



Research Article

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Heavy metals contamination of some polyherbal products from Lagos state, Nigeria

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ABSTRACT

Background: A “heavy metal” refers to any metallic element that has a high density and is toxic or poisonous even at low concentration. **Aim of study:** To investigate the heavy metal contents of thirty used polyherbal products purchased from different vendors in Lagos State. **Materials and methods:** Using an atomic absorption spectrophotometer, the heavy metals Lead (Pb), Zinc (Zn), Cadmium (Cd), Copper (Cu), Mercury (Hg), Chromium (Cr) and Magnesium (Mg) were tested. **Results:** Pb, Cd, Hg and Cr were absent in polyherbals analyzed in this study. The concentrations of Zn(0.025-0.42) ppm, Cu(0.017-0.31 ppm) and Mg (0.02-0.55) were below WHO/FAO permissible limits. **Conclusion:** This study has shown that the heavy metals content of most poly-herbals sold in Lagos State is below WHO/FAO permissible limits.

Keywords: Polyherbal, Heavy metals, Markets, Lagos state, Nigeria.

INTRODUCTION

Herbal medicines may have multiple physiological activities and could be used to treat variety of disease conditions^[1]. They could be administered in most disease states over a long period without proper dosage, monitoring and toxic effects consideration^[2]. Medicinal plants (used in manufacturing herbal medicines) are contaminated with toxic metals during growth, development and processing^[3]. Some herbal medicines are not well researched and their formulation and sales are unregulated. They may be adulterated and at risk for producing adverse effects and toxicity^[4].

The term “heavy metals” refers to any metallic element that has a high density and is toxic or poisonous even at low concentration^[5, 6]. Heavy metals are present in the environment. But, the dynamic development of industry and motorization, and the continuing over-intensive use of various chemical compounds in agriculture, cause toxic heavy metals in the environment to be on the increase^[7]. The uptake and bioaccumulation of heavy metals in herbs/plants are influenced by some factors; such as climate, atmospheric depositions, amount of heavy metals in soil, nature of soil on which herbs are grown and maturity of the plant at the time of harvest^[8-11]. Elevated levels of heavy metals in plants are reported in areas having; long-term use of treated/ untreated wastewater^[12, 13], plants growing along heavy traffic ways^[14, 15] and earlier dumpsites^[16]. Other anthropogenic sources of heavy metals include additional manures, sewage sludge, fertilizers and pesticides. These may affect the uptake of heavy metals by changing the physico-chemical properties of the soil. Farm lands near heavy traffic high ways are exposed to atmospheric pollution as metal containing aerosols. These may be deposited on soil and are absorbed by plants and/or deposited on leaves, barks and fruits. In Nigeria, most Trado-medicine practitioners’ sell their products along busy traffic urban centers. Herbs, barks and roots used for various ailments are displayed outside their stores exposing them to air-borne heavy metals contamination^[17]. Machine blended herbs are exposed to heavy metal contamination^[18]. Urban activity may contribute to elevated heavy metal loads in atmospheric deposits and so, contaminate ready-to-use herbal products^[19].

Study Area

The study was conducted in major markets in Lagos State. The study area is shown in the map.

MATERIALS AND METHODS

Samples collection

Thirty poly-herbal preparation samples were purchased from different markets in Lagos State. Purchased

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samples were taken to the laboratory for heavy metals analysis.

Heavy metals analysis

Equipment used for the heavy metal analysis include; weighing balance (GH-200 AND), Digestion block (Foss SE 263), Digestion tube 100 ml and pipette 20 ml.

The Reagent used for the digestion of sample is Nitric acid (Laboratory grade reidel – de Haen) 65%, 1.40. Methods described by [20, 21 and 22] were followed for heavy metal analysis.

Statistical Analysis

The results are presented as Mean \pm SEM (standard error of mean) and n represents the number of replicates used in each experiment. Data were analyzed with ordinary one-way ANOVA and multiple comparisons using Graph pad computer software version 6.0.

RESULTS

The compositions, indications and dosage of the thirty poly-herbal samples purchased from major markets in Lagos State are shown in Table 1. Some poly-herbals compositions were not disclosed by the sellers and manufacturers. Table 2 tests the concentrations of heavy metals associated with poly-herbal products.

Heavy metals tested in this study included lead (Pb), zinc (Zn), cadmium (Cd), copper (Cu), Mercury (Hg), chromium (Cr) and magnesium (Mg). Pb, Cd, Hg and Cr were absent in polyherbals analyzed while concentrations of Zn ranged from 0.025ppm in H27 to 0.42ppm in H11; Cu concentration in ppm ranged from 0.017 in H5 to 0.31 in H23; while that of Mg ranged from 0.02 ppm in H17 and H23 to 0.55 in H30.

Table 1: Composition, indication and dosage of commonly consumed poly-herbal in Lagos state.

Poly-herbal samples	Compositions	Indications	Dosage
H1	<i>Aloe vera, Allium sativum, Zingiber officinale, Eremomastax speciosa, Garcinia kola.</i>	Malaria, typhoid.	One cup twice a day.
H2	<i>Gossypium hirsutum, Eremomastax speciosa</i>	Irregular and painful menstruation.	One cup daily.
H3	Not disclosed.	Diabetes, chronic ulcer, pile, malaria, typhoid, worm expellant, itching, nettle rash, Eczema. Rheumatic pain, hypertension, eye infection.	Adult; two tablespoons daily.
H4	<i>Callichilia barteri, Pachylobus edulis, Lecaniodiscus cupanioides, Allium sativum, Zingiber officinale, Monodoramyristica, Khaya ivorensis, Piper nigrum, Eugenia caryophyllata</i>	Energy booster, general over all well-being.	Adult; Four tablespoons daily.
H5	Not disclosed.	Acute stomach ache, painful and irregular menstruation	Two tablespoons daily.
H6	Not disclosed.	Typhoid	Two cups twice daily.
H7	Not disclosed.	Typhoid, malaria,	One cup three times daily
H8	<i>Saccharum officinarum, Ocimum basilicum</i>	Diabetes, stomach disorder, pile.	Four tablespoons daily.
H9	Not disclosed.	Dysentery, cough, pile, convulsion, chest pain, snake or scorpion bite, stomach ache, rheumatism	One tablespoon daily.
H10	Not disclosed	Whooping cough, tuberculosis.	Two tablespoon twice daily.
H11	Not disclosed	Malaria, typhoid, general body well-being.	One cup daily.
H12	Not disclosed	Diabetes	One cup twice daily.
H13	<i>Azadirachta indica, Citrus aurantiifolia, Eremomastax speciosa, Garcinia kola, Newbouldia leavis.</i>	Malaria and typhoid.	One cup three times daily.
H14	Not disclosed.	Malaria	One cup twice daily
H15	<i>Newbouldia leavis, Eremomastax speciosa, Callichilia barteri.</i>	All sexually transmitted diseases. Immune booster.	Two tablespoons twice daily.
H16	<i>Gossypium hirsutum, Eremomastax speciosa and Aloe vera</i>	Irregular menstruation.	Four tablespoons daily.
H17	Not disclosed	Pile, hemorrhoid, waist pain, constipation, irregular menstruation, obesity.	Four tablespoons daily, after meal.
H18	<i>Ocimum gratissimum, Vernonia amygdalina, Eremomastax speciosa.</i>	Vagina itching, burning and irritation, chronic pile and aids digestion.	Two cups daily.
H19	<i>Psidium guajava, Carica papaya, Mangifera indica, Citrus aurantiifolia.</i>	Malaria	Two cups twice daily.
H20	<i>Citrus aurantifolia, Garcinia kola, Cymbopogon citratus.</i>	Malaria.	Two cups twice daily.
H21	<i>Moringa oleifera, Zingiber officinale, Allium sativum, Garcinia kola.</i>	Malaria, typhoid.	Two cups twice daily.
H22	Not disclosed.	Pile, dysentery.	Four tablespoons daily.
H23	Not disclosed	Vagina discharge, itching, burning sensation.	Five tablespoons daily
H24	<i>Sorghum bicolor, Khaya grandifoliola, Cassia</i>	Pile, dysentery, constipation, diarrhea, irregular	Adult; four tablespoons twice

	<i>sieberiana, Staudtia stipitata, Alstonia cognensis, Ocimum basilicum, Mangifera indica, Cyathula prostrate, Securidaca longepedunculata, Saccharum officinarum.</i>	menstruation, men turgidity, withdraws protruding rectum, waist and stomach ache.	daily, children; two tablespoons twice daily.
H25	<i>Cassia siamea, Cassia alata, Cassia augustifolia, Aloe vera, Gongronema latifolium, Anthocleista djalensis, Moringa lucida, Citrus lanatus, Xylopha aethiopica, Khaya senegalensis, Garcinia kola.</i>	Infertility, Menstrual dysfunction, STD, Mouth odor, Acute stomach ache, Obesity,	Adult; Four tablespoons daily. Children; One tablespoon daily
H26	<i>Eremomastax speciosa, Garcinia kola, Allium sativum, Irvingia gabonensis,</i>	Stomach disorder, indigestion, peptic/duodenal ulcer, improves blood circulation, eliminates pain and discomfort.	Adult; two tablespoons daily after meal. Children; one tablespoon daily after meal.
H27	Not disclosed.	Chronic hypertension, palpitation of the heart.	One cup daily.
H28	<i>Syzygium aromaticum, Piper nigrum, Microdesmis puberula, Euphorbia lateriflora, Uvaria chamae, Securidaca longepedunculata, Treculia africana, Allium sativum, Zingiber officinale,</i>	Pile, Hemorrhoid, Waist pain, Enhances effective digestion, Reduces fatigue, Improve libido, Strengthens the heart and cardiac muscles, Reduces blood sugar.	Adult; Four tablespoons daily. Children; two tablespoons daily.
H29	<i>Khaya grandifolia, Croton lobatus, Anthocleista vogelii, Eremomastax speciosa</i>	Glaucoma, Eye ulcer, cataract.	Adult; Four tablespoons daily every two days. Children; One tablespoon daily every two days.
H30	<i>Oxalyscorpioidea, Kigelia africana, Securidaca longepedunculata, Allium ascalonicum, Eremomastax speciosa.</i>	Purifies the blood, Eliminates toxic material from the blood vessels, Reduces accumulated fat and cholesterol level, prevents oedema, numbness, and reduces blood sugar.	Adult; five tablespoons daily. Children; one tablespoon every two days.

Table 2: Heavy metals evaluation of polyherbal products.

Polyherbal products	Pb (ppm)	Zn (ppm)	Cd (ppm)	Cu (ppm)	Hg (ppm)	Cr (ppm)	Mg (ppm)
H1	ND	0.24±0.03	ND	0.25±0.10	ND	ND	ND
H2	ND	0.16±0.01	ND	0.04±0.05	ND	ND	0.30±0.01
H3	ND	0.26±0.10	ND	0.21±0.02	ND	ND	ND
H4	ND	0.40±0.05	ND	0.20±0.04	ND	ND	ND
H5	ND	0.34±0.02	ND	0.02±0.02	ND	ND	ND
H6	ND	0.16±0.02	ND	0.22±0.03	ND	ND	0.30±0.01
H7	ND	0.41±0.10	ND	0.24±0.03	ND	ND	0.24±0.02
H8	ND	0.35±0.01	ND	0.13±0.02	ND	ND	ND
H9	ND	0.25±0.05	ND	0.16±0.03	ND	ND	ND
H10	ND	0.35±0.01	ND	0.40±0.00	ND	ND	0.10±0.04
H11	ND	0.42±0.02	ND	0.25±0.01	ND	ND	0.06±0.05
H12	ND	0.33±0.01	ND	0.03±0.02	ND	ND	0.30±0.01
H13	ND	0.27±0.02	ND	0.21±0.01	ND	ND	ND
H14	ND	0.14±0.02	ND	0.15±0.03	ND	ND	ND
H15	ND	0.15±0.02	ND	0.05±0.03	ND	ND	ND
H16	ND	0.18±0.00	ND	0.14±0.01	ND	ND	0.05±0.10
H17	ND	0.22±0.03	ND	0.01±0.06	ND	ND	0.02±0.03

H18	ND	0.41±0.03	ND	0.12±0.06	ND	ND	0.05±0.01
H19	ND	0.24±0.03	ND	0.14±0.01	ND	ND	0.14±0.01
H20	ND	0.28±0.02	ND	0.24±0.01	ND	ND	ND
H21	ND	0.31±0.05	ND	0.23±0.02	ND	ND	ND
H22	ND	0.4±0.02	ND	0.15±0.01	ND	ND	ND
H23	ND	0.25±0.30	ND	0.31±0.15	ND	ND	0.02±0.10
H24	ND	0.30±0.03	ND	0.20±0.01	ND	ND	ND
H25	ND	0.40±0.12	ND	0.60±0.06	ND	ND	ND
H26	ND	0.20±0.07	ND	0.10±0.35	ND	ND	ND
H27	ND	0.03±0.02	ND	0.15±0.01	ND	ND	0.50±0.03
H28	ND	0.25±0.02	ND	0.10±0.10	ND	ND	0.50±0.04
H29	ND	0.20±0.06	ND	0.15±0.03	ND	ND	ND
H30	ND	0.35±0.05	ND	0.25±0.01	ND	ND	0.55±0.10

Key: Values are Mean±SEM; n=3. ND= Not Determined.

DISCUSSION

Atmosphere and soil are being polluted with chemicals and heavy metals due to dynamic development of industries and motorization along with extensive use of pesticides and fertilizers. These pollutants and heavy metal deposits get into the human food chain via plant parts and/or extracts

Lead (Pb) is hazardous to plants, animals and microorganisms. Continuous consumption of fertilizers, sewage sludge and fuel combustion are the major reasons leading to escalation in Pb pollution. The permissible limit of lead (Pb) is 10ppm as defined by WHO [23]. In this study, Lead which causes renal failure and liver damage in humans, was not detected in the samples analyzed [24, 25]. Cadmium (Cd) is another hazardous heavy metal which can reduce plant yield at concentrations ranging from 5–30 mg/kg. Cadmium is gaining more attention due to wide occurrence in water, milk, dietary products, soil, herbal products and medicinal plants. The main sources leading to accumulation of cadmium in soil and plants are phosphate fertilizers, lead, non-ferrous smelters, and zinc mines, combustion of fossil fuels and sewage sludge application. The samples analyzed in this study had no Cadmium. The permissible limit of Cadmium is 0.03ppm recommended by WHO [23].

Zinc (Zn) is an essential part of thousands of proteins in plants, although toxic in excess quantities. In this study, the Zinc content in the samples range from 0.03ppm in sample H27 to 0.42ppm in sample H11. These values are low compared to the dietary limit of Zinc, (100ppm). This result corroborates with the research findings of [6, 26]. Zn has a protective effect against both cadmium and lead [27]. A deficiency of Zn is marked by loss of taste, retarded growth and hypogonadism, leading to decrease in fertility. Zn toxicity is rare, but at high concentrations, it may induce toxicity characterized by symptoms of muscular stiffness, irritability, loss of appetite and pain, and nausea.

Concentration of copper ranges from 0.01 ppm in sample H17 to 0.31 ppm in samples H23. The concentrations in the samples were below the permissible limit of 2ppm recommended by WHO [23]. Copper is an essential part of several enzymes and it is necessary for the synthesis

of hemoglobin, deficiency can lead to anemia and hypo-proteinemia [28].

Magnesium (Mg) ranged from 0.02ppm in H17 and H23 to 0.55 ppm in H30. Permissible limit for Mg is 10ppm WHO [23]. Mg deficiency is a detrimental plant disorder that occurs most often in acidic, light sandy soils, where magnesium can be leached away. Without enough magnesium, plants degrade the chlorophyll in the old leaves. This shows the main symptom of magnesium deficiency; chlorosis, or yellowing between leaf veins [24, 25].

Canada adopts 0.2ppm as limit of mercury (Hg) in raw herbal material, since 0.02 mg/day is the permissible limit in finished herbal products. China and Singapore use 0.5ppm limit as proposed by WHO [23]. In this study, Hg was not present in the samples tested. This result complies with a study carried out to assess the level of heavy metal concentration in four selected medicinal plants locally consumed in Kura Local Government Area of Kano State, Nigeria [29]. Mean concentration of heavy metals in the selected medicinal plants were 0.0177mg/Kg for Zn, 0.0385mg/Kg (Ni), 0.0136 mg/Kg (Pb), 0.0192 mg/Kg (Co), 0.0185mg/Kg (Fe), 0.0364mg/Kg (Cu) and 0.0011mg/Kg (Cr). The mean concentration of the heavy metals in the plant samples was within the permissible limit of the recommended range by WHO/FAO. Results in this study corroborates with those reported by [22], in which Mn, Zn, Fe, Cr, Ni, Hg and Pb were analyzed in medicinal plants sold in some local markets in Benin City, Edo state.

In contrast to the result, findings of [19], showed that the Zn, Cd and Pb content of selected ready-to-use herbal remedies in South-east Nigeria were beyond WHO permissible limits. The concentration levels of Pb, Cd were high and above the safe limits set by WHO/FAO [23]. Tunya B. fil, Virgy-virgy worm expellant and Sekin powder had Zn concentrations above international safe limits representing 20% of the tested herbal remedies. The consumers are exposed to heavy metal poisoning. It is important that regulatory bodies should intensify efforts to decrease human exposure risk. Also, the investigation carried out by Kulhari [31] on the chemical profiling of nine heavy metals (Mn, Cr, Pb, Fe, Cd, Co, Zn, Ni and Hg) using the stem and leaf samples of ten medicinal plants is in variance to results in this study. Concentration of heavy

metals apart from Cr, was within permissible limits in the tested stem and leaf samples. Other two most perilous metals Pb (2.64 ± 0.0260) and Cd (0.04 ± 0.0274) were reported in Bahadurgarh region, although below permissible limits. Concentration of Hg remained at low levels in the leaf and stem samples tested. These results suggest that cultivation of medicinal plants and other dietary herbs should be curtailed near polluted industrial areas to avoid health hazards. Contamination of polyherbals by these heavy metals may have resulted from the soil where these herbs are cultivated or from atmospheric deposit on leaves^[19, 32]. The absence of Pb, Cd and Cr in these herbal products reflects the safety of these products as these metals are poisonous to humans^[33].

Twenty four different Nigerian herbal remedies (NHR) in two types of pharmaceutical dosage forms-liquid and capsules were studied using basket market protocol in the Niger Delta, Nigeria. The remedies were digested using concentrated aqua regia HCl: HNO₃ (3:1) and arsenic, cadmium, chromium, cobalt, lead and nickel were tested for using an Atomic Absorption Spectrophotometer. Arsenic, cadmium, chromium, cobalt, lead and nickel contents were matched with the recommended limits of the World Health Organization (WHO), European Union (EU) and United States Environmental Protection Agency (USEPA). The highest concentrations of the heavy metals were noticed in the solid dosage forms while the lowest concentrations were observed in the liquid preparations. The study showed the percentage violation of the WHO and EU limits for the six metals were arsenic (0%); cadmium (58.3%); chromium (4.16%); lead (54.1%). The study highlights the need for pharma-vigilance and in-depth risk assessment with regards to metal-toxicity of Nigerian herbal remedies to understand the severity of the problem^[34]. According to^[35], presence of Cr, Mn, Ni, Co, Cu, Cd, Zn and Pb were tested in four of the most consumed vegetables in the Southern part of Nigeria. The metal analysis results showed the concentrations (mg/kg) as follows; Cr (1.50-10.25), Mn (9.75-62.75), Ni (15.75-19.25), Co (1.75-3.00), Cu (7.75-11.00), Cd (1.25-1.50), Zn (79.75-186.95) and Pb (6.25-8.00). The concentrations of the metals are in the order of Zn>Mn>Ni>Cu>Pb>Cr>Co>Cd^[36]. This trend shows a heavy deposition of heavy metals in vegetables in the Southern part of Nigeria.

CONCLUSION

In conclusion, heavy metal poisoning is a serious risk posed to the public by poly-herbal products sold in the market. Thus, proper regulation and screening of heavy metal contents in herbal remedies before they get to their final consumers is recommended. So, pharmacovigilance of poly-herbals is recommended such that the permissible limits of heavy metals in herbal preparation stay within specification. The absence of Cd, Cr and Pb in poly-herbal products analyzed in this study suggests that the products are safe for consumption.

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CONFLICTS OF INTEREST

The authors declare that there are no conflict of interests as regards publishing this article.

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CONFLICTS OF INTEREST

There are no conflicts of interest.

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