

Gross Alpha and Gross Beta Activity Concentrations and Committed Effective Dose due to Intake of water in Solid Mineral Producing Areas of Enugu State, Nigeria.

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Abstract

Mineral radioactivity is due to alpha, beta and gamma radiation from the unstable isotope in the composition. Gross alpha and beta activity concentration in ground water from solid mineral producing areas of Enugu State has been determined using an IN-20 model gas-flow proportional counter. The measured activity concentration of gross alpha and beta were compared with the reported data from other countries but they exceeded the recommended value of 0.1 and 1.0 Bq⁻¹ respectively. The committed effective dose due to one year intake of drinking water for infants and adults were calculated from the activity concentration in order to estimate the exposure risk arising from intake of water. The recommended reference dose level (RDL) of the committed effective dose of 0.1 mSv was however exceeded both for infant and adult in all the communities. This study has shown that solid mineral deposit in ten communities of Enugu State has radiologically contaminated the ground water which might pose radiation related health risk to the populace. Therefore incorporation of reverse osmosis technology in boreholes will help reduce the radiological burden of the public in the area.

Keywords: Gross alpha, beta, committed effective dose, gas-flow proportional counter, Enugu

INTRODUCTION

Portable safe drinking water has been a global issue for the past decade. Radionuclide in water is primarily a problem for water supplies which extracts water from drilled holes in rocks or from springs flowing through areas endowed with solid mineral

deposits and crystalline rocks which have higher uranium and thorium decay series than the average bedrock (Faanu et al., 2011). Natural radionuclides present in water beyond the recommended level are considered to have potential risks to man from their consumption at a regular rate. This is because of their long environmental half-life, high radiotoxicity and high affinity to biota. Natural radionuclides are classified as radiological toxic agents (Fasae et al., 2015). The ingested radionuclides accumulate in the human body primarily through the intake of food and water. The contribution of drinking water to the total dose intake becomes important when the drinking water sources are from ground water since the radium concentration varies widely. Most of the drinking water producing industries utilizes ground water.

Ground water deposits have varying range of quality and chemistry. The quality depends on the mineralogy and reactivity of the drift material and also the degree of equilibrium that has been attained between water and rock (Fassae et al., 2015). Water resources are contaminated by these radioactive isotopes from naturally decaying series of ^{238}U , ^{232}Th and ^{40}K . Human activities such as mining, milling and processing of uranium ores and mineral sands, smelting of metalliferous ores, manufacturing of fertilizers, drilling of solid minerals, processing and burning of fossil fuels have raised the concentrations of naturally occurring radioactive materials in the environment (Ogundare and Adekoya, 2015).

Enhanced levels of radionuclide in the soil as well as ground water are associated with low-grade radionuclides deposits. Soil acts as a source of transfer of radionuclides through the food chain depending on their chemical properties and the uptake process by the roots to plants and animals (Jabbar et al., 2010, Biswas et al., 2015); hence it is the basic indicator of the radiological status of the environment. These radionuclides take part in several biogeochemical processes that determine their mobility and availability for biological uptake. Water contains a number of both alpha emitters (such as ^{238}U , ^{226}Ra , and ^{210}Po) and beta emitters (such as ^{40}K , ^{228}Ra and ^{210}Pb). Natural isotopes as ^{40}K and the nuclides from the ^{238}U and ^{232}Th series are the greatest source of internal and external exposure of human beings. Among the radionuclide of terrestrial origin, ^{40}K and ^{238}U and ^{232}Th series enter the human body basically through food and water ingestion. The presence of radionuclides in water poses a number of health problems especially when these radionuclides are deposited in the human body via drinking water. Dissolved radionuclide in body emits particles (alpha and beta) and photons (gamma) which gradually interacts with living tissues (Gruber et al., 2009, Ogundare and Adekoya, 2015).

World Health Organization (WHO) guidelines for drinking water quality suggest performing an indirect evaluation of committed effective dose by measuring gross alpha and gross beta radioactivity in drinking water and checking the level of compliance to reference dose level of 0.1 and 1.0 Bq l^{-1} for gross alpha and gross beta activity respectively (WHO, 2004). Mineral radioactivity is due to alpha, beta and gamma radiation from unstable isotope in the composition. Gross alpha and gross beta depends on many factors such as the time intervals between sample collection, preparation and analysis, the radionuclide used as the calibration standards, the counting efficiency and so on. The values of gross alpha radioactivity originating from

these alpha emitters in ground water samples depends on the geological characteristics of the area, content of mineral deposits and the type of human activities in the area.

In environmental monitoring, much attention has been given to gamma emitter's detection and quantification even where it is possible to have alpha and beta emitters (Lu et al., 2012; Mehade et al., 2014). Though gamma rays have the highest penetrating power when compared to alpha and beta particles, effect of alpha and beta particles inside the body are far more detrimental to human health because of their ionizing power.

The aim of this study is to evaluate the gross alpha and gross beta radioactivity in ground water from solid mineral producing areas of Enugu state and also determine committed effective dose of the general public. The result will serve as a radiological baseline data for the area.

MATERIALS AND METHODS

Sample Collection and Preparation :

Thirty water samples used were collected from rivers/streams in close proximity to the mineral sites and from the community public water supply sources (rivers, taps and wells). The samples were collected in 2-litre plastic containers with about 1% air space left for thermal expansion (Agbalagba et al., 2013).

To minimize contamination, the containers were first rinsed three times with sample water before use. Well water samples were collected manually in the early hours of the day from community wells of varying depths. Tap water samples were collected at laminar flow rate after first turn on at full capacity for several minutes to purge the plumbing system of any water which might contaminate sample, to reduce radon loss (Tchokossa, *et al.*, 1999).

The water samples were immediately acidified with 20ml \pm 1ml of nitric acid per litre of sample collected to minimize the absorption of radioactivity into the walls of the containers [ISO, 9697 & 9698: 1992a]. The samples were then tightly covered and kept in the laboratory. These samples were analyzed for gross alpha and beta activity using an IN-20 model gas-flow proportional counter at the Centre for Energy Research and training, Ahmadu Bello University, Zaria, Nigeria.

Each sample was counted three times and the mean used in computing the activity. The operational modes used for the counting were the α -only mode for the alpha counting and the β ($+\alpha$) mode for the beta counting. The count rate of each sample was automatically processed by the computer using the equation (IN-20 Model Technical Manual, 1991);

For purposes of analysis the samples were slowly evaporated without boiling, down to a volume of 50ml at a furnace temperature of 60⁰ C. The residue was then transferred to a stainless-steel planchet, dried and allowed to equilibrate with ambient temperature and weighed. The counting time was 30000s while the screening technique is in agreement with ISO, 9697 and 9698: 1992b guidelines.

The calculated volume of each of the samples was measured into a beaker and evaporated on a hot plate to about 45 to 50ml which was transferred into a weighted petri-dish which was cleaned with acetone to avoid cross contamination and then evaporated

to dryness using infra-red light. 77mg of the residues was washed with distilled water and transferred into counting planchette with the help of a rubber policeman. The sample was evenly distributed using a syringe and some drops of vinyl acetate were added to the sample to act as a binder. It was kept in a desicator until it was ready for counting (about 28 days).

Counting Equipment :

The gas-flow-proportional counter (Eurisys Measure- IN20) eight channel counter at the Material Laboratory, Centre for Energy Research and Training (CERT), Ahmadu Bello University (ABU), Zaria was used for the counting. Each counter channel has a window thickness of $450 \mu\text{gcm}^{-3}$ and a diameter of 60 mm. The chambers are covered with lead whose thickness can be varied. The detectors are operated within the radiation environment of $< 01 \mu\text{radh}^{-1}$. The system is connected to a microprocessor loaded with a spread sheet programme (Quattro-Pro) and graphic programme (Multiplan). The system can be operated at a bias voltage ($\sim 1100\text{V}$ with P10 gas: argonmethane of 10%) where only alpha particles are detected, referred to as 'alpha only' mode.

Detector Calibration :

For the alpha activity measurement the standard used is ^{239}Pu (α -source) with a half life of 24110 years and with activity ranging from 133.29 to 185.51Bq for the eight channels at 2π -steradians. The radionuclide impurity in each of them varied from 0.74 - 0.82% (Fasae, 2013). For beta activity measurement, the standard used are ^{90}Sr (β -source) of diameter 38mm with a half life of 28years and an active film of 12mg.cm^{-3} thick. For the eight channels, eight sources of activities varied from 105.1 Bq to 117.7 Bq at 4π -steradians. The radionuclide impurity in each of them was less than 0.1%. These standards were certified by CERCA LEA Laboratories in France with certification numbers CT 001/1285/001920-1927 and CT 1271/00/1778- 1783, respectively.

The alpha and beta specific activities were calculated using the following expression (Akpa et al., 2004).

Specific Activity (α/β) Bq^{-1}

$$= \frac{\text{counting Rate } (\alpha, \beta) - \text{Backgroundcounting rate } (\alpha, \beta)}{\text{Sample Efficiency} \times \text{channel efficiency} \times \text{weight of the sample}} \quad \text{---- (1)}$$

Estimation of committed effective dose

Radionuclide may reach the gastrointestinal tract directly by ingestion or indirectly by transfer from the respiratory tract. From small intestine (S1) the radionuclide can be absorbed to the body fluids. The annual alpha and beta effective dose due to intake of ground water was determine by averaging the individual annual committed effective doses contributed by the major alpha and beta emitters in the ^{238}U and ^{232}Th series of

the naturally occurring radionuclide (Ogundare and Adekoya, 2015). In this work, the effective dose over one year was calculated using the following relation (IAEA , 2003).

$$E_{\text{avg}} (\alpha/\beta) = \sum A_{i(\alpha/\beta)} \times \text{DCF}_{i(\alpha/\beta)} \times 730 \text{ (for Adult)} \quad \text{----- (2)}$$

$$E_{\text{avg}} (\alpha/\beta) = \sum A_{i(\alpha/\beta)} \times \text{DCF}_{i(\alpha/\beta)} \times 183 \text{ (for Infant)} \quad \text{----- (3)}$$

Where $E_{\text{avg}} (\alpha/\beta)$ is the average gross annual alpha or beta committed effective dose in drinkable water, $A_{i(\alpha/\beta)}$ is the gross alpha or beta activity concentration of individual radionuclides present in water samples and $\text{DCF}_{i(\alpha/\beta)}$ is the dose conversion factor in Sv/Bq for ingestion of the individual radionuclide. From EPA (2005) report, it was assumed that adult consume a minimum of 2 liters of water per day resulting in annual consumption rate of 730 litres per year while infant consumes half litre of water per a day (1/2 l/d) resulting in annual intake rate of 183 litres per year. Damlah et al., (2006) stated that more than 50% of the annual dose from intake of water corresponds to radium (gross alpha radium). This was assumed in this work since the component radionuclides in the gross α and gross β could not be determined due to the limited functions of proportional Counter used.

According to Gortir et al.,(2011), the major contributors to gross α activities is ^{226}Ra while the major contributors for β activities are ^{210}Pb and ^{226}Ra . For calculations, the dose conversion factors of 2.8×10^{-4} Sv/Bq, for ^{226}Ra and 6.9×10^{-4} Sv/ Bq for both ^{210}Po and ^{226}Ra published by the (WHO, 2004) were used.

RESULTS AND DISCUSSION

Table 1: Gross Alpha and Beta Activity Concentration in Ground water from Solid mineral Producing Areas of Enugu state.

S/N	Code Name	Location	Mineral Found	Gross Alpha Activity (Bq/l)	Gross Beta Activity Bq/l
1	OG	Ogugu	Limestone	0.015±0.006	2.153±1.01
2	BN	Bunker	Coal	0.032±0.013	6.515±7.1
3	EE	Enugu-Ekulu	Clay	0.288±0.474	51.813± 6.5
4	AG	Awgu	Bitumen	0.344±0.018	39.472±3.4
5	AK	Akwuke	Ironstone	0.245±0.480	5.099±5.98
6	AE	Ama-Echi	Silica	0.105±0.052	11.947±11.95
7	UU	Uzo-Uwani	Kaoline	0.546±0.012	122.663±21.18
8	EN	EnuguNorth	Gypsum	0.150±0.106	9.654±4.9
9	NS	Nsude	Glass-sand	0.133±0.141	55.718±13.2
10	OK	Okpara	Coal	0.197±0.195	14.045±1.6
Mean Value				0.2174±0.167	32.105±7.441
Control				0.036±0.01	1.153±0.32
WHO,2004				0.1	1.0

Table 2: Committed Effective Dose Due to gross Alpha (^{226}Ra) and gross Beta (^{228}Ra) Activity concentrations in Ground water

S/N	Location	Effective dose due to ^{226}Ra (mSv)		Effective dose due to ^{228}Ra (mSv)		Total Effective Equivalent dose (mSv)	
		Infant	Adult	Infant	Adult	Infant	Adult
1	Ogugu	0.00084	0.00307	0.297	1.084	0.298	1.0871
2	Bunker	0.00179	0.00654	0.899	3.382	0.901	3.289
3	Enugu-Ekulu	0.01613	0.0589	7.150	26.098	7.166	26.157
4	Awgu	0.01926	0.0700	5.446	19.881	5.465	19.951
5	Akwuke	0.01372	0.0501	0.704	2.518	0.718	2.618
6	Ama-Echi	0.0059	0.0215	1.649	6.018	1.655	6.039
7	Uzo-Uwani	0.0306	0.1116	16.927	61.785	16.958	61.897
8	EnuguNorth	0.0084	0.0307	1.332	4.863	1.340	4.894
9	Nsude	0.0075	0.0272	7.413	27.059	7.470	27.086
10	Okpara	0.0110	0.0403	1.938	7.074	1.949	7.1167
	Mean	0.0122	0.0444	4.430	16.171	4.442	16.215

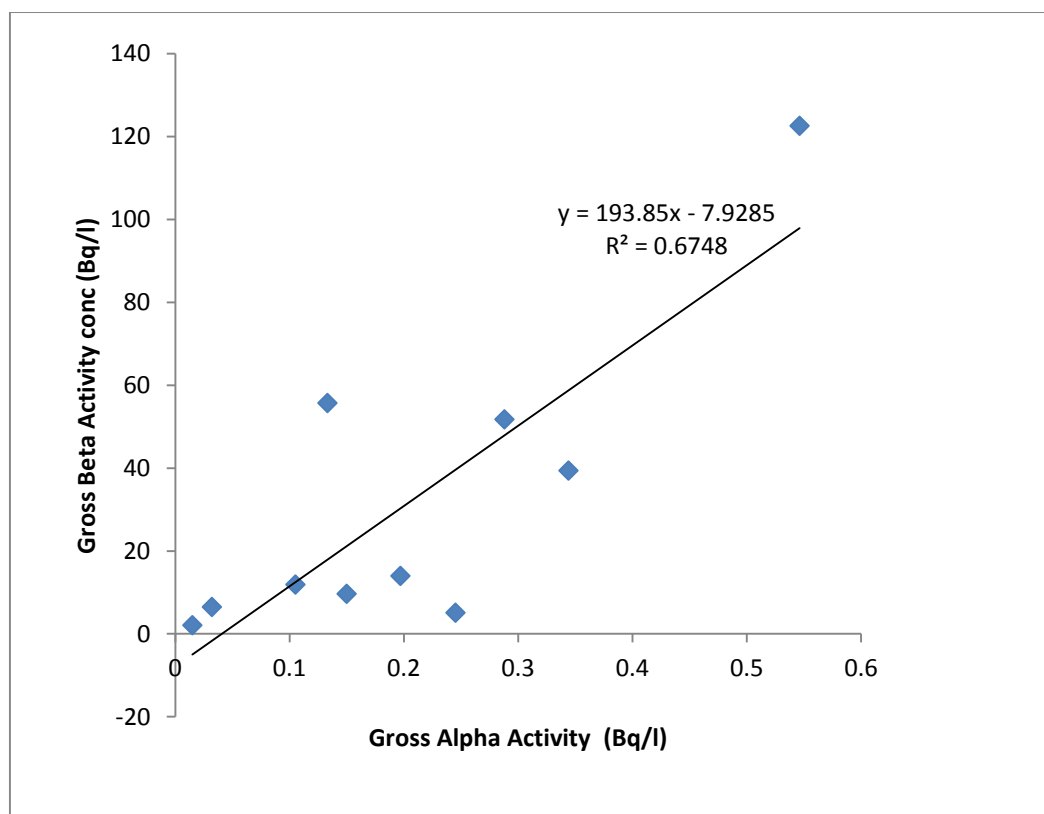
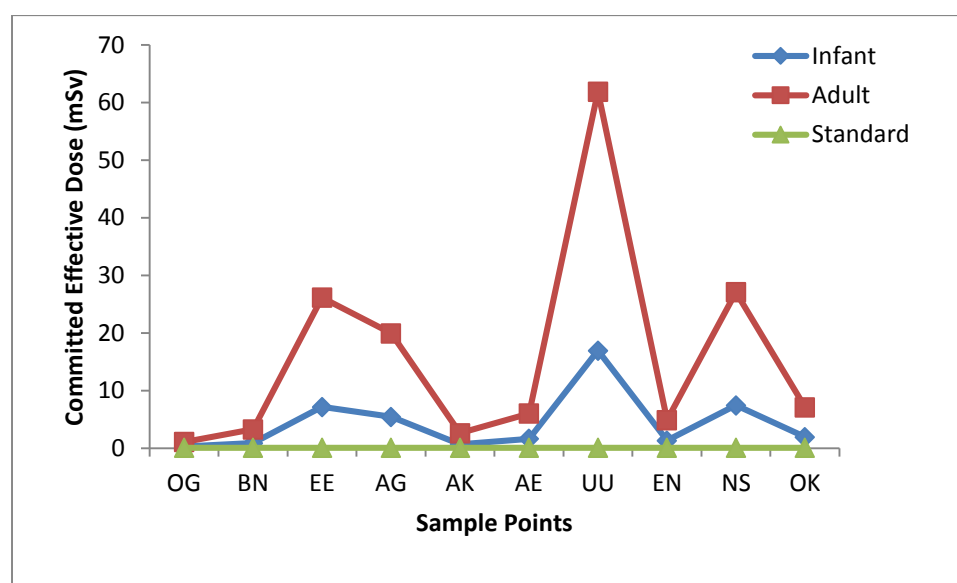


Fig 1: Correlation of gross alpha and gross beta activity concentration in ground water

Table 3: Comparison of the gross α and β concentrations determined in drinking water samples with those reported in other countries.

Country	Gross alpha (mBqL ⁻¹)	Gross beta (mBqL ⁻¹)	References
Greece	82	283	Karamanisa et al., 2007
Turkey	192	579	Damla et al., 2009
Brasil	1- 400*	120 - 860*	Bonotto et al., 2009
Jordan	29 – 3146*	0 – 5014*	Ismail et al., 2009
Nigeria	5.8 – 174*	14.7 – 222.5*	Fasae et al., 2015
Nigeria	15.0 - 546*	2153 – 122,663.0	This study

**Fig 2:** Comparison of the committed Effective dose with WHO (2008) standard

Gross alpha and gross beta activity concentration in ground water from solid mineral producing areas of Enugu State is presented in Table 1 while Table 2 shows the calculated committed effective doses due to alpha emitters and beta emitters in samples of ground water collected from ten communities of solid mineral producing areas of Enugu state. The gross alpha activity concentration ranges from 0.015 ± 0.006 BqL⁻¹ to 0.546 ± 0.012 BqL⁻¹ with average value of 0.2174 ± 0.016 BqL⁻¹. The gross beta activity concentration ranges from 2.153 ± 1.01 BqL⁻¹ to 122.663 ± 21.18 BqL⁻¹ with an average value of 32.105 ± 7.44 BqL⁻¹. The highest gross alpha and gross beta activity concentration was recorded at Uzo-Uwani which might be due to rich Kaolin deposit in the area.

Kaolin is used extensively in the ceramic industry, where its high fusion temperature and white burning characteristics makes it particularly suitable for the manufacturing

of white wares, porcelain and refractory. Studies on radiological impacts of the usability of clay and kaolin as raw material in manufacturing of structural building materials in Turkey showed that ^{226}Ra , ^{232}Th and ^{40}K activity concentration in Kaolin was $82.0\pm 37.3 \text{ Bqkg}^{-1}$, $94.8\pm 47.2 \text{ Bqkg}^{-1}$ and $463.6\pm 55.3 \text{ Bqkg}^{-1}$ respectively (Turhan, 2009). ^{226}Ra is an alpha emitter and ^{228}Ra which is a daughter product of ^{232}Th is a beta emitter. This gives the reason why the highest alpha and beta activity was measured in this community with abundance Kaolin. The least gross alpha and gross beta activity concentration was recorded at Ogugu community which is endowed with limestone. The study carried out in Northern Iraq shows that natural radioactivity levels of ^{238}U , ^{226}Ra and ^{40}K in limestone are considered to be a typical level of natural background (Laith et al., 2011).

There is a good linear relationship between the gross alpha and gross beta activities concentration in ground water as shown in figure 1 with correlation coefficient (R^2) of 0.674. This implies that radionuclides (alpha and beta emitters) present due to Kaolin natural deposit in the area might be responsible for the radioactive contamination of the ground water (drinking water) of the communities sampled. The results obtained in all the communities sampled are higher than the safe value of 0.1 and 1.0 Bq l^{-1} recommended by the World Health Organization (WHO, 2004) as shown in figure 2. The gross alpha activity and gross beta activity measured is higher than those recorded in tap water from Niger Delta region where average activity values are $0.100\pm 0.013 \text{ Bq l}^{-1}$ and $8.9\pm 0.2 \text{ Bq l}^{-1}$ respectively for gross alpha and gross beta (Agbalagba et al., 2013) and for ground water in Ado-Ekiti where gross alpha and gross beta activities are $0.589\pm 0.36 \text{ Bq l}^{-1}$ and $0.236\pm 0.019 \text{ Bq l}^{-1}$ respectively (Fassea, 2013). Table 3 shows the comparison of the results obtained with reported data from other countries. Gross alpha and beta radioactivity obtained in this work was higher than that obtained in Greece, Turkey, Brazil and Jordan. This may be due to varying degrees of solid mineral deposits that constitute the geology of the areas studied.

The committed effective dose due to intake of water calculated from gross alpha and gross beta activity concentration for infant and adult is presented in Table 2. The committed effective dose calculated for infants' ranges from 0.298 to 16.958 mSv with an average value of 4.442 mSv while for adult it ranges from 1.0871 to 61.890 mSv with average value of 16.215 mSv. The highest value was recorded at Uzo-Uwani due to the presence of solid mineral Kaolin which is rich in alpha and beta emitter radionuclides. The least effective dose both for infant and adult were recorded at Ogugu which is endowed with limestone. Ekulu –Enugu, Nsude and Awgu communities recorded high values of effective dose for infant and adult. These areas are endowed with solid minerals; clay, glass – sand (radioactive glass) and Bitumen respectively. The recommended reference dose level (RDL) of committed effective dose of 0.1 mSv from one year intake of drinkable water was however exceeded in all the communities studied. Thus there could be radiation health risk for long term exposure mostly at Uzo-Uwani community.

CONCLUSION

Gross alpha and gross beta activity concentration in ground water from solid mineral producing areas of Enugu state was measured using gas flow proportional counter. The gross alpha and beta activity concentration exceeded the safe value of 0.1 and 1.0 BqL⁻¹ respectively recommended by WHO, (2004). Community (Uzo-uwani) endowed with Kaolin recorded the highest value.

Committed effective dose was calculated for infants and adults from the gross alpha and beta activity concentration. The results showed that the reference dose level of 0.1 mSv from one year consumption of water was exceeded in all the communities sampled. This implies that ground water from solid mineral producing areas of Enugu has been contaminated by the radionuclides in the solid mineral deposit in the area. We therefore recommend incorporation of reverse osmosis technology or ion exchange technology in boreholes to remove the dissolved mineral radionuclides from drinkable water in order to reduce radiological burden to the populace.

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