CONTAMINATION OF PET BOTTLED CARBONATED SOFT DRINKS SOLD IN NIGERIA AFTER LONG STORAGE DUE TO ANTIMONY LEACHING: RISK ASSESSMENT

C. E. Umeocho[1,2], T. U. Onuegbu[1], E. N. Nwosu[2]. R. O. Ohakwe[2]

1. Department of Pure and Industrial Chemistry, Nnamdi Azikiwe university, Awka
2. Bioresource Development Centre Abagana, National Biotechnology and research Development Agency.

Chinyereumeocho@gmail.com

ABSTRACT

This research was carried out to ascertain the rate of leaching of antimony (Sb)from PET bottled soft drinks after long storage and to calculate the health risk associated with this leaching. Six different brands of soft drinks sold in Nigeria (eleven bottles of each) were purchased from 9th mile area of Enugu State. One bottle of each brand was analysed for antimony before storage to serve as control. Five bottles of each brand were exposed to sunlight while five bottles of each brand were stored at room temperature and they were analysed for antimony concentration after 3, 6, 9, 12 and 15 weeks of storage. For sample 1, Sb concentration ranged from 0.004 to 0.010 and 0.004 to 0.011, sample 2 ranged from 0.001 to 0.006 and 0.001 to 0.010, sample 3 ranged from 0.001 to 0.008 and 0.001 to 0.014, sample 4 ranged from 0.002 to 0.007 and 0.002 to 0.010. sample 5 ranged from 0.003 to 0.006 and 0.003 to 0.011, sample 6 ranged from 0.001 to 0.006 and 0.001 to 0.010 for room temperature and sunlight exposed samples respectively. The antimony concentration on the samples were found to exceed the WHO limit of 0.006 at 15 weeks of exposure to sunlight apart from sample 2. It was observed that the cancer risk increased with increase in storage time, temperature and amount of leached antimony increases.

Key words: antimony, leaching, cancer risk, chronic daily intake

INTRODUCTION

Soft drinks consumption is still a controversial issue for public health and public policy. Over the years, numerous studies have been conducted into the possible links between soft drink intake and medical problems, the results of which, however, remain highly contested. Plastic bottles made from poly(ethylene terephthalate) (PET) are increasingly used for beverages such as soft drinks, mineral water,\ beer, among others; most of the PET resins are sold as food grade material for beverage packaging [1]. Soft drink containers are often stored under unpredictable conditions for several months before consumption. Poly (ethylene terephthalate) bottles have been shown to contaminate water with Sb, with concentrations increasing with storage time [2]. Antimony toxicity is dependent on the exposure dose, duration, route (breathing, eating, drinking or skin contact), exposure to other chemicals, age, sex, nutritional status, family traits, lifestyle, and state of health [3].

There is an association of lung cancer with Sb exposure, although smoking greatly exacerbates the chances [4]. Organic compounds such as toluene, cyclohexane,

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dichloromethane, pentane, benzene, phthalate, esters and ethers with tumour inducing properties may leach from plastic packaging, polystyrene cap liners or unknown sources [5,6].

MATERIALS AND METHODS

Sample collection

Six different brands of PET bottled carbonated soft drinks (eleven bottles of each) all made in Nigeria were purchased from ninth mile area of Enugu State. The analysed samples were sixty-six in number. Each brand name, batch number, manufacturing and expiry dates are given below

Table 3.1: carbonated soft drinks used.

Samples code and names	Batch number	Manufacturing date	Expiry date	Number of drinks purchased	Producers
1 (Swan orange)	1047090413	23/04/13	08/10/13	11	UAC foods
2 (Pepsi)	830413R14:49	23/04/13	22/10/13	11	7-up bottling company
3 (Fanta)	A0601:4839	17/02/13	16/08/13	11	Cocacola company
4 (Mirinda pineapple)	160413B23:11	16/04/13	15/10/13	11	7-up bottling company
5 (Mirinda orange)	AA413B2137	10/04/13	09/10/13	11	7-up bottling company
6 (Teem bitter lemon)	560413808:52	26/04/13	25/10/13	11	7-up bottling company

Sample preparation

One bottle of each brand of carbonated PET bottled soft drink was analysed before storage to serve as control. Five bottles of each of the brands (30 bottles altogether) were kept at room temperature while five bottles of each brand (30 bottles) were exposed to the sunlight and they

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were analyzed after 3, 6, 9,12 and 15 weeks of storage in both conditions.

Leaching test

The method used by Umeocho *et al.,* (2021) was adopted in conducting the leaching test.

A portion (100ml) of the beverage was boiled on a hot plate till the volume reduced to about 30ml. 10ml of perchloric acid and 10ml of HNO_3 were added to the solution. The solution was boiled for 10mins after which 20ml of H_2O_2 was added to it. The solution was further boiled until it became colourless and less than 40ml. The solution was cooled and added 5ml of saturated boric acid solution. The cooled solution was filtered through no12cm Whatmann filter paper. The filtrate was received in a 50ml volumetric flask and subsequently made up to mark with deionized water and labeled accordingly for AAS analysis.

Risk Assessment/ calculation

In order to estimate the risk caused by long time exposure to Sb , chronic daily intake (CDI) was calculated. The equation used in calculating is as follows:

CDI - chronic daily intake

$$CDI_{injection} = \frac{Cs\ x\ IRS\ XEf\ X\ ED\ X\ CF}{BW\ X\ AT}\ x\ CF$$

Cs = Exposure point concentration [7]

IRS = ingestion rate: 100mg.d^{-1}[7]

Ef = exposure frequency: 350d//a [7]

ED = exposure Duration: 30a [7]

BW = Body weight: 70kg [8]

AT = averaging time for carcinogens = 365 x 70d [9]

Cf = unit conversion factor: 10^{-6} kg mg^{-1}[9]

Cancer risk = $CDI\ xSf$

Total cancer risk = $\sum^{n}_{k=1} CDI_k$ x SF_k CDI x SF_K

$SF = \frac{1}{6}$

Statistical Analysis

The effect of two factors, storage condition (room temperature and sunlight) and storage time (0, 3, 6, 9, 12, and 15 weeks), was analysed using a two-way analysis of variance (ANOVA). The analysis was done using R statistical software, version 3.5.3.

RESULTS AND DISCUSSION

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Table1: antimony concentration on the carbonated soft drinks

S/No	Sample	Exposure Duration	PARAMETERS									
			Sb		pH		TSSC		TITRABLE ACIDITY		SPECIFIC GRAVITY	
			room temp	sunlight	room temp	Sunlight	room temp	sunlight	room temp	sunlight	room temp.	Sunlight
1	1	0	0.004	0.004	2.93	2.93	7.800	7.800	0.120	0.120	1.0313	1.0313
		3	0.006	0.007	2.92	2.92	8.530	8.530	0.120	0.120	1.0345	1.0345
		6	0.006	0.009	2.92	2.92	8.880	9.010	0.120	0.120	1.0353	1.0364
		9	0.006	0.009	2.91	2.91	8.886	9.010	0.120	0.121	1.0359	1.0366
		12	0.008	0.010	2.89	2.89	8.900	9.260	0.121	0.122	1.0362	1.0368
		15	0.010	0.011	2.88	2.88	9.000	9.260	0.121	0.122	1.0365	1.0368
		WHO	0.006	0.006	-	-	-	-	-	-	-	--
2	2	0	0.001	0.001	3.09	3.09	9.50	9.50	0.093	0.093	1.0338	1.0338
		3	0.003	0.003	3.09	3.08	9.51	9.53	0.097	0.098	1.0383	1.0383
		6	0.003	0.003	3.07	3.06	9.60	9.74	0.098	0.098	1.0384	1.0387
		9	0.006	0.007	3.06	3.05	9.68	9.74	0.098	0.100	1.0388	1.0389
		12	0.006	0.008	3.05	3.03	9.70	9.98	0.101	0.102	1.0390	1.0400
		15	0.006	0.010	3.03	3.02	9.61	9.98	0.102	0.103	1.0403	1.0406
		WHO	0.006	0.006	-	-	-	-	-	-	-	-

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3	3	0	0.001	0.001	3.08	3.08	0.00	0.00	0.069	0.069	1.0001	1.0001
		3	0.002	0.002	3.07	3.07	0.24	0.26	0.071	0.071	1.0005	1.0005
		6	0..02	0.005	3.07	3.07	0.24	0.26	0.072	0.072	1.0007	1.0007
		9	0.005	0.005	3.05	3.05	0.24	0.26	0.072	0.072	1.0010	1.0012
		12	0.005	0.009	3.05	3.04	0.46	0.51	0.072	0.072	1.0015	1.0016
		15	0.008	0.014	3.04	3.03	0.49	0.51	0.073	0.073	1.0018	1.0020
		WHO	0.006	0.006	-	-	-	-	-	-	-	-
4	4	0	0.002	0.002	3.61	3.61	10.40	10.40	0.066	0.066	1.0429	1.0429
		3	0.002	0.003	3.60	3.60	10.50	10.70	0.066	0.066	1.0430	1.0430
		6	0.002	0.003	3.60	3.59	10.53	10.70	0.066	0.066	1.0434	1.0434
		9	0.004	0.005	3.58	3.58	10.61	10.94	0.067	0.069	1.0440	1.0440
		12	0.007	0.008	3.58	3.57	10.62	10.94	0.067	0.069	1.0440	1.0440
		15	0.007	0.010	3.57	3.56	10.63	10.94	0.068	0.069	1.0440	1.0442
		WH0	0.006	0.006	-	-	-	-	-	-	-	-
5	5	0	0.003	0.003	2.90	2.90	11.18	11.80	0.115	0.115	1.0458	1.0458
		3	0.003	0.005	2.89	2.89	12.00	12.14	0.131	0.131	1.0476	1.0476
		6	0.003	0.008	2.88	2.88	12.00	12.37	0.135	0.135	1.0486	1.0503
		9	0.005	0.010	2.84	2.84	13.40	14.02	0.139	0.139	1.0501	1.0503
		12	0.005	0.011	2.84	2.83	13.60	14.02	0.146	0.146	1.0515	1.0573

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		15	0.006	0.011	2.82	2.82	14.00	14.20	0.146	0.149	1.0520	1.0577
		WHO	0.006	0.006	-	-	-	-	-	-	-	-
6	6	0	0.001	0.001	2.92	2.92	0.000	0.000	0.110	0.110	1.0497	1.0497
		3	0.003	0.002	2.91	2.92	0.000	0.000	0.115	0.115	1.0500	1.0500
		6	0.003	0.006	2.92	2.91	0.000	0.015	0.120	0.120	1.0502	1.0503
		9	0.003	0.006	2.90	2.90	0.019	0.260	0.120	0.120	1.0509	1.0513
		12	0.005	0.006	2.88	2.88	0.223	0.260	0.125	0.126	1.0512	1.0515
		15	0.006	0.010	2.88	2.88	0.231	0.260	0.127	0.130	1.0516	1.0608
		WHO	0.006	0.006	-	-	-	-	-	-	-	-

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The antimony (Sb) concentration for samples 1, 4 and 5 were initially 0.004, 0.002 and 0.003 and changed to 0.010, 0.007 and 0.006 (room temp) and 0.011, 0.010and 0.011 (sunlight) after 15 weeks of storage. The Sb was found higher under sunlight than room temperature. This increase was tested statistically at both storage conditions and different storage times and the following were obtained:

The change in the Sb of sample 1 was statistically significant (P<0.05) between (0-12 and 0-15) weeks of storage.

The change in Sb of sample 4 was significant (P<0.05) between (0-12,0-15, 3-15, 6-15, 9-12 and 9- 15) weeks of storage.

Change in Sb for sample 5 was statistically significant (P<0.05) throughout the different storage times and at different storage conditions.

The change in the Sb of samples 2, 3 and 6 were initially 0.001, 0.001 and 0.001 and changed to 0.006, 0.008 and 0.006(room temp) and 0.010.0.014 and 0.010 (sunlight) respectively after 15 weeks of storage. This change in the level of Sb were tested statistically and the following were observed:

Statistically, a significant change was observed in the Sb of sample 2 between (0-9, 0-12 and 6-15) weeks of storage. The Sb concentration for sample 6 was found to statistically significant (P<0.05) between 0- 15 weeks of storage.

The higher migration level observed for the sunlight exposed samples may be due to the degradation of PET bottle with temperature as was observed by Takahashi et al.,(2008). The migration behaviour may depend on the total antimony concentration of the PET bottle, bottle volume and wall thickness, activation energy and diffusion coefficient of antimony [10].

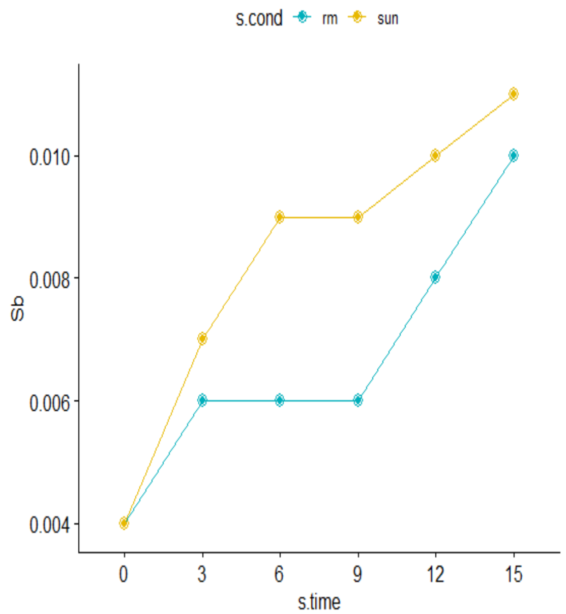

Fig 1: Effects of storage time and storage on antimony (Sb) levels for sample 1

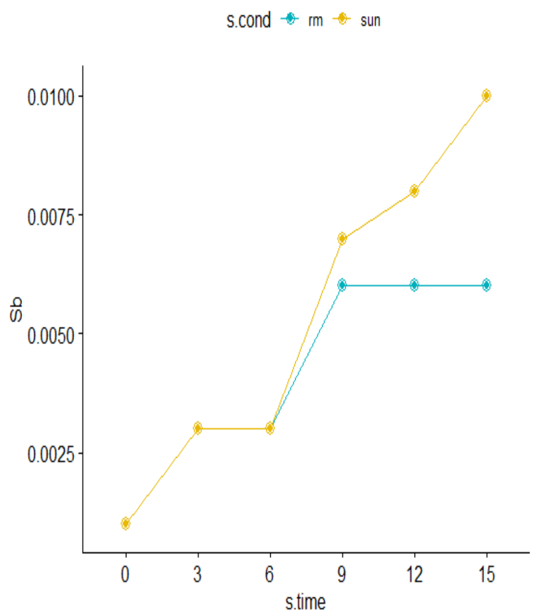

Fig 2: Effects of storage time and condition storage condition on antimony (Sb) levels for sample 2

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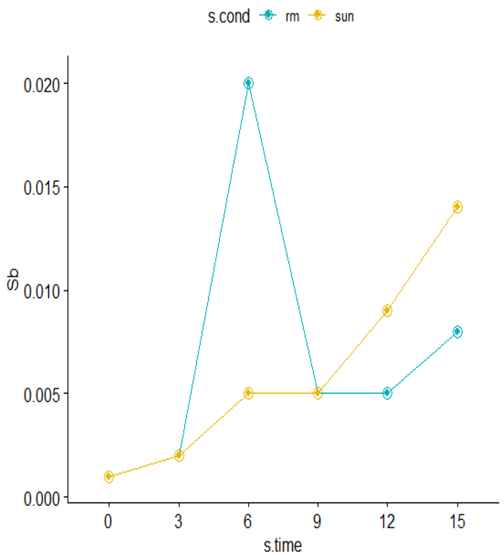

Fig 3: Effects of storage time and storage condition on antimony (Sb) levels for sample 3

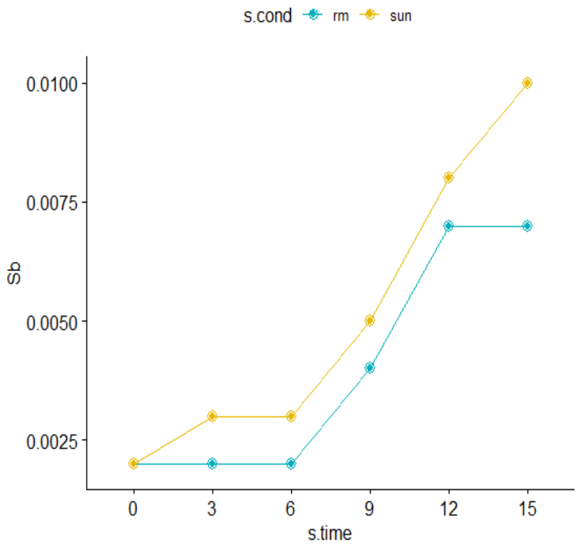

Fig 4: Effects of storage time and storage condition on antimony (Sb) levels for sample 4

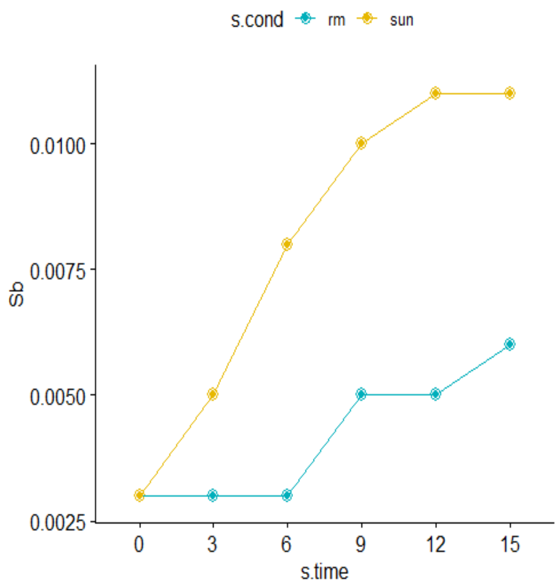

Fig 5: Effects of storage time and storage condition on antimony (Sb) levels for sample 5

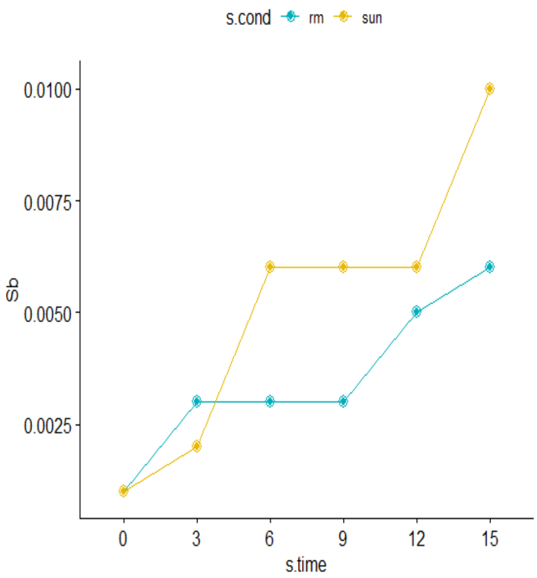

Fig 6 Effects of storage time and storage condition on antimony (Sb) levels for sample 6

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Table2: The chronic daily intake (CDI) and the calculated cancer risk of antimony

S/No	Sample	Exposure Duration	Sb					
			Conc at room temp. storage (ppm)	Conc on Exposure to sunlight(ppm)	$CDI_{injection}$(roomtemp)== Csx IRSxEfxED / BW x AT x CF	$CDI_{injection}$(sunlight) = CsxIRSxEfxED / BW x AT x CF	Cancer risk @ room temp= CDI x Sf	Cancer risk @ sunlight exposure = CDI x Sf
1	1	0	0.004	0.004	2.34×10^{-9}	2.34×10^{-9}	3.4×10^{-10}	3.4×10^{-10}
		3	0.006	0.007	3.52×10^{-9}	4.1×10^{-9}	5.6×10^{-10}	6.6×10^{-10}
		6	0.006	0.009	3.52×10^{-9}	5.28×10^{-9}	5.6×10^{-10}	8.4×10^{-10}
		9	0.006	0.009	3.52×10^{-9}	5.28×10^{-9}	5.6×10^{-10}	8.4×10^{-10}
		12	0.008	0.010	4.69×10^{-9}	5.87×10^{-9}	7.5×10^{-10}	9.4×10^{-10}
		15	0.010	0.011	5.87×10^{-9}	6.45×10^{-9}	9.4×10^{-10}	1.0×10^{-9}
2	2	0	0.001	0.001	5.8×10^{-10}	5.8×10^{-10}	9.3×10^{-11}	9.3×10^{-11}
		3	0.003	0.003	1.76×10^{-9}	1.76×10^{-9}	2.8×10^{-10}	2.8×10^{-10}
		6	0.003	0.003	1.76×10^{-9}	1.76×10^{-9}	2.8×10^{-10}	2.8×10^{-10}
		9	0.006	0.007	3.52×10^{-9}	4.1×10^{-9}	5.6×10^{-10}	6.6×10^{-10}
		12	0.006	0.008	3.52×10^{-9}	4.69×10^{-9}	5.6×10^{-10}	7.5×10^{-10}

		15	0.006	0.010	3.52×10^{-9}	5.87×10^{-9}	5.6×10^{-10}	9.4×10^{-10}
3	3	0	0.001	0.001	5.8×10^{-10}	5.8×10^{-10}	9.3×10^{-11}	9.3×10^{-11}
		3	0.002	0.002	1.17×10^{-9}	1.17×10^{-9}	1.9×10^{-10}	1.9×10^{-10}
		6	0..02	0.005	1.17×10^{-9}	2.93×10^{-9}	1.9×10^{-10}	4.7×10^{-10}
		9	0.005	0.005	2.93×10^{-9}	2.93×10^{-9}	4.7×10^{-10}	4.7×10^{-10}
		12	0.005	0.009	2.93×10^{-9}	5.28×10^{-9}	4.7×10^{-10}	8.4×10^{-10}
		15	0.008	0.014	4.69×10^{-9}	8.21×10^{-9}	7.5×10^{-10}	9.9×10^{-10}
4	4	0	0.002	0.002	1.17×10^{-9}	1.17×10^{-9}	1.9×10^{-10}	1.9×10^{-10}
		3	0.002	0.003	1.17×10^{-9}	1.76×10^{-9}	1.9×10^{-10}	2.8×10^{-10}
		6	0.002	0.003	1.17×10^{-9}	1.76×10^{-9}	1.9×10^{-10}	2.8×10^{-10}
		9	0.004	0.005	2.34×10^{-9}	2.93×10^{-9}	3.74×10^{-10}	4.7×10^{-10}
		12	0.007	0.008	4.1×10^{-9}	4.69×10^{-9}	6.6×10^{-10}	7.5×10^{-10}
		15	0.007	0.010	4.1×10^{-9}	5.87×10^{-9}	6.6×10^{-10}	9.4×10^{-10}
5	5	0	0.003	0.003	1.76×10^{-9}	1.76×10^{-9}	1.8×10^{-10}	1.8×10^{-10}
		3	0.003	0.005	1.76×10^{-9}	2.93×10^{-9}	1.8×10^{-10}	4.7×10^{-10}
		6	0.003	0.008	1.76×10^{-9}	4.69×10^{-9}	1.8×10^{-10}	7.5×10^{-10}
		9	0.005	0.010	2.93×10^{-9}	5.87×10^{-9}	4.7×10^{-10}	9.4×10^{-10}
		12	0.005	0.011	2.93×10^{-9}	6.45×10^{-9}	4.7×10^{-10}	10×10^{-9}

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		15	0.006	0.011	3.52×10^{-9}	6.45×10^{-9}	5.6×10^{-10}	10×10^{-9}
6	6	0	0.001	0.001	5.8×10^{-10}	5.8×10^{-10}	9.3×10^{-11}	9.3×10^{-11}
		3	0.003	0.002	1.76×10^{-9}	1.17×10^{-9}	1.8×10^{-10}	1.9×10^{-10}
		6	0.003	0.006	1.76×10^{-9}	3.52×10^{-9}	1.8×10^{-10}	5.6×10^{-10}
		9	0.003	0.006	1.76×10^{-9}	3.52×10^{-9}	1.8×10^{-10}	5.6×10^{-10}
		12	0.005	0.006	2.93×10^{-9}	3.52×10^{-9}	4.7×10^{-10}	5.6×10^{-10}
		15	0.006	0.010	3.52×10^{-9}	5.87×10^{-9}	5.6×10^{-10}	9.4×10^{-10}

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For sample 1, CDI room temperature and sunlight storage ranged from 2.34E-9 – 5.87E-9 and 2.34E-9 – 9.4E-10 but the cancer risk for room temperature storage ranged from 3.4E-10 – 9.4E-10 while that of sunlight storage ranged from 3.4E-10 – 1.0E-9.

In sample 2, the CDI of both room temperature and sunlight storage ranged from 5.8E-10 – 3.52E-9 and 5.8E-10 – 5.87E-9 while the cancer risk for both storage ranged from 9.3E-11 – 5.6E-10 and 9.3E-11 – 9.4E-10.

Sample 3, CDI room temp and CDI sunlight ranged from 5.8E-10 – 4.69E-9 and 5.8E-10 – 8.21E-9 but the cancer risk of both room temp and sunlight storage ranged from 9.3E-11 – 7.5E-10 and 9.3E-11 – 9.9E-10.

The 4th stored sample had CDI for room temp and sunlight ranging from 1.17E-9 – 4.1E-9 and 1.17E-9 – 5.87E-9. The respective cancer risk rangedfrom 1.9E-10 – 6.6E-10 and 1.9E-10 – 9.4E-10.

The 5th sample had CDI of both room temperature and sunlight storage ranging from 1.76E-9 – 3.52E-9 and 1.76E-9 – 6.45E-9 but the cancer risk of both ranged from 1.8E-10 - 5.6E-10 and 1.8E-10 – 1.0E-9.

The 6thstored sample had a CDI at roomtemp and sunlight ranging from 5.8E-10 - 3.52E-9 and 5.8E-10 – 5.87E-9. While the cancer risk also ranged from 9.3E-11 – 5.6E-10 and 9.3E-11 – 9.4E-10.

Exposure duration (weeks of storage) and storage temperature in addition to the concentration of leached antimony control the cancer risk. The result showed that the cancer risk increased as the storage duration, temperature, and amount of leached antimony increases

Leaching of volatile and semi volatile organic compounds from packaging material into the soft drink has been shown to increase with length of storage time, temperature and exposure to sunlight.

Because sellers of soft drinks buy in large quantities and store for a long time, at the time of purchase and consumption, the consumers are already consuming large quantities of leached antimony, additional effect is that because Nigeria is within the tropic legion, high evapo-transpiration and perspiration, the populace especially children consume large volume of soft drinks implying large quantities of leached antimony [1].

According to [11] an excess cancer risk can be negligible for values lower than 1.0E-6 whereas values above 1.0E-4 need serious attention.

The value of the present study can be considered very negligible, but the cumulative effect of long-term consumption and additional effect other tumor – inducing organic contaminants can be detrimental to health.

The total cancer risk for the analyzed samples was calculated and the results were shown in Table 3.

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Table 3: The total cancer risk for the analyzed samples

s/n	Sample	Total cancer risk$_{room\ temp}$= $\sum^{n}_{k=1}$ x CDI_k x SF_k	Total cancer risk $_{sunlight}$ = $\sum^{n}_{k=1}$ x CDI_k x SF_k
1	1	3.71E-9	4.62E-9
2	2	2.33E-9	3.003E-9
3	3	3.87E-9	3.053E-9
4	4	2.26E-9	1.81E-9
5	5	6.9E-10	3.44E-10
6	6	1.09E-9	1.22E-9

The total cancer risk of all the samples exposed to room temperature and sunlight correlated with each other. Samples exposed to sunlight showed higher cancer risk than those kept at room temperature.

According to [11] an excess cancer risk can be negligible for values lower than1.0 E-6 whereas above 1.0 E-4 are of health risk concern. In this study, Table 3 showed that the values were within the low risk or no risk range with a range of 1.216E-9 to 5.670E-9 (room temp exposed samples) and 2.542E-9 to 1.305E-8 (sunlight exposed samples), implying that there is low appreciable risk of cancer from exposure to antimony by drinking bottled beverages through leaching from PET bottles but long term accumulation can pose a health risk.

CONCLUSION

The antimony concentration for the tested samples were found to be higher under sunlight than room temperature. This increase was statistically significant ($P < 0.05$) between some weeks of storage at both storage conditions and different storage times, apart from the change in the antimony of sample 3 which showed no significant change under the different storage conditions and time.

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