JASEM ISSN 1119-8362

Full-text Available Online at www.ajol.info and www.bioline.org.br/ja

J. Appl. Sci. Environ. Manage. *Sept.* 2016
Vol. 20 (3) 681-688

Heavy metal contamination of selected spices obtained from Nigeria

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ABSTRACT: In this study, the levels of trace metals (Cd, Cr, Cu, Co, Fe, Mn, Ni, Mo, Pb, Zn) in twenty two spices representing four spice groups (seeds, bulbs, leaves, fruit pods and rhizome) from a major market in Northern Nigeria were determined using atomic absorption spectroscopy, and assessed based on regulatory standards. Garlic exhibited the highest zinc concentration (21.733 ±0.044 mg/kg), which falls lower than the upper toxic level for most plants. Lead concentrations peaked in African nutmeg (4.717 ± 0.017 mg/kg) but are lower than the normative for the final dosage form of medicinal plants. The daily intake-based hazard of the spices was assessed using the minimum risk levels set by WHO, and the FAO/WHO maximum tolerable intake limits. All the spices contain excessive cobalt and copper with maximum levels (mg/kg) in ginger cobalt (11.117±0.069) and African nutmeg (15.300 ± 0.041), respectively. The estimated daily intake values (µg/kg day) of onion (1.10), ginger (12.00), utazi (1.30), alligator paper (1.20), garlic (0.89), Ashanti leaves (0.88), castor seeds (1.20), and shallot (0.86)were higher than the provisional maximum tolerable intake set by FAO/WHO, constituting a potential risk to human health. These results show that spices can accumulate exceeding levels of toxic metals whose potential risk to human health should be given priority. © JASEM

http://dx.doi.org/10.4314/jasem.v20i3.23

KEYWORDS: heavy metal; contamination; Nigeria; spices; health; daily intake

Over the millennia, spices have been used in changing world's cuisine and medicine (Dukes, 2003). Regrettably, significant quantities of heavy metals have been detected in natural food spices such as pepper and mustard (Krejpcio, 2007; Khan *et al.,* 2014)].Although low levels of some heavy metals such as chromium, iron, manganese, cobalt, zinc and copper are considered essential, even low levels of other metals such as cadmium and lead can have toxic effects in human biochemical reactions (Järup , 2003; Cao *et al.,* 2010). The accumulation of these important hazardous metals can breed middle-term or long-term health effects manifesting *inter alia* as depression, chronic asthma, liver damage, insomnia, kidney damage and neurological disorders (Mandal and Suzuki, 2002; Barakat, 2011).

Generally speaking, Cd, Pb, Cr, Cu, Zn, As and Ni are the most hazardous heavy metals in the environment (Brimer, 2011). Despite being the most ecotoxic metal, lead resides naturally in plants. Recently, Pb and Cd were listed by the Agency for Toxic Substances and Disease Registry (ATSDR, 2015) as second and seventh priority toxic substances. Consequent upon this, it is imperative to monitor the hazardous effect of the heavy metal in common condiments. In a study, Egyptian spices were found to contain alarming levels of Pb, Cd, Cr, Sn, Mn, Zn, Co, Cu, Fe and Ni (Abou-Arab & Abou Donia, 2000).

Admittedly, the extent of contamination of the spices with heavy metal varies from one plant to another. Reasons for this variation have been revealed by the work of Chizzola *et al*. (2003),which determined the levels of Cd, Fe, Cu, Zn and Pb in spices, aromatic and medicinal plants from different regions of Austria and confirmed that the tendency to accumulate Cd is species dependent while Pb uptake occurs rather by chance.

Nigerians are prone to spicy foods which lends them to the associated consequences. Previously, the levels of Cd, Pb, Cu, Cr, Ni, Fe and Zn have been determined in Nigerian sesame (Obiajunwa *et al.,* 2005) and some Nigerian spices from Warri (Iwegbue *et al.,* 2011).In this study, we investigate various common spices for possible metal (Pb, Cu, Cd, Zn, Cr, Co, Ni, Mo, Fe and Mn)contamination.

MATERIALS AND METHODS

Sample Collection and Preparation: Twenty two different spices were obtained from Yankura, Abubakar Rimi market, Kano metropolis (lat. 11° 30N, long. 8° 38E). The samples were categorized as bulbs, fruit pods, leaves, rhizome or seeds. The scientific name of the plants and the plant part used in the study are listed in
Table 1

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Table 1: List of the spices investigated with their groups, and botanical names of the source plants.

Spice group	Plant	Botanical name
Bulbs	Onions	*Allium cepa*
	Shallot	*Allium aggregatum*
	Garlic	*Allium sativum*
Rhizome	Ginger	*Zingiber officinale*
Seeds	Alligator Pepper	*Aframomum melegueta*
	Cloves	*Syzygium aromaticum*
	African nutmeg	*Monodora myristica*
	Ashanti Pepper	*Piper guineense*
	Efu	*Monodoro Sp*
	African locust bean	*Parkia biglobosa*
	Melon	*Citrullus lanatus*
	Castor seed	*Ricinus communis*
	Nutmeg	*Myristica fragrans*
Leaves	Uziza	*Piper guineense*
	Curry	*Murraya koenigii*
	Scent leaves	*Ocimum gratissimum*
	Utazi	*Gongronema latifolium*
	Bay leaves	*Lauras noiblis*
Fruit pods	Red chili pepper	*Capsicum annum*
	Yellow pepper	*Capsicum annum*
	Red bell pepper	*Capsicum annum*
	African pepper	*Xylopia aethiopica*

In order to ensure that metals determined are exclusively those uptaken by the plants, each sample was thoroughly washed with tap water and then with deionized water. The samples were then dried in air, followed by oven drying at 80 ^oC until constant weight. The dry samples were ground to fine particle and stored in airtight nylons.

Sample Preparation and Digestion: Metal extraction was accomplished by the wet digestion. Exactly 1 g of dry spice powder was weighed into 250 cm^3beakers, mixed with 20 cm^3 of 2:1 $HNO_3/HClO_4$ and heated in a fume cupboard for 5-10 min using hot plate. Completion of digestion was marked by the evolution of white fumes. The digests were allowed to cool, diluted with deionized water (to avoid chemical attack of filter paper), then filtered into 50cm^3 standard volumetric flask and made up to the mark with the deionized water. Calibration standards as well as blank were prepared at the same time as the samples. All standards were prepared from nitrates (in concentrations of 0, 0.1, 0.2, 0.4, 0.6, 0.8, and 1 mg/dm^3) except for molybdenum and manganese which were prepared from $(NH_4)_2MoO_{24}.4H_2O$ and $MnCl_2$, respectively.

Heavy Metal Determination: The filtrate resulting from wet digestion was subsequently analyzed for Pb, Cu, Fe, Cd, Zn, Cr, Co, Ni, Mo, and Mn using Hitachi Z-5000 flame atomic absorption spectrophotometer(AAS). The AAS was fueled by acetylene. Standards were analyzed accordingly but devoid of the spice. Chromium was determined in the +3 oxidation state and the standard used was Cr $(NO_3)_3.9H_2O$. The actual concentrations were extrapolated from calibration curves. Analytical concentrations in mg/dm^3were converted to mg/kg. Each analysis was repeated twice and standard deviations from the mean values were calculated.

Human Health Risk Assessment: The health risk associated with the consumption of the spices under study was estimated based on daily intake as estimated daily intake (EDI). This parameter was calculated on the uniform basis of 10 g raw spice per 60 kg body weight.

$$EDI = \frac{(C \text{ x } AC)}{bw}$$

C (mg/kg) is the concentration of heavy metals in the raw spice, AC is the average dry weight of the spice consumed by local inhabitants based on g day^{-1} $person^{-1}$ and bw is the average adult body weight (60g).

Quality Control: The method of AAS analysis was validated by recovery method. Samples of onions, melon, garlic, scent leaves and African pepper were spiked with Fe, Cu, Mn, Zn, and Mo doses, respectively. In a typical recovery test, exactly 1 $mg/dm^3$of the metal was added to 1g of each of the samples in a 250cm^3 beaker and digested with 20cm^3 2:1 $HNO_3/HClO_4$. The digest was treated as described above. Percentage recovery was calculated using the relation:

$$\text{Recovery } (\%) = \frac{\text{Amount determined}}{\text{Amount added}} \text{ x } 100$$

The results of the recovery of spikes of Fe, Cu, Mn, Zn, and Mo from onions, melon, garlic, scent leaves and African pepper, which were 90%, 89%, 90%, 95% and 97% respectively, which validates the method and assures its quality (Table 2). In other words all the metal recoveries are within 11 % of the true concentrations, thus attesting the suitability of the AAS method.

Table 2: Metal recovery efficiencies as 5 mg/dm^3 heavy metal is augmented.

Metals	Spice sample (1g)	Initial conc. (mg/kg)	Final conc. (mg/kg)	Amount recovered	Recovery (%)
Fe	Onion	275.90	280.65	0.90	90
Cu	Melon seed	2.00	6.01	0.89	89
Mn	Garlic	2.28	6.78	0.90	90
Zn	Scent leaves	8.88	13.65	0.95	95
Mo	African pepper	7.67	12.55	0.97	97

RESULTS AND DISCUSSION

Assessment of Heavy Metals: The results of heavy metal analysis are presented according to the spice group (seeds, bulbs, leaves, fruit pods and rhizome; Tables 3-7).

Table 3: Descriptive statistics of analytical concentrations (mg/kg) of heavy metals in seeds

Spices	Statistics	Cd	Cr	Cu	Co	Fe	Mn	Ni	Mo	Pb	Zn
Castor	Mean	7.800	3.150	9.450	10.550	16.667	3.750	2.733	3.233	2.700	13.050
	SD	0.054	0.010	0.027	0.019	0.084	0.025	0.014	0.033	0.011	0.022
Baobab	Mean	3.200	4.067	8.100	7.217	9.733	5.000	4.100	3.783	3.433	13.050
	SD	0.021	0.011	0.027	0.020	0.048	0.043	0.014	0.018	0.023	0.022
Efu	Mean	3.550	4.383	9.450	9.433	13.883	5.833	3.883	4.300	3.250	14.867
	SD	0.012	0.011	0.027	0.051	0.048	0.014	0.021	0.049	0.022	0.045
Ashanti pepper	Mean	4.250	3.150	7.200	9.433	18.067	6.667	4.117	6.983	2.517	14.500
	SD	0.021	0.010	0.041	0.051	0.096	0.029	0.015	0.018	0.006	0.012
Cloves	Mean	3.550	2.830	9.450	8.333	20.833	8.333	3.417	5.383	3.800	13.050
	SD	0.012	0.019	0.027	0.034	0.084	0.052	0.014	0.049	0.011	0.038
Melon	Mean	4.250	4.030	9.000	7.217	18.050	7.083	3.700	2.700	3.250	3.250
	SD	0.021	0.029	0.041	0.051	0.048	0.014	0.010	0.019	0.022	0.022
African Pepper	Mean	6.383	3.450	7.650	5.567	19.450	4.167	2.283	5.383	3.983	2.533
	SD	0.022	0.010	0.016	0.051	0.048	0.014	0.008	0.049	0.006	0.033
African Nutmeg	Mean	3.900	3.150	15.300	6.667	13.900	6.667	2.733	5.383	4.717	14.867
	SD	0.012	0.010	0.041	0.034	0.127	0.038	0.014	0.037	0.017	0.025
Alligator pepper	Mean	7.083	3.760	8.100	8.333	19.433	6.250	2.733	7.000	3.067	3.250
	SD	0.044	0.019	0.027	0.034	0.096	0.066	0.014	0.049	0.017	0.022
Nutmeg	Mean	4.250	3.760	9.900	10.000	18.050	5.000	2.717	6.983	3.800	3.250
	SD	0.021	0.019	0.031	0.033	0.127	0.025	0.024	0.018	0.011	0.022
	Min.	3.200 ±0.021	2.830 ±0.019	7.200 ±0.041	6.667 ±0.034	9.733 ±0.048	3.750 ±0.025	2.717 ±0.024	2.700 ±0.019	2.700 ±0.011	2.533 ±0.033
	Max.	7.800 ±0.054	4.383 ±0.011	15.300± 0.041	10.550± 0.019	20.833± 0.084	8.333 ±0.052	4.117 ±0.015	7.000 ±0.049	4.717 ±0.017	14.867 ±0.045

Table 4: Heavy metal concentration (mg/kg) in bulbs

Bulbs	Cd	Cr	Cu	Co	Fe	Mn	Ni	Mo	Pb	Zn
Garlic	5.317±0.022	3.450±0.010	10.350±0.031	13.333±0.067	12.500±0.083	6.250±0.025	4.100±0.014	5.383±0.018	5.083±0.017	21.733±0.044
Onion	6.383±0.022	5.650±0.019	9.450±0.027	8.333±0.034	10.550±0.145	5.833±0.038	3.650±0.021	5.383±0.037	4.167±0.006	2.883±0.013
Shallot	5.317±0.022	3.767±0.019	7.200±0.016	5.000±0.033	13.900±0.127	5.833±0.014	3.633±0.008	4.850±0.032	3.250±0.011	3.250±0.022

Table 5: Heavy metals concentration (mg/kg) in leaves

Leaves	Cd	Cr	Cu	Co	Fe	Mn	Ni	Mo	Pb	Zn
Bay leaves	4.967±0.044	2.683±0.006	12.150±0.027	7.783±0.051	12.333±0.083	5.417±0.058	2.733±0.014	4.850±0.032	3.433±0.006	12.317±0.034
Curry leaves	4.250±0.021	2.533±0.011	8.550±0.041	7.783±0.020	12.500±0.083	4.583±0.014	2.733±0.010	3.733±0.050	3.617±0.013	14.500±0.012
Uziza leaves	5.317±0.022	3.767±0.019	9.900±0.041	6.117±0.039	13.883±0.048	5.833±0.038	3.233±0.008	4.300±0.049	3.250±0.011	3.250±0.022
Scent leaves	7.450±0.021	5.650±0.019	12.000±0.027	9.453±0.019	20.833±0.144	6.250±0.025	2.733±0.014	4.300±0.049	2.883±0.006	2.883±0.013
Utazi leaves	5.317±0.022	2.533±0.011	9.900±0.041	8.333±0.034	12.500±0.083	6.250±0.025	4.333±0.008	6.450±0.032	4.167±0.017	2.167±0.022

Table 6: Heavy metals concentration (mg/kg) in fruit pods

Fruit pod	Cd	Cr	Cu	Co	Fe	Mn	Ni	Mo	Pb	Zn
Red bell pepper	6.033±0.013	4.700±0.019	12.150±0.011	5.550±0.019	13.900±0.127	5.000±0.050	3.417±0.014	3.783±0.018	4.717±0.023	3.983±0.013
Red pepper chili	5.317±0.022	3.767±0.019	11.700±0.041	7.217±0.020	16.667±0.084	5.833±0.029	2.050±0.014	5.383±0.049	3.617±0.023	3.250±0.022
Yellow pepper	4.600±0.024	4.700±0.019	8.100±0.027	5.567±0.051	13.883±0.048	6.250±0.025	4.333±0.008	5.383±0.049	3.067±0.006	2.517±0.013

Table 7: Heavy metals concentration (mg/kg) in rhizome

Rhizome	Cd	Cr	Cu	Co	Fe	Mn	Ni	Mo	Pb	Zn
Ginger	7.450±0.021	5.650±0.019	13.500±0.027	11.117±0.069	16.667±0.084	5.000±0.043	3.417±0.014	5.383±0.049	2.700±0.011	10.133±0.025

The levels of Cd, Zn, Ni, Mn, Cu, Pb, Fe, Cr, Co, Mo in seeds and their descriptive statistical parameters are shown in Table 3. Cadmium ranged from 3.200 ± 0.021 in baobab seeds to 7.800 ± 0.054 mg/kg in castor seeds. Even the lower limit of the range is above the value set for foods or products by the most regulatory bodies. A maximum permissible limit of 1 mg/kg was prescribed by the FAO/WHO (2002) for food additives. Threshold values (mg/kg) for cereal grains, leafy vegetables and fruiting vegetables are 0.1, 0.2 and 0.05 (FAO/WHO, 2012). Differently, a value of 0.03 mg/kg was prescribed by (WHO, 1999a) for medicinal plants in their final dosage form. The level of Cd in the bulbs, leaves, fruit pods and rhizome are displayed in Table 4, 5, 6and7. The tables show that all Cd values are evidently above the FAO/WHO limits. The high levels of Cd may not be unconnected with the agricultural practice, the plant and the soil properties (such as calcerous nature of soil) that affect uptake of the metal (Kabata-Pendias, 2011). A few decades back, McGrath (1993) has reported variability in Cd content (mg/kg) with plant, showing ranges of < 0.1 and 0.4 in beans, 0.9 and 8.2 in carrot and 3.6 and 91.

The mean levels of manganese (mg/kg) in the seeds (Table 3) ranged from 3.750 ±0.025 in castor seeds to 8.333 ±0.052 in cloves. The normative limit for Mn in foods has yet to be exactly specified due to non-toxicity of the metal (WHO, 1999b). In fact, plant foodstuff may contain up to 113 mg/kg of this metal and deficiency manifests within 15-25 mg/kg (Kabata-Pendias, 2011).Comparatively, the levels of manganese (mg/kg) in the bulbs (Table 4), leaves (Table 5), fruit pods (Table 6) and rhizome (Table 7) lies within the range of 5.000 ± 0.043 to 6.250 ± 0.025. Generally speaking, the levels of Mn are low in all the spices studied and do not pose any hazard risk.

Lead concentrations (mg/kg) in the spice seeds range from 2.700 ±0.011 in castor seeds to 4.717 ± 0.017 in African nutmeg (Table 3). The range covers the levels found bulbs, leaves, fruit pods and rizhome (Tables 4, 5, 6 and 7, respectively). All the values are above the 2 mg/kg allowable for food additives (FAO/WHO, 2002) and for most foods (0.1-2 mg/kg) (FAO/WHO, 2012) but lower than the prescribed values for the final dosage form of medicinal plants (10 mg/kg), which shows that the lead levels in the spice seeds are within tolerable limits. In fact, lead levels above those reported in this study (6.19 ± 1.88 mg/kg) were detected in Egyptian chamomile (Abou-Arab & Abou Donia, 2000).

The mean Ni levels in the spice seeds ranged from 2.717 ±0.024 mg/kg in nutmeg to 4.117 ±0.015 mg/kg in Ashanti pepper (Table 3) while levels in the bulbs and leaves (mg/kg) are in the ranges of 3.633 ±0.008 to 4.100 ±0.014 and 2.733 ±0.010 to 4.333 ±0.008 (Table 4 and 5), respectively. The levels of this metal (mg/kg) in fruit pods are between 2.050 ±0.014 and 4.333 ±0.008 (Table 6). Ginger had chromium content of 3.417 ±0.014 mg/kg. The levels of Ni determined in this study represent potential health risk being well above the requirement of monogastric animals (0.2 mg/kg of diet) (WHO, 1999b).Egyptian spicy leaves contained 0.61 ± 0.26 to 2.85 ± 1.85 mg Ni/kg (Abou-Arab & AbouDonia, 2000), which both exceed the limit of 0.6 mg/day beyond which skin reaction may result in nickel-sensitive humans (WHO, 1999b).

Food is the major source of exposure to chromium (WHO, 2008). While Cr(III) is essential in human body for the improvement of glucose tolerance (Mertz, 1993; Mandiwana *et al.,* 2006) excessive intake, especially of the more oxidizing Cr(VI), can harm biological systems (Michalski, 2004). The Cr(III) (mg/kg) in spice seeds (Table 3) ranged from 2.830 ±0.019 in cloves to 4.383 ±0.011 in efu. The chromium ranges in bulbs, leaves and fruit pods (Table 4, 5 and 6) fall within these limits. However, ginger (Table 7) records the highest level of this metal (5.650 ± 0.019 mg/kg). Generally, the accumulation of chromium in edible plants may constitute hazard to animals (Oliveira, 2012). In short, investigation has shown the possibility of phytotoxic effect as chromium levels exceed 1 mg/kg (Kabata-Pendias, 2011), which clearly shows possible Cr hazard with consumption of the spices.

The mean concentration of Cu in the spice seeds ranged from 7.200 ±0.041 mg/kg in Ashanti pepper to 15.300 ±0.041 mg/kg in African nutmeg (Table 3). These levels of Cu exceed the maximum acceptable daily limit of the metal in crude (fatty) foods (0.4 mg/kg) (FAO/WHO, 2012). The levels of this heavy metal in other spice groups such as bulbs, leaves, fruit pods and rizhome are displayed in Tables 4 to 7. Generally, as adjudged by the joint FAO/WHO standards (FAO/WHO, 2012), all the spices contain excessive copper levels. It is not however surprising as the mean Cu levels (mg/kg) in Egyptian chamomile and saffron were found to contain Cu levels within the same ranges (8.88 ± 1.38 and 11.3 ± 2.4, respectively) (Abou-Arab & Abou Donia, 2000).
The mean concentration of iron in the seed samples ranged from 9.733 ±0.048 mg/kg in baobab seeds to 20.833 ±0.084 mg/kg in cloves (Table 3). The values of this metal in bulbs, leaves, fruit pods and rhizome (Table 4-7) are also within the same bracket. In actual fact, several workers have shown that natural Fe content of plants can reach orders of 1000 mg/kg (WHO, 1999a). Various medicinal plants of leaves and herbs category from a number of regions of Austria have been reported to contain levels of iron well above 1000 mg/kg (Chizzola *et al.,* 2003).

There has been dramatic increase in the use of Zn-based fertilizers and addition of sludge to the soil under plants, so it is important to monitor the zinc levels in spices. In this study, zinc levels in seeds group ranged from 2.533 ± 0.033 in African pepper to 14.867 ± 0.045 in efu (Table 3). The highest Zn content (mg/kg) was obtained in garlic 21.733 ±0.044 (Table 4). Among leaves group, curry leaves have the highest zinc content (14.500 ± 0.012 mg/kg; Table 5). For a high number of plants the upper toxic level of zinc is 100 to 500 mg/kg (Kabata-Pendias, 2011) which indicates that all the zinc values determined are within tolerable limits.

Molybdenum is an essential constituent of many enzymes. Molybdenum concentration in the seeds group (mg/kg) ranged from 2.700 ±0.019 in water melon to 7.000 ±0.049 in alligator pepper (Table 3). Curry leaves had the lowest molybdenum content (3.733 ±0.050 mg/kg) where as utazi leaves accumulated the largest amount of this metal (6.450 ±0.032; Table 5). Ginger contains 5.383±0.049 mg Mo /kg (Table 7). The molybdenum concentrations of bulbs and fruit pods are close to those of the leaves and seeds groups. All the values of Mo determined in this study are below the threshold of 10 mg/kg above which most grazing animals encounter serious problems (Kabata-Pendias, 2011).

In the seeds group, cobalt concentrations (mg/kg) ranged from 6.667 ±0.034in African nutmeg to 10.550 ±0.019 in cloves (Table 3) while in the bulbs the levels of this metal ranged from 5.000 ± 0.033 in shallot to 13.333 ± 0.067 in garlic (Table 4). In leaves, the cobalt concentration ranged from 6.117 ± 0.039 in uziza to 9.453 ± 0.019 in scent leaves (Table 5).On the other hand, cobalt in fruit pods ranged from 7.217 ± 0.020 in red chili pepper to 9.450 ± 0.027 in red bell pepper (Table 6). Ginger accumulated 11.117±0.069 mg/kg of cobalt (Table 7).The symptoms of phytotoxicity of cobalt become obvious as concentration (mg/kg) ranges between 30 and 40, and hazard to animals is likely when concentrations exceed 60 mg/kg (Kabata-Pendias, 2011).Cobalt is an essential trace element being a component of vitamin B_{12} precursor and applicable in treatment of anemic patients (ATSDR, 2004). Therefore, the levels of cobalt determined in all the spice groups may not represent any potential risk.

Human Health Risk: The estimated daily intake (EDI) values of all the heavy metals are displayed in Table 8.The EDI levels of Cu and Pb in all the spices were lower than the provisional maximum tolerable intake (PMTDI)of 0.5 mg/kg day and 0.02 - 3 µg/kg day set by the joint FAO/WHO committee (FAO/WHO, 2012). Because Zn is an essential trace element and absorption depends on some other dietary factors (with average intake of 20 mg/day), the lower limit of the joint FAO/WHO standard (0.3-1 mg/kg bw) (FAO/WHO, 2012) is often exceeded. In fact, some elemental body formulas contain at least 30 mg Fe per capsule. The EDI values of cadmium for most of the spices was lower than the PMTDI for this metal (0.8µg/kg day) (FAO/WHO, 2012). However, were faced with intolerable EDI values of cadmium, associated with the consumption of onion, ginger, utazi, alligator paper, garlic, Ashanti leaves, castor seeds, and shallot.

As seen from Table 8, cobalt levels in all the spices is above the minimal risk levels for oral intake (MRLOI) (0.01 mg/kg day) recommended by ATSDR (ATSDR, 2016). Similarly, copper levels exceed the risk levels of 0.005 mg/kg day WHO (1996), meaning potential health risk for the consumers. Zinc with MRLOI of 0.3 mg/kg day, represents some risk in all the spices except in ginger, locust beans, African nutmeg, cloves, efu, melon seeds, bay leaves and castor seeds. Even though Cr in all the spices has exceeded the prescribed MRLOI of 0.1 µg/kg day (ATSDR, 2016), tolerable Cr level could be up to 250 µg/day (WHO, 1996). Because the risk level of Mo is 0.14 to 0.20 mg/kg bw (WHO, 1996), adverse effects are therefore likely to occur with consumption of all the spices except for nutmeg, alligator pepper and utazi. The Ni levels in all the spices are below the WHO permissible limit of 600 µg/day (WHO, 1996), which confirms their safety as food materials.

Table 8: Estimated daily intake of each metal (µg/kg day)

Spices	Cr	Cu	Co	Fe	Mn	Ni	Mo	Pb	Zn	Cd
Ginger	0.94	2.3	1.9	2.8	0.83	0.57	0.90	0.45	0.17	12
Onion	0.94	1.6	1.4	1.8	0.97	0.61	0.89	0.70	0.48	1.1
Garlic	0.57	1.7	2.2	2.1	0.10	0.68	0.90	0.85	0.36	0.89
Nutmeg	0.63	1.7	1.7	3.0	0.83	0.45	0.12	0.63	0.54	0.71
Locust Beans	0.68	1.4	1.2	1.6	0.83	0.68	0.63	0.57	0.22	0.53
African nutmeg	0.53	2.6	1.1	2.3	0.11	0.46	0.89	0.79	0.25	0.65
Alligator pepper	0.63	1.4	1.4	3.2	0.10	0.46	0.12	0.51	0.54	1.2
Cloves	0.47	1.6	1.4	3.5	0.14	0.57	0.90	0.63	0.22	0.59
Ashanti pepper	0.53	1.2	1.6	3.1	0.11	0.69	1.1	0.42	2.4	0.71
Efu	0.73	1.6	1.6	2.3	0.97	0.65	0.72	0.54	0.25	0.49
Ashanti leaves	0.63	1.7	1.0	2.3	0.97	0.54	0.71	0.24	0.54	0.88
Melon	0.67	1.5	1.2	3.0	0.11	0.62	0.45	0.42	0.24	0.70
Curry leaves	0.42	1.4	1.3	2.1	0.76	0.46	0.62	0.60	0.17	0.82
Utazi	0.42	1.7	1.4	2.1	0.11	0.72	0.12	0.70	0.36	1.3
Bay leaves	0.45	2.0	1.3	2.1	0.90	0.46	0.81	0.57	0.21	0.88
Castor seed	0.53	1.6	1.8	2.8	0.63	0.46	0.54	0.45	0.12	1.2
Shallot	0.63	1.2	8.3	2.3	0.97	0.61	0.81	0.50	0.54	0.86
Scent leaves	0.94	2.0	1.6	3.4	0.10	0.46	0.72	0.48	0.48	0.77
Red chili pepper	0.63	1.9	1.2	2.8	0.97	0.34	0.89	0.60	0.40	0.10
Yellow pepper	0.78	1.4	9.2	2.3	0.10	0.72	0.78	0.51	0.42	0.60
Red bell pepper	0.78	2.0	9.3	2.3	0.83	0.57	0.63	0.79	0.67	0.31
African pepper	0.58	1.3	9.3	3.2	0.70	0.38	0.75	0.66	0.42	0.12

Conclusion: The levels of Cd, Cr, Cu, Co, Fe, Mn, Ni, Mo, Pb, Zn in a spectrum of Nigerian spices was successfully determined using AAS and the health risk associated with intake of these heavy metals has been assessed. The accumulation of heavy metals by spice groups followed the order seeds>leaves>bulb>rhizome>fruit pods. For all the spices investigated, cobalt and copper levels have exceeded the risk levels set by regulatory agencies. African nutmeg contains the highest copper concentration, 15.300 ± 0.041 mg/kg, while ginger contains the highest cobalt value 11.117±0.069 mg/kg. The cadmium hazard was associated with only a few species, and cuts the cross-section of the groups.

Acknowledgement: The authors are grateful to Mal. Muhammad Musa of Research Laboratory, Department of Pure and Industrial Chemistry for his assistance to SA. Ikechukwu.

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