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Radioactivity in some sachet drinking water samples produced in Nigeria

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Background: Activity concentrations of ^{40}K, ^{226}Ra and ^{228}Ra were measured in some sachet drinking water samples produced in Nigeria. ***Materials and Methods:*** The measurement was done by using high-purity germanium (HPGe) detector (Canberra Industries Inc.). The measured activity concentrations for ^{226}Ra (U-series) and ^{228}Ra (Th-series) were used with their ingested dose conversion factors to estimate annual effective doses for the International Commission on Radiological Protection (ICRP) age groups 0 – 1y, 1 – 2y, 2 – 7y, 7 – 12y, 12 – 17y and >17y from consumption of the water samples. ***Results:*** Measured activity concentration values varied from 0.57±0.21 to 34.08±5.61 Bq l^{-1}, 2.22±0.97 to 15.50±4.51 Bq l^{-1} and 0.04±0.01 to 7.04±1.16 Bq l^{-1} for the radionuclides respectively. Estimated total annual effective doses varied from 4.73 to 49.13, 1.21 to 12.26, 0.86 to 8.54, 1.22 to 11.66, 3.40 to 28.98 and 0.68 to 5.04 mSv y^{-1} for the age groups respectively. The highest total annual effective dose was found in Tisco sample while the lowest was found in Focar sample. ***Conclusion:*** The total annual effective dose in all samples considerably exceeded the average worldwide ingestion exposure dose value of 0.12 mSv y^{-1} from uranium and thorium series reported by the United Nations Scientific Committee on Effects of Atomic Radiations (UNSCEAR). Therefore the Nigeria populace is advised to consume less of these water samples. **Iran. J. Radiat. Res., 2009; 7 (3): 151-158**

Keywords: *Natural radioactivity, sachet water, effective dose, water quality parameter, ingested dose conversion factors.*

INTRODUCTION

Radioactivity, the term used to describe the decay of atomic nucleus, has existed since time began on earth. Its origin has been traced to (i) a number of naturally occurring radioactive materials (NORM), which are present in soils, rocks, the floors and walls of dwellings, offices or schools, in the food humans eat and drink, in the air humans breathe and in human bodies and (ii) man-made or artificial sources. Humans have always been exposed to natural radiation arising from the earth as well as from outer space. Naturally occurring radioactive materials enter the human body through two main pathways – by inhalation of radioactive gases like radon and dust, and ingestion of primordial radionuclides ^{40}K, ^{232}Th and ^{238}U as well as their radioactive progenies. The decay of inhaled or ingested radionuclides gives rise to internal exposure of the tissues and organs in the human body.

The United Nations Scientific Committee on Effects of Atomic Radiation (UNSCEAR) [1] has reported that the average worldwide exposure to natural sources in foods and drinking water (ingestion exposure) is 0.29 mSv y^{-1} (about 0.17 mSv y^{-1} from ^{40}K and about 0.12 mSv y^{-1} from Uranium and Thorium).

In Nigeria, consumption of sachet drinking water has widely increased in recent years. This is partly due to growing awareness that the consumption of unsafe or untreated drinking water is the cause of many diseases especially water-borne diseases. Therefore it is important to set an enhanced trend of sachet water consumption in both rural and urban areas of the country. The World Health Organisation (WHO) has recommended safe values for various drinking water quality parameters in its general guidelines [2]. These guidelines have been used by different countries, e.g the USA and EU to formulate their own national water quality guidelines. The increasing consumption of sachet drinking

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water by people of all ages – infants, children and adults alike – calls for evaluation of its suitability for consumption since its quality varies from source to source. Such evaluation will assist in assessing potential radiation doses so that remedial action or steps can be taken if necessary to avoid undue exposure of consumers. UNSCEAR [1] report provides information on natural radionuclides in drinking water for some countries in North America, Asia and Europe but no such information exists for African countries.

In Nigeria, the National Agency for Food and Drug Administration and Control (NAFDAC) has formulated sachet drinking water quality standards and has been enforcing the standards. The standards emphasise the physico-chemical parameters (non-radioactive contaminants) at the expense of natural activity concentration limits. Many of the producers of sachet drinking water in Nigeria have not been processing and sealing their water as recommended by WHO.

Radioactivity contents have been measured in drinking water in different countries by using various analytical methods [3 – 9].

In this work, we used the method of gamma spectrometry to determine the activity concentrations of ^{40}K, ^{226}Ra and ^{228}Ra in various sachet drinking water samples processed in Akure, Nigeria. The measured activity concentrations were used to estimate annual effective doses from intakes of ^{226}Ra and ^{228}Ra by the population living in the city. The data generated through the study will provide information on natural radioactivity in sachet drinking water for Nigeria and add to the existing sparse data on radioactivity in drinking water in UNSCEAR data bank.

MATERIALS AND METHODS

Analysis of Chemical parameters

Two samples per brand of fifteen different sachet water samples, making a total of 30 samples, were bought from their different producers in Akure, Southwestern Nigeria. All the samples were first analyzed for non-radioactive contaminants. The pH of the water samples was measured using Ph211 meter (Hanna Instruments Inc.) with combined electrodes. The calibration of the instrument was carried out using 4, 7 and 9 buffer standard solutions. The samples were analysed for sodium (Na), magnesium (Mg), and calcium (Ca) using the Model 200 Atomic Absorption Spectrophotometer equipment. Quantitative analysis technique [10] was used to analyse the samples for chloride (Cl^-) and bicarbonate (CO_3^{2-}) while colorimetric method at an absorbance of 420 nm was used to analyse them for nitrate (NO_3^-) and sulfate (SO_4^{2-}).

Radioactivity measurement

The activity concentrations of the water samples were measured using high-resolution high purity germanium (HPGe) vertical co-axial detector (Canberra Model GC 2018-7500, series number 87063) coupled to a Canberra Multichannel Analyzer (MCA) computer system. The detector was enclosed in a 100 mm thick lead shield to reduce the natural external background radiation. The energy and efficiency calibrations of the spectrometer were carried out using standard water sources emitting gamma rays in the energy range 200 – 1500 keV, covering all gamma energies of radionuclides of interest. The calibration techniques are described in IAEA [11]. Each water sample was placed on top of the HPGe detector and counted for 36,000 s. An already washed empty 1-l Marinelli beaker was kept on the detector for the same counting time (36,000 s) under identical geometry to determine the background radiation level of the laboratory environment. The prominent photo peaks observed with regularity in the spectra of the samples were identified as those of the radionuclides in the natural decay series of ^{238}U and ^{232}Th, and the non-series ^{40}K. The analysis of the precursor radionuclides depends on the daughter gamma ray energy

peaks of the decay products in equilibrium with their precursor nuclides. ^{226}Ra activity concentrations were estimated from its gamma ray peak of 609.3 keV from ^{214}Bi. The 583.1 keV gamma ray of ^{208}Tl was used to estimate the activity concentration of ^{228}Ra. The activity concentration of ^{40}K was estimated by using its own gamma ray photopeak at 1460.2 keV. The activity concentration analysis was carried out using a user-friendly SAMPO 90 spectra analysis software, which matched various gamma-energy peaks to a library of possible radionuclides.

Calculation of annual effective dose

Estimation of annual effective dose E_d (Sv y^{-1}) to an individual due to the consumption of ^{226}Ra and ^{228}Ra present in the sachet drinking water was done using the following relationship:

$$E_d = A_c A_i C_f \qquad (1)$$

by Alam *et al.* [12] where A_c is the activity concentration of the radionuclide in the sachet water (Bq l^{-1}), A_i is the annual intake of sachet drinking water (l y^{-1}) and C_f is the ingested dose conversion factor for radionuclides (Sv Bq^{-1}), which varies with both radionuclides and the age of individuals ingesting the radionuclides. The total effective dose D (Sv y^{-1}) to an individual was established by summing contributions from all radionuclides present in the water samples i.e.

$$D = \sum A_c A_i C_f \qquad (2)$$

The conversion factors used in the estimations were taken from DWAF [13]. The annual effective dose was calculated for the six ICRP [14] age groups 0 - 1y, 1 – 2y, 2 – 7y, 7 – 12y, 12 – 17y and >17y old with annual average water intake of 200, 260, 300, 350, 600 and 730 litres respectively.

RESULTS AND DISCUSSION

The results of the measurement of the non-radioactive contaminant in the sachet water samples are presented in table 1. The pH of the samples varied from 7.1 to 7.6. Primus, Tisco and DPC with pH of 7.1 are nearly pure water. Other samples are alkaline with Febtolene being the most alkaline sample. The most variable of the non-radioactive contaminants was Chloride that ranged from 104.4 to 362.8 mg/ l. The least variable was Nitrate that varied from 10 to 23 mg/l.

Measured activity concentrations of ^{226}Ra and ^{228}Ra, which are the decay product of ^{238}U and ^{232}Th, respectively as well as that of ^{40}K in the sachet water samples are presented in table 2. The activity concentrations of ^{40}K, ^{226}Ra and ^{228}Ra ranged from 0.57±0.21 to 34.08±5.61 Bq kg^{-1}, 2.22±0.97 to 15.50±4.51 Bq kg^{-1} and 0.04±0.01 to 7.04±1.16 Bq kg^{-1} respectively. Zion sample had the highest concentration of ^{40}K while Josta had the lowest concentration. The highest and lowest activity concentration values of ^{226}Ra were found in Chiva sample and Focar sample respectively. While the highest concentration of ^{228}Ra was found in Tisco, the lowest was found in C.P.W. These variations are attributable to the different sources of the water samples. The mean activity concentration values were 19.09±10.05, 7.75±4.28 and 2.03±1.95 Bq kg^{-1} for ^{40}K, ^{226}Ra and ^{228}Ra respectively. Thus ^{40}K contributed the largest activity concentration while ^{228}Ra contributed least activity in the water samples. The ± values associated with the mean values represent the variability (standard deviation) in the activity concentration values of the radionuclide. The table shows that the activity concentrations of ^{226}Ra in all the samples exceeded the limit of 1.00 Bq l^{-1} recommended by IAEA [11]. This is found to be greater than values reported for the U.S.A and Poland as shown in table 3.

The mean activity concentration of 2.03±1.95 Bq kg^{-1} obtained for ^{228}Ra in the water samples was above the value of 1.06±0.31 Bq l^{-1} reported for commercialized drinking water from Tunisia [7].

The results of the calculated age-dependent annual effective dose (mSv y^{-1})

Table 1. Concentration of chemical parameters (mg^{-1}) of sachet water samples.

Sample	pH	Na	Mg	Ca	Cl^-	SO_4^{2-}	NO_3	CO_3^{2-}
Amuye	7.6	39.5	40.3	30.6	185.3	88.0	22.5	62.4
Nitokeba	7.5	32.9	51.4	40.4	242.1	55.0	11.5	61.0
Josta	7.3	50.5	60.5	69.5	205.9	67.0	23.0	58.2
Febtolene	7.6	51.4	55.6	70.5	362.1	59.5	16.0	62.2
Bofa	7.4	60.5	60.1	60.1	220.1	72.5	10.5	81.8
Primus	7.1	42.7	45.4	30.3	135.6	60.0	10.0	22.2
Feotamy	7.3	45.5	56.8	56.4	230.8	80.5	11.5	46.6
Dabmot	7.5	55.1	50.5	60.9	347.9	55.0	23.0	63.0
C.P.W	7.3	31.3	64.2	70.6	362.8	55.0	10.5	82.0
Focar	7.4	40.8	48.3	80.5	323.1	80.5	12.0	101.0
Chiva	7.2	29.5	35.6	37.9	220.1	88.0	22.5	62.2
Tisco	7.1	10.9	38.9	40.5	152.7	52.5	16.0	23.0
DPC	7.1	19.5	30.5	25.5	104.4	52.5	16.0	22.4
Yem-Saw	7.3	40.7	51.3	50.7	227.2	59.5	11.5	42.0
Zion	7.5	53.5	60.1	60.2	239.3	80.5	10.5	63.0

Table 2. Activity concentration of different radionuclides present in the water samples.

Sample No.	Sample name	Activity concentration ($Bq\ l^{-1}$)		
		^{40}K	^{226}Ra	^{228}Ra
1.	Amuye	10.53± 3.78	4.11±1.98	0.74±0.32
2.	Nitokeba	9.58± 2.96	4.32±2.04	3.93±1.07
3.	Josta	0.57± 0.21	4.30±1.87	3.52±1.20
4.	Febtolene	4.97± 1.31	2.90±0.66	0.43±0.17
5.	Bofa	19.12± 4.32	7.70±2.30	1.13±0.72
6.	Primus	33.85±11.23	9.11±3.12	0.40±0.13
7.	Feotamy	17.42± 6.07	8.62±2.64	1.79±0.67
8.	Dabmot	31.46±12.08	7.04±2.13	0.56±0.22
9.	C.P.W	27.71± 7.60	14.40±3.42	0.04±0.01
10.	Focar	20.57± 6.34	2.22±0.97	0.44±0.08
11.	Chiva	22.01± 0.26	15.50±4.51	2.70±1.30
12.	Tisco	14.10± 3.92	7.33±1.01	7.04±1.16
13.	DPC	16.33± 0.11	11.02±2.11	1.37±0.09
14.	Yem-Saw	24.02± 4.11	13.45±3.39	3.02±0.64
15.	Zion	34.08± 5.61	4.27±1.48	4.04±1.40
Range		0.57 – 34.08	2.22 – 15.50	0.04 – 7.04
Mean		19.09	7.75	2.03
Standard deviation		10.05	4.28	1.95

Table 3. Comparison of ^{226}Ra activity concentration range in drinking water from different countries.

Country	Concentration range (mBq l^{-1})
U.S	0.4 – 1.8[a]
France	7.0 – 700[a]
Finland	10.0 – 49000[a]
Germany	1.0 – 1800[a]
Italy	0.2 – 1200[a]
Poland	1.7 – 4.5[a]
Spain	<20 – 4000[a]
Turkey (Istanbul)	11 – 36[b]
Turkey (Eastern Black Sea)	3 – 45[c]
Nigeria	2220 – 15500[d]

[a]UNSCEAR, 2000 (1)
[b]Karahan *et al.* 2000 (15)
[c]Cevik *et al.* 2006 (8)
[d]This study

are presented in table 4. The table shows that the annual effective dose due to the intake of ^{226}Ra in the water samples varied from 2.09±0.91 to 14.57±4.24 mSv y^{-1}, 0.55±0.24 to 3.87±1.13 mSv y^{-1}, 0.41±0.18 to 2.88±0.84 mSv y^{-1}, 0.62±0.27 to 4.34±1.26 mSv y^{-1}, 2.00±0.87 to 13.95±4.06 mSv y^{-1}, and 0.45±0.20 to 3.17±0.92 mSv y^{-1} for age group 0 – 1 y, 1 – 2 y, 2 – 7 y, 7 – 12 y, 12 – 17 y and >17 y respectively. The annual effective dose due to the intake of ^{228}Ra ranged from 0.24±0.06 to 42.24±6.96 mSv y^{-1}, 0.06±0.02 to 10.43±1.72 mSv y^{-1}, 0.04±0.01 to 7.18±1.18 mSv y^{-1}, 0.05±0.01 to 9.61±1.58 mSv y^{-1}, 0.13±0.03 to 22.39±3.69 mSv y^{-1} and 0.02±0.01 to 3.55±0.58 mSv y^{-1} respectively for age group 0 – 1 y, 1 – 2 y, 2 – 7 y, 7 – 12 y, 12 – 17 y and >17 y. The mean annual effective doses from ^{226}Ra to the six age groups are 7.29±3.86, 1.93±1.03, 1.44±0.77, 2.17±1.15, 6.98±3.70 and 1.58±0.84 mSv y^{-1} respectively and that due to ^{228}Ra in the samples are 25.79±53.36, 3.08±2.79, 2.12±1.92, 2.84±2.57, 6.60±5.99 and

Table 4. Annual effective dose (mSv y^{-1}) for the six age groups.

SAMPLE	^{226}Ra					
	0 - 1y	1 - 2y	2 – 7y	7 - 12y	12 – 17y	≥ 17y
Amuye	3.86±1.86	1.03±0.49	0.76±0.37	1.15±0.55	3.70±1.78	0.84±0.41
Nitokeba	4.06±1.92	1.08±0.51	0.81±0.38	1.21±0.57	3.89±1.84	0.88±0.42
Josta	4.04±1.76	1.07±0.47	0.80±0.35	1.20±0.52	3.87±1.68	0.88±0.38
Febtolene	2.73±0.62	0.72±0.17	0.54±0.12	0.81±0.19	2.61±0.59	0.59±0.14
Bofa	7.24±2.16	1.92±0.57	1.43±0.43	2.16±064	6.93±2.07	1.57±0.47
Primus	8.56±2.93	2.27±0.78	1.69±0.58	2.55±0.87	8.20±2.81	1.86±0.64
Feotamy	8.10±2.48	2.15±0.66	1.60±0.49	2.41±0.74	7.76±2.38	1.76±0.54
Dabmot	6.62±2.00	1.76±0.53	1.31±0.40	1.97±0.60	6.34±1.92	1.44±0.44
C.P.W	13.54±3.22	3.59±0.85	2.68±0.64	4.03±0.96	12.96±3.08	2.94±0.70
Focar	2.09±0.91	0.55±0.24	0.41±0.18	0.62±0.27	2.00±0.87	0.45±0.20
Chiva	14.57±4.24	3.87±1.13	2.88±0.84	4.34±1.26	13.95±4.06	3.17±0.92
Tisco	6.89±0.95	1.83±0.25	1.36±0.19	2.05±0.28	6.60±0.91	1.50±0.21
DPC	10.34±1.98	2.75±0.53	2.05±0.39	3.09±0.59	9.92±1.90	2.25±0.43
Yem-Saw	12.64±3.19	3.36±0.85	2.50±0.63	3.77±0.95	12.11±3.05	2.75±0.69
Zion	4.04±1.39	1.07±0.34	0.79±0.28	1.20±0.41	3.84±1.33	0.87±0.31
Range	2.09 – 14.5	0.55 – 3.87	0.41 – 2.88	0.62 – 4.34	2.00 – 13.95	0.45 – 3.17
Mean	7.29	1.93	1.44	2.17	6.98	1.58
SD	3.86	1.03	0.77	1.15	3.70	0.84

SD - Standard Deviation

Table 4: Continues

SAMPLE	^{228}Ra					
	0 - 1y	1 - 2y	2 – 7y	7 - 12y	12 – 17y	≥ 17y
Amuye	4.44±1.92	1.10±0.47	0.76±0.33	1.01±0.44	2.35±1.02	0.37±0.16
Nitokeba	23.58±6.42	5.82±1.59	4.01±1.09	5.36±1.46	12.50±3.40	1.98±0.54
Josta	21.12±7.20	5.22±1.78	3.59±1.22	4.81±1.64	11.19±3.82	1.77±0.61
Febtolene	2.58±1.02	0.64±0.25	0.44±0.17	0.59±0.23	1.37±0.54	0.22±0.09
Bofa	6.78±4.32	1.68±1.07	1.15±0.73	1.54±0.98	3.59±2.29	0.57±0.36
Primus	2.40±0.78	0.59±0.19	0.41±0.13	0.55±0.18	1.27±0.41	0.20±0.07
Feotamy	10.74±4.02	2.65±0.99	1.83±0.68	2.44±0.92	5.69±2.13	0.90±0.34
Dabmot	3.36±1.32	0.83±0.33	0.57±0.22	0.76±0.30	1.78±0.70	0.28±0.11
C.P.W	0.24±0.06	0.06±0.02	0.04±0.01	0.05±0.01	0.13±0.03	0.02±0.01
Focar	2.64±0.48	0.65±0.12	0.45±0.08	0.60±0.11	1.40±0.25	0.22±0.04
Chiva	16.20±7.80	4.00±1.93	2.75±1.33	3.69±1.78	8.59±4.13	1.36±0.66
Tisco	42.24±6.96	10.43±1.72	7.18±1.18	9.61±1.58	22.39±3.69	3.55±0.58
DPC	8.22±0.54	2.03±0.13	1.40±0.09	1.87±0.12	4.36±0.29	0.69±0.05
Yem-Saw	18.12±3.84	4.48±0.95	3.08±0.65	4.12±0.87	9.60±2.04	1.52±0.32
Zion	24.24±8.40	5.99±2.08	4.12±1.43	5.52±1.91	12.85±4.45	2.04±0.71
Range	0.24 – 42.24	0.06 – 10.43	0.04 – 7.18	0.05 – 9.61	0.13 – 22.39	0.02 – 3.55
Mean	25.79	3.08	2.12	2.84	6.60	1.05
SD	53.36	2.79	1.92	2.57	5.99	0.95

SD - Standard Deviation

1.05±0.95 mSv y^{-1} respectively. The age group with the highest exposure dose is 0 – 1 y (babies) followed by the 12 – 17 y age group. The annual effective doses from ^{226}Ra to all age groups are higher than the reported worldwide average value of 0.12 mSv y^{-1} for Uranium and Thorium [1]. Those due to ^{228}Ra in the samples are lower than the reported worldwide average value only in C.P.W and only for age groups 1 – 2 y, 2 – 7 y, 7 – 12 y and >17 y.

The total annual effective dose to the six age groups considered in this study is presented in table 5 and pictorially in figure 1. The figure shows that babies (0 – 1 y old) are most exposed internally to radium in the water samples. Radium is highly radiotoxic and it builds up in the growing bones of babies and children where it can cause bone cancers. Therefore the use of these sampled sachet drinking water for babies should be discouraged. The figure shows that people of ages 12 to 17 years also suffer substantial internal exposure from these sampled sachet drinking waters. In fact people of all ages that consume the water face the risk of some health effects that may result from the significant accumulation of radium in their bones and other vulnerable or radiosensitive soft body tissues. Hence remedial measures should, as a matter of urgency, be taken to reduce radium in these sachet drinking waters. Their sources must be treated using conventional water-treatment methods that remove radium from ground water.

Figure 2 shows a bar graph comparison of the total annual effective dose from each of the sampled sachet drinking water. The figure shows that Tisco sample gave the highest internal exposure, followed by Chiva and Yem-Saw samples. Focar and Febtolene sachet water gave the least internal exposure to consumers.

CONCLUSION

This study shows that it not radiologically safe to consume any of the sachet drinking water samples that have been investigated in this study since they all gave

Table 5. Total annual effective dose (mSv y^{-1}) to the six age groups.

Sample	0-1y	1-2 y	2-7 y	7-12 y	12-17 y	≥ 17 y
Amuye	8.30	2.12	1.52	2.16	6.05	1.21
Nitokeba	27.64	6.90	4.81	6.57	16.39	2.86
Josta	25.16	6.29	4.39	6.01	15.06	2.65
Febtolene	5.31	1.36	0.98	1.40	3.98	0.81
Bofa	14.02	3.60	2.58	3.70	10.52	2.14
Primus	10.96	2.87	2.10	3.10	9.47	2.06
Feotamy	18.84	4.80	3.43	4.86	13.45	2.66
Dabmot	9.98	2.59	1.88	2.74	8.12	1.72
C.P.W	13.78	3.65	2.72	4.09	13.09	2.96
Focar	4.73	1.21	0.86	1.22	3.40	0.68
Chiva	30.77	7.87	5.64	8.03	22.54	4.53
Tisco	49.13	12.26	8.54	11.66	28.98	5.04
DPC	18.58	4.78	3.45	4.96	14.27	2.94
Yem-Saw	30.76	7.83	5.58	7.89	21.71	4.27

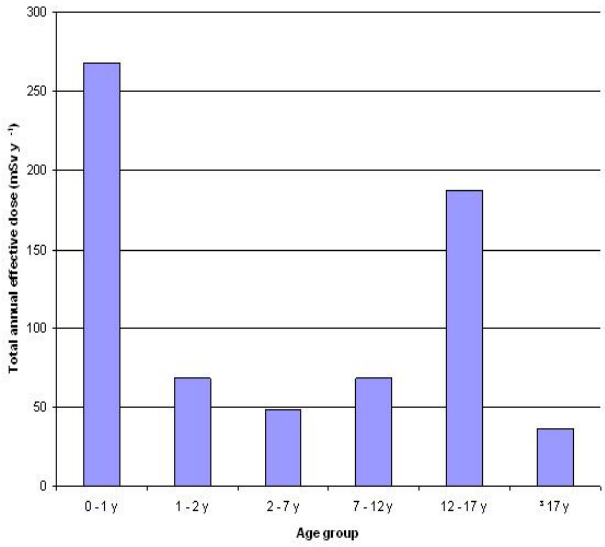

Figure 1. Total annual effective dose (mSv/y) to the six age groups.

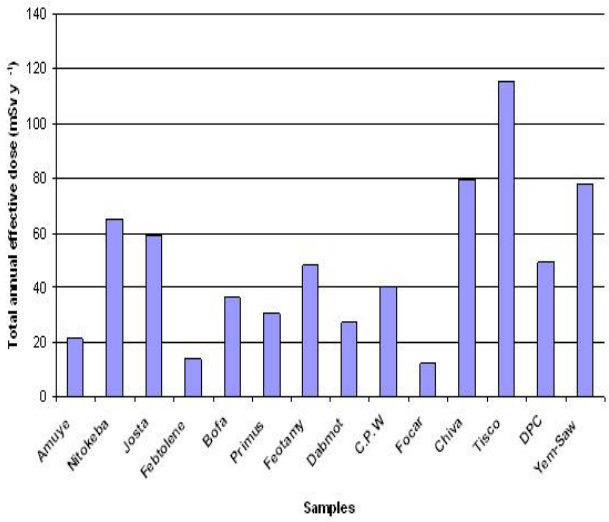

Figure 2. Comparison of total annual effective dose (mSv/y) from different samples.

much larger internal exposures than the UNSCEAR reported world average value of 0.12 mSv y^{-1} and the WHO and ICRP reference limits of 0.1 mSv y^{-1} and 1.0 mSv y^{-1} respectively. We recommend all the sachet drinking water samples for treatment that will remove radium from them and advise that less of these water samples should be consumed by the Nigeria populace.

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