

Department of Clinical Pharmacy, Faculty of Pharmaceutical Sciences, University of Port Harcourt, Rivers State, Nigeria

Academic Editor: Dr. Qaisar Mahmood, COMSATS Institute of Information Technology, Abbottabad, Pakistan

\*eorish@aol.com

### DOI: 10.5339/connect.2011.2

Published: 17 November 2011 © 2011 Roberts & Orisakwe, licensee Bloomsbury Qatar Foundation Journals. This is an open access article distributed under the terms of the Creative Commons Attribution License CC BY 3.0 which permits unrestricted use, distribution and reproduction in any medium, provided the original work is properly cited.





**Research article** 

# Evaluation of potential dietary toxicity of heavy metals in some common Nigerian beverages: A look at antimony, tin and mercury

I.I. Roberts and O.E. Orisakwe\*

# ABSTRACT

There is currently little information on the composition of heavy metals in beverages imported and locally produced in Nigeria. The study quantitatively determined the composition of antimony (Sb), tin (Sn) and mercury (Hg) in 50 different beverage samples and evaluated the extent of violation of guideline values. Analysis of the beverage samples for the presence of Sb, Sn, and Hg was carried out using an atomic absorption spectrophotometer (AAS) 929. The mean values detected for mercury, tin and antimony ( $\pm$ SE) in fruit juices and soft drinks were 2.39  $\pm$  0.25, 3.66  $\pm$  0.22 and 0.49  $\pm$  0.048 µg/l; 2.93  $\pm$  0.34, 3.60  $\pm$  0.46 and 0.49  $\pm$  0.10 µg/l in dairy drinks and 0.94  $\pm$  0.02, 4.34  $\pm$  0.48 and 0.48  $\pm$  0.05 µg/l in bottled water samples respectively. While antimony detected in all products was below guideline values, mercury and tin were above the acceptable levels established by the World Health Organization, United States Environmental Protection Agency and European Union in most samples tested.

Keywords: beverage, heavy metal, food contaminants, toxicology, risk assessment, public health

Classifications: biology, chemistry, medicine

Cite this article as: Roberts II & Orisakwe OE. Evaluation of potential dietary toxicity of heavy metals in some common Nigerian beverages: A look at antimony, tin and mercury, *QScience Connect* **2011**:2 doi: 10.5339/connect.2011.2

# INTRODUCTION

The industrial processes of extraction and distribution of mineral resources which undergo synthetic changes yield chemicals such as heavy metals that have an atomic weight and density almost five times that of water. The pollution of our soil, atmosphere, underground and surface water with effluents of such industrial activities inevitably results in chemicals finding their way into our food chain and consequently into our foods and beverages. The presence of heavy metals in beverages has been documented in a number of worldwide studies. A recent study of beverages produced in Poland detected increased heavy metal content in fruit juice samples (Krejpcio, Sionkowski& Bartela, 2005). This study highlighted the fact that beverages are rarely free from heavy metal contamination regardless of the environment they are produced. In particular, foodstuffs produced in regions suffering from high levels of pollution and environmental problems such as Nigeria, are thought to be more prone to violating guideline values of the permissible amounts of heavy metals in locally produced products. The recent proliferation of a number of different brands of Nigerian beverages has also necessitated the study of the extent of heavy metal contamination of these products.

Heavy metals such as mercury (Hg), antimony (Sb) and tin (Sn) are known to have toxic effects when administered acutely and chronically to laboratory animals. Although studies of heavy metal contamination in beverages have shown that concentrations are generally too low to cause any immediate toxic effects, chronic exposure has the potential to cause severe adverse effects to human health.

Antimony a suspected carcinogen with unknown biological function and effects on long term exposure is used as a catalyst in the production of polyethylene terephthalate (PET) plastics, with many applications including its use in the packaging of beverages and foodstuffs (Hansen et al., 2010). Low doses of antimony is known to cause headaches, dizziness and depression, but in larger doses can cause violent and frequent vomiting.

Food and water are the main sources of mercury in non-occupationally exposed populations. According to Galal-Gorchev (1991), the average daily intake of mercury from food and water ranges from  $2-20 \ \mu g$ . Acute administration of mercury was found to induce severe vacuolation of the renal tubular epithelium and nephropathy in the kidneys (Carmignani, Boscolo & Preziosi, 1989) and caused a decrease in seminiferous tubule diameter, spermatogenic cell counts and leydig's cell nuclear diameter in the male reproductive structure (Lamperti and Printz, 1973). A study carried out by the WHO (1990), found mercury to be a neurotoxin particularly affecting the developing brain and causing mental retardation to the unborn foetus due to its ability to cross the placenta.

Tin was found to cause testicular degeneration in rats at a dose of 10 mg/kg daily for 13 weeks (de Groot, Feron & Til, 1973) and was carcinogenic at doses of between 0–2000 mg in B6C3F1 mice with increased incidence of hepatocellular adenomas (ATSDR, 2003). Higher concentrations of tin are typically found in canned foodstuffs as a result of dissolution of the tin coating or tin plate, the levels depending largely on the type and acidity of the food, the presence of oxidants, the duration and temperature of storage and the presence of air in the can headspace (Vannoort, Cressey & Silvers, 2000).

Several studies of Nigerian soft drinks and juice drinks (Orisakwe, Oragwu, Maduabuchi, Nzegwu & Nduka, 2009; Maduabuchi et al., 2008), dairy drinks and drinking water (Orisakwe et al., 2006) have detected the presence of trace quantities of heavy metals that have exceeded the maximum contaminant levels (MCL) recommended by the United States Environmental Protection Agency (USEPA), European Union (EU) and World Health Organization (WHO).

A recent renowned study of the concentration of antimony in 42 commercial juices (Hansen et al., 2010), found eight of the products to have antimony concentrations that were up by a factor of 2.7 above the EU limit for drinking water. Studies of mercury and tin in various foodstuffs have also indicated the presence of these heavy metals in excess of safety limits.

Many studies to date have described environmental exposure of humans to heavy metals in African populations, however little is known about the exposure to heavy metal toxins from processed or unprocessed foods consumed in Africa. No data currently exists on the concentrations of antimony, tin and mercury in Nigerian beverages and findings from studies conducted worldwide are varied.

This study determined the concentrations of antimony, tin and mercury in a number of commonly sold and consumed beverages in Nigeria and assessed the extent of the violation of recommended limits set by the USEPA, EU and WHO guidelines.

## MATERIALS AND METHODS

Fifty different samples of beverages were utilized in the study including dairy drinks, soft drinks, water, and various types of juice drinks such as fresh juice, juice concentrates and mixed fruit juices, which were purchased in May 2010 in the Niger Delta area of Nigeria. The samples chosen were considered to be a fair representation of the beverages available on the Nigerian market, with the minor exception of a few products that were newly introduced to the market such as energy drinks.

The samples were prepared for analysis by hydrolysing 20 ml of the beverage with 10 ml of hydrochloride (HCL) and 20 ml water (H<sub>2</sub>O). The hydrolyzed samples were well shaken and transferred to a centrifuge tube for centrifugation at the rate of 3000 rpm to remove solid particles. The resulting homogenised samples were thoroughly mixed before sub-samples were taken for analysis to ensure homogeneity of the mixture. The presence of antimony and tin were analysed in samples using the Unicam Atomic Absorption Spectrophotometer (AAS) Model 929, employing an air-acetylene oxidizing (lean, blue) flame at the recommended wavelength of 217 nm and a nitrous-oxide-acetylene reducing (rich red) flame of wavelength 286.3 nm for antimony and tin respectively. Determining the presence of tin and antimony in dairy drinks involved precipitation of the milk proteins including casein by adding trichloroacetic acid (TCA) to the samples. All other determinations were carried out in malt drinks, soft drinks, energy drinks and the table waters by the direct aspiration method.

Mercury was determined by the cold vapour technique after reduction with stannous chloride  $(SnCl_2)$ , in order to release the mercury in the sample solution. Precaution was taken at all times due to the toxic nature of mercury. A Stock Standard Solution was prepared by dissolving 1.08 g of mercury (II) oxide, in a minimum volume of 1:1 HCL and diluted to 1 litre with de-ionised water. This solution was then analysed by the AAS using an air-acetylene, oxidizing (lean, blue) flame at a wavelength of 253.7 nm.

The concentration of the contaminant was calculated using the arithmetic mean according to Parkhurst (1998), by multiplying the amount of the chemical element/volume of the sample beverage. The volume of one and a half litres was assumed in the calculations of the concentration of antimony, tin and mercury, as it was considered the average intake volume for adults.

Appropriate quality procedures and precautions were carried out to assure the reliability of the results. Reagents used to calibrate the instrumentation were of analytical grades. A spike-and-recovery analysis was performed to assess the accuracy of the analytical techniques used. Post-analysed samples were spiked and homogenized with varying amounts of the standard solutions of the different metals. The spiked samples were then processed for the analysis by the dry ashing method.

## RESULTS

The results of the analysed samples were grouped into three subgroups: fruit juices and soft drinks, dairy drinks and bottled waters, as shown in Tables 1-3. The group with the highest and broadest concentration ranges of the heavy metals studied were of the fruit juices and soft drinks group.

The results of the analyses of mercury, tin and antimony in 37 fruit juice and soft drink samples are shown in Table 1. Tin had the highest concentration recorded amongst all heavy metals analysed. The concentration range of tin within the fruit juices and soft drinks group was  $0.97 - 6.33 \,\mu$ g/L, which represented a mean concentration of  $3.70 \pm 0.22 \,\mu$ g/L. Both mercury and antimony were also of the highest concentrations recorded within the fruit juice and soft drinks group. Mercury ranged from  $0.61 - 6.04 \,\mu$ g/L, giving a mean concentration of  $2.39 \pm 0.25 \,$ g/L and antimony ranged between  $0.21 - 1.86 \,\mu$ g/L with a mean concentration of  $0.49 \pm 0.05 \,\mu$ g/L.

Table 2 shows the levels of mercury, tin and antimony detected in ten dairy drinks. Amongst all the beverage samples, mercury was calculated as having the highest mean concentration of  $2.93 \pm 0.34 \,\mu$ g/L, owing to the lesser variation of concentrations detected,  $1.44 - 4.88 \,\mu$ g/L. The mean concentration of tin was the lowest detected within this group and levels of antimony recorded were similar to that of the fruit juices and soft drinks group.

In the bottled water sample group tin had a narrower range of concentrations recorded, however the mean concentration calculated for tin was the highest of all the beverage samples. The mean concentration of  $4.34 \,\mu$ g/L was detected for tin, while conversely the mean values of mercury and antimony were the lowest recorded amongst all the samples, as shown in Table 3.

Table 1. Antimony, tin and n	Table 1. Antimony, tin and mercury (ug/L) levels in fruit juices and soft drinks.	nd soft drinks.					I
Product name	Producer	Place of manufacture	Batch number	Antimony (µg/l)	Tin (µg/l)	Mercury (µg/l)	
GRAND MALT	PABOD BREWERIES LTD.	Nigeria	C2262130	0.57	5.43	3.16	1
COCA-COLA	NIGERIAN BOTTLING COMPANY NIG	Nigeria	R300:37072F	0.41	4.22	0.61	
GINGER BEER	SUN MARK LTD	UK	L22730	0.22	6.33	3.11	
FANTA ORANGE	NIGERIAN BOTTLING COMPANY	Nigeria	AB305:12051F	0.52	0.97	0.96	
SPRITE	NIGERIAN BOTTLING COMPANY	Nigeria	AB301:42005F	0.24	3.06	0.88	
SCHWEPPES BITTER LEMON	NIGERIAN BOTTLING COMPANY	Nigeria	AB301:48007F	0.24	4.21	1.03	
RUBICON GUAVA	RUBICON DRINKS LTD	NK	9324	0.33	4.22	4.63	
FAYROUZ PREMIUM SOFT DRINK	NIGERIAN BREWERIES PLC	Nigeria		0.44	5.06	1.59	
CAPRI-SONNE ORANGE DRINK	CHI LTD	Nigeria	Z130910A5	0.46	5.22	1.21	
DANSA FRUIT DRINKS	DANSA FOODS LTD	Nigeria	01414:11:08	0.41	4.26	2.83	
DELITE ORANGE FRUIT JUICE	UAC DAIRES	Nigeria	24/2D	0.61	4.06	1.22	
CHIVITA PREMIUM PINEAPPLE	CHI LTD	Nigeria	Z010710B9	0.61	3.14	1.01	
CHI EXOTIC PINEAPPLE AND COCONUT NECTAR	CHI LTD	Nigeria	Z160610C9	0.36	1.66	1.14	
TROPICAL PURE FRUIT JUICE	DANSA FOODS LTD	Nigeria	00703:50:26	0.21	1.26	1.87	
ICE TEA LEMON	CHI LTD	Nigeria	Z020610A12	1.86	4.16	1.33	
LUCOMALT	GLAXOSMITHKLINE	Nigeria	ETSILN	0.54	3.64	4.03	
5 ALIVE BERRY BLAST	NIGERIAN BOTTLING COMPANY	Nigeria	ADJ322:44063	0.46	2.45	1.57	
LUCOZADE BOOST	GLAXOSMITHKLINE	Nigeria	DT2IER	0.25	3.74	1.87	
RIBENNA BLACKCURRENT	GLAXOSMITHKLINE	Nigeria	ET170R	0.36	3.27	1.59	
FRUCHTEGUT	MERTINGER	Germany	EHM4	0.87	3.51	5.87	
LA CASERA APPLE DRINK	CLASSIC BEVERAGES NIG. LTD	Nigeria	522032511	0.39	4.01	1.33	
BOBO APPLE MILK DRINK	BOBO FOOD AND BEVERAGE LTD	Nigeria	A-03	0.31	3.66	2.14	

(continued on next page)

Table 1 (continued)						
Product name	Producer	Place of manufacture	Batch number	Antimony (µg/l)	Tin (µg/l)	Mercury (ug/l)
V8 SPLASH BERY BLEND FUZE HEALTHY INFUZIONS SPRITE SANS CREAM SODA SCHWEPPES BITTER LEMON COCA-COLA FANTA ORANGE TANGO OLD JAMAICA GINGER BEER AMSTEL MALTA BETAMALT NESTLE MILO ENERGY DRINK ROBUST SUGAR FREE SEVEN UP COCA COLA (Glass Bottle) % (number) of products Violating t Mercury Range: 0.6 1–6.04; Mean Value =	V8 SPLASH BERRY BLEND CAMPBELL SOUP COMPANY USA CUBTZL0503 0.44 3.321 FUZ EHCITHY INEUZIONS FUZ BEVERAGES LLC USA UTDU01576756 0.26 0.26 3.64 SABRITE AN BOTTLING COMPANY USA UTDU01576756 0.26 0.26 3.64 SANS CREAM SODA PHARAM DEKO PLC NIgeria A328 0.69 0.51 4.73 SANS CREAM SODA NIGERIAN BOTTLING COMPANY Nigeria A5511:10166F 0.26 0.36 SANS CREAM SODA NIGERIAN BOTTLING COMPANY Nigeria A511:10166F 0.26 1.34 ANTA ORANGE NIGERIAN BOTTLING COMPANY Nigeria A511:101699 0.51 4.17 TANGO CAC-COLA NIGERIAN BOTTLING COMPANY Nigeria A511:101089 0.51 4.17 TANGO CAC-COLA NIGERIAN BOTTLING COMPANY Nigeria A511:101089 0.51 4.17 ANGO COCA-COLA NIGERIAN BOTTLING COMPANY Nigeria A511:101089 0.51 4.17 AMGO COTACOLA NIFERVARIES PLC Nigeria 0.35155621316 0.79 5.79 AMSTLE MALTA NICERIAN BOTTLING COMPANY Nigeria 0.31081017:02 0.26 5.79 NITERNATIONAL BREWERIES PLC Nigeria 0.31081017:02 0.26 5.35 NICERIAN BOTTLING COMPANY Nigeria 0.31081017:02 0.26 5.36 AMSTLE MALTA NICERIAN BOTTLING COMPANY Nigeria 0.31081017:02 0.26 5.35 NICERIAN BOTTLING COMPANY Nigeria 0.30 0.31 0.31081017:02 0.26 0.39 5.36 COCA COLA (Glass Bottle) NICEN UP BOTTLING COMPANY Nigeria 0.201014A 0.39 5.35 KOBUST SUGAR FREE SUCA UP BOTTLING COMPANY PLC Nigeria 0.201014A 0.39 5.35 COCA COLA (Glass Bottle) NICERIAN BOTTLING COMPANY PLC Nigeria 2.77 0.26 0.201014A 0.39 5.36 KOUND) OF products Violating the WHO, USEPA and EU Limits for Heavy Metals in drinking water 0.57 0.26 0.2010 0.96 (0.00000000000000000000000000000000000	USA USA USA Nigeria Nigeria Nigeria Nigeria Nigeria Nigeria Nigeria Nigeria Nigeria Si Mean Value: 0.49 ±	CUBTZL0503 UTDU01576756 A0621:16038 A328 A5511:10166F Ab611:01089 R03L0134 5814148 0:35556Z1316 31081017:02 0U20155BA 8E01D14A - - - - - - - - - - - - - - - - - - -	0.44 0.26 0.69 0.33 0.26 0.54 0.54 0.51 0.54 0.51 0.54 0.51 0.54 0.51 0.54 0.51 0.54 0.51 0.79 0.79 0.79 0.26 0.39 0.26 0.39 0.26 0.39 0.26 0.39 0.26 0.39 0.26 0.39 0.26 0.39 0.26 0.39 0.26 0.39 0.26 0.54 0.51 0.54 0.51 0.54 0.51 0.54 0.51 0.54 0.54 0.51 0.54 0.51 0.54 0.54 0.51 0.54 0.51 0.54 0.51 0.54 0.54 0.51 0.54 0.57 0.54 0.57 0.54 0.57 0.54 0.57 0.56 0.57 0.57 0.57 0.57 0.57 0.57 0.57 0.56 0.57 0.57 0.57 0.57 0.56 0.57 0.56 0.57 0.57 0.56 0.57 0.56 0.57 0.56 0.57 0.56 0.57 0.56 0.57 0.56 0.57 0.56 0.57 0.56 0.57 0.56 0.57 0.56 0.57 0.56 0.57 0.56 0.57 0.56 0.57 0.56 0.57 0.56 0.57 0.56 0.57 0.56 0.56 0.56 0.56 0.56 0.56 0.56 0.56	3.21 3.64 2.44 4.36 1.33 4.17 5.79 1.34 5.79 1.34 5.79 2.79 2.79 2.79 2.79 2.79 2.79 2.79 2	5.41 3.84 1.06 2.41 1.63 1.63 1.24 3.16 4.88 3.26 4.26 4.26 4.26 6.04 0.84 1.38 2.53 6.04 0.84 1.38 1.38 USEPA (2 μg/L) 43.2%

Product name	Producer	Place of manufacture	Batch number	Antimony (µg/l)	Tin (µg/l)	Mercury (µg/l)
THREE CROWNS EVAPORATED MILK	FRIESLAND COMPINIA WAMCO NIG.	Nigeria	11:47:39	0.32	6.17	1.44
HOLLANDIA YOGHURT DRINK	CHILTD	Nigeria	Z190810C9	0.47	3.22	1.84
YO! YOGHURT FLAVOURED DRINK	PZ CUSSONS NIG PLC	Nigeria	175P	0.49	2.46	2.67
ZIZA DAIRY PRODUCTS	DANSA FOODS LTD	Nigeria	01313:32:58	0.37	3.44	2.34
DICTA YOGHURT PLAIN	DICTACHI FOODS NIG. LTD	Nigeria	526XXXXXX	0.44	2.64	2.06
GARDEN CITY YOGHURT	SIMPLEX PACKAGES AND ALLIED	Nigeria	21217	1.35	3.54	2.95
	KESOURCES LID					
VIJU MILK	VIJU INDUCIRIES (NIG.) LID	Nigeria	A2-03	0.33	5.14	3.85
VITAMILK	GREEN SPOT LTD	Thailand	31-1-11135-2-001	0.34	2.47	4.88
PEAK CONDENSED MILK	FRIESLAND COMPINIA	Holland	NPOLIA07:21	0.49	5.22	4.06
NUTRICIMA YOGHURT FLAVOURED	NUTRICIMA INDUSTRIES	Nigeria	NA	0.28	1.69	3.16
				EU (5 mg/L) 0% (0)	EU(No Limit)*	EU (1 ug/L) 100%
% (number) of products Violating the	% (number) of products Violating the WHO, USEPA and EU Limits for Heavy Metals in drinking water	etals in drinking <sup>,</sup>	water	WHO (20 μg/L) 0% (0) USEPA (6 μg/L) 0% (0)	WHO (2 μg/L) 90% USEPA (No Limit)*	WHÔ (1 μg/L) 100% USEPA (2 μg/L) 80%

Table 3. Antimony, tin and mercury $(\mu g/L)$ levels in bottled water samples.	ury (µg/L) levels in bottled wate	r samples.				
Product name	Producer	Place of manufacture	Batch number	Antimony (µg/l)	Tin (µg/l)	Mercury (µg/l)
EVA TABLE WATER LA VOLTIC TABLE WATER PURE LIFE PREMIUM DRINKING WATER	NIGERIAN BOTTLING COMPANY VOLTIC LTD NESTLE NIGERIA PLC	Nigeria Nigeria Nigeria	A0313:52 29/06/RSP 102323087	0.55 0.37 0.51	5.26 4.11 3.66	0.91 0.97 0.94
% (number) of products Violating the WHO, USEPA and EU Limits for Heavy Metals in drinking water	VHO, USEPA and EU Limits for Heavy I	Metals in drinking v	vater	EU (5μg/l) 0% (0) WHO (20 μg/L) 0% (0) USEPA (6 μg/L) 0% (0)	EU (No Limit)* WHO (2 µg/L) 100% (0) USEPA (No Limit)*	EU (1μg/l) 0% (0) WHO (1 μg/L) 0% (0) USEPA (2 μg/L) 0% (0)
Mercury (Range: 0.91–0.97; Mean Value = 0.94 $\pm$ 0.02 ( $n = 3$ ), Tin (Range: 3.66–5.26; Mean Value = 4.34 $\pm$ 0.48 ( $n = 3$ ), Antimony (Range: 0.37–0.55; Mean Value = 0.48 $\pm$ 0.05 ( $n = 3$ ).	$_{4}\pm$ 0.02 ( $\eta=3$ ), Tin (Range: 3.66–5.26; Mear	) Value = 4.34 ± 0.48 (	n=3), Antimony (Range	:: 0.37-0.55; Mean Value = 0.48	± 0.05 ( <i>n</i> = 3).	

ä
Ы
Ē
a
l water samples.
F
Ĕ
ž
÷.
ð
Ŧ
Б
р
$(\mu g/L)$ levels in bottled wate
5
-
۳
e
~
-
ug/L) l
Ŀ
~
ž
cury
ercury
mercury
d mercury
nd mercury
and mercury (
in and mercury
tin and mercury.
/, tin ;
ntimony, tin and mercury
/, tin ;

Page 7 of 10 Roberts II & Orisakwe OE. QScience Connect 2011:2 On comparison of the levels of the three heavy metals in the fruit juices and soft drinks, dairy drinks and bottled water samples with standards set by the WHO, USEPA and EU, as shown in Tables 4–6, the dairy drinks group were found in most violation of mercury 100%, 100% and 80% violating the EU, WHO and USEPA guidelines respectively. Fruit juices and soft drinks were found 89.2%, 43.2% and 89.2% in violation of WHO, USEPA and EU permissible levels of mercury respectively.

Tin however was in most violation in the bottled water group with 100% of samples violating the WHO permissible levels and 90% and 86.5% estimated violations within the dairy drink and fruit juices and soft drink groups respectively. Antimony on the other hand was found to be generally low in comparison to the other heavy metals detected, with none of the samples violating the guideline values.

The 'worst case scenario' estimation of the intake of the heavy metals as a result of consumption of beverages from each sample group, on an average weekly basis volume of 4.5 L (1.5 L of the three most contaminated beverage products) was 26.63  $\mu$ g/L, 5.64  $\mu$ /L and 17.79  $\mu$ g/L respectively, as shown in Table 7.

Heavy metal	Average	Range	WHO Lower	limits Upper		A Limits Upper	EU L Lower	imits Upper	Percentage of samples in violation (WHO/EPA/EU)
Mercury	2.35	0.61 - 6.04	1 µg/L	1 µg/L	-	2 μg/L	-	1 μg/L	89.20% /, 43.24% / & 89.20%
Tin	3.88	0.97 - 6.33	2 µg/L*	-	-	-	-	-	86.50% *
Antimony	0.50	0.22 - 1.86	-	20 µg/L	-	2 μg/L	-	5μg/L	Nil

## Table 4. Heavy metal content of different brands of fruit juice and soft drinks.

## Table 5. Heavy metal content of different brands of dairy drinks.

Heavy metal	Average	Range	WHO lower	limits upper	USEP/ lower	A Limits upper	EU L lower	imits upper	Percentage of samples in violation (WHO/EPA/EU)
Mercury	2.93	1.44 - 4.88	1 µg/L	1 μg/L	-	6 μg/L	-	1 μg/L	100% /, 80% & 100%
Tin	3.60	1.69 - 6.17	2 µg/L*	-	-	-	-	-	90%*
Antimony	0.49	0.28 - 1.35	-	20 µg/L	-	2 μg/L	-	5 μg/L	Nil

#### Table 6. Heavy metal content of different brands of bottled water.

Heavy metal	Average	Range	WHO lower	limits upper	USEP/ lower	A Limits upper	EU L lower	imits upper	Percentage of samples in violation (WHO/EPA/EU)
Mercury	0.94	0.91 - 0.94	1 μg/L	1 μg/L	-	6 μg/L	-	1 μg/L	Nil
Tin	4.34	3.66 - 5.25	2 μg/L	-	-	-	-	-	100%*
Antimony	0.48	0.37 - 0.55	-	20 µg/L	-	2 μg/L	-	5 μg/L	Nil

## Table 7. Example of calculating heavy metal weekly intake.

True Sb intake =  $1.5 \times 1.86 + 1.5 \times 0.55 + 1.5 \times 1.35 = 5.64 \,\mu$ g/L Sb True Hg intake =  $1.5 \times 6.04 + 1.5 \times 0.94 + 1.5 \times 4.88 = 17.79 \,\mu$ g/L Hg True Sn intake =  $1.5 \times 6.33 + 1.5 \times 5.25 + 1.5 \times 6.17 = 26.63 \,\mu$ g/L Sn

\*(1.5 L is assumed the weekly beverage consumption, which is multiplied by the highest concentration of heavy metal contaminant from each beverage group: the volume of the each beverage was assumed to be one litre).

# DISCUSSION

On analysis of the levels of antimony, tin and mercury detected in the beverage samples, it was found that the concentration of heavy metals varied considerably between each sample group. Some samples were below the recommended safety limits, others within this and some exceeding the threshold limits established by the European Union, World Health Organization and the US Environmental Protection Agency guidelines.

The levels of antimony recorded in this study were found to be within the limits stipulated by the WHO ( $20 \mu g/L$ ), EU ( $5 \mu g/L$ ) and USEPA ( $2 \mu g/L$ ) for antimony in drinking water (WHO, 2003; USEPA, 2011). These levels reported however, were much lower than the antimony levels recorded in the study by Hansen et al. (2010), despite the beverage samples similarly having been packed in PET containers. The different instrumentation techniques may have accounted in part for the difference in antimony levels detected.

The intake estimates for antimony calculated using the arithmetic mean for a Nigerian consuming one and half litres of beverages per week was 5.64  $\mu$ g/L, which was found to be in line with the World Health Organization recommended daily tolerable intake of antimony at 6.0  $\mu$ g/kgbw/day (Egan, Tao, Pennington & Bolger, 2002).

Alarmingly the mercury levels analysed in 100% of dairy drinks and 89.2% fruit juices and soft drinks were found to violate both the WHO ( $1 \mu g/L$ ), EU ( $1 \mu g/L$ ) and USEPA ( $2 \mu g/L$ ) regulations (WHO, 2003; USEPA, 2011). These results are contrary to findings by Cheung Chung, Kwong, Yau & Wong (2008), who reported extremely low mercury concentrations in Nigerian beverages. Considering the daily intake of these beverages in particular dairy drinks by children, young adults and pregnant women, it is feared that long-term exposure may pose significant health risks.

Although fish and other seafood products have been documented as being the main source of mercury in the diet (WHO, 1990), this has been disproved by our findings due to the high levels of total mercury that were detected in both the dairy drinks group and fruit juices and soft drink samples.

Although previous authors have recorded detectable concentrations of tin in beverages such as tea, carbonates, fruit drinks, bottled water, herbal drinks and iced tea (MAFF UK 1998) these levels were all below 1 mg/kg and lower than the regulatory limit set by the WHO The Tin in Food Regulations (1992) of 200 mg/kg. In this study however, the levels of tin were found to violate 100%, 88.9% and 90% of bottled water samples, fruit juices and soft drinks and dairy products respectively, exceeding the WHO ( $2 \mu g/L$ ) regulation.

The present study has confirmed that contrary to common belief, beverages sold and consumed in Nigeria are not free of contamination of the toxic heavy metals tin and mercury. The values of tin and mercury detected were both found in severe violation of WHO, USEPA and EU safety standards.

Possible routes that are believed to result in the contamination of beverages during the production process include the use of contaminated water, poor assaying of raw materials, poor hygiene, low quality packaging materials and inadequate storage conditions.

Imported beverages were also found to contain high levels of the contaminants, which implies that heavy metals are yet to be regulated and consequently Nigeria has become a dumping ground for beverages untested for these toxic heavy metals.

## CONCLUSION

As the heavy metal content in beverages constitutes a significant source of exposure to the general population, increased consumption of beverages unregulated for such contaminants represents a worrying problem. Limited studies of the chronic effects of some of these heavy metals warrants further investigation to determine blood concentrations of highly susceptible population groups such as in children and pregnant women.

This study also calls for mandatory testing of heavy metals in beverages and food as a way of monitoring the extent of violation of guideline values. Tighter regulation on the composition of beverages locally produced and imported into Nigeria would help to greatly reduce the extent of this problem.

#### References

Agency for Toxic Substance and Disease Registry. (2003). Toxicological Profile for Mercury, U.S. Department of Health and Humans Services, Public Health Service, Centers for Disease Control, Atlanta, GA.

Blunden, S., & Wallace, T. (2003 Dec). Tin in canned food: a review and understanding of occurrence and effect. Food Chem Toxicol, 41(12), 1651–1662.

- Boogaard, P.J., Boisset, M., Blunden, S., Davies, S., Ong, T.J., & Taverne, J.P. (2003 Dec). Comparative assessment of gastrointestinal irritant potency in man of tin(II) chloride and tin migrated from packaging. Food Chem Toxicol, 41(12), 1663–1670.
- Carmignani, M., Boscolo, P., & Preziosi, P. (1989). Renal ultrastructural alterations and cardiovascular functional changes in rats exposed to mercuric chloride. Archives of Toxicology, 13, 353–356. Supplement.
- Chung, Cheung, Kwong, Stephen Wai, Yau, Ka Ping, & Wong, Joan (July 2008). Waiky Dietary exposure to antimony, lead and mercury of secondary school students in Hong Kong. Food Additives and Contaminants, Volume 25, Number 7. 831–840.
- de Groot, A.P., Feron, V.J., & Til, H.P. (1973). Food Cosmet Toxicol, 11, 19–30.
- Egan, S.K., Tao, S.S.H., Pennington, J.A.T., & Bolger, P.M. (2002). US Food and Drug Administration's Total Diet Study: intake of nutritional and toxic elements, 1991–96. Food Additives and Contaminants, 19(2), 103–125.
- Galal-Gorchev, H. (1991). Dietary intake of pesticide residues, cadmium, mercury, and lead. Food Additives and Contaminants, 8, 793–806.
- Hansen, C., Tsirigotaki, A., Bak, S.A., Pergantis, S.A., Stürup, S., Gammelgaard, B., & Hansen, H.R. (2010 April 9). Elevated antimony concentrations in commercial juices. J Environ Monit, 12(4), 822–824. Epub 2010 Feb 17.

Iyengar, G.V., Tanner, J.T., & Wolf, W.R. et al. (1987). Sci Total Environ, 61, 235–252. JECFA, 1989 Evaluation of certain food additives and contaminants: thirty-third report of the Joint FAO/WHO Expert Committee on Food Additives. Geneva, World Health Organization (WHO Technical Report Series No. 776).

Krejpcio, Z., Sionkowski, S., & Bartela, J. (2005). Safety of fresh fruits and juices available on the Polish market as determined by heavy metal residues. Polish Journal of Environmental Studies, 14, 877–881.

Lamperti, A.A., & Printz, R.H. (1973). Effects of mercuric chloride on the reproductive cycle of the female hamster. Biology of Reproduction, 8, 378–387.

Maduabuchi, J.-M.U., Nzegwu, C.N., Adigba, E.O., Oragwu, C.I., Agbo, F.N., Agbata, C.A., Ani, G.C., & Orisakwe, O.E. (2008). Iron, Manganese and Nickel exposure from beverages in Nigeria: a public health concern? I Health Sci. 34, 335–338.

Orisakwe, O.E., Igwilo, I.O., Afonne, O.J., Maduabuchi, J.U., Obi, E., & Nduka, J.C. (2006). Heavy Metal Hazards of Sachet Water in Nigeria Archives of Environmental & Occupational Health, 61(XXXX).

- Orisakwe, O.E., Oragwu, C.I., Maduabuchi, J.M.U., Nzegwu, C.N., & Nduka, J.K.C. (February 2009). Copper, selenium and zinc content of canned and non-canned beverages in Nigeria. African Journal of Environmental Science and Technology, 3(1), 042–049.
- Parkhurst, D.F. (1998). Arithmetic versus geometric means for environmental Concentration data. Environ Sci Technol, 32, 92A–98A.

The Tin in Food Regulations 1992 (S.I. [1992] No. 469). The Stationery Office.

US Environmental Protection Agency (2011). Drinking Water Standards and Health Advisories.

Vannoort, R., Cressey, P., & Silvers, K. (2000). 1997/1998 New Zealand Total Diet Survey. Part 2: Elements. Ministry of Health, World Health Organization, Geneva.

World Health Organization (WHO). Methylmercury.Environmental Health Criteria 101. Geneva: WHO;1990. Available from: http://www.inchem.org/documents/ehc/ehc/ehc101.htm.

World Health Organization. (2003). http://www.who.int/watersanitationhealth/dwq/chemicals/antimony/en/accessed on May 1, 2006.

WHO. Guidelines for Drinking-Water Quality. 2nd ed., Vol 1. Recommendations, 2003. World Health Organization, Geneva.