#### © 2010 Cenresin Publications www.cenresin.org

# FOOD GRINDING STONES AS A SOURCE OF HEAVY METALS CONTAMINATION OF DIETS

### Richard C. Ehiri; Ugochukwu G. Megwa and Omaka N. Omaka Department of Industrial Chemistry Ebonyi State University Abakaliki, Nigeria E-mail: richcee2003@yahoo.com

# ABSTRACT

The heavy metals contamination of diets by using food grinding stones in ten families in Nkalagu, Ebonyi State, Nigeria is reported in this study. Melon seeds were used in both the experimental and control samples and heavy metal analysis was carried out using atomic absorption spectrometry (AAS). Results show that the mean concentration of heavy metals added by the stones to diets were 0.184, 0.259, 0.204, 0.790, 2.390 and 0.648 ppm for arsenic, cadmium, lead, copper, iron and zinc respectively. These values were all below the recommended dietary allowance (RDA) and tolerable upper intake levels (UL) for copper, iron and zinc. Furthermore, the proportion of these metals added by the grinding stones is significant for copper (p>0.05) and not significant for both iron and zinc (p<0.05) at the 5% level of significance when compared with their RDA values. Adequate care should be taken in using food grinding stones to avoid heavy metals loading of diets and their associated toxicity.

Keywords: Heavy Metal; Contamination; Diet; Food Grinding Stone.

## INTRODUCTION

Food grinding stones (FGC) are slabs of stones made from abrasive rocks that people use to grind, crush or pound food materials such as seeds, nuts, bulbs and berries during food preparation<sup>1</sup>. The use of dietary food grinding stones or rocks is a common practice in food preparation and processing in Nigeria and many other developing nations. These stones, invariably, contain varying amounts of mineral nutrients that regulate many vital biological processes. Heavy metals can be associated with pollution and toxicity but some may be essential for humans and other living organisms at approved concentrations<sup>2</sup>.

The determination of trace elements particularly heavy metals such as arsenic, mercury, chromium, iron, lead, cadmium, cobalt, copper, manganese, nickel, zinc, etc. has received increasing attention in food chemistry, nutrition, and pollution studies<sup>3</sup>. Flame, Zeeman and graphite furnace atomic absorption spectrometry (AAS) are the most common techniques for heavy metals estimation<sup>4-7</sup>. These techniques are very sensitive, precise, robust, highly accurate and have high detection limits.

Heavy metals have geochemical origin where they occur as trace constituents of rocks<sup>8, 9</sup>. Small amount of these elements are common in our diets and some are actually necessary for good health, but large amount of any of them may cause acute, severe or chronic toxicity. Some may, outrightly, not be needed at all for body processes and functioning<sup>10, 11</sup>. Heavy metals have the potential to bio-accumulate in food chain or web. Bio-accumulation of these metals has great significance if the metal's toxicity threshold limit is exceeded<sup>12, 13</sup>. Furthermore, bio-availability of metals depends on such factors as pH, metal valence state,

#### Food Grinding Stones as a Source of Heavy Metals Contamination of Diets

type of chelating agents present, the pattern and amounts of other elements present<sup>14</sup>. Toxicity of metals is associated with their undergoing complexation reactions with organic molecules whereby they are incorporated into structural forms in the biological systems<sup>15</sup>. Essentially a modified bio-molecule is formed in the reaction and this may result in malfunctioning and eventually death of the affected molecules<sup>15, 16</sup>.

The objective of the present study is to evaluate the effect of using food grinding stones with special regards to the stones' contaminating diets with heavy metals namely arsenic (As), cadmium (Cd), copper (Cu), iron (Fe), lead (Pb) and zinc (Zn).

Arsenic, cadmium and lead are toxic metals and non-essential to the human body. They are not required by the body in any amount<sup>17</sup>. Arsenic is a toxic element that has high potential risk especially its inorganic forms. Exposure to inorganic arsenic increases the risk of skin, bladder and kidney cancer<sup>18, 19</sup>. Also cardiovascular and neurological effects have been attributed to inorganic arsenic<sup>18</sup>. Cadmium derives its toxicological properties from its chemical similarity to zinc, an essential micro-nutrient for animals, plants and human beings<sup>20</sup>. Cadmium easily displaces zinc in metalloenzymes and consequently alters the stereochemistry of the latter. It also produces bone defects in human and animals<sup>11</sup>. Cadmium also causes poor reproductive capacity, hypertension and hepatic failure<sup>11, 20</sup>. Lead is a toxic metal that is well-known. It has severe deleterious effects on human even at low concentrations. It causes renal and liver dysfunction and failure<sup>11, 21</sup>.

Copper, iron and zinc are essential heavy metals. However, at higher concentrations than the recommended dietary allowances (RDA)<sup>22, 23</sup>, these elements can be harmful to the body. Copper works hand in hand with iron. It is involved with the storage and release of iron to form haemoglobin for red blood cells<sup>24</sup>. The RDA of copper is 900µg/day and its tolerable upper intake level (UL) is 10000µg/day<sup>23</sup>. An excess amount of copper in the human body causes anemia, liver and kidney damage as well as stomach and intestinal irritation<sup>25</sup>. Iron is an important trace element in the body's chemistry. It transports oxygen and carbon dioxide in the body as well as mediation in electron-transfer reactions. The RDA and UL of iron are 8mg/day and 45mg/day respectively<sup>23</sup>. Excess amount of iron in the body may be deposited in the liver, lungs, pancreas and heart causing hemmosiderosis<sup>26, 27</sup>. Zinc is an important physiological trace element in the human body. It plays vital roles in the biosynthesis of enzymes, auxins and some proteins<sup>27</sup>. It is widely distributed in various parts of the body respectively<sup>23</sup>. Studies show that high zinc intake may depress platelet activity in animals and humans<sup>17,20</sup>.

Accurate information on the concentration of heavy metals in diets and food supplements and their contributory factors is needed for health risk assessment due to toxicity and harmful effects of heavy metals<sup>28</sup>. The right amounts of minerals in our diets are necessary for good health and growth and insufficient supply of minerals to the body causes various types of deficiency diseases or even death.

### Journal of Sciences and Multidisciplinary Research

This paper therefore presents the results of the analysis carried out on melon seeds ground on selected FGCs in order to determine the amounts of heavy metals added into diets by grinding stones. Melon seeds were chosen for the study because it has been reported that these seeds do not contain significant amounts of heavy metals<sup>29</sup>. Graphite furnace atomic absorption spectrometry (GFAAS) was used as the analytical procedure. We hypothesize that significant amounts of heavy metals are added into diets by the food grinding stones used to grind the melon seeds.

# MATERIALS AND METHOD

## Sample Collection

The study area for sample collection was Nkalagu town in Ebonyi State, Nigeria. A large proportion of the families in this town use grinding stones in food processing. These stones consist of a larger lower stone slab and a smaller upper stone. The smaller stone is held on both hands while being used to crush the melon seeds. Ten families, namely, Okeagu, Ezeoha, Ngele, Uzuu, Nnaji, Ogbunkwu, Aliede, Ogba, Eneh and Udeh were randomly selected for use in the study.

10 g of the melon seeds were ground on the food grinding stones for 30 min and thereafter scrapped from both stones and put in clean black polythene bags for storage pending digestion and analysis.

# Reagents

Analar grade reagents and chemicals were used. These are nitric acid, sulphuric acid and deionized water

# Sample Digestion

Digestion was carried out using the method of the Analytical Methods Committee<sup>30</sup>. 5g of dry the ground melon seeds was put in the digestion flask placed on a hot plate. 5 cm<sup>3</sup> of concentrated nitric acid was added. A vigorous initial reaction and foaming occurred but later subsided. The mixture was heated gently on the hot plate and cooled. 8 cm<sup>3</sup> of concentrated sulphuric acid was added to the mixture at a rate that did not cause frothing. The mixture was heated until it began to darken owing to incipient charring.

1 cm<sup>3</sup> of concentrated nitric acid was slowly added to the mixture and heated. This procedure of adding 1 cm<sup>3</sup> of concentrated nitric acid and heating thereafter was continued until the sample solution failed to darken on prolonged heating to fuming for 10 minutes. At the end of digestion the resulting solution was cooled and diluted with 10 cm<sup>3</sup> of de-ionized water, evaporated and cooled. This treatment was repeated with 5 cm<sup>3</sup> of de-ionized water. Finally the sample solution was cooled and diluted with 5 cm<sup>3</sup> of de-ionized water<sup>30</sup>. The control sample was pure melon seeds. 10g of pure melon seed was weighed and ground on a crucible mortar and pestle for 30 min and thereafter scrapped from both stones and put in clean black polythene bags for storage pending digestion and analysis. The digestion procedure used for the stone-ground samples was carried out for the control sample.

# Heavy Metal Analysis

The heavy metal analysis of the control and experimental samples was done at Project Development Institute (PRODA), Enugu, Nigeria. The sample solution was placed in Parking Elmer A Analyst 400 atomic absorption spectrometer to aspirate for the presence of heavy metals. All analyses were done in triplicates. Standard addition technique (SAT) was used as calibration method to check and correct matrix effects.

# **RESULTS AND DISCUSSION**

The amount of heavy metal added into diets as a consequence of grinding melon seeds on the stones in the ten different families in Nkalagu is presented in Table 1. Generally, the increase in concentration of the heavy metal in the experimental sample over the control sample represents the amount of heavy metal added by the stone. Results show that the mean amount of heavy metals added by the grinding stones to diets were 0.184, 0.259, 0.204, 0.790, 2.39 and 0.648ppm for As, Cd, Pb, Cu, Fe and Zn respectively. The highest concentration of metal added by grinding stone was 2.390  $\pm$  0.370 ppm iron while the least was 0.184  $\pm$  0.033 ppm arsenic.

The melon seeds showed a great tendency to stick to the surfaces of the stones during grinding action and thus greater friction occurred. This effect may be responsible for the increase in the amount of heavy metals added. Nevertheless, the geochemical composition of the stones or rocks will affect the amount of metals added. Other factors that may affect the amount of trace element added by stones include type of food material ground and time taken or duration of the grinding process or action.

Family	Arsenic	Cadmium	Lead	Copper	Iron	Zinc
name						
Okeagu	0.24	0.28	0.20	1.20	0.40	0.80
Ngele	0.28	0.08	0.22	1.00	1.20	0.40
Ezeoha	0.08	0.64	0.40	0.60	4.20	1.00
Uzuu	0.04	0.04	0.10	0.60	2.00	0.80
Ogbunkwu	0.28	0.48	0.20	0.80	2.40	0.60
Nnaji	0.06	0.30	0.20	0.70	2.20	0.48
Aliede	0.24	0.05	0.10	0.60	2.60	0.50
Ogba	0.08	0.40	0.20	0.80	3.10	0.40
Eneh	0.28	0.08	0.30	1.00	4.00	0.80
Udeh	0.26	0.24	0.12	0.60	1.80	0.70
Range	0.04 - 0.28	0.04 - 0.28	0.10 -	0.60 –	0.40 –	0.4 - 1.00
			0.40	1.20	4.20	
Std Dev	0.104	0.203	0.092	0.213	1.178	0.203
Mean + SE	0.184±0.033	0.259±0.064	0.204 ±	0.79 ±	2.39 ±	0.648 ±
Mean			0.029	0.067	0.370	0.064

# Table 1: Heavy metal concentration (ppm) added to diets by grinding stones from differentfamilies studied

SE Mean: Standard error of the mean; Std Dev: Standard Deviation.

## Journal of Sciences and Multidisciplinary Research

## Volume 2, September 2010

Table 2 shows the amount of heavy metal added by the stones being compared with International Standards for daily dietary intakes and tolerable limits for these trace elements for a 25-year old male. Results indicate that the values of heavy metals added by the stones are all below the RDA and very far below the UL values.

Table 2: Comparison of heavy metal added by stones with the RDA and UL <sup>23</sup> for a 25 year old male							
Element		Arsenic	Cadmium	Lead	Copper	Iron	Zinc
Amount added stone mg/L	by in	0.184	0.259	0.204	0.790	2.390	0.648
RDA mg/day	in	ND	ND	ND	0.900	8.000	11.000
UL mg/day	in	ND	ND	ND	10.000	45.000	40.000

**RDA:** Recommended Dietary Allowance; UL: Tolerable Upper Intake Level; ND: Not Determined, element not essential to body.

Table 3: Significant tests<sup>31</sup> on difference between mean of heavy metals added compared with the RDA for copper, iron and zinc

Elemen t	Variabl e	Numbe r	Mea n	Std Dev.	SE Mea n	t- value	Probabilit Y	95% CI
Copper	ppm	10	0.790	0.21 3	0.067	–1.63 <sub>NS</sub>	2.26	(0.638,0.942 )
Iron	ppm	10	2.39	1.17 8	0.370	_ 15.06*	2.26	(1.554,3.226 )
Zinc	ppm	10	0.648	0.20 3	0.064	161.10 *	2.26	(0.503,0.793 )

# **Std Dev: Standard deviation; SE Mean: Standard error of the mean; CI: Confidence Interval**.

The result of the significance tests on difference of means<sup>31</sup> carried out on the data obtained at the 5% level of significance is presented in Table 3. The hypothesis that significant amount of heavy metals are added by the grinding stones to diets is accepted for copper at the 5% level of significance. The value specified by the null hypothesis, 0.900 ppm is contained in the 95% CI (Confidence Interval) for the mean ppm copper. However no significant amounts of iron and zinc were added by the stones because 8.000 ppm and 11.000 ppm are not contained in the 95% CI for the mean ppms of iron and zinc respectively.

# CONCLUSION

Food grinding stones have been used over the ages in food processing by families. In this study the contribution of heavy metals – As, Cd, Pb, Cu, Fe and Zn – to diets was carried out. Results show that grinding stones in no small measures contaminate diets when they are used in food processing. Grinding stones should therefore be taken into account when evaluating the dietary composition and pattern of the people that use stones in food processing with special regards to heavy metals contamination and toxicity.

# REFERENCES

- 1. State Government of Victoria (2008) Aboriginal Grinding Stones, Department of Planning and Community Development, Melbourne, ISBN 978 1 9 21331 60 2, pp1-2.
- Njoku P. C. and Ayoka A. O. (2007) Evaluation of Heavy Metal Pollutants from Soils at Municipal Solid Waste Deposit in Owerri, Imo State, Nigeria, *J. Chem. Soc. Nigeria* 32 (1), 57-60.
- 3. Yamini Y. and Amiri N. (2001) Solid-Phase Extraction, Separation and Visible Spectrophotometric Determination of Trace Amounts of Iron in Water Samples, *Journal of AOAC International* **84 (3)**, 713-717.
- 4. Welz B. (1985) Atomic Absorption Spectroscopy, VCH, Amsterdam, The Netherlands.
- 5. Marczenco Z. (1986) Separation and Spectrophotometric Determination of Elements, Ellis Harwood, London, UK.
- Greenberg A. E., Clesceri L. S. and Eaton A. T. (1995) ' Standard Methods for Examination of Water and Waste Water" 19<sup>th</sup> Ed., *American Public Health Association*, Washington DC, pp. 3-67.
- 7. Ademoroti C. M. A. (1996) Standard Methods for Water and Effluents Analysis, Foludex, Ibadan, Nigeria, pp. 97-145.
- 8. Brush G. J. (1988) Determinative Mineralogy and Blowpipe Analysis 16<sup>th</sup> Ed., John Wiley & Sons, New York, pp. 2-4.
- 9. Koauskopf K. B. (1967) Intoduction to Geochemistry, McGraw Hill, New York, pp. 166-168.
- 10. Mayer J. (1971) 'Trace Minerals: Fact or Fad", *Family Health*, March 1971, pp. 54-55.
- 11. Biney C. A., Calamari A. T. D., Kaba N., Naeve H. and Saad M. A. H. (1991) Review of Heavy Metals in the African Aquatic Environment, *Ecotoxicology and Environmental Safety*, 134-159.

- Tulonen T., Pihlström M., Arvola L. and Rask M. (2006) Concentration of heavy metals in food web components of small, boreal lakes, *Boreal Environment Research*, Helsinki 11: 185-194.
- 13. World Health Organization (WHO) Guideline for Drinking Water Quality, Geneva, USA, 1 (7) 52-73.
- 14. Doisy E. A. (Jr.) (1972) "Micronutrient Control on Biosynthesis of Clotting Proteins and Cholesterol" In D. Hemphil (Ed.), Trace Substances in Environmental Health – VI. Proceedings of University of Missouri's 6<sup>th</sup> Annual Conerence on Trace Substances in Environmental Health, University of Missouri, Columbia, MO, p.193.
- 15. Miethinen J. K. (1975) The Accumulation and Excretion of Heavy Metals in Aquatic Environment, A. Peter (Ed.) Krental Pergamon pp. 153-156.
- 16.Underwood E. J. (1977) "Trace Elements in Human and Animal Nutrition", 4<sup>th</sup> Ed., Academic Press, New York, p. 208.
- 17. Ferguson J. E. (1980) The Heavy Elements Chemistry: Environmental Impact and Health Effects, Pergamon, Oxford, pp. 251-254.
- 18. Goessler W. and Pavkov M. (2003) Accurate quantification and transformation of arsenic compounds during wet ashing with nitric acid and microwave assisted heating, *The Analyst*, **128**, 796-802.
- 19. Francesconi K. A. and Kuehnelt D. (2002) In *Environmental Chemistry of Arsenic*, W. T. Frankenberger, Jr. (Ed.), Marcel Dekker, New York, pp. 51-116.
- 20. Williams D. R. (1976) An Introoduction to Bio-Inorganic Chemistry, Charles Thomas, Springfield, Illinois, pp. 152-158.
- 21. Akporhonor, E. E., Iwegdue, C. M. A., Egwaikikhide P. A. and Emua S. A. (2007) Levels of cadmium, lead and mercury in organs of some fish species from Warri River, Nigeria. *J. Chem. Soc. Nigeria*, **32 (1)** 221-226.
- 22. Nestle M. (2002) Food Politics, Berkley, University of California Press.
- 23. Dietary Reference Intakes (DRIs): *Recommended Intakes for Individuals*, Food and Nutrition Board, Institute of Medicine, National Academies, USA, 2004.
- 24.Orton J. M. and Neuhaus O. W. (1975) Human Biochemistry 9<sup>th</sup> Ed., C V Mosby, Louis, pp. 529-554.

#### Food Grinding Stones as a Source of Heavy Metals Contamination of Diets

- 25. Budinova T. K., Gergova K. M., Petrov H. V. and Minka V. N. (1994) "Removal of metal ions from aqueous solution by activated carbon obtained from different raw materials" *J. Chem. Tech and Biotech.* **60**: 179-180.
- 26. Debruin E. J. P. (1970) Iron Absorption in the Bantu, J. Am. Diet. Assoc. 57, 129-31.
- 27. Committee on Iron efficiency, Council on Foods and Nutrition (1968) "Iron Deficiency in the United States, *J. A. M. A.* 203: 407-14.
- 28. Janssen C. R., De Schamphelaere K., Heijerick D., Muyssen B., Lock K., Bossuyt B. Vangheluwe M. and Van Sprang P. (2000) Uncertainties in the environmental risk assessment of metals, *Hum. Ecol. Risk Assess.* 6: 1003-1018.
- 29. Adekunle A. A. and Oluwo O. A. (2008) The Nutritive Value of Cucumis melo var. agrestis serad (Cucurbitaceae) Seeds and Oil in Nigeria. *Am J. Food and Tech.* **3 (2)**: 141-146.
- 30. Analytical Methods Committee (1960) "Methods for the Destruction of Organic Matter", *The Analyst*, **85**: 646-647.
- 31.Spiegel M. R. and Stephens L. J. (1999) "Schaum's Outlines Statistics", 3<sup>rd</sup> Ed. Mc Graw Hill, New York, pp. 216- 227.