

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

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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 work, 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 - 220.20mg Kg⁻¹ baking duration 60 - 180mins beef, 1.05 - 1.42mg g⁻¹ drugs, and 0.171- 0.481mg g⁻¹ 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 60mg for an average body weight of 60kg 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

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 [1]. 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 [2]. The neurotoxicity of Al to patients with chronic renal disease is well-established [3], 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 [4] and preterm infants receiving intravenous feedings [5]. It has been suggested that low-level long-term exposure to Al may be a contributing factor in Alzheimer's disease [2].

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 [6]. Methods using atomic absorption with electrothermal atomization have been used for Al analysis in beverage [7], environmental samples [8]. Many of the recently published works on Al determination are spectrofluorometric methods. For the analysis of water and foodstuffs [9], and biological fluids [10]. 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 [11].

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 [12,13]. Although the provisional weekly intake of Al established by the FAO/WHO Expert Committee on Food Additives is quite high, (60 mg per day for an adult of average body weight 60 Kg) [14]. 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 work, three 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

Materials: To eliminate possible contamination from detergents or other sources, all glassware and polyethylene material was treated with a 30%v/v HNO₃ solution for 24 h, and then rinsed several times with distilled water immediately before use. Concentrated HCl and HNO₃ 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 (250mg), vitamin C (100mg), flagyl (200mg), paracetamol (500mg), and aspirin (300mg). The brands of beverage samples studied were coded A,B,C,D,E, and F with their corresponding canned product labeled A¹,B¹,C¹,D¹,E¹, and F¹. They were also purchased from different selling points.

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

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 ₂ O – C ₂ H ₂
Burner height (mm)	11
Fuel gas flow rate (L/min)	7.0
Zero intercept	Yes

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 30g; three of these portions were transferred into a stainless steel pan and labeled SSBB, which is to serve as control. Five (5) of the remaining portions were separately wrapped in Al foils and labeled UBB. The last five 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 mins. kept in the oven at 60°C for 72h until dried, and grounded into homogenized fine powder. 1g each of sample was weighed and completely transferred into a 250ml beaker, 10ml 1; 3 concentrated HCl/HNO₃ acid was added, and was heated on a hot plate under fume cupboard to dryness. Warm distilled water was added, filtered, into a 100ml 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. 1g each of sample was weighed and completely transferred into a 250ml beaker, 10ml 1; 3 concentrated HCl/HNO₃ acid was added, and was heated on a hot plate under fume cupboard to dryness. Warm distilled water was added, filtered, into a 100ml volumetric flask and made up to mark with distilled water. Al was determined by GFAAS.

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

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

n = 3

Sample Preparation and Determination of Al in Beverages:

Beverages containing CO₂ 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 48h. 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 1g of Al foil in 25ml concentrated HCl and few drops of concentrated HNO₃. Standard solutions of 1ppm, 2ppm, 3ppm, 4ppm, and 5ppm were made by diluting appropriate volume of the stock solution to 100ml 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 150-180min, Al content increased sharply from 84.94 - 204mg Kg⁻¹ 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 work by Sadetin [15]. Within the same time range the increase for the control is 10.26-12.88mg Kg⁻¹ which is not comparatively significant. A drop in Al content was observed for the spiced beef from 221.04-220.20mg Kg⁻¹, 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 three observations, to be from 0.32 – 0.61g Table 2. The highest was 0.61g/tablet of flagyl and the least of 0.32g/tablet of aspirin. Al content of drug samples are in Table 4. Result indicated a mean range of 0.96 – 1.59mg Kg⁻¹. Chloroquine has the highest mean of 1.42mg Kg⁻¹; aspirin with the least of 1.05mg Kg⁻¹. 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 – 0.481mg L⁻¹ while for canned beverage is 0.197 – 0.442mg L⁻¹. Group mean and standard deviation of the means are 0.329mg L⁻¹, ±0.117, and 0.343mg L⁻¹ ±0.105 for bottled and canned beverages respectively. Comparatively, no significant difference exists between the results obtained for both bottled and canned drinks. 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.

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

Sample	Baking duration (min)	Aluminium mg kg ⁻¹ (n = 3)	Group mean mg kg ⁻¹	SD of means
SSBB				
1	120	11.63		
2	150	10.26	11.50	± 1.31
3	180	12.88		
UBB				
1	60	59.83		
2	90	68.14		
3	120	78.94	99.25	± 59.57
4	150	84.94		
5	180	204.41		
SBB				
1	60	84.76		
2	90	95.56		
3	120	102.21	144.75	± 69.54
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^{-1})

Sample	Al mg g^{-1} (n = 3)	Mean of the means mg g^{-1}	SD of means
chloroquine			
1	1.22		
2	1.45	1.42	± 0.187
3	1.59		
Flagyl			
1	1.16		
2	1.04	1.10	± 0.060
3	1.11		
Paracetamol			
1	1.15		
2	1.31	1.30	± 0.150
3	1.45		
Aspirin			
1	1.10		
2	0.96	1.05	± 0.080
3	1.10		
Vitamin C			
1	1.25		
2	1.14	1.19	± 0.055
3	1.20		

Table 5: Aluminium content in beverages

Sample	Al (mg L^{-1}) n = 3	Group Mean (mg L^{-1})	SD of means
Bottled beverages			
A	0.433		
B	0.481		
C	0.171	0.329	± 0.117
D	0.359		
E	0.276		
F	0.254		
Canned beverage			
A ¹	0.442		
B ¹	0.197		
C ¹	0.442	0.343	± 0.105
D ¹	0.354		
E ¹	0.236		
F ¹	0.389		

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

Sample	Al intake (mg/day ⁻¹)
Baked beef	

UBB	

50g	4.96
100g	9.93
150g	14.89
SBB	
50g	7.24
100g	14.48
150g	21.72
Drugs	

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
Beverage	

Bottled	

35cl	0.12
50cl	0.17
Canned	

33cl	0.11

UBB = baked beef without spice, SBB = Spiced baked beef

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 60mg/day Al for an average adult weighing 60Kg 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: In 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 work. 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

1. Tria, J., E.C.V. Butler, P.R. Haddad, A.R. Bowie, 2007. Determination of aluminium in natural water samples, *Anal. Chim. Acta*, 588: 153.
2. Domingo, J.L., 1995. Reproductive and developmental toxicity of aluminium: a review, *Neurotoxicol. Teratol.*, 17: 515-521.

3. Daugirdas, J.T., T.S. Ing, 1994. Handbook on Dialysis, Little Brown, Boston..
4. Klei, G.I., 2005. Aluminum: new recognition of an old problem, *Curr. Opin. Pharmacol.*, 5: 637-640.
5. Klein, G.I., 2003. Aluminium Contamination of parenteral nutrition solutions and its impact on the Pediatric patient, *Nutr. Clin. Pract.*, 18: 302-307.
6. Lopez-Gonzalvez, A., M.A. Ruiz, C. Barbas, 2008. Valid flow-injection method for rapid aluminium determination in anti-perspirants, *J. Pharm. Biomed. (In press)*
7. de Amorim, F.R., C. Bof, M.B. Franco, da J.B.B. Silva, 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.
8. Narin, I., M. Tuzen, 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.
9. Tabrizi, A.B., 2007. Cloud Point Extraction and Spectrofluorimetric determination of aluminium and zinc in foodstuffs and water samples, *Food Chem.*, 100: 1698-1703.
10. Buratti, M., C. Valla, O. Pellegrino, F.M. Rubino, A. Colombi, 2006. Aluminium determination in biological fluids and dialysis concentrates via chelation with 8-hydroxyquinoline and solvent extraction/fluorimetry. *Anal. Biochem.*, 353: 63-68.
11. Shokrollahi, A., M. Ghaedi, M.S. Niband, 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.
12. Gramiccioni, L., G. Ingraio, M.R. Milana, R. Santaroni, G. Tomassi, 1996. Aluminium levels in Italian diets and selected foods from aluminium utensils. *Food Additives and Contaminants*, 13: 767-774.
13. Sherlock, J.C., 1989. Aluminium in foods and the diets. In Massey.R.C, Taylor.D (Eds), *Aluminium in food and the environment* (pp.68-76) Cambridge, U, K; The Royal Society of Chemistry.
14. World Health Organization. Evaluation of certain food additives and contaminants. Thirty-third Report of the Joint FAO/WHO Expert Committee on food Additives. WHO Technical Report Series, 776: 26-27(Geneva: WHO).
15. Sadetin, T., 2006. Aluminium contents of baked meats wrapped in Aluminium foil, *Meat Science*, 74: 644-647.