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### **Research Article**

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# Quantitation of Heavy Metals in Herbal Medicinal Drinks and Herbal Alcoholic Drinks in Lagos, Nigeria

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#### **Abstract**

Herbal medicinal plants are the starting material for herbal preparations such as herbal medicines, herbal oil, herbal teas etc. Recently, herbal alcoholic drinks are being added to the list. The present study was done to check the presence of lead, cadmium, nickel and copper in different herbal medicinal mixtures and herbal alcoholic drinks, sold in the local markets in Lagos, Nigeria. The mixtures were digested and subjected to analysis by Atomic Absorption Spectrophotometer (AAS). Results showed that lead ranged in the Herbal drinks and Mixtures from 0.00 mg/ml to 0.11 mg/ml, copper ranged from 0.00 mg/mi to 0.61mg/ml, chromium ranged from 0.00 mg/ml to 0.02 mg/ml, cadmium ranged from 0.00mg/ml to 0.01mg/ml and nickel ranged from 0.00 mg/ml to 0.02mg/ml. Some of these values are above the permissible levels for heavy metals intake by the World Health Organization. Consumption of these herbal mixtures could pose a serious health risk if consumed over a long period of time.

Keywords: Cadmium, lead, nickel, chromium, copper.

# Introduction

Herbal plants have been used for thousands of years to treat various diseases, despite significant developments in modern medicine. These herbal remedies contain active pharmacological components including minerals, trace elements and heavy metals, about 80% of the world population still depends on herbal medication [1, 2, 3]. Herbal medicine is widely accepted among the populace probably due to its low costs and easy accessibility [4]. Herbal medicine is used to treat cough, malaria, HIV, infection, infertility, pain etc [5]. Amongst the populace, there is a misconception that herbal formulations are safe despite different reports on the adverse effects of herbal medicine in the world [6, 7].

Herbal plants can be contaminated by absorbing heavy metals from the air, water and soil. Their toxicity can be related to contaminants such as pesticides, microbes, heavy metals, chemical toxins etc [8, 9].

The geography of the soil, the chemical components of the soil, contaminants in the soil, air and water can affect the quality of herbal plants [9]. Toxic metals from effluents can also contaminate the soil, water and air and hence the food chain. The plants can also accumulate high levels of toxic metals within them. Plants growing along the roadside and previous dumpsites usually have high levels of heavy metals. After the transformation of herbal plants into medicinal mixtures, the toxic metals present in such plants will finally enter the human body and may lead to various diseases such as ulcer, diabetes, hypertension, cancer etc [10].

Herbal medicinal mixtures and herbal alcoholic drinks are likely to be contaminated with different substances such as pesticides and heavy metals [11]. These herbs absorb the heavy metals and store them in their tissues. Other sources of herbal plants contamination include water contaminated with heavy metals used in the preparation of the mixtures/drinks or the use of utensils containing heavy metals [11, 12]. Heavy metals are widely distributed in nature. They occur freely in water and soil. Mercury, cadmium, lead, arsenic and chromium are toxic and they have mutagenic effects even at low concentrations. Human beings, animals and plants are strongly affected by the presence of minute concentrations of heavy metals in them [13].

The concentrations of toxic metals in herbal medicines are not known and this may cause serious risks to consumers [14]. Excessive amounts are toxic to consumers and concentrations above the WHO Maximum Permissible Limits (MPL) are hazardous to human health [15, 16]. This study aims to investigate the concentrations of heavy metals in herbal medicinal mixtures and herbal alcoholic drinks in Lagos, Nigeria

# Materials and Methods Collection of Samples

Traditionally used herbal medicinal mixtures and herbal alcoholic drinks were purchased from local vendors in the market. Each sample was mixed thoroughly to obtain a homogenous sample. The samples were taken to the laboratory and the pH levels were gotten with the use of a pH meter.

#### **Digestion of Samples**

100mls of each sample was measured and 50mls of concentrated nitric acid (HNO $_3$ ) and perchloric acid in the ratio 9:4 was added to each sample in a 250mls digestion flask. Each solution was heated on the hot plate inside a fume cupboard, until the brown fumes of the nitric acid ceased and the solution becomes clear. The solutions were cooled and each digest was made up to 100mls with deionized water. The solutions were further filtered into 200rnls conical flasks with Whatman filter paper to obtain a particle free solution.

#### **Analysis of Samples**

Determination of various metals (triplicates) in each sample was performed using the Perkin Elmer Atomic Absorption Spectrophotometer.

#### **Results**

Results are presented in Tables 1-7. Table 1 illustrates the names of the herbal medicinal mixtures and the diseases they are used to treat. These ailments range from skin problems, diabetes, cough, ulcer to gonorrhea and typhoid. Table 2 also illustrates the functions of the herbal alcoholic drinks and what they are meant to do in the body. Tables 3 and 4 shows the pH levels of the herbal medicinal mixtures and the herbal alcoholic drinks. pH values ranges from 5.1 (agbo iko) to 6.8 (agbo eje) in herbal medicinal mixtures and from 2.9 (koboko drink) to 5.4 (kodurosoke drink) in herbal alcoholic drinks. The herbal mixtures used to treat diarrhea, typhoid, cough and impotency are all in the alkaline range while that of boosting the blood level is closer to 7.0. Kodurosoke and Alomo bitters are the only alcoholic drinks within the range of 5,3-5.4. All other herbal alcoholic drinks are between 2.9 and 4.6. All these drinks are being used to improve sexual performance as well as detoxify the body. Imagine a drink supposedly used to detoxify the body, containing a pH level of 2.9.

Tables 4 and 5 shows the concentrations of heavy metals in herbal medicinal mixtures and herbal alcoholic drinks. In herbal medicinal mixtures, lead concentration ranged from 0.00mg/mi to 0.09mg/mi. Lead was detected in herbal mixtures used to treat skin problems, diarrhea, malaria, expel worms and treat sexual impotence. Lead was significantly high in agbo ale and agbo kokoro, (p0.05). Cadmium was detected in four herbal medicinal mixtures while chromium was detected in nine herbal mixtures. These levels are below the WHO maximum permissible limits. The highest level of chromium was found in mixtures used to enhance sexual performance and treat urinary problems. Chromium was significantly high in agbo ale and agbo atosi. Nickel was only detected in the herbal medicine used to treat diarrhea while copper was found in all the herbal mixtures. These levels were below WHO MPL. The highest level of copper

was found in the herbal mixture used to treat sexual impotence. Lead was present in seven alcoholic drinks and it ranged from 0.001mg/ml (Orijin drink) to 0.110 (Ale drink). Lead was significantly high in Ale drink. Ale drink is used to enhance sexual performance. Cadmium was only present in Koboko, Ale and Kodurosoke drinks and it ranges from 0.002mg/ml to 0.007mg/ml0.05). These levels are above the MPL limits of the WHO. Chromium was present in seven drinks, ranging from 0.001mg/ml (Ibile bitters) to 0.020mg/ml (Osomo bitters). These levels are below the MPL limits of the WHO. Chromium was significantly high in Osomo bitters and Koboko drink. Nickel was present in Origin, Ibile bitters, Alomo bitters, Campe bitters and Skelewu bitters in minute concentrations. Copper was present in all the alcoholic drinks except Ale drink and it ranges from 0.030mg/ml to 0.610mg/ml. Copper was significantly high in Origin drink.

Tables 6 and 7 shows the concentrations of trace metals in herbal medicinal mixtures and herbal alcoholic drinks respectively. Manganese was found in four medicinal mixtures and the concentration ranges from 0.07mg/ml to 1.95mg/ml. The highest level of manganese was found in the medicinal mixture used to expunge worms from the body where it is significantly high. High levels of iron were found in all the medicinal mixtures and they ranged from 0.455mg/ml (in typhoid medicinal mixture) to 4.1 9mg/ml (in the blood booster). These levels are extremely high and they show that herbal plants are a good source of iron. However, continuous intake of them without regulation or dosage can lead to excessive levels of iron in the body. Zinc was present in all the medicinal mixtures and they ranged from 0.92mg/ml (in skin problem medicinal mixture) to 9.89mg/ml (ulcer medicinal mixture). Magnesium was present in all the medicinal mixtures and it ranges from 12.37mg/ml (worm expeller) to 13.52mg/ml (skin problems medicinal mixture). Calcium was absent in the herbal mixture used as a worm expeller and in the mixture used to treat ulcer.

For the herbal alcoholic drinks, manganese was found in six alcoholic drinks and it ranges from 0.02mg/ml to 8.03mg/ml. The highest value of manganese was found in skelewu drink while the lowest was found in alomo bitters. Iron was found in all the alcoholic drinks and it ranges from 0.23mg/ml (skelewu drink) to 5.32mg/ml (ibile bitters). Zinc was found in all the alcoholic drinks and it ranges from 0.89mg/ml (alomo bitters) to 9.89 mg/ml (koboko drink). The level of zinc in koboko drinks is significantly high. Magnesium was found in all the drinks and it ranges from 12.52mg/ml (baby oku) to 13.56mg/ml (alomo bitters). Calcium was found in all the drinks and it ranges from 41.63mg/ml (koboko drink) to 141.20mg/mi (ale bitters).

Table 1: Names of Herbal Medicinal Mixtures and their uses

Sample Label	Names of Herbal Samples	Uses
A	Agbo kokoro	Used to treat skin problems
В	Agbo iba	Used to treat fever and malaria
С	Agbo jedi	Used to treat diarrhea
D	Agbo aran	Used to expunge worms from the body
E	Agbo iko	Used to treat cough
F	Agbo ulcer	Used to treat ulcer
G	Agbo ale	Used to treat erectile dysfunction
Н	Agbo atosi	Used to treat gonorrhea
I	Agbo typhoid	Used to treat typhoid
J	Agbo eje	Used as a blood tonic

Table 2: Names of Alcoholic Herbal Drinks collected for the study

Sample Label	Samples Names	Uses
K	Osomo bitters	Used to detoxify the body
L	Koboko drinks	Used to enhance sexual performance
M	Skelewu drinks	Used to enhance sexual performance
N	Kodurosoke	Used to enhance sexual performance
O	Baby oku	Used to enhance sexual performance
P	Ibile bitters	Used to detoxify the body
Q	Campe bitters	Used to detoxify the body
R	Alomo bitters	Used to detoxify the body
S	Ale drinks	Used to enhance sexual performance
Т	Orijin	Herbs and alcohol drinks

Table 3: pH values of Herbal Medicinal Mixtures

Names of Herbal Samples	рН
Agbo kokoro	6.1
Agbo iba	6.4
Agbo jedi	5.0
Agbo aran	5.3
Agbo iko	5.1
Agbo ulcer	6.5
Agbo ale	5.3
Agbo atosi	6.0
Agbo typhoid	5.7
Agbo eje	6.8

Standard pH: 6.5-8.5

Table 4: pH values of Herbal Alcoholic Drinks

Names of Herbal Alcoholic Drinks	рН
Osomo bitters	4.2
Koboko drinks	2.9
Skelewu drinks	4.6
Kodurosoke	5.4
Baby oku	3.3

Ibile bitters	3.7
Campe bitters	4.4
Alomo bitters	5.3
Ale drinks	4.1
Orijin	3.9

Standard pH: 6.5-8.5

**Table 5: Concentrations of Heavy Metals in Herbal Medicinal Mixtures** 

Sample Label	Lead (mg/ml)	Cadmium (mg/ml)	Chromium (mg/ml)	Nickel (mg/ml)	Copper (mg/ml)
A	0.077±0.003*	0.001±0.001	0.006±0.002	ND	0.003±0.001
В	0.016±0.001	0.002±0.001	ND	ND	0.012±0.002
С	ND	ND	0.011±0.005*	0.012±0.004	0.064±0.007
D	0.025±0.002	ND	0.011±0.003*	ND	0.041±0.003
Е	ND	0.003±0.004*	0.007±0.006	ND	0.021±0.005
F	ND	0.001±0.001	0.009±0.008	ND	0.082±0.007*
G	0.087±0.007*	ND	0.019±0.005*	ND	0.088±0.004*
Н	ND	ND	0.015±0.002	ND	0.079±0.002*
I	ND	ND	0.011±0.002*	ND	0.002±0.001
J	ND	ND	0.006±0.005	ND	0.038±0.004
MPL	0.01	0.003	0.05		2.00

<sup>\*</sup>p0.05

**Table 6: Concentrations of Heavy Metals in Herbal Alcoholic Drinks** 

Sample Label	Lead (mg/ml)	Cadmium (mg/ml)	Chromium (mg/ml)	Nickel (mg/ml)	Copper (mg/ml)
K	0.031±0.002	ND	0.021±0.007	ND	0.199±0.05
L	ND	0.005±0.002*	0.018±0.004	ND	0.136±0.03
M	0.006±0.005	ND	ND	0.011±0.003	0.082±0.02
N	0.016±0.003	0.002±0.001	0.013±0.005	ND	0.142±0.08*
O	ND	ND	0.015±0.001	ND	0.034±0.04
P	0.012±0.002	ND	0.001±0.001	0.010±0.001	0.083±0.01
Q	ND	ND	0.016±0.004*	0.019±0.007*	0.144±0.05*
R	0.072±0.003*	ND	$0.009\pm0.007$	$0.009 \pm 0.002$	0.139±0.08*
S	0.111±0.009*	0.007±0.004*	0.012±0.001*	ND	
T	0.001±0.001	ND	ND	0.021±0.002*	0.606±0.04*
MPL	0.01	0.003	0.05	0.01	2.00

<sup>\*</sup>p0.05

Table 7: Concentrations of Trace Metals in Herbal Medicinal Mixtures

Sample Label	Manganese (mg/ml)	Iron (mg/ml)	Zinc (mg/ml)	Magnesium (mg/ml)	Calcium (mg/ml)
A	0.548+0.90	4.036+0.33*	0.920+0.05	13.520+1.89*	69.920+3.98
В	ND	3.869+0.67	9.422+0.75*	13.090+1.77	68.110+3.73
С	ND	3.941±0.61	9.441±0.67*	13.290±1.56	75.560±3.09
D	1.953±0.88*	0.132±0.10	2.412±0.23	12.370±1.78	ND
Е	ND	0.786±0.21	8.581+0.56*	13.310±1.73	132.530±5.88*
F	0.130±0.76	1.244±0.55	9.894+0.42*	12.920±1.98	ND
G	ND	4.075±0.49*	7.561±0.99	13.500±1.66*	121.100±4.21*
Н	0.073±0.11	5.215±0.11*	9.448±0.87*	12.840±1.54	1.770±0.05
I	ND	0.450±0.02	7.512+0.42	12.920+1.44	59.510±3.99
J	0.473±0.56	4.191±0.54*	7.611±0.65	13.420+1.59	140.040±5.45*
MPL	0.5	0.3	5.00		

<sup>\*</sup>p0.05

**Table 8: Concentrations of Trace Metals in Herbal Alcoholic Drinks** 

Sample Label	Manganese (mg/ml)	Iron (mg/ml)	Zinc (mg/ml)	Magnesium (mg/ml)	Calcium (mg/ml)
K	0.323±0.07	4.005±0.11*	2.061±0.41	13.280±1.57	123.500±3.20*
L	ND	1.244±0.56	9.894±0.87*	12.920±1.37	41.630±2.22
M	8.029±0.45*	0.227±0.05	6.700±0.49	13.350±1.44*	101.030±5.98*
N	ND	4.029±0.77*	6.988±0.34	13.200±1.29	88.070±4.29
О	ND	5.200±0.42*	7.420±0.44	12.520±1.50	53.070±2.50
P	ND	5.320±0.79*	9.767±0.79*	13.270±1.31	60.460±2.88
Q	4.974±0.88*	5.226±0.42*	8.201±0.41	13.310±1.70	47.660±3.11
R	0.009±0.001	3.982±0.71	0.891±0.09	13.560±1.48*	158.40±5.99*
S	0.556±0.36	0.078±0.03	9.201±0.78*	13.240±1.33	141.200±5.01*
Т	7.405±0.53*	3916±076	5.119±0.12	13.140±1.68	90.060± 4.03
MPL	0.5	03	5.0		

<sup>\*</sup>p0.05

#### **Discussion**

The overall results show clearly that Pb, Cd, Cr, Zn and Ni are all present in some of the samples examined. Lead was detected in eleven samples out of the twenty samples analyzed. Out of these 11 samples, 7 samples had levels that exceeds the WHO recommended limit of 0.01mg/ml for lead [17]. This result is similar to the findings of who detected lead in all herbal tablets analyzed [18]. 91% of the analyzed samples exceeded the maximum recommended limit of 10µgPb/g. High concentrations of lead has been reported in the Middle Eastern countries [19]. Maximum levels of 14.40mg/kg and 21.70mg/kg of lead were reported in spices and medicinal plants gotten from Egypt and Iran [20, 21]. Lead is highly toxic and it can complex with various molecules, thereby affecting their functions. Exposure to lead may lead to adverse effects on the brain, kidney, muscles, liver, ovaries, testes etc [19, 22]. In children, exposure to lead, leads to slow cognitive developments and learning deficits [22].

Cadmium was present in 7 samples out of the 20 samples examined. Koboko drink and Ale dink which were being used to enhance sexual performance, has the highest concentrations of 0.01mg/ml which

is above WHO limit of 0.003 mg/ml for cadmium. [23] reported cadmium concentrations of 1.012mg/ml and 0.671 mg/ml in herbal mixtures obtained from selected suburbs of Kumasi in Ghana. These levels were higher than the levels of cadmium obtained in this study. In a study on herbal drugs in the Iranian market, cadmium was present in all the samples analyzed and about 64% of the herbal drugs examined had cadmium levels above  $0.30\mu g/g$ . The high levels of cadmium pose a serious effect on the health of human beings. The kidney is the target organ in cadmium exposed populace. Cadmium accumulates in the kidneys for a long time; resulting in the damage of the kidney tract because its excretion is slow. At high concentrations, cadmium affects the liver, the immune system and the vascular system [24].

Chromium was present in 17 of the samples analyzed. Although, these levels were below the maximum permissible limits for chromium, there is still cause for concern. Copper was present in all the samples analyzed in significantly high amounts, except in Ale alcoholic drink. The highest level of copper was found in Origin drink (0.610mg/ml). Although, this level is below the permissible level for copper by

the WHO in drinking water. Copper is an important component of many enzymes and it plays a significant role in melanin production, free radical elimination, iron utilization etc. However, excessive intake of copper can cause diarrhea, vomiting, liver damage, nausea, abdominal pain etc [25].

Zinc is a powerful antioxidant. It is an essential trace element for proper growth, blood clotting, DNA synthesis, protein synthesis etc. However, the excessive intake of zinc produces toxic effects on the blood lipoprotein levels, copper levels and on the immune system [26]. Zinc was present in all the 20 samples analyzed and it ranged from 0.89mg/ml in Alomo bitters, to 9.89mg/ml in Koboko drink. Zinc was significantly high in 15 samples, out of the 20 samples examined. An adult man/woman needs about 5mg of zinc on a daily basis. Sixteen samples out of all the samples examined exceeded the WHO permissible limit of 3.00mg/ml for zinc in drinking water [1, 2].

Iron has several important functions in the body including energy production, immunity and oxygen supply. Excessive amounts of iron in the body leads to nausea, vomiting, diarrhea, joint pain, dizziness, shock, liver damage etc. Iron toxicity has adverse effects on various metabolic functions and on the cardiovascular system of the body [27]. From the study carried out, excessive amounts of iron were found in all the samples analyzed most especially the medicinal mixtures. The concentrations of iron in the samples analyzed ranged from 0.08mg/ml (Skelewu drink) to 5.32mg/ml (Ibile bitters). Iron was significantly high in 15 samples. The WHO limit for iron in drinking water is 0.30mg/ml.

Results from various studies have shown that herbal market samples are more contaminated with heavy metals when compared with herbs from the natural sources [16]. Herbal plants can be contaminated in. different ways and by exposure to industrial effluents, polluted soils, polluted water, fertilizers, pesticides etc. Sources of heavy metals can also be linked to harvesting, transportation and storage processes [16]. Generally, the health risk in herbal plants depends on the average daily intake and in Nigeria, the general populace consumes these herbal mixtures and alcoholic drinks as if they are consuming water. The level of patronage together with the consumption rate is very high especially in Lagos State.

#### **Conclusion**

The results of the study show s that there is potential health risk of heavy metals to the general populace in Lagos, Nigeria over long term consumption of these contaminated herbs.

#### Recommendation

- 1. Safe and hygienic practices must be put in place, starting from the harvesting of these plants until they reach the consumers.
- 2. The sources of pollution should be identified and eradicated.
- There should be regular monitoring of the quality of herbal mixtures and herbal alcoholic drinks sold in the Nigerian market.
- 4. There should be public awareness on the dangers inherent in the long term consumption and over dose of these herbs.

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