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ORIGINAL ARTICLE

HEAVY METALS HAZARDS FROM NIGERIAN SPICES

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ABSTRACT

Background. Natural spices are commonly used by the people in Nigeria. They may be easily contaminated with heavy metals when they are dried and then pose a health risk for the consumers.

Objective. The aim of this study was to determine the levels of heavy metals in some commonly consumed natural spices namely *Prosopis Africana, Xylopia aethiopica, Piper gineense, Monodora myristica, Monodora tenuifolia* and *Capsicum frutescens* sold in the local markets of Awka, Anambra state, South East Nigeria to estimate the potential health risk.

Results. The range of heavy metal concentration was in the order: Zn (14.09 – 161.04) > Fe (28.15 – 134.59) > Pb (2.61 – 8.97) > Cr (0.001 – 3.81) > Co (0.28 – 3.07) > Ni (0.34 – 2.89). Pb, Fe and Zn exceeded the maximum allowable concentrations for spices. The Target Hazard Quotient (THQ) of the spices varied from 0.06 – 0.5. Estimated daily intakes (EDI) were all below the tolerable daily intake (TDI). The lead levels in *Prosopis africana, Xylopia aethiopica, Piper gineense, Monodora myristica* and *Capsicum frutescens* which are 8-30 times higher than the WHO/FAO permissible limit of 0.3 mg/kg.

Conclusions. Lead contamination of spices sold in Awka (south east Nigeria) may add to the body burden of lead. A good quality control for herbal food is important in order to protect consumers from contamination.

Key words: *food products, spices, potential toxic metals, risk assessment, public health*

INTRODUCTION

Spices are dried parts of plants which are used to improve colour, aroma, palatability and acceptability of food [25]. Most common spices have been documented to possess outstanding microbial, antidiabetic, anti-inflammatory, antioxidant and antihypertensive potential [9, 15, 37]. On the other hand, the method of preparation and handling of spices can make them a source of food poisoning [34]. Currently there is an increased emphasis on eating healthy foods which are low in fat and salt [27], although they belong to condiments, spices are substances which do not contain nutritive components [21] but their ability to improve taste and sometimes appearance of food has led to increased usage among locals in Nigeria. Spices are a potential link to transfer contaminants and heavy metals from the environment to humans through the food chain due to their plant origin. These spices may easily be contaminated by heavy metals from the soil or aerial depositions as these spices are dried on the ground or on roof tops [36]. Exposures to toxic metals are associated with severe health problems with varying degrees of severity and conditions: kidney problems, neurobehavioral and developmental disorders, high blood pressure and possibly cancer [5, 12, 17]. Natural spices used in Nigeria are mostly used by the population in the rural areas although different local spices have ethnic and regional peculiarities, which may be as a result of distinct preparation methods.

Most of these spices are produced from natural sources and therefore the wide spread public opinion that they are harmless and free from adverse effect. Nevertheless, a good quality control for herbal food is important in order to protect consumers from contamination [41].

Although spices represent a small fractions of the total food intake, their use in the preparation of popular daily Nigerian meals such as jollof rice,

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beans, moi-moi, and a variety of soups, results in the need to determine its safety. Spices have now been considered a potential non-paint, non-petrol source of lead poisoning within apparently lead safe home environments [13].

The aim of this study is to determine the levels of heavy metals contamination in some commonly consumed natural spices namely *Prosopis africana, Xylopia aethiopica, Piper gineense, Monodora myristica, Monodora tenuifolia* and *Capsicum frutescens* sold in the local markets of Awka, Anambra state, South-East Nigeria and to estimate their potential health risk.

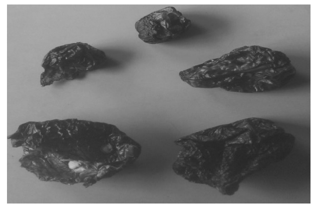
MATERIALS AND METHOD

Sample collection, preparation and analysis

Samples of spices namely *Prosopis africana, Xylopia aethiopica, Piper gineense, Monodora myristica, Monodora tenuifolia* and *Capsicum frutescens* (Figure 1) sold in different parts of Awka, Anambra state, south-east region of Nigeria, were taken from the open markets within the state in March 2015 and kept in plastic bags. Only edible parts of the samples were used for the analysis. The sources of spices were authenticated from the seller at the point of purchase to validate they were locally produced in the area. The samples were cutting into small pieces before drying in the oven to the constant weight. The samples were then pulverized with a ceramic mortar and pestle to fine powder. About 0.5 g of dried powdered sample was weighed and transferred into beaker and were subjected to wet digestion with 10 mL HNO_3:$HClO_4$ (at a volume ratio of 4:1) at 150 ^oC for four hours. Following this, samples were left to cool; filtered using a filter paper, transferred into a 50 mL volumetric flask and a final volume of 25 mL was made by adding deionized water. For each of the samples, triplicate digestion were carried out together with blank reagent and kept in refrigerator until analysis. The samples were analysed for the presence of Pb, Cr, Ni, Zn, Fe and Co using Flame Atomic Absorption Spectrometer (FAAS). The limit of detection of the heavy metals were 0.005 μg/g, with blank values reading below this value for the metals in deionized water with an electrical conductivity value of <5 μS cm^{-1}. Standard quality control measures were carried out to reduce the risk of contamination and assure reliability of the results.

Prosopis Africana

Xylopia aethiopica



Capsicum frutescens

Monodora myristica

Piper gineense

Monodora tenuifolia

Figure 1. Samples of tested spices

HEAVY METAL HEALTH RISK ASSESSMENT

Calculation of Estimated Daily Intake of Metals (EDI)

The daily intake of metals depends on both the metal concentration in food and the daily food consumption. In addition, the body weight of humans can influence the tolerance of contaminants. The EDI was calculated based on the following formula [35].

$$EDI = \frac{C_{metal} \times D_{food\ intake}}{BW_{average}}$$

Where:
C - the metal concentration in spices in mg/kg,
D - the daily intake of food in kg $person^{-1}$
BW - average body weight in kg $person^{-1.}$

An average daily consumption of 0.01 kg of spices was assumed in this study. This method was adapted because spices are widely consumed as a major part of the diet. Average adult body weight was considered to be 65 kg.

Non carcinogenic risk estimation of heavy metals consumption was determined using THQ values. THQ is a ratio of the determined dose of a pollutant to a reference level considered harmful. THQ values were determined based on the following formula [35].

$$THQ = \frac{Efr \times ED \times FIR \times C}{RfDo \times B_{average\ weight} \times ATn \times 10^{-3}}$$

Where:
Efr - exposure frequency in 156 days $year^{-1}$
ED - exposure duration in 70 years (equivalent to an average lifetime) [3]
FIR - average daily consumption in kg $person^{-1}day^{-1}$
C - concentration of metal in food sample in mg/kg
RfDo - reference dose in mg/kg day^{-1}
ATn - average exposure time for non – carcinogens in days.

The following reference doses were used (Pb = 4.0 x 10^{-3}, Cd = 0.001, Cu = 0.04, Fe = 0.7, Zn = 0.3, Mn = 0.014, Co = 0.043). THQs were calculated according to the methodology described by the Environmental Protection Agency (EPA) in the USA [39]. Doses were calculated using the standard assumption for an integrated risk analysis and an average adult body weight of 65 kg [39, 42]. In addition, based on EPA guidelines, it was assumed that ingested doses were equal to absorbed contaminant doses [7].

RESULTS

The concentration (mg/kg) of heavy metals (Pb, Cr, Co, Zn, Fe and Ni) analysed in the local spices collected from various location in Awka are presented in Table 1 with the following ranges Pb (2.61 – 8.97), Cr (0.001 – 3.81), Fe (28.15 – 134.59), Zn (14.09 – 161.04), Co (0.28 – 3.07) and Ni (0.34 – 2.89).

Table 1. Concentration (mg/kg) of heavy metals in studied spices

Spices	Nickel	Cobalt	Zinc	Iron	Chromium	Lead
Prosopis Africana	0.34	1.69	15.31	134.59	1.2	8.51
Xylopia aethiopica	2.34	3.07	14.32	28.15	<0.001	6.1
Mondora myristica	1.42	0.28	161.04	84.45	0.92	8.97
Piper gineense	0.98	1.77	16.1	55.9	1.48	2.67
Capiscum frutescens	1.37	1.16	14.09	87.68	0.08	2.61
Mondora tenuifolia	2.89	1.55	23.4	83.63	3.81	<0.001
Permissible limits (WHO/FAO)	5.0	3.5	50	20	-	0.3

<0.001 = Below detectable limit
WHO/FAO - World Health Organization/ Food and Agricultural Organization, (2009) permissible limit.

The heavy metals with the highest concentration (mg/kg) was Zinc (161.04) in *Mondora myristica* and Iron (134.59) in *Prosopis africana.* While the least concentration of heavy metals was in *Xylopia aethiopia* while *Mondora tenuifolia* had Lead and Chromium levels below the detectable limit. The level of iron in all the samples was relatively high when compared with other heavy metals present in different samples. Also the least concentration of heavy metal was chromium with the highest concentration of 3.81 mg/kg in *Mondora tenuifolia*. The lead levels in *Prosopis Africana, Xylopia aethiopica, Piper gineense, Monodora myristica* and *Capsicum frutescens* were higher than the World Health Organization/ Food and Agricultural organization permissible limit of 0.3 mg/kg. The combined heavy metal contamination (mg/kg) in the local spices followed this trend *Mondora myristica* (257.08) > *Prosopis Africana* (161.64) > *Mondora tenuifolia* (115.28) > *Capiscum frutescens* (106.99) > *Piper gineense*(78.9) and *Xylopia aethiopica* (53.98). The FAO/WHO permissible limits of Pb, Zn, Fe, Co and Ni is shown in Table 1 and when compared with the result of our present work were lower than the permissible limit for spices, except for Pb, Fe and Zn (*Mondora tenuifolia*) which was above the set standard.

Table 2. Provisionally tolerable daily intake of heavy metals

Heavy metals	Concentration (mg/kg day^{-1} bw)	Regulatory body
Ni	0.0028	EFSA, 2015
Pb	0.0005	WHO/FAO, 2010
Cr	0.3	EFSA, 2014
Co	0.023	FSA, 2003
Zn	0.43	SCF, 2003
Fe	0.8	EFSA, FAO/WHO, 2010

Table 2 shows the Estimated Daily Intake (EDI) of metals from the consumption of spices. All samples were below the Tolerable Daily Intake (TDI) of the different metals when compared with the calculated result in Table 2. The THQ values of Pb, Zn, Fe, Co, Cr and Ni for the investigated samples had THQ values less than one. Also the Total Target Hazard Quotient (TTHQ) of all samples ranged from 0.06 – 0.5 (Figure 2).

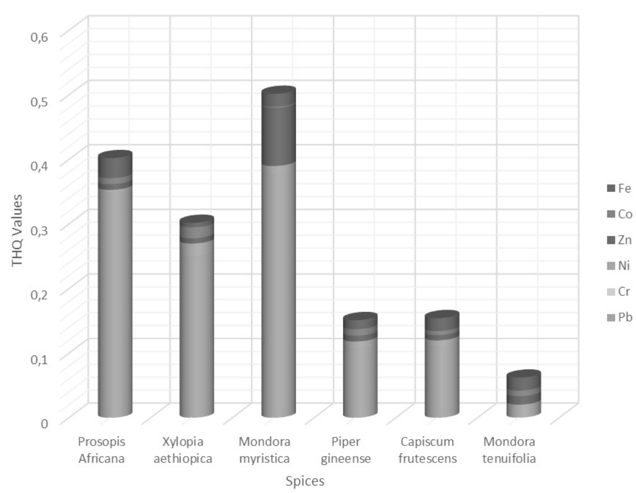


Figure 2. Target Hazard Quotient

DISCUSSION

Spices include the leaves, seeds, flowers, and/or other plant parts of herbs containing oils and other chemicals, which impart the characteristic taste and aroma give to the spices. Spices used in foods are purchased after grinding has produced a powdered substance, and mixtures of individual spices are commonly used to enhance the flavouring of prepared foods. There has been an increasing concern over the years on the adverse health effect resulting from heavy metal exposure through food consumption in Nigeria. Trace food components such as spices which are contaminated by heavy metals from the atmosphere, soil and water pose health risk to humans and animals [18]. The present study investigated the presence of toxic and essential heavy metals in local spices which is a major component of the diet among the population in South Eastern Nigeria. The growing use of these spices as flavouring, colouring and preservative agent gotten/prepared from variety of plant seed, fruit, root and vegetables which could have been contaminated by heavy metals during agricultural cultivation, industrial or human activities and from leaded gasoline [20]. However there should be strict regulation or sensitization on the level of contaminants present in locally produced spices in Nigeria, arising from food preparation processes. This may support export as there is an increased use of spices in most regions of the world including Europe, North America and Asia [27].

The concentration (mg/kg) in the present study with Pb range (< 0.001 – 8.97) were higher than those reported by others [21, 27] in Ghana, Poland and Libya. They are also lower than the level reported by *Umar* and *Salihu* [38] and *Mubeen* et al. [25] in Abuja and Pakistan respectively. In studies of heavy metal levels in artificial spices in Nigeria, *Nnorom* et al. [28] reported lead and cadmium (µg/g) levels which ranged from 3.60-3.65 for bouillon cubes, 3.9-5.05 for chicken seasoning, nd-1.80 for curry powder, 0.85- 4.80 for beef seasoning, 1.10-1.15 for thyme and 0. 80-4.90 for mixed species. Although the Estimated Daily Intake (EDI) of metals from the consumption of spices were below Tolerable Daily Intake (TDI) and the Total Target Hazard Quotient (TTHQ) were <1, the lead levels in *Prosopis africana, Xylopia aethiopica, Piper gineense, Monodora myristica* and *Capsicum frutescens* were 8-30 times higher than the World Health Organization/ Food and Agricultural organization permissible limit of 0.3 mg/kg [43]. The public health implication of this should not be overlooked. The high consumption of these natural spices in Eastern Nigeria may explain at least in part the higher blood lead levels in rural than urban pregnant women in Eastern Nigeria reported by *Njoku* and *Orisakwe* in 2012 [26]. Although most cases of lead poisoning are caused by contaminated paint and dust in older homes, a variety of unusual sources of lead exposure are occasionally found. Woolf and Woolf [44] reported families whose children were poisoned by lead-contaminated spices. A middle aged German suffered clinically severe lead intoxication that required chelation with intravenous Na-CaEDTA after using "red lead" adulterated paprika [24]. In 1994, Hungarian health officials reported that the intentional adulteration of paprika with red lead had resulted in widespread poisoning of 141 adults, many of whom were symptomatic and required chelation therapy [19]. Mixed spices generally exhibited higher value for trace metals specially lead (6.6–9.2 µg/g), cadmium (0.65–1.34 µg/g), iron (142.3–285.0 µg/g) and zinc (64.2–65.8 µg/g) [32].

The presence of heavy metals in spices has been reported from different places [1, 14, 45]. Contamination may occur accidentally through contaminated irrigation water and fertilizer or deliberately when weight and colour are deceptively enhanced for profit especially to the artificial spices.

Lead forms complexes with oxo-groups in enzymes used in haemoglobin synthesis and porphyrin metabolism [2]. Lead is a toxic metal of public health concern with no known biological function and report to induce toxicity at concentrations as low as 10 µg/kg [43].

Exposure to Cr can occur through food, it is considered an essential metal for carbohydrate and lipid metabolism [29]. The level of chromium in this study with range (0.001 – 3.81), is in contrast with levels reported for spices in Northern Nigeria; *Singh* et al. [35] reported a range of (3.87 – 7.87) in spices gotten from plants irrigated with effluent water, while *Umar* and *Salihu* [38] reported Cr levels which were below detectable limit. Although Cr exist in two forms Cr^{3+} and Cr^{6+}, the essential Cr^{3+} is mainly found in food and dietary supplements while the carcinogenic Cr^{6+} form is found in water [33].

It is known that zinc is an essential trace element not only for humans, but for all organisms, it is a component of over 300 enzymes and even greater number of other proteins, which emphasizes its indispensable role for human health [31]. The observed range of Zn in the current study was between (14.09 – 161.04 mg/kg) with *Mondora myristica* having the highest concentration of 161.04 mg/kg. The result indicates that only *Mondora myristica* exceeded the FAO/WHO permissible limit of Zinc among all the studied samples. This result is comparable with the results of *Dghaim* et al [8] and *Krejpcio* et al. [21].

There is relatively high level of Fe recorded in the present study ranging from 28.15 – 134.59 when compared to other metal concentrations across all samples. Fe serves as a constituent in proteins e.g. haemproteins: haemoglobin, myoglobin; non-haemproteins: ferritin, transferrin and as a co-factor for many important iron dependent enzymes (Cytochromes A, B, C; peroxidases, catalases) [10]. The levels reported in UAE, Egypt and Pakistan [1, 8, 25, 36] were significantly higher in comparison with our present study thus confirming that the heavy metal contents in spices varied depending on the country of origin, environmental pollution levels, plant parts and technological processes [21].

The cobalt concentration (mg/kg) varied slightly between 0.28 – 3.07. The maximum concentration is in *Xylopia aethiopica* with 3.07. Cobalt is a component of the essential Vitamin B12, important for the functioning of red blood cells, with the largest source of cobalt in humans through the diet. Although environmental exposure to toxic levels may occur in industrial settings [22] and this has been shown to cause adverse health effects which includes liver toxicity, dermatitis, endocrine and reproductive toxicity [11, 23, 30]. Although there is no permissible limit for the levels of Cobalt in food, a 1.5 mg/kg maximum permissible limit of this metal has been reported because higher levels can be related with health problems [40].

In humans nickel toxicity is influenced by the route of exposure, dose and solubility [4]. Samples with concentrations ranging from 0.34 – 2.89 in this study which were below the WHO/FAO permissible limit of 50 mg/kg in spices, suggests the levels currently detected in the various samples are within the safe limit. The most common effect of Nickel exposure is allergic reaction [16] from products like jewellery, coins and stainless steel but dietary exposure is responsible for the high intake of Nickel in the general population [6].

The toxic metals investigated in this study which includes Pb, Ni and Cr are known food and environmental contaminants, continuous ingestion from spices and other sources of heavy metal exposure taken into account is likely to result in health risk challenges. The estimated daily intake (EDI) of metals based on the assumption that an adult consumes at least 10 g of spices daily suggests that all metals were below the tolerable daily intake with the essential metals below the recommended daily intake in the investigated spices. Ingestion of heavy metals from other food sources could either augment for this concentration in essential metals or increase the body burden in toxic metals.

CONCLUSION

The lead levels in *Prosopis africana, Xylopia aethiopica, Piper gineense, Monodora myristica* and *Capsicum frutescens which* are 8-30 times higher than the World Health Organization/ Food and Agricultural Organization permissible limit of 0.3 mg/kg may contribute to the body burden of lead even amongst the rural population that consume lots of natural spices.

Conflict of interest

The authors declare no conflict of interest.

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