Determination of gross Alpha, Beta radioactivity in sachet drinking water

Gado A.A.[1,2]*, Muthukumar M.[2], Gwani M.[1], Jonathan L.A.[1], Kazuga S.[1], Umar A.B.[1], Jafar A.B.[3] and Anbazhagi M.[2]
[1]Department of Physics, Kebbi State University of Science and Technology, Aliero, Nigeria
[2]Department of Environmental Science, School of Earth Science Systems, Central University of Kerala, Kasaragod, India
[3]Department of Mathematics, Kebbi State University of Science and Technology, Aliero, Nigeria
gadoabubakar@cukerala.ac.in

Abstract

Radioactivity found in underground water has become a serious issue of health concern across the globe in recent years. In this study gross alpha and beta radioactivity in package drinking water were analysed using Eurisys measures IN20 eight-channels-gas–filled proportional counter. Seven samples were collected from different package drinking water producing companies across Aliero metropolis, Kebbi state, North-western Nigeria using alpha and beta sampling procedure. The results obtained shows that, the range of alpha activity concentration in the analysed samples is 0.007 to 0.080Bq/L with a geometric mean of 0.041Bq/L which gives average yearly effective dose equivalent for gross alpha 0.011mSv/yr. Further results indicate that, the range of beta activity concentration in the samples ranges from 0.120 to 0.98Bq/L with a geometric mean of 2.66Bq/L giving average yearly effective dose equivalent for gross beta of 0.695mSv/yr. The findings of the research reveals that, the beta activity concentration in most of the samples is above the minimum level set by World Health Organization (WHO) and United States Environmental Protection Agency (USEPA) hence the water are not safe for drinking and other domestic activities.

Keywords: Gross alpha, gross beta, radioactivity, drinking water, gas-flow proportional counter, aliero.

Introduction

The occurrence of radioactivity in drinking water is among the major issues of global concern in this century especially in under-developed countries, where the agencies in charge are not giving much attention to the level of radioactive elements as well as the quality of drinking water. The usage of plastic package water popularly known as pure-water in Nigeria, has increased significantly in recent years, however, most of this package water source is from ground-water mainly boreholes because of the existing law about the use of surface water for private businesses[1].

Most of the sources of water supply in Nigeria are from upland surface water or boreholes and hand dug wells passing through rocks and abundant deposit of crystalline rocks, which are known for higher uranium and thorium decay series than average rocks. The earth's crust contains naturally occurring radioactive materials that increase with depth and of most concern are the uranium series, thorium series, and their progeny (radon and thoron)[2]. Drinking water most especially package water, which are mainly from boreholes, will most likely contain a high concentration of radioactive elements[3]. However, to quantify the level of radioactivity in drinking water, investigation were carried by various researchers to determine the level of radioactivity in drinking water due to the importance of water to human life and the increased consumption of package drinking water[4,5].

Radioactivity in water bodies is mainly caused as a result of uranium and thorium nuclear activities, which are natural processes and can change with location due to cosmic rays in the atmosphere[6]. These rays produce cosmogenic radio nuclides which diffuse to lower atmosphere where they mixed with water and living matter. Similarly, rainwater flowing over hills, could influence the radioactivity level of drinking water if flows into wells or sinks to the ground[7]. This study is aimed to evaluate gross alpha and beta radioactivity of package drinking water in a town of Kebbi state, North-western Nigeria as a useful guide for the improvement of the health and well being of the Nigerians at the study site.

Natural Alpha and Beta: Natural alpha and beta are found in air, rocks, soil, water and oceans, in our building materials and even found in humans as by-products of our environment[8]. Most food has some small amount of alpha and beta in it. The common radio nuclides in food are Potassium–40 (^{40}K) Radium–226 (^{226}Ra) and Uranium-238(^{238}U) and the associated progeny. Ingestion of these radioactive elements by humans is toxic and dangerous especially the radioactive elements found in the common foods materials such as potassium-40 (^{40}K), radium-226 (^{226}Ra)[9].

Elements that are naturally radioactive in nature include ^{238}U, ^{232}Th and ^{40}K, Carbon, as well as Radon and Radium[10]. Uranium is the first element in a long series of decay that produces Radium and Radon. As such, Uranium is referred to as

the parent element while Radium and Radon are refers to as daughters. Table-1 presents the natural alpha and beta level in some common food materials. When alpha-emitting radioisotopes are ingested, they are 20 times more effective in damaging cells in comparison to gamma rays and X-rays. Among the highly poisonous alpha-emitters are all isotopes of radium, radon and polonium mainly due to the amount of decay that occur in their short half-life[11].

Table-1: Natural Alpha and Beta in some food materials[12].

Food	$^{\alpha}$ PCi/kg	$^{\beta}$PCi/kg
Banana	3520	1
Brazil Nuts	5600	1000-7000
Carrot	3400	0.6-2.0
White Potatoes	3400	1-2.5
Beer	390	-------
Red Meat	3000	0.5
Lima Bean raw	4640	2-5
Drinking water	--------	0-0.17

Materials and methods

Rivers, rainwater flowing over hills, rocks, agricultural waste and animal waste are the main sources of alpha and beta. Aleiro town is characterized with most of these sources and hence the underground water in Aliero town may likely contain highalpha and beta activity.

Study site: Aliero town is the headquarters of Aliero local government in Kebbi state, North-western Nigeria. Agriculture is the main occupation of the people in Aliero of which irrigational farming, animal-rearing being the dominant activities. The town is located along the latitude $12^{0}16'42''$N and on longitude $4^{o}7'6''$E of the equator. Aliero town has a total area of 412 square kilometers with estimated population of one hundred and twenty-five thousand, seven hundred and eighty three people (125,783) based on the 2006 census data[13].

The town enjoy tropical climate characterized by wet and dry seasons. The study site is located in the south east of the state, bordered in the North-East by Gwandu Local government area, in the South-West by Jega Local Government, and in the North-West by Birnin Kebbi Local Government area[13].

Sample collection: Seven different portable package water producing companies labelled 1, 2, 3, 4, 5, 6 and 7 with codes ALR001, ALR002, ALR003, ALR004, ALR005, ALR006 and ALR007 were selected for this study. The volume of each package is 500ml per package collected from these companies located at different areas (refer to Table-2 for the site information).

The sampling procedure adopted for the research is as follows: i. Three package water from each sampling point (package water production factory) are collected and one of the package was used to thoroughly wash the sample container to minimize contamination. The remaining two packages were mixed together to give a true representation of the water samples from each sampling point. ii. Conductivity and pH of the water were measured at the site itself. iii. About 10ml of dilute nitric acid were added to the samples (for preservation) immediately after collection to reduce the pH and to minimize precipitation and the absorption on the walls of the container. iv. Only 0.9% space of the container capacity was left for air and thermal expansion, v. The sample containers were labelled and tightly covered with container cover and kept in the laboratory until analyses.
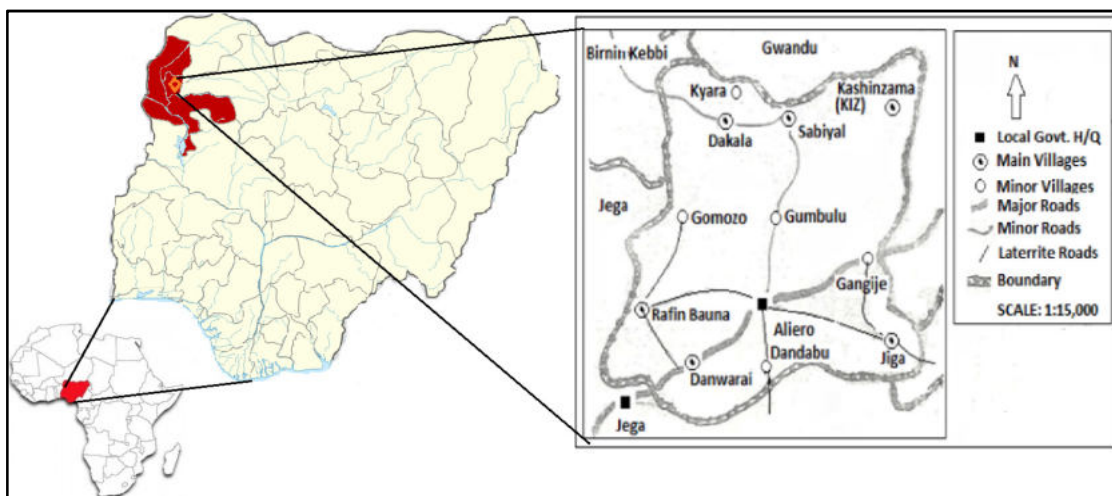

Figure-1: Map of Aliero local government[14].

Sample preparation: To adhere to the ISO-Standard for radioactivity counting, the collected samples were evaporated using hot plates without stirring at moderate heat in an open beaker of 600ml. On average, it took about 24 hours to complete the evaporation of one liter of the each sample. In the evaporation process, when the level of the sample in the beaker reached 50ml, it was then transferred into a Petri dish and placed under infrared light source to completely dry the residue. The samples were then allowed to cool before weighing. The weight of the residue was obtained by subtracting the weight of empty Petri-dish from the weight of Petri-dish plus sample residue. An empty planchette was weighed after which about 0.077g of the residue was transferred to the planchette. 0.077g is arrived from the fact that, ISO-Standard requires about 0.1Amg of residue should be placed on the planchette for counting, where A is the area of the planchette in mm^2. The planchette plus residue was then weighed. A few drops of vinyl acetate is added on the sample to make them stick to the planchette to prevent scattering of the residue during counting. The sample was then kept in a desiccator until ready for counting.

Samples analysis: Determination of the alpha and beta activity in sample package water was performed at the Centre for Energy Research and Training Zaria (CERT) Kaduna State Nigeria. ISO 9696 (Alpha Analysis) and ISO 9697 (Beta Analysis) are the method of analysis employed in this research using _Eurisys Measures IN20_ instrument[15]. The gross alpha and beta counting equipment used is a low background multiple (eight) channels gas flow proportional type counter with $450\mu g/cm^2$ window of 60 mm diameter each. The counting gas in the instrument is Argon-Methane at the ratio of 90% and 10% respectively. The inside dimensions are $480 \times 105 \times 280mm^3$ and a thick lead shielding of 10cm with a pair of oversized guard counter in the instrument helping in reducing the background noise.

The system is connected to a micro-processor IN-SYST, a spread sheet programme, Quattro-pro and a graphic programme Multiplan that are used for signal processing. The alpha and beta counting system has eight gas circulation proportional type detectors, each measuring in one or a pair of electronic channels. For each counter, there are two coincident counter system located above and separated by a thick stainless steel lining to maximize counter sensitivity by stopping low background noise.

Radionuclide when ingested may reached the intestines where it can be absorbed in to the body fluids there by having tendency of reaching all the delicate internal organs. Due to the effects of radionuclide to our body, it is necessary to quantify the effective dose of alpha and beta due to intake of ground water to ascertain the contributed doses by the major alpha and beta emitters. In this study, the effective dose over one year was calculated using,

$$DRw = Aw \times IRw \times IDw \tag{1}$$

Where: DRw=effective dose equivalent (mSv/yr), Aw=Activity Bq/L, IRw=Intake of water for a person in a year (730 L/year), IDw=Ingestive equivalent dose factor, IDw=3.58x10-7Sv/yr/10- = 3.58x10-4 mSv/yr.

The calculation is based on the World Health Organization (WHO) and United State Environmental Protection Agency (USEPA) estimate that, adult consume 2L/day thus resulting to the annual consumption rate of 730 L/year.

Results and discussion

The results obtained from this study were based on the characterization of the detector and the measurement of radioactivity (gross alpha and beta activity) in the seven (7) samples of the package water collected from different package water producing factories within the study site.
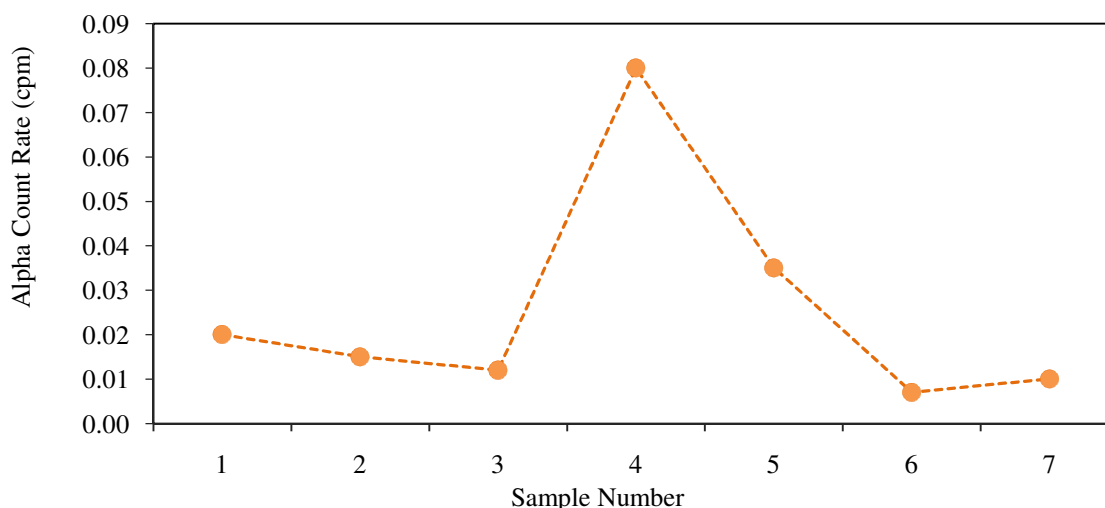

Figure-2: Alpha Background Count Rate of the samples.

Alpha count rate: Figure-2 presents the alpha background counts rate obtained in the alpha only mode. In alpha only mode, the range of the background count rate observed is from 0.007 to 0.080 with an arithmetic mean of 0.026cpm. Sample 4 shows higher alpha background count rate of 0.08 although it is within the accepted limit for drinking water set by World Health Organization (WHO) of 0.5cpm. The low alpha background count rate recorded at the study site is due to the higher limestone of the area. Studies from the literature revealed that, natural radioactivity in limestone rich areas, are typically at levels of natural background which in most cases falls below the standard vales[22].

The background count rates in the only beta mode is shown in Figure-3. In the beta only mode, the range of the background count rate observed is from 0.120 to 0.98 with an arithmetic mean of 0.58cpm. The background count observed on alpha only mode and beta only mode of the samples analysed, all falls within the accepted limit for drinking water and water for domestic uses set by World Health Organization (WHO) i.e alpha =0.1 and beta=1.0[16].

Table-2: Conductivity and pH Values for all the Collected Samples.

Sample code	Brand	pH	Conductivity(μS/cm)
ALR001	Sa'a	6.9	1.74×10^2
ALR002	Arziki	6.9	1.33×10^2
ALR003	Namaki	6.8	1.33×10^2
ALR004	Zamfarawa	7.2	0.88×10^2
ALR005	Agaji	7.3	0.63×10^2
ALR006	Awadata	7.2	1.23×10^2
ALR007	Atai	6.9	1.88×10^2

The conductivity and pH values for all the collected samples for the research are presented in Table-2, the pH values of the samples analysed shows that, the samples have pH values within the standard values accepted for drinking water. The variation of the pH and conductivity observed, could be related to the source of the water and the pre-treatment processes it undergoes before packaging by the different package water producing companies. As observed during the sampling, different water treatment steps are followed by different sachet water producing companies before final packaging, and this might affect several water quality parameters.

Gross Alpha and Beta Radioactivity in Samples: The result of the alpha and beta activity concentration on the analysed samples is presented in Figure-4. The results of the activity concentration confirm that, most of the samples have low alpha

activity. Higher alpha activity concentration is observed in sample 6, and sample 2 is having the lowest alpha activity in comparison to all the samples analysed. However in terms of beta activity of the analysed samples, higher beta activity is observed on most of the samples. The results of beta activity in the samples shows that, most of the analysed samples has higher beta activity concentration beyond the accepted level for drinking water set by United States Environmental Protection Agency (USEPA) and World Health Organization (WHO). The highest beta activity concentration is observed in ascending order on sample 7, 4, 1, 2 respectively, while the lowest beta activity is recorded on sample 5. The highest beta activity concentration observed on sample 7 (ALR007 Atai) might be due to the higher animal waste observed around the site, as the water company is deep inside the town in comparison to sample 5 (ALR007 Agaji), which is located at the GRA area of the study site.

Determination of Effective Dose Equivalent: Using equation (1), the effective dose due to intake of water is calculated, from the estimation of World Health Organization and United State Environmental Protection Agency that, adult consume 2L/day thus resulting to the annual consumption rate of 730L/year.

DR_w (Gross Alpha)=$0.041 \times 730 \times 3.58 \times 10^{-4}$mSv/yr=$107.149 \times 10^{-4}$
DR_w (Gross Alpha)=0.011mSv/yr
DR_w (Gross beta)=$2.66 \times 730 \times 3.58 \times 10^{-4}$mSv/yr=$6951.644 \times 10^{-4}$
DR_w (Gross beta)=0.695mSv/yr

Based on the calculations, it can be observe that, the values are beyond the worldwide accepted per capita effective dose of 0.4 mSv/yr of one year intake of drinkable water. Thus there could be radiation related hazards for consuming the sachet water in Aliero as well as the water for domestic use. Although the range of the accepted value varies depending on the level of health care of the country[16,22,23].

Discussion of the Results and Comparison with the Standard and Studies in the Literature: Although, this studies can be considered as first of its kind in determine the gross alpha and beta activity in drinking water at the study site, however, validation and comparison is require with the studies conducted across the country and with the standards. For the studies reported in this section, all the analyses of the samples were carried out with the same sampling technique except the work of Kolo[17]. The comparison reveled that, there is high alpha and beta activity concentration in the water samples from Aliero Metropolisin comparison with samples from Zaria, Kano, Jos, Minna and Sokoto. However, the ratio of samples collected in the study site is low compared to the values obtained from the studies traced in the literatures as reported in Table-4. Evidence from Table-4 shows that, there is high alpha and beta activity concentration in the analysed water samples compared to the studies traced in the literature. The values of beta activity observed in this study are beyond the accepted standard level in the water for drinking and domestic usage set by USEPA and WHO.
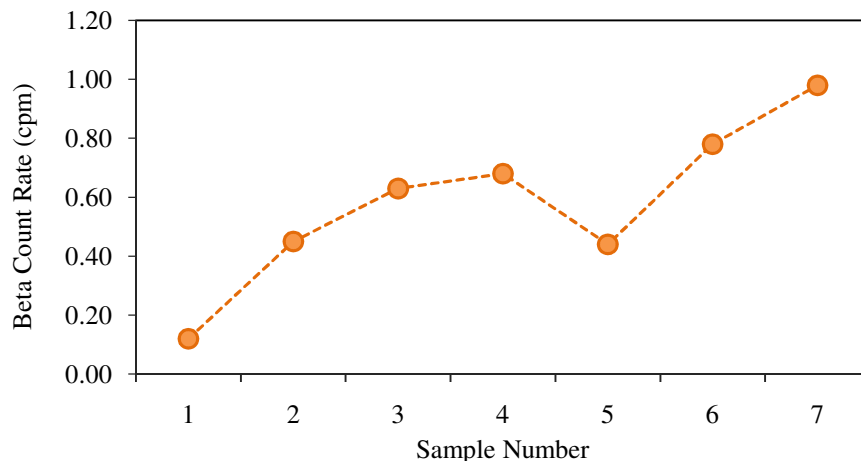
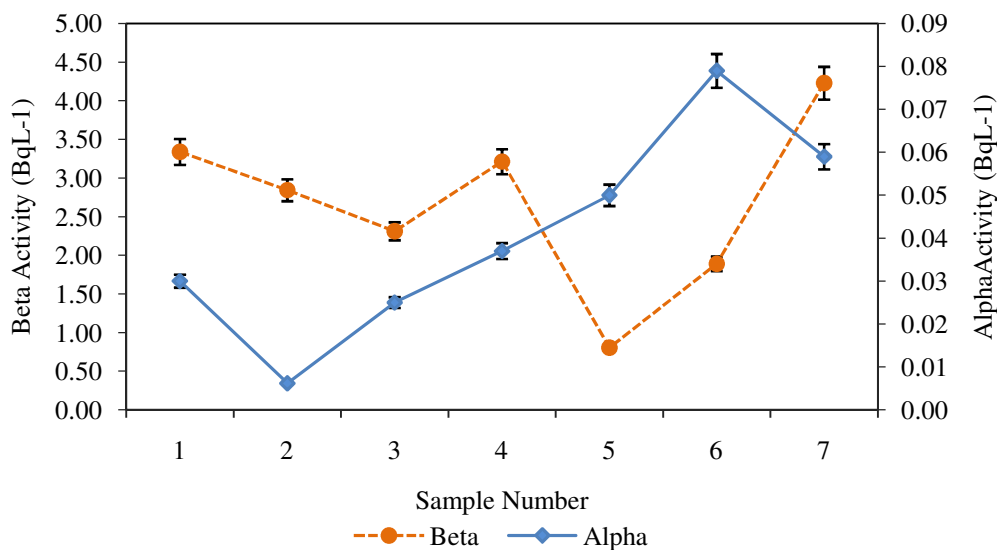**Figure-3:** Beta Background Count Rate of the samples.

Figure-4: Alpha and Beta activity concentration in the analysed samples; the whiskers displays error bars.

Table-4: Comparison between the sample collected and those in literature.

Location	Measured Activity (Bq/L)						Source
	Alpha			Beta			
	Min	Max	No. of sample	Min	Max	No. of sample	
Zaria	5.6×10^{-4}	0.043	32	3.6×10^{-3}	0.62	32	18
Kano	0.002	0.010	40	1×10^{-5}	1.8×10^{-4}	40	19
Jos	0.100	6.050	48	0.250	9.640	48	20
Minna	0.001	0.024	20	0.072	0.558	20	17
Sokoto	0.010	6.000	40	0.350	49.850	40	2
Aliero	0.0062	0.0790	7	0.804	4.230	7	Study Site
USEPA		0.550			1.850		21
WHO		0.500			1.000		22

Conclusion

In this study, the gross alpha and beta activities in 7 package drinking water (pure water) in Aliero metropolis, Kebbi state North western Nigeria were measured and the effective doses were calculated. Based on the outcome of the research, the range of alpha activity was found to be from 0.007 to 0.080 Bq/L with geometric mean of 0.041 ± 0.026 Bq/L that gives the average yearly effective dose equivalent of 0.011 mSv/yr. The average yearly effective dose observed for gross beta is 0.695 mSv/yr with geometric mean of beta activity concentration of 2.66 ± 0.58 Bq/L. Based on the findings of the research, it can be concluded that, some package water from Aliero town meet the USEPA and WHO standards while quite a reasonable number of the samples package water does not meet the USEPA and WHO standards and therefore there is a need for further radionuclide screening of the package drinking water hawked in Aliero metropolis to avoid radiation related hazards to the general population. It is hoped that, the results from this work would send information to the concern authorities for necessary action.

Acknowledgement

References

1. Igwilo I.O., Afonne O.J., Obi E.A. and Orisakwe O.E. (2006). Heavy metal hazards of sachet water in Nigeria. *Toxicology Letters*, 164(20), 12-14.

2. Saidu A., Baba-Kutigi A.N. and Buda S. (2012). Determination of Gross Alpha Radioactivity in Underground Water in Usmanu Danfodiyo University Permanent Site, Sokoto. *International Journal of Science and Advance Technology*, 1, 1-4.

3. Baeza A., Salas A., Guillén J., Muñoz-Serrano A., Ontalba-Salamanca M.Á. and Jiménez-Ramos M.C. (2017). Removal naturally occurring radionuclides from drinking water using a filter specifically designed for Drinking Water Treatment Plants. *Chemosphere*, 167, 107-113.

4. Batt A.L., Furlong E.T., Mash H.E., Glassmeyer S.T. and Kolpin D.W. (2017). The importance of quality control in validating concentrations of contaminants of emerging concern in source and treated drinking water samples. *Science of the Total Environment*, 579, 1618-1628.

5. Shygonskyj Valerij and Shygonska Natalia (2016). Assessment of drinking water quality as a factor affecting human health in Zhytomyr Region. *Kontakt,* 18(2), 120-126.

6. Chmielewska I., Chałupnik S. and Bonczyk M. (2014). Natural radioactivity in drinking underground waters in Upper Silesia and solid wastes produced during treatment. *Applied Radiation and Isotopes*, 93, 96-100.

7. Vasile M., Loots H., Jacobs K., Verheyen L., Sneyers L., Verrezen F. and Bruggeman M. (2016). Determination of 210Pb, 210Po, 226Ra, 228Ra and uranium isotopes in drinking water in order to comply with the requirements of the EU 'Drinking Water Directive'. *Applied Radiation and Isotopes*, 109, 465-469.

8. Ogundare F.O. and Adekoya O.I. (2015). Gross alpha and beta radioactivity in surface soil and drinkable water around a steel processing facility. *Journal of Radiation Research and Applied Sciences*, 8(3), 411-417.

9. Brandhoff P.N., Van Bourgondiën M.J., Onstenk C.G.M., Van Avezathe A.V. and Peters R.J.B. (2016). Operation and performance of a National Monitoring Network for Radioactivity in Food. *Food control*, 64, 87-97.

10. Semat H. and Albright J.R. (1978). An Introduction to Atomic Nuclear Physics. Chapman and Hall, London.

11. Senthilkumar G., Raghu Y., Sivakumar S., Chandrasekaran A., Prem Anand D. and Ravisankar R. (2014). Natural radioactivity measurement and evaluation of radiological hazards in some commercial flooring materials used in Thiruvannamalai, Tamilnadu, India. *J. Radiat Res.Appl. Sci.* 7, 116-122.

12. ISU. (2017). Idaho State University, Radioactivity in Nature. *Radiation Information Networks*, *ISU Health Physics Radinf*, Pocatello, Idaho USA

13. Illo A.I., Kaka Y., Hassan U., Umar S. and Bamidele A.A. (2016). Marketing Of Onion in Aliero Central Market, Aliero Local Government Area of Kebbi State. *J.IOSR of Hum.Soc. Sci. (IOSR-JHSS),* 21(1), 42-49.

14. Salah A. (2010). Map of Aliero Local Government. *Department of Geography,* Usmanu Danfodiyo University Sokoto. UDUS Sokoto Nigeria.

15. ORTEC (2016). Alpha/Beta Counting Automatic Systems. South Illinois Ave, Oak Ridge USA.

16. WHO (2008). World Health Organization, Guidelines for Drinking-water Quality Third Edition Incorporating the First and Second Addenda. Geneva.

17. Kolo M.T., Kutigi A.N., Olarinoye I.O. and Sharifat I. (2009). Survey of Gross Alpha and Gross Beta Radioactivity in package Water Hawked in Minna, Niger State. *J Sci Educ Technol.,* 2(1), 404-413.

18. Onoja R.A. (2004). Survey of Gross Alpha and Beta Radioactivity in Wells from Zaria. A.B.U. Zaria Kaduna, Nigeria.

19. Tajudeen H.V. (2006). Survey of Radioactivity in Wells and Bore holes from Gwammaja Area of Kano City, Nigeria. A.B.U., Zaria Kaduna Nigeria.

20. Habila Nuhu (2008). Survey of Gross Alpha and Beta Radioactivity in Jos City, University of Jos, Jos Plateau State, Nigeria.

21. USEPA-United State Environmental Protection Agency (1980). National Primary Drinking Water Regulations Agency Report EPa-570/9-76-003. *Federal Register* 45(231), 79347-79357.

22. Koz1owska B., Morelli D., Walencik A., Dorda J., Altamore I., Chieffalo V. and Giammanco S. (2009). Radioactivity in waters of Mt. Etna (Italy). *Radiat Meas*, 44, 384-389.

23. Najam L.A., Al-Jomaily F.M. and Al-Farha E.M. (2011). Natural radioactivity levels of limestone rocks in northern Iraq using gamma spectroscopy and nuclear track detector. *Journal of radioanalytical and nuclear chemistry*, 289(3), 709-715.