

## Determination of Aluminium in Different Sources and its Contribution to Daily Dietary Intake in Nigeria

<sup>1</sup>E.J. Ekanem, <sup>1</sup>J.A. Lori, <sup>1</sup>F.G. Okibe, <sup>1</sup>G.A. Shallangwa,

<sup>2</sup>B.A. Anhwange, <sup>3</sup>M. Haliru and <sup>4</sup>A.A. Moyosore

<sup>1</sup>Department of Chemistry, Ahmadu Bello University, Zaria, Nigeria

<sup>2</sup>Department of Chemistry, Benue State University, Makurdi, Nigeria

<sup>3</sup>Department of Chemistry, Federal College of Education, Zaria, Nigeria

<sup>4</sup>Department of Chemistry, Federal College of Education, Katsina, Nigeria

**Abstract:** Aluminium (Al) is well known to be a toxic metal, particularly in patients with chronic renal dysfunctions. It is therefore, crucial to determine the levels of the element in dietary matrices with a view to estimating the daily dietary intake. In the present research, the total content of Al from different sources is present. Analytical technique employed is Graphite Furnace Atomic Absorption Spectrometry (GFAAS). Samples analyzed were beef baked in Aluminium foil, drugs (tablets) and beverages. Results indicate Al in the range 59.83-220.20 mg kg<sup>-1</sup> baking duration 60-180 min beef, 1.05-1.42 mg g<sup>-1</sup> drugs and 0.171-0.481 mg g<sup>-1</sup> for both bottled and Al canned beverages. The standard deviation of the means is from ±1.31 ±69.54, ±0.055 ±0.187 and ±0.105 ±0.117; baked beef, drugs and beverages, respectively. Al content is lower than the daily dietary intake of 60 mg for an average body weight of 60 kg as set by WHO/FAO as tolerable based on product packaging/serving or doses normally prescribed. However, a pool of the metal from all the sources investigated and other sources may significantly increase the daily dietary intake above the WHO/FAO specification, which may become deleterious to health.

**Key words:** Aluminium, daily dietary intake, baked beef, drugs, beverages, Nigeria

### INTRODUCTION

Aluminum (Al) is the third most abundant element in the Earth's crust (8.1% by weight) and is a non-essential element, to which humans are frequently exposed (Tria *et al.*, 2007). Al is widespread throughout nature, air, water, plants and consequently in all the food because of its uses. The metal enters the human system mainly through food, drugs, cosmetics, drinking water and beverages.

Aluminum has been shown to have deleterious effects on the central nervous, skeletal and hematopoietic systems of humans (Domingo, 1995). The neurotoxicity of Al to patients with chronic renal disease is well-established (Daugirdas and Ing, 1994) and its presence in the bloodstream leads to Al accumulation in bone and brain causing an encephalopathy called dementia dialysis. It is also associated with neurological disorders in patients on long-term parenteral nutrition (Klei, 2005) and preterm infants receiving intravenous feedings (Klein, 2003). It has been suggested, that low-level long-term exposure to Al may be a contributing factor in Alzheimer's disease (Domingo, 1995).

Many methods have been reported in the literature for the determination of Al. Methods applied for aluminium analysis in natural waters and for the speciation of Al in environmental samples have been reviewed (Lopez-Gonzalvez *et al.*, 2008). Methods using atomic absorption with electrothermal atomization have been used for Al analysis in beverage (De Amorim *et al.*, 2006), environmental samples (Narin *et al.*, 2004). Many of the recently published works on Al determination are spectrofluorometric methods. For the analysis of water and foodstuffs (Tabrizi, 2007) and biological fluids (Buratti *et al.*, 2006). However, Graphite Furnace Atomic Absorption Spectrometry (GF-AAS) and Inductively Coupled Plasma Atomic Emission Spectrometry (ICP-AES) are accepted reference methods for determining total Al in virtually all sample matrices (Shokrollahi *et al.*, 2008).

Studies of the average Al concentration in 24 h diets indicated that under normal circumstances the average dietary intake of Al is about 6 mg per day (Gramiccioni *et al.*, 1996; Sherlock, 1989). Although, the provisional weekly intake of Al established by the FAO/WHO expert committee on food additives is quite high (60 mg day<sup>-1</sup> for an adult man of average body

weight 60 kg). It is important to check the Al concentration in various foods, drugs and beverages since it may sometimes vary depending on the region of production.

In the present research, 3 different sources of Al; beef baked in aluminium foil, drugs and beverages commonly found in Nigerian eateries and pharmacy shops were analyzed for Al by GFAAS. This was done with a view to evaluating their contribution to the daily dietary intake of Al in Nigerian diet.

### MATERIALS AND METHODS

To eliminate possible contamination from detergents or other sources, all glassware and polyethylene material was treated with a 30% v v<sup>-1</sup> HNO<sub>3</sub> solution for 24 h and then rinsed several times with distilled water immediately before use. Concentrated HCl and HNO<sub>3</sub> used were analar grade (Sigma, St Louis, MO, USA). Distilled water produced by the physical chemistry Laboratory of the Department of Chemistry, Ahamdu Bello University, Zaria, Nigeria, was used for all washings and dilutions.

**Instrumentation:** Determination of Aluminium concentrations in treated samples and standard solutions were carried out using a graphite furnace atomic absorption spectrometer Shimadzu 6800 (Izasu, Madrid, Spain). The operation settings and the analytical conditions are presented in Table 1. Memmert D-91126 oven (schwabach, Germany) was used for drying samples. Weighing was done using *E. mettler* Zurich analytical balance (Switzerland).

**Sample collection:** The entire samples investigated were purchased from local retail outlets in Nigeria. Fresh red meat samples were collected from different selling points in the market to get a representative sample. The drugs (tablets) investigated were obtained from pharmacy shops, they are chloroquine (250 mg), vitamin C (100 mg), flagyl (200 mg), paracetamol (500 mg) and aspirin (300 mg). The brands of beverage samples studied were coded A, B, C, D, E and F with their corresponding canned product labeled A<sup>1</sup>, B<sup>1</sup>, C<sup>1</sup>, D<sup>1</sup>, E<sup>1</sup> and F<sup>1</sup>. They were also purchased from different selling points.

**Sample preparation and determination Al in baked beef:** Fresh red meat samples were trimmed to remove bone and fat, cut into small pieces and rinsed in distilled water. The samples were divided into small portions of about 30 g; 3 of these portions were transferred into a stainless steel pan and labeled SSBB, which is to serve as control. Five

Table 1: Operational settings and the analytical conditions of GFAAS for Al determination

Operational settings	Analytical conditions
Lamp current low (mA)	10
Lamp current high (mA)	600
Wavelength (nm)	309.3
Slit width	0.5
Lamp mode	BGC - SR
Sample volume (µL)	20
Concentration unit	Ppm
Repetition sequence	Sm - Sm ...
Pre-Spray time (sec)	3
Integration time (sec)	5
Response time (sec)	1
Temperature low (°C)	100
Temperature high (°C)	3000
Flame type:	N <sub>2</sub> O - C <sub>2</sub> H <sub>2</sub>
Burner height (mm)	11
Fuel gas flow rate (L min <sup>-1</sup> )	7.0
Zero intercept	Yes

Table 2: Average weight of a tablet of drug sample (g)

Samples	Average weight (g)
Chloroquine (250 mg)	0.33
Vitamin C (100 mg)	0.40
Flagyl (200 mg)	0.61
Paracetamol (500 mg)	0.58
Aspirin (300 mg)	0.32

n = 3

of the remaining portions were separately wrapped in Al foils and labeled UBB. The last 5 portions were spiced with seasoning by immersion in a mixture of knor seasoning and sodium Chloride paste and wrapped in Al foils this was labeled SBB. They were all transferred into an oven and baked at 100°C for between 60-180 min kept in the oven at 60°C for 72 h until dried and grounded into homogenized fine powder. One gram each of sample was weighed and completely transferred into a 250 mL beaker, 10 mL 1; 3 concentrated HCl/HNO<sub>3</sub> acid was added and was heated on a hot plate under fume cupboard to dryness. Warm distilled water was added, filtered, into a 100 mL volumetric flask and made up to mark with distilled water. Al was determined by GFAAS.

**Sample preparation and determination of Al in drugs:**

The average weight of the samples were determined, Table 2. The samples were separately crushed and grounded into fine powder. One gram each of sample was weighed and completely transferred into a 250 mL beaker, 10 mL 1; 3 concentrated HCl/HNO<sub>3</sub> acid was added and was heated on a hot plate under fume cupboard to dryness. Warm distilled water was added, filtered, into a 100 mL volumetric flask and made up to mark with distilled water. Al was determined by GFAAS.

**Sample preparation and determination of Al in beverages:**

Beverages containing CO<sub>2</sub> were degassed before analysis opening the bottles and cans, few drops of concentrated

nitric acid added and allowing them to stand in the refrigerator at 4°C for 48 h. Al in beverages was determined without further treatment by GFAAS.

**Preparation of standard solutions for GFAAS:** A 1000-ppm stock solution was prepared by dissolving 1 g of Al foil in 25 mL concentrated HCl and few drops of concentrated HNO<sub>3</sub>. Standard solutions of 1-5 ppm were made by diluting appropriate volume of the stock solution to 100 mL with distilled water.

**RESULTS AND DISCUSSION**

**Aluminium foil baked beef:** Al content in baked beef increased with baking duration and spicing Table 3. This expected because of the differences in contact time between the beef and the aluminium foil. The longer the contact time the more the Al leached into beef. The seasonings used also contain some amount of Al, which will become added on to the beef during baking. The significantly large difference between the Al content of beef baked in stainless steel pan and aluminium foil prove that the use of aluminum cooking utensils will normally increase the Al content of food. For the beef without spice between baking duration 150-180 min, Al content increased sharply from 84.94-204 mg kg<sup>-1</sup> an indication that leaching of Al from foil is at maximum within that time range for baking temperature of 100°C. This is in agreement with results of similar research by Sadetin (2006). Within the same time range the increase for the control is 10.26-12.88 mg kg<sup>-1</sup>, which is not comparatively significant. A drop in Al content was observed for the spiced beef from 221.04- 220.20 mg kg<sup>-1</sup>, systematic error in the analysis may have accounted for this slight drop.

**Determination of Al in drugs:** Average weight of each tablet of the drug samples was determined after 3 observations, to be from 0.32-0.61 g Table 2. The highest was 0.61 g tablet<sup>-1</sup> of flagyl and the least of 0.32 g tablet<sup>-1</sup> of aspirin. Al content of drug samples are in Table 4. Result indicated a mean range of 0.96-1.59 mg kg<sup>-1</sup>. Chloroquine has the highest mean of 1.42 mg kg<sup>-1</sup>; aspirin with the least of 1.05 mg kg<sup>-1</sup>. All the values obtained are below the FAO/WHO tolerable limit. Al as consequence of raw material used for production and handling during processing.

**Determination of Al in beverages:** Presented in Table 5 are results for Al content of bottled and canned beverages. The mean range of Al in bottled beverage is 0.171-0.481 mg L<sup>-1</sup>, while for canned beverage is 0.197-0.442 mg L<sup>-1</sup>. Group mean and standard deviation

Table 3: Al content of beef baked in Aluminium foil and stainless steel pan at different baking time and at 100°C

Samples	Baking duration (min)	Aluminium mg kg <sup>-1</sup> (n = 3)	Group mean mg kg <sup>-1</sup>	SD of means
<b>SSBB</b>				
1	120	11.63	11.50	±1.31
2	150	10.26		
3	180	12.88		
<b>UBB</b>				
1	60	59.83	99.25	±59.57
2	90	68.14		
3	120	78.94		
4	150	84.94		
5	180	204.41		
<b>SBB</b>				
1	60	84.76	144.75	±69.54
2	90	95.56		
3	120	102.21		
4	150	221.04		
5	180	220.20		

SSBB = Stainless Steel Baked Beef, UBB = Baked Beef Without Spice, SBB = Spiced Baked Beef

Table 4: Aluminium content of drugs (mg g<sup>-1</sup>)

Samples	Al mg g <sup>-1</sup> (n = 3)	Mean of the means (mg g <sup>-1</sup> )	SD of means
<b>Chloroquine</b>			
1	1.22	1.42	±0.187
2	1.45		
3	1.59		
<b>Flagyl</b>			
1	1.16	1.10	±0.060
2	1.04		
3	1.11		
<b>Paracetamol</b>			
1	1.15	1.30	±0.150
2	1.31		
3	1.45		
<b>Aspirin</b>			
1	1.10	1.05	±0.080
2	0.96		
3	1.10		
<b>Vitamin C</b>			
1	1.25	1.19	±0.055
2	1.14		
3	1.20		

Table 5: Aluminium content in beverages

Samples	Al mg L <sup>-1</sup> (n = 3)	Group mean (mg L <sup>-1</sup> )	SD of means
<b>Bottled beverages</b>			
A	0.433	0.329	±0.117
B	0.481		
C	0.171		
D	0.359		
E	0.276		
F	0.254		
<b>Canned beverage</b>			
A <sup>1</sup>	0.442	0.343	±0.105
B <sup>1</sup>	0.197		
C <sup>1</sup>	0.442		
D <sup>1</sup>	0.354		
E <sup>1</sup>	0.236		
F <sup>1</sup>	0.389		

of the means are 0.329±0.117 and 0.343±0.105 mg L<sup>-1</sup> for bottled and canned beverages, respectively. Comparatively, no significant difference exists between the results obtained for both bottled and canned drinks.

**Table 6: Daily dietary intake of Al from baked beef, drugs and beverages**

Sample	Al intake (mg day <sup>-1</sup> )
<b>Baked beef</b>	
<b>UBB</b>	
50 g	4.96
100 g	9.93
150 g	14.89
<b>SBB</b>	
50 g	7.24
100 g	14.48
150 g	21.72
<b>Drugs</b>	
Chloroquine (6 tablets)	2.81
Flagyl (6 tablets)	4.03
Paracetamol (6 tablets)	4.52
Aspirin (3 tablets)	1.01
Vitamin (6 tablets)	2.86
<b>Beverage</b>	
<b>Bottled</b>	
35 cl	0.12
50 cl	0.17
<b>Canned</b>	
33 cl	0.11

UBB = Baked Beef Without Spice, SBB = Spiced Baked Beef

Therefore, leaching of Al from aluminium can into beverages is not significant. The values obtain in this sample is the least compare to the other samples investigated.

On the basis of quantity of baked beef served, daily dose of drugs prescribed and the quantity of beverage presentation to the public for consumption, the possible daily dietary intake may be as presented in Table 6. The results indicated values below the 60 mg day<sup>-1</sup> Al for an average adult weighing 60 kg as recommended by FAO/WHO. However, a consumer is most likely to ingest the recommended amount or more depending on the quantity and frequency of food consumption.

### CONCLUSION

It is clear that during baking of beef wrapped in aluminium foil, some Al migrates from the foil into the beef. The migration is probably dependent on baking duration and other factors not investigated in this research. Al content in the samples analyzed is in order; baked beef >drugs >beverages. Amount of Al in the sample are below the FAO/WHO recommendation. However, amount of metal ingested will depend on the quantity and frequency of food consumption.

### REFERENCES

Buratti, M., C. Valla, O. Pellegrino, F.M. Rubino and A. Colombi, 2006. Aluminium determination in biological fluids and dialysis concentrates via chelation with 8-hydroxyquinoline and solvent extraction/f luorimetry. *Anal. Biochem.*, 353: 63-68. DOI: 10.1016/j.jpba.2008.01.029.

Daugirdas, J.T. and T.S. Ing, 1994. *Handbook on Dialysis*, Little Brown, Boston.

De Amorim, F.R., C. Bof, M.B. Franco, J.B.B. Da Silva and C.C. Nascentes, 2006. Comparative study of conventional and multivariate methods for aluminium determination in soft drinks by graphite furnace atomic absorption spectrometry. *Microchem. J.*, 82: 168-173. DOI: 10.1016/j.jpba.2008.01.029.

Domingo, J.L., 1995. Reproductive and developmental toxicity of aluminium: A review. *Neurotoxicol. Teratol.*, 17: 515-521.

Gramiccioni, L., G. Ingraio, M.R. Milana, R. Santaroni and G. Tomassi, 1996. Aluminium levels in Italian diets and selected foods from aluminium utensils. *Food Additives Contamin.*, 13: 767-774.

Klein, G.I., 2003. Aluminium contamination of parenteral nutrition solutions and its impact on the pediatric patient. *Nutr. Clin. Pract.*, 18: 302-307.

Klei, G.I., 2005. Aluminum: New recognition of an old problem. *Curr. Opin. Pharmacol.*, 5: 637-640.

Lopez-Gonzalvez, A., M.A. Ruiz and C. Barbas, 2008. Valid flow-injection method for rapid aluminium determination in anti-perspirants. *Journal of Pharmacology and Biomedical Analysis*. DOI: 10.1016/j.jpba.2008.01.029.

Narin, I., M. Tuzen and M. Soylak, 2004. Aluminium determination in environmental samples by graphite furnace atomic absorption spectrometry after solid phase extraction on Amberlite XAD-1180/pyrocatecol violet chelating resin. *Talanta*, 63: 411-418. DOI: 10.1016/j.jhazmat.2008.05.044.

Sadatin, T., 2006. Aluminium contents of baked meats wrapped in Aluminium foil. *Meat Sci.*, 74: 644-647.

Sherlock, J.C., 1989. Aluminium in Foods and the Diets. In: Massey, R.C. and D. Taylor (Eds.). *Aluminium in Food and the Environment* Cambridge, U.K. The Royal Soc. Chem., pp: 68-76.

Shokrollahi, A., M. Ghaedi, M.S. Niband and H.R. Rajabi, 2008. Selective and sensitive spectrophotometer method for determination of sub-micro-molar amounts of aluminium ion. *J. Hazard. Mater.*, 15: 642-648.

Tabrizi, A.B., 2007. Cloud Point Extraction and Spectrofluorimetric determination of aluminium and zinc in foodstuffs and water samples. *Food Chem.*, 100: 1698-1703. DOI: 10.1016/j.jpba.2008.01.029.

Tria, J., E.C.V. Butler, P.R. Haddad and A.R. Bowie, 2007. Determination of aluminium in natural water samples, *Anal. Chim. Acta.*, 588: 153-165. DOI: 10.1016/j.jpba.2008.01.029.