

## Evaluation of Effects of Heavy Metal Contents of Some Common Spices Available in Odo-Ori Market, Iwo, Nigeria

Peter Olusakin Oladoye<sup>1\*</sup> and David Olaoluwa Jegede<sup>2</sup><sup>1</sup>Chemistry Unit, Department of Science Laboratory Technology, Wolex Polytechnic, Iwo, Nigeria<sup>2</sup>Chemistry Unit, Department of Basic Sciences, Babcock University, Ilishan-Remo, Ogun State, Nigeria

### Abstract

This study is aimed at assessing the levels of Fe, Cu, Cd, and Pb in some common spices available in Odo-ori Market, Iwo, Osun State, Nigeria. Four samples each of natural spices (ginger, garlic, onion and locust beans) and processed spices (curry, thyme, nutmeg and beef spicy) were bought and analysed for Fe, Cu, Cd, and Pb using Atomic Absorption Spectrophotometry. 2 g of each of the samples was digested using 30 mL of concentrated HNO<sub>3</sub> and heated until digestion was complete. The digests were filtered into standard 30 mL volumetric flask using Whatmann filter paper and made up to mark with distilled water. The digested samples were analysed for Fe, Cu, Cd, and Pb using Buck Scientific Model 210 VGP Atomic Absorption Spectrophotometer. Results showed that the concentrations of heavy metals, such as Fe, was present at level ranging from 33.4 mg/kg – 107 mg/kg and Cu, was present at level ranging from 4.35 mg/kg - 8.40 mg/kg in natural spices. Cd was above the permissible limit set by WHO (0.30 mg/kg). The Fe and Cu levels in natural spices were below maximum permissible limit set by WHO while Cd was above the permissible limit. Therefore, it can be concluded that majority of these spices were not contaminated with the studied heavy metals except Cd which was present in natural and processed spices above MPL and indicating Cd pollution in the natural and processed spices. To avoid chronic effect (which involved accumulation of the metals in the body system and which can be harmful to human health), too much of the spices (processed) and continuing/routine use should be avoided.

**Keywords:** Spices; Heavy metals; Minimum risk levels; Spectrophotometry; Iwo; Nigeria; Maximum permissible level; Correlation coefficients

### Introduction

A spice is a dried seed, fruit, root, bark, or vegetable substance primarily used for flavouring, colouring or preserving food. Sometimes a spice is used to hide other flavours. Many spices have antimicrobial properties. This may explain why spices are more commonly used in warmer climates, which have more infectious diseases, and why use of spices is especially prominent in meats, which is particularly susceptible to spoiling [1]. Natural food spices such as pepper and mustard have been reported to contain significant quantities of some trace metals [2]. Spices have played an important role in the history of civilization, exploration and commerce as these had a universal acceptance as condiment and flavours in human diet as well as in treatment of ailments. There are evidence of plant derived aromatic compounds especially spices being used by almost all ancient civilizations, the Indian, the Egyptian, the Babylonian, the Persian, the Jews, the Chinese, the Greek and the Roman [3]. Many common spices have outstanding antimicrobial effects. On the other hand, the process of preparation and handling can make them a source of food poisoning [4]. Moreover, in the last three decades, mainly because of their medicinal values, the use of spices has increased markedly in most regions of the world. Several researches have shown that heavy metals could be present in spices and the addition of contaminated spices to food may result in accumulation of these metals in human organs. Heavy metals above the permissible levels affect human health and may result in illness to human foetus, abortion and preterm labour, and mental retardation to children. Adults also may experience high blood pressure, fatigue kidney and neurological disorder [5]. Due to the significant amount of spices consumed, it is important to know the toxic metal contents in these spices. Natural spices (ginger, garlic, onion and locust beans) and processed spices (curry, thyme, nutmeg and beef spicy) are therefore assessed for lead, cadmium, copper and iron contents and possible health risk(s).

### Materials and Methods

Eight samples of spices (ginger, onion, garlic, beef spicy, curry, thyme, locust beans, nutmeg) commonly consumed in Iwo were bought from a retail shop in Odo-Ori Market, Iwo Nigeria in October 2015. The samples were classified as natural (ginger, garlic, onion and locust bean) and processed (curry, thyme, beef spicy and nutmeg). The gross sample was dried in oven, pounded to powder and a representative sample taken. 2 gm each of the samples was digested using 30 mL of concentrated HNO<sub>3</sub> on hot-plate, filtered and made up to 30 mL. The concentration of iron, lead, copper and cadmium were determined using Buck Scientific 210 VGP Flame Atomic Absorption Spectrophotometer at the respective wavelength of the metals after calibrating the instrument with standards of Fe, Cu, Cd and Pb. Working standards of the metals were prepared from instrument's stock standard for instrument's calibration: 2 ppm, 4 ppm, 8 ppm and 10 ppm for iron; 0.2 ppm, 0.4 ppm, 0.8 ppm and 0.16 ppm for copper; 1 ppm, 2 ppm, 3 ppm and 4 ppm for cadmium and 1 ppm, 2 ppm, 3 ppm and 4 ppm for lead. The calibration curve (plot of concentration against absorbance) gave r<sup>2</sup> value of 1.00, 1.00, 1.00 and 0.99 for iron, copper, cadmium and lead respectively.

$$\text{The level of heavy metal in the sample} = \text{mg/kg} \frac{\text{Concentration of metals from instrument} \times \text{volume of digest}}{\text{mass of sample taken for digestion}} \text{mg/kg}$$

### Quality assurance

As part of the quality control measures:

- All the glasswares used for the analysis were soaked in chromic acid for 48 hours, thoroughly washed with laboratory

**\*Corresponding author:** Peter Olusakin Oladoye, Chemistry Unit, Department of Science Laboratory Technology, Wolex Polytechnic, Iwo, Nigeria, Tel: 23407038308138; E-mail: [doyeinfo@gmail.com](mailto:doyeinfo@gmail.com)

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detergents, rinsed copiously with distilled water.

- Recovery study was also carried by spiking the sample with standards of iron, lead, cadmium and copper (96.3% for copper, 98.5% for cadmium, 95.7% for iron and 91.8% for lead).

## Results and Discussion

### Distribution of heavy metals in natural spices

**Iron:** Iron concentrations in natural spices collected in Iwo vary. The iron content of natural spices consumed in Iwo ranged between the lowest concentration, 33.5 mg/kg in onion and the highest, 107 mg/kg, see Table 1. They were generally below the MPL (maximum permissible limit) and don't pose a health threat. Daily human exposure calculations estimate risks associated with hazardous substances. The adverse non-carcinogenic health effects for iron were below World Health Organization's Minimum Risk Level in all the spices (Table 2) [6]. Salihu reported iron concentrations in spices, ginger had the highest concentration, 1266 ± 140 mg/kg and garlic 115 ± 11 mg/kg the lowest. These results differ from ours, garlic and ginger had 51 and 107 mg/kg iron respectively due to the polluted soil where they grew. Compared with the WHO's Maximum Permissible Level of iron (300 mg/kg), only curry powder with 502 mg/kg and ginger with 1265 mg/kg had iron level above the limit. Calculations based on our results to determine daily spice intake showed no risk. Iron facilitates carbohydrate, protein and fat oxidation controlling body weight, an important factor in some diseases [7].

**Copper:** Analysis of copper concentrations in natural spices consumed in Iwo revealed maximum values ranged from 8 and 4 mg/kg in locust beans and onion respectively. The WHO/FAO's Maximum Permissible Limit for copper is 50 mg/kg and the Minimum Risk Level for daily intake is 0.01 mg/kg/day. The calculated daily copper intake ranged between 0.0008 mg/kg/day in onion and 0.0015 mg/kg/day in locust beans (Table 2). No natural spices contain more copper than the maximum permissible or pose an acute effect [6]. Salihu et al. revealed the copper content of spice samples analysed ranged from 9 ± 0.6 to 33 ± 3 mg/kg in cinnamon and black pepper respectively. All samples were below the WHO/FAO's (WHO/FAO MPL) maximum permissible limit except locust beans above their 20 mg/kg MPL. Excess copper can cause oily skin, loss of skin tone (it blocks vitamin C) and can lead to hair loss especially in women [2].

**Cadmium:** The cadmium content of natural spice samples ranged from 0.45 mg/kg, in garlic, locust beans and onion and 0.3 mg/kg in ginger, see Table 3. Their cadmium levels exceeded the MPL (maximum permissible limit) of 0.3 mg/kg except in ginger. Calculated daily intakes from 10 g of the spices for 55 kg body weight showed no appreciable risk of adverse non-cancer health effects. Excess cadmium targets the liver, placenta, kidneys, brain and bones [8].

**Lead:** Lead wasn't detected in the natural spices, probably it was below the instrument's detection limit. The MPL (maximum permissible limit) for lead is 10 mg/kg and its MRL (Minimum Risk Level) is 0.0002 mg/kg. Lead is the most recognised toxic environmental pollution. Toxic levels of lead in man are associated with encephalopathy seizures and mental retardation [9].

### Distribution of heavy metals in processed spices

**Iron:** The WHO MPL 300 mg/kg iron limit wasn't exceeded but values ranged from 29 to 136 mg/kg for beef spicy and curry powder respectively, see Table 4. All are below the limit and may be considered tolerable. Calculated daily iron intakes ranged from 0.005 mg/kg/day in

S/N	Spice name	Samples code	Fe mg/kg	Cu mg/kg	Cd mg/kg	Pb mg/kg
1.	Garlic	Spice -1	51.0	4.65	0.45	<DL*
2.	Ginger	Spice-2	107	5.10	0.30	<DL
3.	Locust beans	Spice-3	59.3	8.40	0.45	<DL
4.	Onion	Spice-4	33.5	4.36	0.45	<DL
Range			33.5 - 107	4.35 - 8.40	0.30 - 0.45	<DL
W.H.O (Maximum Permissible Limit)			300	50	0.3	10

\*DL: Detection limit for heavy metals are Cu 0.005 mg/L, Fe 0.05 mg/L, Pb 0.08 mg/L, Cd 0.01mg/L

**Table 1:** Concentration of heavy metals in natural spices in mg/kg.

S/N	Spice name	Samples code	Fe mg/kg	Cu mg/kg	Cd mg/kg	Pb mg/kg
1.	Garlic	Spice -1	$0.9 \times 10^{-2}$	$0.8 \times 10^{-3}$	$8.2 \times 10^{-5}$	<DL*
2.	Ginger	Spice-2	$1.9 \times 10^{-2}$	$0.9 \times 10^{-3}$	$5.4 \times 10^{-5}$	<DL
3.	Locust beans	Spice-3	$1.0 \times 10^{-2}$	$1.5 \times 10^{-3}$	$8.2 \times 10^{-5}$	<DL
4.	Onion	Spice-4	$0.6 \times 10^{-2}$	$0.8 \times 10^{-3}$	$8.2 \times 10^{-5}$	<DL
Range			$0.6 \times 10^{-2}$ - $1.9 \times 10^{-2}$	$0.8 \times 10^{-3}$ - $1.5 \times 10^{-3}$	$5.4 \times 10^{-5}$ - $8.2 \times 10^{-5}$	<DL
W.H.O (Minimum Risk Level)			$70 \times 10^{-2}$	$10 \times 10^{-3}$	$20 \times 10^{-5}$	$2 \times 10^{-4}$

**Table 2:** Calculation of daily intake level of each natural spice for various metals (mg/kg/day) on assumed 10 g consumed by 55 kg body weight.

S/N	Spice name	Sample code	Iron	Copper	Cadmium	Lead
1.	Garlic	Spice-1	Chronic effect	Chronic effect	Chronic effect	No effect
2.	Ginger	Spice-2	Chronic effect	Chronic effect	Chronic effect	No effect
3.	Locust beans	Spice-3	Chronic effect	Chronic effect	Chronic effect	No effect
4.	Onion	Spice-4	Chronic effect	Chronic effect	Chronic effect	No effect

**Table 3:** Effects of daily intake of heavy metal (mg/kg/day) on consumption of 10 g of spices, effect based on 55kg of human body weight.

S/N	Spice name	Sample code	Fe mg/kg	Cu mg/kg	Cd mg/kg	Pb mg/kg
1.	Thyme	Spice-5	124	5.40	0.45	<DL*
2.	Curry	Spice-6	136	6.75	0.45	6.60
3.	Nutmeg	Spice-7	39.3	6.90	0.30	<DL
4.	Beef Spicy	Spice-8	29.0	1.35	<DL	<DL
Range			29.0 - 136	1.35-6.90	0.00-0.45	0.00 - 6.60
W.H.O MPL (Maximum Permissible Limit)			300	50	0.3	10

\*DL: Detection limit for heavy metals are Cu 0.005 mg/L, Fe 0.05 mg/L, Pb 0.08 mg/L, Cd 0.01mg/L.

**Table 4:** Concentration of heavy metals in processed spices in mg/kg.

beef spicy to 0.025 mg/kg/day in curry powder, see Table 5, also below the MRL, 0.7 mg/kg/day and posing no health threat or hazard. They don't affect health [10]. Gulzar et al. reported relatively high iron levels in foods, citing spice sample iron contents ranging from 56 mg/kg for cardamom to 650 mg/kg for mint. Calculated daily spice intakes based on their results showed no health risk. Iron also has important roles in various bodily functions.

**Copper:** The results of heavy metal determination in processed spice samples consumed in Iwo showed spice copper concentrations ranging from 1.35 to 6.90 mg/kg, see Table 4. Generally copper levels were below the MPL and the calculated daily intakes ranged from 0.002 mg/kg/day in beef spicy to 0.0013 mg/kg/day in nutmeg, see Table 5,

also below the 0.01 mg/kg/day minimum risk level [11]. Ozkutlu et al. reported 21 mg/kg copper in locust bean, ginger and garlic were both low, 9 mg/kg. All spices contained less than half the WHO limit for copper and were relatively tolerable [12]. Nkansah et al. reported 3.11 mg/kg copper in some spices; previous work 6–17 mg/kg. Excess copper damages liver and kidney and can be lethal [13].

**Cadmium:** Cadmium concentrations in processed spices were undetectable in the beef spicy sample, levels in other spices ranged from 0.30 mg/kg in nutmeg to 0.45 mg/kg in thyme and curry, see Table 4. Those in thyme and curry spices exceeded the MPL and calculated daily intakes ranged between 0.000055 mg/kg/day in nutmeg and 0.000082 mg/kg/day in thyme and curry, see Table 5, below WHO's minimum risk level [11]. Ozkutlu et al. revealed cadmium concentrations ranging between 0.012 mg/kg for turmeric and 1.30 mg/kg for ginger. Cadmium targets blood vessels, heart tissue, kidneys, lungs and brain causing heart disease, hypertension, liver damage and suppressing the immune system with other nasty symptoms [14].

**Lead:** The 6.60 mg/kg of lead found in curry didn't exceed the 10 mg/kg Maximum Permissible Limit. The calculated daily lead intake from curry was 0.0002 mg/kg/day, the same as WHO's figure [6]. Salihu et al. reported the highest lead concentration,  $22 \pm 10$  mg/kg in ginger, the lowest,  $5 \pm 1$  mg/kg in negro pepper. Those in ginger, long pepper, cherry pepper and curry powder exceeded the 10 mg/kg MPL. A 60 kg human's estimated daily lead intake from 10 gm of spice would be risky. According to WHO/FAO, lead's MRL is 0.0002 mg/kg/day.

### Metal-metal correlation study

**Natural spices:** A correlation coefficient indicates the strength of relationship between two variables. A Microsoft Excel 2007 was used to determine multiple correlation coefficients for metals in natural spices, see Table 6. According to Salihu [6], there's no relationship if it's 0.0; a value of 1.0 indicates absolute dependency and if negative they're said to oppose one another. Correlation coefficients <0.50 are less significant than those >0.50. Table 7 shows insignificant correlations ranging from -0.939 to 0.187, implying no relationship, they're from different sources. Fe-Cu are oppositely correlated with a negative correlation coefficient.

**Processed spices:** Table 8 shows the metal-metal correlation coefficients for processed spices. Those for processed spices are all significant, >0.50 except for Cu-Pb. In decreasing order, the correlation coefficients are 0.930, 0.766, 0.645, 0.522, 0.517 and 0.424 for Fe-Cd, Cu-Cd, Fe-Pb Cd-Pb and Fe-Cu respectively. Fe-Cd (0.930) and Cu-Cd (0.766) are strongly correlated, suggesting they came from the same source.

S/N	Spice name	Sample code	Iron	Copper	Cadmium	Lead
1.	Thyme	Spice-5	$2.3 \times 10^{-2}$	$0.9 \times 10^{-3}$	$8.2 \times 10^{-5}$	
2.	Curry	Spice-6	$2.5 \times 10^{-2}$	$1.2 \times 10^{-3}$	$8.2 \times 10^{-5}$	$2.0 \times 10^{-4}$
3.	Nutmeg	Spice-7	$0.7 \times 10^{-2}$	$1.3 \times 10^{-3}$	$5.5 \times 10^{-5}$	
4.	Beef Spicy	Spice-8	$0.5 \times 10^{-2}$	$0.2 \times 10^{-3}$	<DL	
Range			$0.5 \times 10^{-2}$ - $2.5 \times 10^{-2}$	$0.2 \times 10^{-3}$ - $1.3 \times 10^{-3}$	$9.5 \times 10^{-5}$ - $8.2 \times 10^{-5}$	$0.00 \times 10^{-4}$ - $2.0 \times 10^{-4}$
W.H.O MRL (Minimum Risk Level)			$70 \times 10^{-2}$	$10 \times 10^{-3}$	$20 \times 10^{-5}$	$2.0 \times 10^{-4}$

**Table 5:** Calculation of daily intake level of each processed spice for various metals in mg/kg/day on assumed 10 g consumed by 55 kg human body weight.

S/N	Spice name	Sample code	Iron	Copper	Cadmium	Lead
1.	Thyme	Spice-5	Chronic effect	Chronic effect	Chronic effect	No effect
2.	Curry	Spice-6	Chronic effect	Chronic effect	Chronic effect	Chronic Effect
3.	Nutmeg	Spice-7	Chronic effect	Chronic effect	Chronic effect	No effect
4.	Beef Spicy	Spice-8	Chronic effect	Chronic effect	Chronic effect	No effect

**Table 6:** Effect of daily intake of heavy metals in mg/kg/day on consumption of 10 g of spices, effect based on 55 kg of human body weight.

	Iron	Copper	Cadmium	Lead
<b>Iron</b>	1			
<b>Copper</b>	0.089	1		
<b>Cadmium</b>	-0.939	0.187	1	
<b>Lead</b>	0	0	0	1

**Table 7:** Multiple Metal Correlation Coefficients for Natural Spices.

	Iron	Copper	Cadmium	Lead
<b>Iron</b>	1			
<b>Copper</b>	0.517	1		
<b>Cadmium</b>	0.930	0.766	1	
<b>Lead</b>	0.645	0.424	0.522	1

**Table 8:** Multiple Metal Correlation Coefficients for Processed Spices.

### Conclusion

Generally, heavy metal concentrations in both natural and processed spices were below WHO maximum permissible limits. Consuming them won't have any acute effect, calculations based on a 55 kg person consuming 10 gm daily confirmed they were below the minimum risk level. About 62.5% of eight samples exceeded the tolerable limit for cadmium. In conclusion, most spices used in Iwo, Osun State, Nigeria weren't contaminated with heavy metals but Cd in natural and processed spice exceeded the MPL (Maximum Permissible Limit). To avoid the chronic effect of their bodily accumulation, which can harm health, their excessive and/or routine use should be avoided.

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