Original Research

Heavy Metal Concentrations in Selected Herbal Drinks Sold in Abeokuta, Ogun State, and Their Toxicological Risk Assessment

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Abstract

Herbal beverages are one of the most popular drinks in Africa, especially in Nigeria. Nevertheless, most herbal beverages do not adhere to World Health Organization (WHO) standards. This study aims to examine the levels of potentially toxic metals (PTMs) in selected herbal beverages and their toxicological risk assessment in children and adults in Abeokuta. Ten (10) samples of herbal drinks were obtained from vendors and delivered to the chemistry laboratory for examination. 20 mL of each herbal sample was digested using the standard method and an atomic absorption spectrophotometer for PTM estimation. The study's findings revealed a higher level of cadmium (Cd) in Black Horse, Action, Jigijigi, and Ogbonge (0.007±0.001, 0.005±0.001, 0.006±0.001and 0.004±0.000 mg/L), Iron (Fe) in Otoge and Kondo (11.1±0.107 and 1.83±0.089 mg/L), Manganese (Mn) in Otoge, Kondo, Jigi-jigi, Jaye, and Ogbonge (0.060±0.008, 0.123±0.014, 0.080±0.010, 0.134±0.015, and 0.070±0.009 mg/L) than the WHO limit, while none of the herbal beverages contained copper (Cu). Iron (Fe) appears to be the most dosed metal, while the child population is more vulnerable to PTM exposure according to the average daily intake. The studied PTMs showed a negligible non-carcinogenic risk (<1) based on human health risk evaluations. Hence, the study offers pertinent data for evaluating the indirect consumption of PTMs through their availability in regularly consumed herbal drinks in Abeokuta and its environs.

Keywords: herbal drinks, potentially toxic metal, spectroscopy, toxicology, risk assessment

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Introduction

Foods and beverages supply the nutrients needed for healthy growth [1], development, and maintenance [2]. Various kinds of beneficial drinks are being consumed by Nigerians [3]. There are mainly two types of drinks: alcoholic and non-alcoholic beverages. The non-alcoholics are subdivided into general, local, and herbal non-alcoholics, while the alcoholic types involve gin, spirit liquor, palm wine, and beer. General non-alcoholic drinks are soft drinks, yogurt, juice, and wine. Examples of non-alcoholic local drinks are Pito, Kunnu, Zobo, etc., while those of herbal origin are bitter concoctions that are claimed to possess anti-malarial, anti-pile, and anti-jaundice properties, among others [4].

Herbal beverages are a type of beverage, generally alcoholic, that is flavored with herbal essences and characterized by a bitter or bitter-sweet taste. Herbal drinks are made from secondary metabolites and biologically active phytochemicals from medicinal plants, narcotic components of tropical and subtropical plants, spices, herbs, and root extracts [5, 6]. They are usually dark in color as well as prized due to their capacity to stimulate appetite and enhance digestion, which is why they are used in patent medicine, as a digestive aid, and also in cocktail flavoring. Herbal drinks comprise diverse kinds of naturally occurring chemically-oriented constituents taken from medicinal plants that function to stimulate the essential energy center of the body [7].

In view of this, herbal drinks are generally acceptable to the general public because of their potency, cost-effectiveness, and ease of availability. It has been reported to be used traditionally in treating cough, worm infestation, malaria, hemorrhoids, infection, infertility, and pain, among other ailments in children and adults [4, 6, 8]. Herbal drinks are sold in Nigeria as a "cureall" patent medicine; however, the majority of them have not been subjected to scientific testing [9]. Furthermore, they are in high demand because of their well-known health benefits, and they have become ubiquitous in many Nigerian households [10], including Abeokuta. Despite various global studies regarding the detrimental effects of herbal drinks, there is a widespread belief that medicinally formulated herbal drinks are safe for human consumption [11]. One way in which this detrimental effect could occur is through the adverse effect of PTMs gaining access to herbal plant materials [12].

Human diets or drinks can contain varying amounts of PTMs, much of which could be affected by the quantity in the planting soil since these soils can be found in populated areas globally, including sub-Saharan Africa [13], such as Nigeria, where Abeokuta is a city. The PTMs are known to be potentially harmful [14] because of their densities and/or nucleon numbers [15]. Drinking potentially harmful chemical-containing beverages can cause serious health problems such as cardiac, renal, neurological, etc. [16]. Furthermore, the PTM content of the drinks is significant because of

the effects it has on their quality and consumer health due to prolonged intake. Potentially toxic metals can bioaccumulate in the human body, resulting in both carcinogenic and non-carcinogenic consequences [4]. Due to weak regulation and monitoring by appropriate organizations in third-world nations, herbal drinks enter the market with little or no data on their safety and efficacy. Despite the increased usage of herbal drinks, scientific evaluations are particularly required to ensure safety by following regulations and standardization.

Abeokuta is the state capital of Ogun State, southwestern Nigeria, which is in the southwestern part of Nigeria. It is not news that the people of this locality are used to consuming various herbal drinks due to their perception of their therapeutic potential. As a result, evaluating PTM concentrations in accordance with the WHO standard is critical and beneficial in the provision of informative and relevant data, which may result in essential solutions as well as the formulation of the right policy for health and safety. Thus, it is conceivable to evaluate the PTM concentrations in selected herbal drinks and their toxicological risk assessment for environmental and health safety.

Materials and Methods

Study Area

The study area where the herbal drink samples were collected was Abeokuta metropolis and its environs. Abeokuta is located at coordinates 7.1475°N, 3.3619°E. It is the capital of Ogun State, which is in the southwestern part of Nigeria. The extracted map of Abeokuta from that of Ogun State in Nigeria is shown in a modified version in Fig. 1 [17]. The geopolitical regions of the state's Abeokuta North and Abeokuta South Local Government Areas are included by the city of Abeokuta. Although, the Egbas were the first people to settle in the area and build the historic city, the Yoruba tribe makes up the majority of the population in Abeokuta, but many other ethnic groups have also settled there as a result of industry and urbanization. In the region, there are dialectical groups such as Ijebu, Egba, Remo, Oyo (Owu), Awori, Ikale, and Ilaje. The folks are well-known for their traditional sculpture and carving. Abeokuta is home to an estimated four million people and is encircled by a substantial mass of rocks.

Sample Collection, Digestion, and PTM Estimation

A total of ten (10) herbal drink samples were purchased and transported to the chemistry laboratory for sample analysis. 20 mL of the herbal sample was measured into a 100 mL volumetric flask with 10 mL of concentrated nitric acid (HNO₃), gently heated on a hot plate for 45 minutes inside a fume hood. The flask was allowed to cool to ambient temperature,

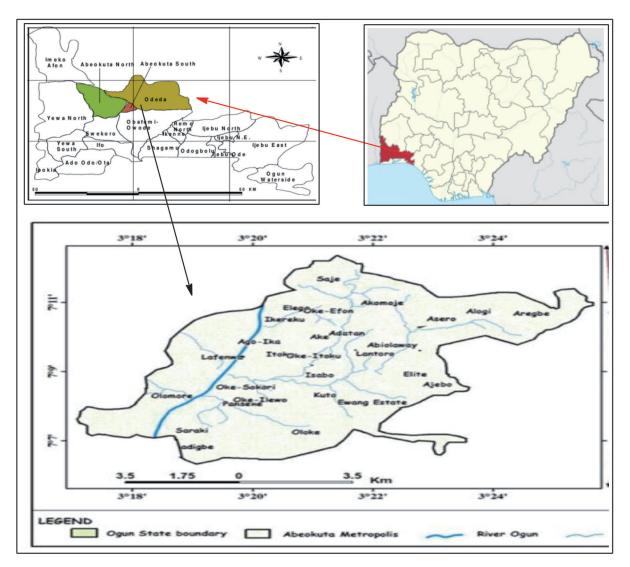


Fig. 1. Map of Abeokuta City from Ogun State and Nigeria.

then the digest was filtered using Whatman No. 42 filter paper and diluted to mark with deionized water in a 250 mL standard volumetric flask [18]. The Atomic Absorption Spectrometer (AAS) method was used to analyze the concentrations of PTMs in the samples.

Toxicological Risk Assessment

Human health risks from the consumption of the herbal drink sample were evaluated via the ingestion route based on the equation provided by the United States Environmental Protection Agency [19]. The average daily intake (ADI) for potentially toxic metals in the sample was calculated using Equation (1).

$$ADI_{ing} = C \times \frac{IngR \times EF \times ED}{BW \times AT}$$
 (1)

where ADIing stands for average daily intake through ingestion per kilogram of body weight [20]. Potentially toxic metal concentration in herbal beverage C is

measured in mg/L, ingestion rate IngR is measured in liters per day, and exposure duration ED is measured in years, which is equal to life expectancy. Exposure frequency EF is measured in days per year, body weight BW is measured in kilograms, and AT is measured in hours (ED x EF). By applying equation 2, the hazard quotient was calculated by comparing the average daily intake of each PTM to their reference dose (RfD). The hazard quotient is used to determine if a PTM presents a significant non-carcinogenic risk to humans after consumption [21].

$$HQ = \frac{ADI}{RfD} \tag{2}$$

Where HQ represents the hazard quotient through ingestion and RfD is the oral reference dose in mg/L/day. HQ values greater than 1 indicate a significant non-carcinogenic side effect, whereas values below 1 indicate no side effect [21].

Data Analysis

The Microsoft Excel 2013 version was utilized for data analysis, graphical presentation, and also for computing the toxicological risk assessment.

Results and Discussion

Potentially Toxic Metal Concentration

The damaging influence of PTMs on humans and their environment cannot be overemphasized. These non-degradable elements in the earth's crust find their way into human life through diverse means. Due to the toxicity of PTMs, their availability in food and drink is of immense concern to public health due to their biotoxic effects [22]. It is sad to note that even herbal drinks people consume for medicinal purposes to stay healthy are now gradually becoming a source of adverse health concern because of the presence of PTMs above the tolerable limit in such drinks. This causes a paradox, hence, it is conceivable to determine the concentration of PTMs in some herbal drinks sold commercially in the Abeokuta metropolis of Ogun State and to carry out the toxicological risk assessment in order to ascertain consumers' safety.

Therefore, PTM concentrations in some of the herbal drinks were determined in this present study using an atomic absorption spectrophotometer. Potentially toxic metal concentrations in the analyzed herbal drinks compared to the World Health Organization (WHO) standard for PTMs in drinking water are presented in Table 1. Generally, Cd, Cu, Fe, and Mn were present in the selected herbal drinks, while Pb was absent or not detected (ND). The result showed that the Cadmium (Cd) concentration ranged from not detected in sample E to 0.007 mg/L in sample C. The highest Cd concentration was recorded in the Black Horse (0.007 mg/L), followed by Jigi-jigi (0.006 mg/L), while the lowest concentration was recorded in Kondo and Jabra (0.001 mg/L), respectively. The Cd concentration in Corner Kick was not detected or was below the detectable limit. Copper (Cu) concentrations ranged from 0.048 mg/L to 1.88 mg/L. The highest Cu concentration was recorded in Jaye (1.88 mg/L), followed by Corner Kick (0.786 mg/L), while the lowest concentration was recorded in Black Horse (0.048 mg/L) (Table 1).

In addition, Fe concentrations ranged from 0.139 mg/L to 11.1 mg/L. The highest Fe concentration was recorded in Otoge (11.1 mg/L), followed by Kondo (1.83 mg/L), while the lowest concentration was recorded in Black Horse (0.139 mg/L). Manganese (Mn) concentration ranged from 0.027 mg/L to 0.134 mg/L. Jaye (0.134 mg/L) exhibited the highest Mn concentration, followed by Kondo (0.123 mg/L), whereas Black Horse (0.027 mg/L) had the lowest concentration (Table 1). The concentrations of Cu in all samples were below the WHO limit of 2.00 mg/L, while Cd, Fe, and Mn in 40%, 20%, and 50% of the

samples, respectively, were recorded to be higher than the WHO standard of 0.003 mg/L, 1.0 mg/L, and 0.05 mg/L, respectively.

Furthermore, the concentrations of Cadmium in 40% (04 of 10) of the total samples, such as Black Horse (0.007 mg/L), Jigi-jigi bitters (0.006 mg/L), Action bitters (0.005 mg/L), and Ogbonge (0.004 mg/L), respectively, were higher than the WHO standard (0.003 mg/L). This is also higher than the concentration of the investigated herbal mixture (0.001 mg/l) from Kumasi, Ghana [23], but lower than the concentration of bitters (0.012 mg/l) in an herbal drink studied in Abeokuta, Nigeria [4]. A high concentration of Cd causes a hazardous effect on human health, whereby the kidney serves as a target organ for exposure. Prolonged accumulation of Cd leads to damage to the kidney tract due to its slow excretion and also affects the liver, the immune system, and the vascular systems [24]. Based on its mechanism of action, cadmium causes damage to cells primarily through the generation of reactive oxygen species (ROS). The role of Cd in neurodegenerative disorders is further emphasized by its effect on diverse CNS cell types [25].

Moreover, the concentrations of Cu in 20% (02 of 10) of the herbal samples were higher than the WHO standards of 1.0 mg/L. The Cu concentrations in Jaye bitters (1.88 mg/L) and Corner kick (0.786 mg/L), respectively, from the study were higher than the Cu concentrations reported in Alomo bitters (0.14 mg/L), Baby Oku (0.03 mg/L), and Origin Bitter (0.61 mg/L) in Lagos, Nigeria [12]. More so, an estimated 80% of analyzed herbal drinks have a Cu concentration lower than the WHO standard (2.00 mg/L) (Table 1). Cu is a vital metal that is a part of many enzymes and is crucial for the synthesis of melanin and the removal of free radicals. On the other hand, consuming too much Cu can result in nausea, vomiting, liver damage, diarrhea, and abdominal pain [26].

Fe concentrations in 20% (02 of 10) of the investigated herbal drink samples were higher than the WHO standard of 0.03 mg/L. Moreover, a very high concentration of Fe, multiple times higher than the WHO standard, was recorded in the Otoge herbal drink (11.1 mg/L). The concentration of Fe in the studied herbal drinks was higher than the maximum Fe concentration reported in some imported canned energy drinks (0.026 mg/L) investigated in Lagos, Nigeria [27], while a majority (80%) was lower than the mean Fe concentration reported in bitters (1.06 mg/L), antijaundice (3.93 mg/L), and anti-malaria (7.63 mg/L); but similar to Otoge herbal drink (11.1 mg/L), a very high mean Fe concentration was detected in anti-pile (20.01 mg/L) in an investigation from Abeokuta, Nigeria [4]. The high Fe content in the herbal drink is traceable to the chemical properties of the soils used to grow the plant materials used for the production of the herbal drink [28]. However, a trace concentration of Fe in the herbal drink is essential in biological systems such as those of co-factors for co-transporters, ligands, enzymes, and electrolytes. However, when the concentration of

Herbal Drinks	Potentially Toxic Metals (mg/L)					
	Cadmium (Cd)	Copper (Cu)	Iron (Fe)	Lead (Pb)	Manganese (Mn)	
A	0.002±0.000	0.063±0.009	11.1±0.107	ND	0.060±0.008	
В	0.003±0.000	0.114±0.012	0.318±0.026	ND	0.040±0.006	
С	0.007±0.001	0.048±0.008	0.139±0.015	ND	0.027±0.003	
D	0.001±0.000	0.079±0.009	1.83±0.089	ND	0.123±0.014	
E	ND	0.786±0.049	0.215±0.021	ND	0.041±0.007	
F	0.005±0.001	0.055±0.008	0.244±0.023	ND	0.043±0.007	
G	0.006±0.001	0.064±0.009	0.321±0.027	ND	0.080±0.010	
Н	0.003±0.000	1.88±0.097	0.494±0.032	ND	0.134 ±0.015	
I	0.001±0.000	0.060±0.008	0.729±0.055	ND	0.038±0.007	
J	0.004±0.000	0.109±0.005	0.65±0.043	ND	0.070±0.009	
WHO (2011)	0.003	2.00	1.0	0.01	0.05	

Table 1. The concentration of potentially toxic metals in selected herbal drinks compared to the WHO guidelines values.

Mean±Standard Deviation (SD) of triplicate measurements. There was no significant difference (P>0.05). Concentrations in bold are higher than the WHO Standard for Potentially toxic metals in Drinking water. **A** = Otoge herbal drink; **B** = Gbe body e; **C** = Black Horse; **D** = Kondo; **E** = Corner kick; **F** = Action; **G** = Jigi-jigi; **H** = Jaye; **I** = Jabra; **J** = Ogbonge; ND = Not Detected.

iron exceeds its safe limits, it may also be unpalatable [27]. Raising the population of bacteria that depend on iron, for example, can lower the quality of herbal drinks by preventing slime or unattractive color [29].

Besides, as an element, Manganese functions as a component of several enzymes, including mitochondrial superoxide dismutase [30]. The concentrations of Manganese in 50% (5 of 10) of the selected herbal samples (Otoge (0.060 mg/L), Kondo (0.123 mg/L), Jigi-jigi (0.080 mg/L), Jaye (0.134 mg/L), and Ogbonge (0.070 mg/L)) were higher than the WHO standard (0.05 mg/L), while others were within the limit. In comparison to related studies, it is higher than the mean Mn concentration detected in alcoholic herbal drinks such as Baby Oku (0.04 mg/L) in Abeokuta, Nigeria [4], and Alomo bitters (0.009 mg/L), but lower than that of Origin bitters (7.41 ppm) in Lagos, Nigeria [12].

Prolonged exposure to a high Mn concentration can be linked to neurological disorders [31].

In addition, the comparative study of potentially toxic metal concentrations to that of the WHO standard in each herbal drink sample was evaluated. The result is given in Fig. 2. It is important to note that there is no basis for a comparative study regarding lead (Pb) concentration since it was not detected in any of the ten samples. It was obvious in Fig. 2 that Fe/WHO and Mn/WHO were outrageously higher in sample A (Otoge) than in the rest of the samples. Both Fe and Mn experienced a sharp drop in sample B (Gbe body e). On the contrary, there was an increase in Cu/WHO from sample A to sample B. The trend of the four metals/WHO fluctuated from sample C (Black Horse) to sample J (Ogbonge). Parameters used for the toxicological risk assessment are as presented in Table 2 [4, 32].

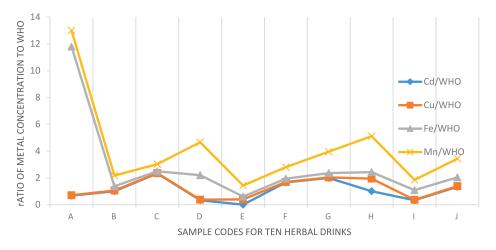


Fig. 2. Comparative study of heavy metals concentrations to WHO Standard.

Table 2. Parameters u	and for	Tarriaglagiagl	mials agassamant
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Parameter	Units	Adult	Children	
Body weight (BW)	Kg	60	15	
Exposure frequency (EF)	days/year	365	180	
Exposure duration (ED)	Years	30	6	
Ingestion rate (IngR)	mg/day	200	20	
Average time (AT)	Days	25550	25550	
Oral reference doses (RfD) in mg k				
Cd	1.0 ×10 ⁻³			
Cu	4.0 ×10 ⁻²			
Fe	7.0 ×10 ⁻¹			
Mn	1.4 ×10 ⁻¹			

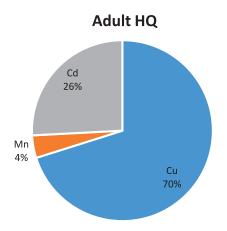
Taiwo et al. [4] and Iwegbue et al. [36]

Table 3. Average Daily Intake of PTMs in Herbal Drinks.

PTMs	Adult	Children	
Cd	3.65×10 ⁻³	2.34×10 ⁻³	
Cu	3.35×10 ⁻¹	2.14×10 ⁻¹	
Fe	1.65×10	1.05×10	
Mn	6.74×10 ⁻²	4.31×10 ⁻²	
Pb	-	-	

Health Risk Assessment

The human health risk of exposure to potentially toxic metals in the investigated herbal drinks was evaluated using the non-carcinogenic risk hazard quotient denoted HQ, which was also used as the HI due to the absence of other routes of exposure (inhalation and dermal contact) over a lifetime exposure for adults and children. The children population was included in this estimation because they have been sighted to be given these drinks by parents/ guardians in this vicinity, and acknowledging this phenomenon appears to be an important part of the investigation. Tables 3 and 4 show the summary of the human health risk assessment data. According to Table 3, the average daily dosage showed that Fe is the most dosed PTM (1.65×10, 1.05×10), followed by Cu (3.35×10⁻¹, 2.14×10⁻¹) in adults and children, respectively. ADI for Cd, Cu, Fe, and Mn for the human population was lower than the provisionally



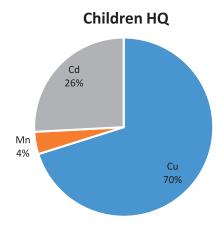


Fig. 3. Percentage hazard quotient of each heavy metal in herbal drink to an adult and children population respectively.

Table 4. Non-carcinogenic risk of the PTMs in herbal drinks.

Human population	Exposure	Cd	Cu	Fe	Mn
Adult	HQ_{ing}	3.65×10	8.37×10	2.35×10	4.81×10 ⁻¹
Children	HQ _{ingt}	2.34×10	5.36×10	1.51×10	3.08×10 ⁻¹

tolerable daily dosage for Cd (6.40 x 10⁻²) [33], Cu (9 x 10⁻¹) [34], Fe (8.0 x 10⁻¹) [35], and Mn (8.33 x 10⁻²) [36]. The child population appears to be more susceptible to potentially toxic metals in herbal drinks than the adult population. This indicated that the act of giving herbal drinks to children should be carefully controlled or, if need be, an alternative measure should be employed. The average daily dose appeared in the decreasing order of Fe>Cu>Mn>Cd for the human population.

The hazard quotient (HQ) value revealed that all estimated PTMs have a value less than 1, indicating a non-significant, non-carcinogenic risk for the human population. The HQ value for the studied PTMs appeared in descending order of Cu>Cd>Mn (Fig. 3). This observable trend was true and similar for both adult and children's populations.

Conclusions

The presence of potentially toxic metals (Cd, Cu, Fe, and Mn) in some selected herbal drink samples was above the WHO limit. Prolonged exposure to Cd toxicity leads to several health issues, such as damage to the kidney, liver, and cardiovascular system. Mn toxicity is linked to neurological degradation. The exposure of children and adults to those brands is detrimental, and care should be taken by the consumer when consuming these drinks. High Fe concentrations can affect the quality of herbal drinks, making them unpalatable for consumption. This contamination may occur during transportation from place to place as a result of processing and handling, and also due to the status of the raw materials used for production. The average daily doses for all metals were below the permissible daily dose limit. Hazard Quotient estimated potentially toxic metals pose a non-significant, non-carcinogenic risk for the human population. Therefore, the study provides useful information for the assessment of PTM intake from selected herbal drinks. As a result of this study, it is therefore recommended that good and hygienic practices be followed during the production of these bitter herbal drinks, and it is important to locate and remove pollution sources.

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Conflict of Interest

The authors declare no conflict of interest.

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