their leaves. According to Naidu (2008), the last fate of As is that it is taken up by plants from top soil. Soils contaminated through anthropogenic activity may cause As contents to exceed 50 mg/kg (Ali *et al*., 2003). The As sequestered from soil remained largely in the roots of the plant, making it difficult to harvest for safe disposal.

Cadmium

Apart from Owonta and Odiawaresem which recorded 1.012 mg/L and 0.671 mg/L respectively of Cd concentration, several others including Dua Ma Aduro, Broni Bitters and Nananom Mixture recorded no detectable concentrations. These recorded concentrations were above the WHO MPL of 0.3mg/L and may be due to its natural abundance (at an average concentration of 0.2 mg/kg), causing large quantities to enter the global environment as a result of natural weathering processes (Howari *et. al*, 2004). The range of Cd concentration in the earth's crust has most often been reported from 0.1 to 0.5 ppm, but much higher and much lower values have also been cited depending on a large number of factors (Howari et. al, 2004). Igneous and metamorphic rocks tend to show lower values, from 0.02 to 0.2 ppm whereas sedimentary rocks have much higher values, from 0.1 to 25 ppm.

Phosphate fertilizers contain Cd in amounts of up to 100 mg/kg, which can lead to an increase in the concentration of Cd in soil. In plant products the allowable concentration of Cd ranges between 0.05-0.2 mg/kg. The use of cadmium-containing fertilizers and sewage sludge is most often quoted as the primary reason for the increase in the Cd content of soils over the last 20 to 30 years in Europe (Jensen and Bro-Rasmussen, 1992) Plants may accumulate unwanted substances including Cd (Pavlik, 1997). Leafy plants including herbs used for drugs naturally accumulate higher levels of Cd than do fruits and cereals (Mench *et al*. 1998). Based on the analysis of new data in 2009, The European Food Safety Authority's Panel on contaminants in the food chain has set a reduced tolerable weekly intake (TWI) for Cd of 2.5 micrograms per kilogram of body weight (µg/kg bw). In the absence of scientifically determined dosage for observance as in the present study, the tendency is that Cd concentrations in these unregistered herbal drugs may be beyond this tolerable weekly intake, a situation that calls for immediate concern.

Lead

The concentrations of Pb varied between non-detectable amount (0.000 mg/L) and 0.830mg/L with Kroye Mixture showing the minimum (Figures 4) while Dua Ma aduro (Figure 2) and Nananom Mixture (Figure 6) showed the maximum. Nsuro Herbal Mixture, Obrapa Herbal, Obaatanpa Mixture and Atanfoaffere (Figure 5) also showed quite low levels.

The highest concentration of Pb (0.830mg/L) recorded in the study was lower than the WHO MPL limits of 0.01mg/L. 10.0 mg/L. This therefore suggests that, in their present states, the herbal drugs sampled pose no health threat to human users. Prolonged consumption of these herbal drugs may be detrimental to the users' health. Parker (2001) noted that Pb, a cumulative poison causes damage to the kidney, nervous system, reproductive system, gastrointestinal tract and the blood-forming organs at concentrations in the range 40 – 60 µg/dl in adults and 35 – 50 µg/dl in children. The symptoms include excessive tiredness, weakness, irritability, anorexia, high blood pressure, anxiety, anaemia and Pb colic or acute abdominal pains and impairment of development of intelligence (DWAF, 1996) in children who whom these drugs are administered to. These make such development a matter of concern.

Many governments throughout the world have created appropriate standards for Pb in herbal medicines. For example, Japan allows 20 parts per million (ppm) for total metals in herbal medicines. Lead occurs naturally in soils, typically at concentrations that range from 10 to 50 mg/kg. It occurs naturally in substantial quantities (16 parts per million) in igneous rocks, on the earth's surface (Dharmananda, 2001). Plants are efficient at absorbing soil lead and retain about 7%. Lead is the heaviest of the

non-radioactive metals (atomic number 82; atomic weight 207) and in spite of its weight, stays also in the atmosphere and falls on the soil but due to its low mobility remains in the top inch of soil for absorption by shallow-rooted plants including herbs. Serious source of Pb contamination is the introduction of Pb in the form of tetraethyl and tetramethyl lead into gasoline, most of which are discharged onto the soil and are absorbed particularly by plants that grow along the roadways. The recorded levels of Pb may therefore result from absorption from the soil, weathered rocks, runoff and atmospheric source through the roots and leaves of the herbal plants. Another source that cannot be ruled out is the lead-containing utensils used by manufacturers in the preparation of the herbal drugs.

Zinc

Zinc concentrations in herbal drugs studied ranged from non-detectable (as in Owonta, Asetenapa Bitters and Odiawaresem) to 2.560 mg/L (in Oseefo) (Figure 6). Quite a number (8) of the herbal drugs representing 13.3% had concentrations greater than 1mg/L among which the least is 1.040 mg/L. Concentrations of zinc recorded by others such as Atanfoaffere (Figure 5) and Osiakwan were also quite low (0.001mg/L). All these concentrations were below the Minimum Permissible Level of 100 mg/L (WHO, 2003) suggesting hazardous-metal safety. However, since Zn metabolism and hormonal growth exert significant effect on each other (Nishi, 1996) there is the possibility of impaired or retarded growth due to the low levels of Zn in these herbal drugs if other food and water sources consumed by users are similarly deficient in Zn.

CONCLUSION

The study reveals that, with the exception of Cd, the concentrations of As, Pb and Zn in the studied herbal drugs are below their WHO permissible levels. Viewing these against the health implications associated with deviated levels of hazardous metals, especially those considered in this study, (As, Cd, Pb and Zn) it can be concluded that the selected unregistered herbal drugs manufactured and sold in the Kumasi Metropolis pose health risks particularly in relation to Cd to users.

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