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Abstract: The radioactivity analysis of gross alpha concentrations were comprehensively carried out in portable drinking sachets water samples collected from sixty different locations in Abuja province, Nigeria. Using the low background gas free proportional counter (MPC-2000B-DP model) detector, the gross alpha activity concentrations values measured in the drinking water samples ranged from (0.0025 ± 0.0001) mBqL⁻¹ to (0.0969 ± 0.0001) mBqL⁻¹. The computed mean gross alpha activity concentrations and mean effective equivalent dose of alpha emitters were (0.0338 ± 0.0001) mBqL⁻¹ and 0.0145mSvy⁻¹ respectively. The obtained results revealed that the overall average effective doses for all the measured portable drinking water samples were much below the limit value of 500mBqL⁻¹ and reference level of 0.1mSvy⁻¹ recommended by World Health Organization (WHO) and poses no significant radiological health concerns.

Keywords: Gross alpha, Drinking water, Annual effective dose, Activity concentration, Abuja (Nigeria)

1. INTRODUCTION

Water is essential for sustenance of life and that is why it is very important to ensure an extremely clean environment for water production. For majority of populace portable sachets drinking water offers a convenient choice to stay hydrated due to its affordability, accessibility and portability [2, 29]. The increased demand for these brands of drinkable water products is attributed mostly to factors such as inadequate or non-availability of reliable, safe municipal water in urban areas; impression that high quality natural spring water and portable drinkable water provide a healthy, refreshing and great tasting alternative to high-calorie soft drinks and ordinary tap water; and convenience which has made the products meet the requirements of any lifestyle when needed [29]. The integrity of the hygienic environment and conditions where the majority of these drinking water factories are located and manufactured has been of great concerns. Virtually all the portable drinkable water factories in Abuja, Nigeria exploited groundwater as the sources for their purification processes. The absorbed radionuclide accumulated in the human body is mainly accomplished through the consumption of water and food. Radioactivity in portable drinking water is an easy means for human beings internal contamination with radionuclides. The presence of radionuclide in drinking water beyond the recommended level could present a risk to human health from their consumption at a regular rate [11]. The radionuclides in drinking water are members of three natural radioactive series (²³⁸U, ²³²Th, ²²⁷Ac) and ⁴⁰K in soil and bedrock, ²³⁸U, ²³⁴U, ²³²Th, ²³⁰Th, ²²⁸Th, ²²⁶Ra and ²¹⁰Po are the principal alpha emitting radionuclides in the natural decay series. The nuclides of the uranium series which can be dangerous to health because of their presence in portable drinking water are ²²²Rn, ²²⁶Ra and their decay products are more concentrated in deep ground water than in surface water [4, 35]. They contaminate the water body directly with their radionuclide products; and indirectly through the radon gaseous products which can solidify and attach themselves as aerosols to the air particles and are washed down by rain into water bodies. Furthermore, human activities such as burning of fossils fuels, milling, mining, processing of uranium ores and mineral sands, metal refining, manufacture of fertilizers etc. have raised Naturally Occurring Radioactive Materials (NORMs) concentrations of the earth's crust in the environment; which emits alpha, beta and gamma radiations [5, 32]. Radiation is part of the natural environment and it is therefore estimated that approximately more than ninety-eight percent of all human exposure comes

from NORMs [34]. Hence, the intake of radionuclides in portable drinking water stimulate human internal exposure. Previous work on portable drinking water phenomenon in Nigeria, have indicate that factors responsible for its contamination ranges from sharp practices, poor hygiene of locations, polluted environment and non-adherence to World Health Organization (WHO), International Commission on Radiological Protection (ICRP) and European Union Commission (EC) regulations [9, 17, 37-40]. The WHO guidelines for portable drinking water suggest performing an indirect evaluation of effective dose by measuring gross alpha radioactivity and checking compliance to derived limit values (0.1BqL⁻¹ gross alpha activity, 0.1mSvy⁻¹ effective dose) [11, 23]. Amongst the various factors which gross alpha activity depends on are: type of radionuclides used as the calibration standard, time interval between samples collection, samples preparation and analysis, counting efficiency of the radioactive [11]. If the gross alpha and gross beta are less than 500mBqL⁻¹ and 1000mBqL⁻¹, it can be assumed that the Total Indicative Dose (TID) is less than the parametric indicator value of 0.1mSvy⁻¹ and no further radiological investigation is needed, but if the reference level is exceeded analysis for specific radionuclides is required [40]. Recently, a great interest arose towards the natural radioactivity concentrations in portable drinking water around the world due to the great danger it presents [2-4, 6, 8, 12-13, 16, 20, 24-30, 35, 41]. Portable drinking water manufacturers in Nigeria needs to adhere strictly to the relevant Nigerian Industrial Standards (NIS), WHO and EC regulations in order to avoid potential hazardous health situation and other water related diseases that could arise from the use of contaminated water.

Abuja province is a federal territory in central Nigeria with one of the most interesting and outstanding landscapes in Africa and the fastest-growing city in the world, located just north of the confluence of the Niger River and Benue River. The study area lies with latitude 8°25′N and 9°4′N of the equator and longitude 6°45′E and 7°29′E of Greenwich Meridian. It has a landmass of approximately 7315km², nearly 0.79% of Nigeria total area, and it is situated within the savannah region with moderate climatic conditions. The portable drinking water produced in this area were consumed by approximately 2.4 million people living in the study area and distributed around the country. The public water boards are generally ineffective in supplying portable drinking water in this area, thus most of the populations depends on untreated ground water sources (boreholes and wells) for domestic and industrial purposes. The ground water collected from boreholes and dug wells samples are not entirely free from radioactive pollutants which are hazardous to human health. In this work, we aimed to determine the current activity concentrations of gross alpha radiation and estimate the annual effective dose rate due to the consumption of drinking water by sampling portable drinking water from sixty (60) locations distributed throughout the six (6) local councils of Abuja, Nigeria, which can be used for detecting changes in the radiological features of the portable drinkable water sources.

2. MATERIALS AND METHODS

2.1. Samples Collection

Ten (10) different samples of portable drinking water were obtained directly from the manufacturing companies in each locations in different parts of the six local councils of Abuja, with each location samples coded as A, B, C, D, E and F. The capacity of the collected samples ranges from 0.5L to 2.0L, with a total of sixty (60) samples of portable water sachets collected from points distributed throughout the Abuja metropolis for effective coverage of the study area. The collected samples were preserved in accordance with the ISO standard (20ml of 50% V of HNO₃ per litre of water, pH of 2.0) so as to minimize the loss of radioactive material from solution due to absorption. Stratified random sampling method is the technique applied to the samples [36]. At every point of sample collection the container is first rinsed twice before the water samples is put in the plastic container (with 85mm wide top opening). To avoid possible ionic exchanges between suspended and dissolved matter during storage, samples were transferred to the laboratory for analysis within a short time after collection. The International Standards Organization procedure [17-19] for the measurement of gross alpha and beta activities in drinking water was employed in all the water samples.

2.2. Samples Preparation

10ml of concentrated HNO_3 acid was added into 2.0L of water samples so as to prevent the absorption of the water sample by the wall of the container, minimizing the precipitation, reduce growth of microorganisms and reduce the pH of the water samples below 2.0. About 600cm³ of the samples water

solution was measured and transferred into a beaker. The evaporation was done at 60°C temperature for 8 hour 30 minutes until the volume was reduced to about 50ml and allowed to cool, and then set on hot plate magnetic stirrer at a steady temperature to avoid boiling in order to prevent loss of much residue and unnecessary samples thickness which may lead to self-absorption, the volume evaporated was then taken and recorded. The next process is surface drying in which the residual volume were transferred quantitatively into a petri-dish for further drying until the final dry residue is obtained. Then about 77mg of the residue was weighed using digital analytical weighing balance and transferred into an aluminium planchette, then dried under an infrared lamp until precipitation take place in the detector. The precipitate was then filtered through a filter paper by vacuum pump. Each samples precipitation in planchette was directly applied to the counting systems. Few drops of acetone and vinyl-acetate were applied into the residue and stirred moderately for 30 minutes at room temperature to obtain uniform dispersions. The vinyl-acetate helps in removing the moisture content while the acetone serves as a binder to avoid contamination of any kind. This procedure was repeated for all the samples. The samples were analysed for gross alpha activity using the low background gas free proportional counter (MPC-2000B-DP model) detector at the Centre for Energy Research and Training (CERT), Ahmadu Bello University, Zaria, Nigeria. The counter composed of sample detector with a window made up of $450 \mu \text{g cm}^{-2}$ and diameter of 60mm. There is a small chamber above the detector which serves as a guard against the influence of high energy cosmic radiation that might enter the measuring environment. The counting gas (90% Argon and 10% Methane) with a flow were adjusted to a pressure slightly above atmospheric pressure. The gross alpha concentrations was determined by taking the average result of three counts. The results indicated that the activity was invariant over a period of time, confirming the validity of the sampling and preparation techniques. The background measurements, plateau test and sample efficiency were simultaneously performed to determine the background gross alpha radioactivity concentrations, optimal operational voltage and frequencies using standard methods.

2.3. Gross Alpha Counting

The counting procedure involves entering the pre-set time, number of cycles and counting mode then the counting is automated. The background radiation level within the measuring environment was estimated to be less than 1.0mSvhr^{-1} . The chambers were covered with 0.1m thick lead and inside dimensions of $(0.480m \times 0.280m \times 0.105m)$. The alpha standards are Plutonium-239 alpha sources with activity ranging from 133.3Bq to 185.8Bq for the counter channel at 2π -steradians. The radionuclides impurity in the channel varied from 0.74 percent to 0.82 percent. The results were displayed as raw counts and count rate (cpm). The raw counts (cpm) were repeated three times each for all the sixty samples and the mean values were used in calculating the gross alpha activity. An optimum alpha plateau was achieved at a 585V operating voltage and sample were counted for three cycles of 3600 second per cycle in alpha only mode. The counting system incorporates interference from high energy cosmic radiation into the measuring environment and was calibrated following the ISO calibration standard procedure [17-19]. The samples efficiency for alpha counting and volume for the water samples were determined using equations (1 and 2):

Sample efficiency,
$$S_{\varepsilon(\alpha)} = \frac{S_w}{M_r} \times 100\%$$
 (1)

Sample volume,
$$S_V = \frac{V}{M} \times S_w$$
 (2)

Where, S_w is the sample weight on the planchette in (mg), M_r is the residual sample weight from evaporated water samples, V is the volume of water sample evaporated in litres and M is the residue mass in (mg) from volume V. We obtained the gross alpha sample count rate and activity concentration in Becquerel per litre (BqL⁻¹) for each water samples as follows:

Alpha Count Rate,
$$\alpha_{(CR)} = \frac{R_{(\alpha)} \times 60}{t}$$
 (3)

Activity,
$$A_{(\alpha)} = \frac{N_{(\alpha)} \times U_{(\alpha)}}{S_{\varepsilon(\alpha)} \times V_{(S)} \times D_{\varepsilon(\alpha)}}$$
(4)

Net counts,
$$N_{(\alpha)} = Raw \text{ count rate(cpm)} - Background \text{ count rate(cpm)} = R_{(\alpha)} - B_{(\alpha)}$$
 (5)

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Activity Concentration,
$$C_{(\alpha)} = \frac{\{R_{(\alpha)} - B_{(\alpha)}\}}{\{R_{(S)} - B_{(\alpha)}\}} \times \frac{M}{1000} \times \frac{1.02}{V}$$
 (6)

We then calculate uncertainty associated with the sample activity using equation (7):

Uncertainty,
$$\epsilon_{\rm A} = \pm \sqrt{\frac{N_{(\alpha)} \times B_{(\alpha)}}{\left[B_{t(\alpha)}\right]^2}} \times \frac{U_{(\alpha)}}{60 \times D_{\epsilon(\alpha)} \times S_{\epsilon(\alpha)}}$$
 (7)

Where, $\alpha_{(CR)}$ is the count rate (cpm), $R_{(\alpha)}$ is raw count rate (cpm), $R_{(S)}$ is the standard count rate (cpm), t is the count time, $A_{(\alpha)}$ is the alpha activity, $B_{(\alpha)}$ is the background count rate (cpm), $U_{(\alpha)}$ is the unit coefficient of alpha particle, $S_{\epsilon(\alpha)}$ is the sample efficiency factor for alpha counting, $D_{\epsilon(\alpha)}$ is the detector's efficiency of the alpha counting, ϵ_A is the uncertainty or error associated with the sample activity, $B_{t(\alpha)}$ is the background count time, and 60 is the conversion factor from minute to second. The gross alpha activity concentrations were calculated by applying calibration data of the counting system. The expression for the gross alpha average annual effective dose for samples was then calculated as:

Alpha Average Annual Effective Dose,
$$\alpha_{avg_{(AED)}} = \sum_{i}^{R(\alpha)} (A_{i(\alpha)} \times IEDF_{i(\alpha)} \times IR_w)$$
 (8)

Where, $\alpha_{avg_{(AED)}}$ is the gross alpha average annual effective dose (mSvyr⁻¹) due to the intake of water, IR_w is the intake of water for a person in one year (730L), IEDF_{i(\alpha)} is the ingestion effective dose conversion factor of the individual natural radionuclides (3.58×10^{-7} SvBq⁻¹ for alpha), and A_{i(\alpha)} is the gross activity concentration of individual radionuclides present in the portable drinking water samples.

3. RESULTS AND DISCURSION

The results of the gross alpha radioactivity concentrations and effective equivalent doses values measured in the portable drinking water samples and associated uncertainties used for alpha counting in each water samples according to the six locations are presented in Tables 1. The mean effective equivalent dose of gross alpha activity concentrations for all the sixty (60) samples are computed in Table 2.

Location code	Sample ID	Gross Alpha (mBqL ⁻¹)	Alpha effective equivalent dose
			$\alpha_{avg_{(AED)}}$ (mSv)
A	E-bright	0.0363 ± 0.0001	0.0386
	Jimroose	0.0147 ± 0.0001	0.0156
	Persido	0.0187 ± 0.0001	0.0199
	Ajason	0.0073 ± 0.0001	0.0078
	Mumal	0.0125 ± 0.0001	0.0135
	Chidera	0.0171 ± 0.0001	0.0182
	Ero	0.0569 ± 0.0002	0.0604
	Zaneta	0.0025 ± 0.0001	0.0027
	Righteous	0.0182 ± 0.0001	0.0193
	Frost	0.0060 ± 0.0001	0.0006
В	Vabe Glory	0.0168 ± 0.0001	0.0026
	Zinno	0.0425 ± 0.0002	0.0067
	Nibodas	0.0825 ± 0.0001	0.0129
	Manawes	0.0353 ± 0.0001	0.0055
	Zippos	0.0389 ± 0.0001	0.0061
	Koye	0.0559 ± 0.0001	0.0088
	Giovani	0.0969 ± 0.0001	0.0152
	Aristo	0.0299 ± 0.0001	0.0047
	De-Rehoboth	0.0064 ± 0.0001	0.0011
	Omatola Obida	0.0286 ± 0.0001	0.0046

Table1. Gross alpha activity concentrations and effective equivalent dose for different samples locations code of portable drinking water samples.

С	Cholas	0.0495±0.0002	0.0203
	Balin Crown	0.0137±0.0001	0.0056
	Hajedoc	0.0367±0.0001	0.0151
	BSK	0.0179±0.0001	0.0074
	Qbase	0.0099±0.0001	0.0041
	Peak Fresh	0.0122±0.0001	0.0050
	Dewluk	0.0619±0.0002	0.0254
	Shybof	0.0269±0.0001	0.0111
	Second Adam	0.0378±0.0001	0.0155
	Rock Pool	0.0418±0.0001	0.0172
D	Angvel	0.0725±0.0001	0.0295
	Abayaro	0.0208±0.0001	0.0146
	Ohirehi	0.0144±0.0002	0.0185
	Angus	0.0281±0.0001	0.0124
	Akala	0.0114±0.0001	0.0168
	Success	0.0088±0.0001	0.0071
	Manko	0.0864±0.0002	0.0351
	Bamu	0.0131±0.0001	0.0116
	Omowumi	0.0609±0.0001	0.0248
	Simac	0.0307±0.0001	0.0125
Е	Valento	0.0618±0.0002	0.0253
	Nabila	0.0045 ± 0.0001	0.0018
	Maochem	0.0663±0.0002	0.0271
	Fakubs	0.0136±0.0001	0.0056
	Kalmed	0.0303 ± 0.0001	0.0124
	Jeffco	0.0686±0.0001	0.0280
	Gera Life	0.0399 ± 0.0001	0.0163
	Mojifet	0.0229±0.0001	0.0094
	Fatras	0.0256±0.0001	0.0105
	Victory	0.0314 ± 0.0001	0.0128
F	Chydera	0.0209 ± 0.0001	0.0085
	Maranatha	0.0493±0.0001	0.0200
	Kawo	0.0122±0.0001	0.0049
	Chimex	0.0570±0.0001	0.0232
	Brudan	0.0623 ± 0.0002	0.0254
	Clement	0.0194±0.0001	0.0079
	Danita	0.0115±0.0001	0.0047
	Felvin	0.0516±0.0001	0.0210
	Solap	0.0162±0.0001	0.0066
	Tahur	0.0817+0.0001	0.0333

Table2. *Mean effective equivalent dose of gross alpha activity concentrations for all the portable drinking water samples locations in Abuja.*

Location code	Mean Gross Alpha (mBqL ⁻¹)	Mean alpha effective equivalent dose
		$\alpha_{avg_{(AED)}}$ (mSv)
А	0.0190 ± 0.0001	0.0197
В	0.0434 ± 0.0001	0.0068
С	0.0308 ± 0.0001	0.0127
D	0.0347 ± 0.0001	0.0183
E	0.0365 ± 0.0001	0.0149
F	0.0382 ± 0.0001	0.0156

The gross alpha activity concentrations for the ten different portable drinking water samples in each of the locations are illustrated in figures (1-6). The mean gross alpha activity concentrations and effective equivalent dose in each samples locations are showed in figure 7.



Fig1. Gross alpha activity concentrations in location A.



Fig2. Gross alpha activity concentrations in location B.



Fig3. Gross alpha activity concentrations in location C.



Fig4. Gross alpha activity concentrations in location D.

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Fig6. Gross alpha activity concentrations in location F.





From the above figures (1-6), the results presented indicates that the gross alpha activity concentrations in location A (Gwagwalada) ranges from (0.0025 ± 0.0001) mBqL⁻¹ to (0.0569 ± 0.0002) mBqL⁻¹ with an average value of (0.0190 ± 0.0001) mBqL⁻¹; location B (AMAC) ranges from (0.0064 ± 0.0001) mBqL⁻¹ to (0.0969 ± 0.0001) mBqL⁻¹ with an average value of (0.0434 ± 0.0001) mBqL⁻¹; location C (Bwari) ranges from (0.0099 ± 0.0001) mBqL⁻¹ to (0.0619 ± 0.0002) mBqL⁻¹ with an average value of (0.0308 ± 0.0001) mBqL⁻¹; location D (Abaji) ranges from (0.0088 ± 0.0001) mBqL⁻¹ to (0.0686 ± 0.0001) mBqL⁻¹ to (0.0365 ± 0.0001) mBqL⁻¹ and location F (Kwali) ranges from (0.0115 ± 0.0001) mBqL⁻¹ to (0.0817 ± 0.0001) mBqL⁻¹ with an average of (0.0382 ± 0.0001) mBqL⁻¹. The low level charts illustrate gross alpha activity concentrations of water

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samples with low rate of alpha activity. Comparison of the results of average gross alpha activity concentrations within the reference point of study shows that the highest alpha radioactivity concentrations in portable drinking water samples with their respective alpha effective equivalent dose is obtained in Ero (0.0604mSv), Govani (0.0152mSv), Dewluk (0.0254mSv), Manko (0.0351mSv), Jeffco (0.0280mSv) and Tahur (0.0333mSv) respectively. Figure 7 shows that the mean gross alpha activity increase with increasing distance from the source except at location B, while the effective equivalent dose increases at location A then start decreasing as the distance increased and remains invariant at location E. This pattern of fluctuation indicates that there is a possible contamination of the environment due to sharp practices and poor hygiene of locations. The computed mean gross alpha activity concentrations and mean effective equivalent dose of alpha emitters of the water samples were found to be (0.0338 ± 0.0001) mBqL⁻¹ and 0.0145mSv respectively. The obtained values were much lower than limit value of 500mBqL⁻¹ and reference level of 0.1mSv recommended by WHO, EC and ICRP [9, 17, 34, 37-40]. Figures (8 – 9) illustrate the mean gross alpha activity and effective equivalent percentage distribution for all samples locations.



Fig8. Percentage distribution of mean gross alpha activity concentrations for all samples locations.



Fig9. Percentage distribution of mean alpha effective equivalent dose for all location.

It can be established from the above figures that 30% (Ero), 22% (Giovani), 20% (Dewluk), 25% (Manko), 21% (Angvel) and 21% (Tahur) of the samples for gross alpha activity concentrations were slightly high, this might be due to the different geological origin and chemical composition of the portable drinking water sources. There is need for drinking water manufacturers in this location to ensure that the water were produced in an environment free from contamination by human, animal, agricultural and chemical waste. Figures 10 - 15 illustrates the variation of gross alpha activity concentrations and figure 16 shows the mean gross alpha activity and mean alpha effective equivalent dose.





Fig13. Variation of gross alpha activity concentrations in for location D.



Fig14. Variation of gross alpha activity concentrations in location E.



Fig15. Variation of gross alpha activity concentrations in location F.



Fig16. Mean gross alpha activity and mean alpha effective equivalent dose sample locations.

It is clearly shows in the above figures that forty-six out of the fifty water samples obtained from locations A, B, C, E and F (see figures 10, 11, 12, 14, 15) and eight out of the ten samples in D (see figure 13) satisfy the recommended value which indicated that they are safe for human consumption since it is in accordance with the maximum acceptable gross alpha activity concentrations, while we observed a slight enhancement of gross alpha activity in the remaining six samples. The average gross alpha activity concentrations was greater at location B and least at location A. The effective equivalent dose was higher at location A (0.0197mSv) and D (0.0183mSv), and least at location B (0.0068mSv) as indicated in figure 16. This implies that there is a slight trace of risk associated with drinking water samples from location A and D, than at location B, C, E and F. The major contributors to the slight risk laden alpha concentrations from this water samples may be traced back to their source, or probably from a very deep borehole water source due to the fact that depth of the underground water is directly proportional to activity concentrations. On the other hand, the water sources may be originated from

mountainous environment or directly from beneath the rock. The atmospheric fall out sometimes contributed immensely to the water concentrations measured. This normally occurred as a result of nuclear disaster like disposal of radioactive waste material into the river. Radionuclide particles suspended in air could be deposited on the soil surface which later dissolved and the level of contamination therefore depends on the surface area of the water. Comparison of the results obtained in drinking water with the published data from similar investigations in other parts of the world shows that the results in this study are generally lower than those values obtained from different countries [1, 7, 10, 14-15, 21-22, 26, 31-33]. The obtained results distinctly shows that in all locations the mean gross alpha activity concentrations and alpha annual effective equivalent dose does not exceeded the recommended reference dose level (0.1mSv) of drinking water, thus making the water samples in the study area safe for human consumption and does not constitute any radiological threat to the people consuming them.

4. CONCLUSIONS

The need for quality portable drinking water, packaged under very stringent hygienic condition cannot be over emphasized due to the costly effect of water-borne diseases such as dysentery, diarrhoea, typhoid fever among others. It can be established from the obtained results that the gross alpha activity varies from (0.0025 ± 0.0001) mBqL⁻¹ to (0.0969 ± 0.0001) mBqL⁻¹, and the computed mean gross alpha activity concentrations and mean effective equivalent dose of alpha emitters of the water samples were found to be (0.0338 ± 0.0001) mBqL⁻¹ and 0.0145mSv respectively, which were much lower than the world average. This revealed that the gross alpha activity concentrations in portable drinking sachets water in the region of Abuja (Nigeria) poses no significant radiological health concerns and therefore can be generally acceptable for consumption. The data obtained can provide vital information for the consumers and the authorities regarding the preliminary dose exposure risk due to portable drinking sachets water.

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