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Determination of Radiation Hazard Indices (Annual Gonadal Dose Equivalent) in Water Samples in the Vicinity of Mining Site in Minna, Niger State, Nigeria.

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Abstract
The gross alpha and beta radionuclide activity of source of water from mining site in Minna, Niger state has been carried out. Eleven water samples were collected from the farming, residential/commercial, and industrial zones in the community and were analyzed using the protean instrument corporation (PIC) MPC 2000DP, single channel proportional counters. Annual Gonadal Dose Equivalent (AGDE) was determined. AGDE $_{\alpha \text{ and } \beta}$ 20.254399/3.088977mSv/y. This shows that the calculated values of alpha in BHL3 are above the accepted value for ICRP. Moreover, the calculated values of beta in BHL1 are above the accepted value for ICRP. It can be observed that all the remaining values for alpha and beta are below the ICRP values of 0.5Bq/L for alpha and 1Bq/L for beta.

Keywords: Annual, Gonadal, Dose, Alpha, Beta.

INTRODUCTION
Radionuclides (radioactive nuclide, radioisotope, or radioactive isotope) is an atom that has excess nuclear energy, making it unstable. This excess energy can be used in one of three ways: emitted from the nucleus as gamma radiation; transferred to one of its electrons to release it as a conversion electron; or used to create and emit a new particle (alpha particle or beta particle) from the nucleus. During those processes, the radionuclide is said to undergo radioactive decay (Alabi, 2001). These emissions are considered ionizing radiation because they are powerful enough to liberate an electron from another atom. The radioactive decay can produce a stable nuclide or will sometimes produce a new unstable radionuclide which may undergo further decay (Ali, 2004). Radioactive decay is a random process at the level of single atoms: it is impossible to predict when one particular atom will decay. However, for a collection of atoms of a single element the decay rate, and thus the half-life ($\frac{t}{\frac{1}{2}}$) for that collection can be calculated from their measured decay constants. The range of

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the half-lives of radioactive atoms have no known limits and span a time range of over 55 orders of magnitude (Al-Masri *et al.*, 2005).

More than 2400 radionuclides have half-lives less than 60 minutes. Most of those are only produced artificially and have very short half-lives. For comparison, there are about 252 stable nuclides. (In theory, only 146 of them are stable, and the other 106 are believed to decay (alpha decay or beta decay or double beta decay or electron capture, or double electron capture) (Arpansa et al., 2003).

All chemical elements can exist as radionuclides. Even the lightest element, hydrogen, has a well-known radionuclide, tritium. Elements heavier than lead, and the elements technetium and promethium, exist only as radionuclides. In theory, elements heavier than dysprosium exist only as radionuclides, but the half-life for some such elements, e.g. gold and platinum, are too long to be found (Baratha, 2000).

Unplanned exposure to radionuclides generally harms living organisms including humans, although low levels of exposure occur naturally without harm. The degree of harm will depend on the nature and extent of the radiation produced, the amount and nature of exposure (close contact, inhalation, or ingestion), and the biochemical properties of the element; with increased risk of cancer the most usual consequence. However, radionuclides with suitable properties are used in nuclear medicine for both diagnosis and treatment. An imaging tracer made with radionuclides is called a radioactive tracer. A pharmaceutical drug made with radionuclides is called radiopharmaceutical (Ekpo *et al.*, 2000).

On Earth, naturally occurring radionuclides fall into three categories: primordial radionuclides, secondary radionuclides, and cosmogenic radionuclides (Eisenbud *et a*l., 2009). Many of these radionuclides exist only in trace amounts in nature, including all cosmogenic nuclides. Secondary radionuclides will occur in proportion to their half-lives, so short-lived ones will be very rare. For example, polonium can be found in uranium ores at about 0.1 mg per metric ton (1 part in 10^{10}). Further radionuclides may occur in nature in virtually undetectable amounts as a result of rare events such as spontaneous fission or uncommon cosmic ray interactions (WHO, 2012).

In the optimization and comparison of three different methods for the determination of ^{222}Rn in water, extracted ^{222}Rn from water by degasification method, ^{222}Rn was extracted directly from water and counted in a liquid scintillation counter for one hour. In the third method, water was sampled using a Marinelli beaker and after four hours the water was counted for gamma using NaI(Tl) scintillator. The results obtained by all three methods agreed, with the first method having detection limits of 20 MBq/l, the second method having 200 MBq/l, and the third method having 1.75 Bq/l (Van Briston, *et al.,* 1995). The determination of uranium in surface water using an absorber resin, saturated after agitation in 2m-HNO_3 and saturated with ammonium chloride and directly electrolyzed for alpha measurement (Severijins *et al.*, 2006). Their results showed that uranium obtained from the resin after agitation for 30 minutes produces a recovery of 10-90% with RSD < 15%. The detection limits were 2ppb for uranium using ICPS and 0.4ppb by alpha-particle spectrometry (Williams, 2000). Another analysis of radium isotopes by alpha spectrometry shows the presence of ^{224}Ra and ^{226}Ra with detection limits of 0.3 and 0.2 MBq respectively (Alvarado *et al.,* 2005). The relative error for the determination of 3.45 MBq/l of ^{226}R was 0.9% and the RSD (n-20) was 4.6%.

Radionuclide was detected in drinking water by analysis that shows the presence of ^{226}Ra, ^{210}Po, and ^{224}Ra, which were obtained from an 80ml sample of drinking water, (Surberck, 2005). This was exposed to Ra-absorbing discs for 20 hours and the discs were measured by alpha-particle spectrometry using a silicon surface barrier alpha detector. The results also show that the detection limit for these three radionuclides was 2.0 x 10-5 Bq/m3 (USGS, 2000). In this study, we employed Single channel proportional counters to analyze gross alpha (α) and gross beta (β) activities concentration in the water samples from the mining sites and we calculate for annual Gonal Dose Equivalent (AGDE).

MATERIALS AND METHOD

Sampling Technique

A random sampling method was used for this study. Five (5) water samples were collected from underground water sources, another five (5) from the surface (pond), and one as control water sources. All samples were collected in Jayfi, Pago in the Tungan Goro area of Minna, Niger state.

Methodology

The following procedures were carefully carried out during the collection and preparation of the samples:

Samples of water were collected directly into 2-liters plastic kegs (polyethylene containers) after washing the containers properly and rinsed with the water sample to be collected. About 10ml of concentrated hydrochloric acid (HNO_3) was added at the point of collection. The addition of concentrated HNO_3 helps preserve the radio-nuclides present in the water samples and it also prevents the absorption of the water with the inner wall of the containers among others. The addition of HNO_3 assists in reducing the pH of the water samples below 2. Surface water from boreholes and ponds within the area was collected and treated with the reagent. Care was taken to avoid fetching from the stagnant areas. Normally for bore-holes; electric pumps are used to pump water to the reservoir which is connected to different pipes and taps within the community. The tap boreholes were first turned on at full capacity for three (3) minutes to purge the plumbing system of any water which might have been there for some time. The flow rate was reduced to attain steady turbulence and radon loss while collecting the water into the kegs (Onoja, 2004).

The water samples were transferred to CERT, ABU Zaria in clean condition where they were prepared and analyzed for gross alpha (α) and gross beta (β) activities.

The beakers, crucibles (Petri dishes), planchets, and spatula were washed properly, rinsed with clean water, and sterilized using acetone. Then the apparatus was kept and dried inside the oven. A little quantity of the water sample was used to rinse the beaker twice to ensure that there is no cross-contamination before evaporation. About 500ml of the water sample was measured into the beaker and set on the hot plate with a steady temperature below boiling point to allow gradual evaporation and to avoid excessive loss of the residue. This process continues until when the volume of the water sample is reduced to a very little quantity (about 50ml), then it will be transferred into the petri-dish and evaporated to dryness under an infra-radiator lamp. This process is known as surface drying. Having taken the initial weight (i.e. empty dish), the weight of the residue together with the petri-dish was measured using a digital analytical weighing balance. The weight of the total residue obtained from the total volume evaporated was then calculated by using the relation below.

$$W_r = W_{(d+s)} - W_d \quad (2.1)$$

Where: $W_{(d+s)}$ is the weight of the dish with the sample's residue,
W_d is the weight of an empty dish
W_r is the weight of the total residue.

0.0770g of the residue is transferred in the sterilized planchet and the exact volume that produced this required weight (0.0770g) is calculated by the use of the expression that follows.

$$0.0770g \times V_{tr} = W_{tr} \times V. \quad (2.2)$$

Where: $V_{tr}$is the volume that generated total residue,
$W_{tr}$is the weight of the total residue obtained
V is the volume that yielded the required residue.

For samples with residue obtained greater than or equal to 0.0770g, the sample efficiency is said to be 100%. But for the samples with residue less than 0.0770g, its sample efficiency can be obtained using the expression below;

$$\text{Sample eff.} = \frac{weight\ of\ residue}{0.0770g} \times 100\%. \quad (2.3)$$

Annual gonadal dose equivalent (AGDE) measures the dose of gross alpha and gross beta received by the gonadal surface cells as a result of radiation exposure.

The computation of AGDE for gross α or gross β is given by the formula:

$$AGDE = \frac{AEDE}{R.W.F\ X\ T.W.F} \text{ ---------- 2}$$

Where
R.W.F = Radiation weighting factor
T.W.F = Tissue weighting factor
R.W.F = 2 for α – activity
1 for β – activity
T.W.F = 0.20 (for both α and β activity)

The sum of AGDE for gross alpha and gross beta radiation is given as $AGDE_T$ (α, β)

$$= \sum_i^{(\alpha, \beta)} \frac{AEDE}{R.W.F\ X\ T.W.F} \text{ ---------- 3}$$

Sample Analysis

The international standards organization procedure (ISO 9696 and ISO 9697: 2014E) for the measurement of gross alpha (α) and gross beta (β) activities in water was employed in this analysis. This method provided a screening technique to measure the gross alpha (α) and gross beta (β) radioactivity in water samples. To analyze drinking water for gross alpha (α) and gross beta (β) activities (excluding radon), the most common approach is to evaporate a known volume of the sample to dryness and measure the activity of the residue. As alpha (α) radiation is easily absorbed within a thin layer of solid material, the reliability and sensitivity of the alpha method (α) determination may be reduced in samples with high total dissolved solids (TDS) content.

Samples analysis was done using a proportional counter system; a portable non-filled gas MPC2000B-DP single channel gross alpha and gross beta radiation detector. The equipment was mainly designed purposely for gross alpha (α) and gross beta (β) counting. Each sample was placed on the detector and counted for 2700 seconds (45 minutes).

RESULTS AND DISCUSSION

RESULTS

Table 3.1 show the **gross alpha, gross beta concentration (BqL^{-1})** results of the detector characterization while Table 3.2 shows the AGDE for all samples. The results of the average

efficiencies of the detector in an alpha-only mode for JF pond is ±0.01159 while that of beta in beta-only mode is ±0.02058, thus the average efficiency of beta is greater than the average efficiency of alpha with the alpha-to-beta efficiency ratio.

Table 3.1: Showing the result of gross alpha, gross beta concentration (BqL^{-1})

S/N	Sample ID	Alpha Concentration (Bq/l)	Beta Concentration (Bq/l)
1.	JF 1 POND	0.02059±0.00779	0.06782±0.01183
2.	JF 2 POND	0.12392±0.02272	0.17276±0.02898
3.	JF 3 POND	0.03285±0.00851	0.07792±0.01217
4.	JF 4 POND	0.02274±0.00839	0.04240±0.01114
5.	JF 5 POND	0.00553±0.01037	0.08671±0.01642
6.	BHL 1	0.14628±0.02016	0.29893±0.02737
7.	BHL 2	0.00797±0.00552	0.00988±0.00693
8.	BHL 3	0.03922±0.01005	0.05905±0.01281
9.	BHL 4	0.00552±0.01036	0.16150±0.01877
10.	BHL 5	0.00968±0.01059	0.05781±0.01542
11.	CONTROL	0.06476±0.01159	0.28508±0.02058

Table 3.2 The AGDE for all samples

S/N	Sample ID	AEDEα ($mSvy^{-1}$)	AEDEβ ($mSvy^{-1}$)	AGDEα ($mSvy^{-1}$)	AGDEβ ($mSvy^{-1}$)
1.	JF 1 POND	4.20859×10^{-3}	0.033171	0.0105215	0.165855
2.	JF 2 POND	0.0253	0.0844969	0.05133	0.4224845
3.	JF 3 POND	6.71454×10^{-3}	0.381106	0.0167864	0.190553
4.	JF 4 POND	4.64806×10^{-3}	0.02073784	0.0116202	0.10369
5.	JF 5 POND	1.130332×10^{-3}	0.0424097	2.826×10^{-3}	0.212046
6.	BHL 1	0.02999	0.1462065	0.074975	0.731033
7.	BHL 2	1.629×10^{-3}	4.7934×10^{-3}	4.0725×10^{-3}	0.02397
8.	BHL 3	8.0165	0.0289	20.0414	5.78×10^{-3}
9.	BHL 4	1.128×10^{-3}	0.07898	2.82×10^{-3}	0.3949
10.	BHL 5	1.979×10^{-3}	0.0283	4.955×10^{-3}	0.1415
11.	CONTROL	0.013237	0.139433	0.0336925	0.697165

Table 3.2: The sum of AGDE for gross alpha for all samples is 20.254999$mSvy^{-1}$ and for gross beta for all samples is 3.088977$mSvy^{-1}$ respectively.

The above table shows that the highest reading for alpha is 20.0414 whilst the lowest is 2.82×10^{-3} and the total AGDE is 20.254999. Similarly, the highest reading for beta is 0.731033 whilst the lowest is 5.78×10^{-3} and the total is 3.088977$mSvy^{-1}$. Of all the values obtained above, it is only Sample BHL 3 which is 20.0414 that is far above the IAEA standard.

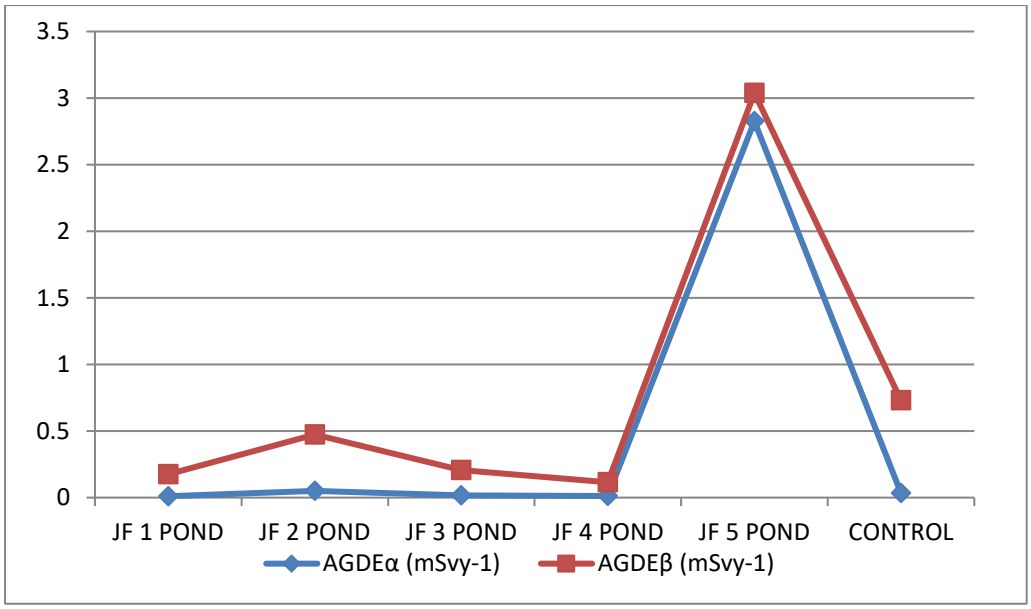

Fig 3.1: Graph of JF POND against **AGDEα (mSvy^{-1})** and **AGDEβ (mSvy^{-1})** activity Concentrations.

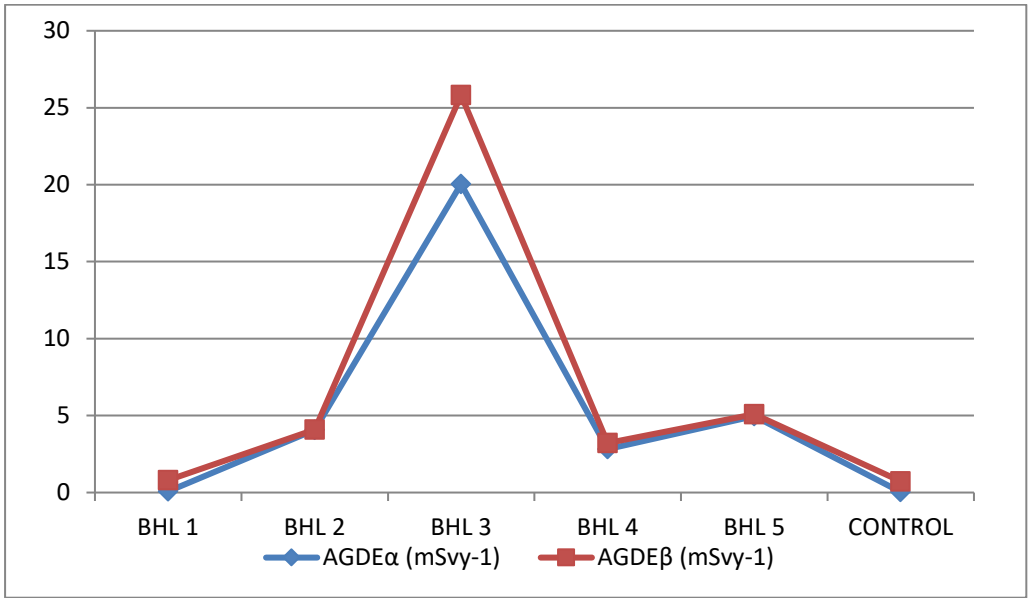

Fig 3.2: Graph of BHL against **AGDEα (mSvy^{-1})** and **AGDEβ (mSvy^{-1})** activity Concentrations.

DISCUSSION

We calculated the Annual Gonadal Dose Equivalent of alpha to be 20.254999mSv/y, and beta to be 3.088977mSv/y. These are good values for this type of counter. Low background activity was also observed with 0.33cpm for alpha and 0.22cpm for beta. This is quite representative of the environment. Similarly, the results obtained from all the counting modes are reproducible and are hence reliable. The gross alpha and beta activity concentrations in the water samples were found to be in the range of 20.254999mSv/y, and beta to be 3.088977mSv/y. This shows that the calculated values of alpha in BHL3 are above the accepted value for ICRP. Moreover, the calculated values of beta in BHL1 are above the accepted value for ICRP. It can be observed that all the remaining values for alpha and beta are below the ICRP values of 0.5Bq/L for alpha and 1Bq/L for beta.

CONCLUSION

The determination of gross alpha and gross beta radionuclide activities from water samples in Jayfi, Tungan Goro village of Pago in Minna Niger State has been studied. The gross alpha and beta activity concentration (Bq/L^{-1}), Annual Gonadal Dose Equivalent (AGDE) in the water samples along the community differ in quantity from sample to sample. This is explained by the heterogeneity of radionuclide deposits, water transportation, and precipitation by organic metabolism, and effluent discharge. The overall low value of gross alpha and gross beta of radionuclide activities observed may be due to the mining activities in the community and the low level of soil formation that constitute the geology of the area except for BHL3 for alpha and BHL1 for beta respectively.

The sum of AGDE of both gross alpha and gross beta of all samples was calculated. However, the values obtained are far below the WHO and ICRP recommended maximum permissible limit except for BHL3 for alpha and BHL1 for beta respectively, and may not pose any serious health side-effects to the public users as their source of drinking water.

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