Original Research Article

EVALUATION OF CHEMICAL AND ~~RADIOLOGICAL~~ IMPACTS OF RADIONUCLIDES IN BOREHOLE WATERS IN IGBAJO TOWN, OSUN STATE, NIGERIA

ABSTRACT

Wateris a universal solvent on earth and sourcessuch as wells, rivers, springs, boreholes, and other freshwater bodies typically serve as a pathway for contaminants to enter the ecosystem while also supplying water for domestic and drinking purposes.The activity concentrations of radionuclides in borehole waterin Igbajo town were assessed using gamma-ray spectroscopy, so as to effectively determine the degree of radiological risk to the environment and its inhabitants, the outcomes were used to calculate all the radiological impact parameters.The activity concentrationsobtained for ^{40}K, ^{238}U and ^{232}Thranged from 7.25±0.60 to 62.15±4.48Bq.L^{-1}; 3.08±0.45 to 15.24±3.07 Bq.$L^{-1}$and 1.08±1.10 to 17.75±1.59 Bq.L^{-1} and with average values of 23.12±1.59 Bq.L^{-1}, 6.27±2.01 Bq.L^{-1} and 7.01±0.89 Bq.L^{-1} respectively.The Annual Effective Dose (AED) for ingested radionuclide in drinking water for an adult was1.4872 µSv.yr^{-1}. The Radium Equivalent Activity Index, Ra_{eq} obtained was 34.9186 Bq.L^{-1}. The estimated hazard indices H_{int} and H_{ext} were 0.04876 and 0.06574 respectively. The Excess Lifetime Cancer Risk, ELCR (x10^{-6}) was 5205.15. With a lifetime expectancy of 70 years, this high value suggests that there is a high chance of developing cancer.The value of the Annual Gonadal Equivalent Dose, AGED in the water was 55.9175µSv.yr^{-1}. The estimatedGamma Index, $I_\gamma$was 0.1273 mSv.yr^{-1}. There is a significant health hazard to the environment and people living in the area owing to the radioactivity contents and radiological impact parameters. Conclude and suggest the wayforward.

Keywords: Borehole waters, radiation hazard~~ parameters~~,radiological impact,toxicity, cancer risk

1.0 INTRODUCTION

The contamination and pollution of nature are results of human activity. These ongoing activities have severely deteriorated the natural ecosystem and caused it to be vulnerable to natural radiation from the earth and space (Edori and Kpee, 2016).

Due to the fact that borehole water quality differs from source to source, examination of the suitability of this water is crucial in Nigeria where drinking of borehole water has significantly increased. Different analytical techniques have been used in different nations to quantify the radioactivity levels in drinking water (El Arabi et al., 2006).Naturally occurring ^{40}K,^{232}Th and ^{238}U series are the sources of radioactivity in geological materials, primarily soil androcks. (UNSCEAR, 2000).

Radiation levels are higher in igneous rocks (granite), and lower in sedimentary rocks.However, rare exceptions abounds, including phosphate and shales rocks that havecomparativelyhigh radioactive concentrations(Uosif, 2007). Depending on the dose ingested, radiation has consequences on people. High radiation doses have the potential to change human DNA, although low doses may not have any ~~discernible~~noticeable effects. Both random and predictable biological effects of radiation exposure exist (Hall, 2000).A deterministic effect takes a dose threshold, and the severity of the effect is dose-related, as in the case of skin reddening, but stochastic effects do not require a dose threshold and are determined by the molecular mechanisms at play, as in the case of cancer or a hereditary defect.

~~NORM —~~Naturally Occurring Radioactive Material (NORM) refers to radioactive elements with a long half-life, such as uranium, thorium, and potassium, as well as any of their decay products, including radium and radon. These elements existed in the Earth's crust and atmosphere but are concentrated in particular places, such as deposits of extractable uranium ore.Industrial with NORM related issues include: oil and gas production;coal mining and combustion; mineral sands (rare earth minerals, titanium, and zirconium); metal mining and smelting;fertilizer (phosphate); recycling;building; and uranium mining and all related fuel cycle activities.(Cooper, 2005).

Uranium series disequilibrium techniques were first applied by Rosholt et al., (1964) and subsequently by Titayeva et al., (1976). Two facts form the basis of these applications. Firstly, ^{234}U and ^{238}U are often under secular equilibrium in rocks. Second, natural levels of ^{238}U differs within a very wide range [101 to 103 mBq/l] (Osmond and Cowart, 1992). These elements work together to provide the foundation of a very effective geochemical instrument. Some daughter radionuclides exhibit preferential leaching and this can result from a variety of physical effects. For duration comparable to their individual half-lives, the initial disequilibrium of other long-lived radionuclides continues. According to geochemical theory,

radium and uranium are soluble species, with natural activity concentrations typically higher than 1 mBq.L^{-1}. (José et al., 2007).

The purpose of this study is to assess the radioactivity and evaluate the risk parameters for the samples of water collected from Igbajo town, to ascertain the level to which the inhabitants are disposed to radiation hazards.

2.0 MATERIALS AND METHODS

2.1 ***The Study Area:*** It is situated in Igbajo town, Boluwaduro Local Government, Osun State, South-west Nigeria on latitude $7^0$54'24"N and longitude $4^0$48'44"E.

The community comprises range of mountains and adjacent basins and its environs are strategically defensible due to surrounding rockytopography with stalwart outcrops. The major tectonic actions range to hornblende-granite-biotite, muscovite-granite-tourmaline-gneiss, gneiss,biotite-gneiss-granite, variably migmatized gneisses and pegmatite intrusions. The dominant rock is quartz schist and quartz and variably biotite-garnet-schist gneiss and biotite-garnet-schist (Bello et al., 2019).

2.2 ***Sampling***: A collection protocol was established and was strictly adhered to. These include a collection procedure, usingsuitablebottles and employing appropriate methods for preservation so as to minimize the influence of adsorption.

A total of six (6) samples were collectedeach with 2L-sized plastic bottles, which was washed and rinsed with dilute hydrochloricacid (0.1M HCl). The samples collected were acidified with 1M hydrochloric acid to attain a pH < 2 so as to prevent adsorption of the radionuclide on the walls of the container (AS/NZS, 1998). The samples were then transported to the laboratory for further processingprior to instrumental analysis.

2.3 ***Measurement and Analysis:***The procedure involved using a thallium activated Canberra vertical high purity 3" x 3" Sodium Iodide [NaI(Tl)] detector connected to ORTEC 456 amplifier. The detector was protected by about 15cm thick lead on the four sides and 10cm thick at the top. About 2.0KeV resolution and 33% efficiency at 1.33 MeV was accomplished in the system with 27000scounting time. For calibration, the usual sources recommended by the International Atomic Energy Agency (IAEA) were employed (IAEA, 2003). From the counting spectra, the activity concentrations of ^{40}K, ^{238}U and ^{232}Th were determined using computer program. Various peaks were taken into account. prior to arriving

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at the calculation of each activity levels, in comparison to 1460 KeV (40K) for 40K, 1764.5 KeV (Bi-214) for 238U, and 2614.5 KeV (Ti-208) for 232Th.

The activity concentration (C) of the radionuclide can be evaluated after subtracting decay correction ~~with~~(Avwri, et al., 2014);

$$C_a = \frac{C_n}{\varepsilon P_\gamma M_s t_c} \quad (i)$$

Where C_a = Activity concentration of radionuclide ($Bq.L^{-1}$)

C_n= net counts of radionuclide in the samples

ε = absolute counting efficiency of the detector system

P_γ= gamma ray emission probability (gamma ray yield)

M_s = the mass of the sample (kg)

t_c = total counting time

2.3.1 Radiological hazard Parameters:

Absorbed Dose Rate (D):The rate, D (nGy/h) with respect to activity concentration of ^{238}U, ^{232}Th and ^{40}K is calculated using (Orosun et al., 2016);

$$D = 0.462C_U + 0.604C_{Th} + 0.0417C_K \quad (ii)$$

Annual Effective Dose (AED):The annual effective dose due to ^{40}K, ^{238}U, and ^{232}Th ingestion was calculated using (Meltem&Gursel, 2010; US-EPA, 1991).

$$AED = \sum (C_A \times AI \times IDF) \quad (iii)$$

Where, AED = Annual effective dose ($mSvyr^{-1}$),

C_A= Activity concentration of the radionuclides (BqL^{-1})

AI = Average person's intake of water per year ($730Lyr^{-1}$ for adult)

DF = Dose conversion factors (ingestion dose coefficient);

DF for adults for ^{40}K, ^{238}U, and ^{232}Th is 6.2×10^{-9}, 2.3×10^{-7}, and $4.5 \times 10^{-8}Sv.Bq^{-1}$ for respectively.

Radium Equivalent Activity Index (Ra_{eq}):It represents the weighted sum of activities concentration of ^{40}K, ^{238}U and ^{232}Th. It is usually calculated to estimate the radiological risksrelated with the three radionuclides. It is assumed that $1Bq.Kg^{-1}$ of ^{238}U, $0.7Bq.Kg^{-1}$ of ^{232}Th and $13Bq.Kg^{-1}$ of ^{40}K produce the same gamma-ray dose. It can be defined empirically using (Issa~~,~~ et al., 2013) as:

$$Ra_{eq} = C_U + 1.43C_{Th} + 0.077C_K \quad (iv)$$

Where C_U, C_{Th} and C_K are the radioactivity concentration of ^{40}K, ^{238}U and ^{232}Th respectively.

Excess Lifetime Cancer Risk (ELCR):The ELCR was assessed using equation (v) below.

$$ELCR = AED \times DL \times RF \quad (v)$$

Where AED = Annual Effective Dose

DL = Average Duration of Life (70 years)

RF = Risk Factor, for stochastic effects, ICRP uses 0.05 for public

Radiation Hazard Indices:Both the external radiation hazard index (H_{ext}) and the internal radiation hazard index (H_{int}) were estimated using (Avwri~~,~~ et al., 2014):

$$H_{ext} = \frac{1}{370} C_U + \frac{1}{259} C_{Th} + \frac{1}{4810} C_K \quad (via)$$

$$H_{int} = \frac{1}{185} C_U + \frac{1}{259} C_{Th} + \frac{1}{4810} C_K \quad (vib)$$

Where C_U, C_{Th} and C_K are the radioactivity concentrations of ^{40}K, ^{238}U and ^{232}Th respectively.

The values of both the $H_{ext}$and H_{int} should be below unity for the radiation risk to be negligible.Internal exposure to radon is very dangerous thus lead to respiratory diseases like lung cancer, asthma etc.

Gamma Index (I_γ):It is used for the estimation of gamma radiation hazard related with the natural radionuclide in specific samples.It can be calculated using:

$$I_\gamma = \frac{1}{150} C_U + \frac{1}{100} C_{Th} + \frac{1}{1500} C_K \quad (vii)$$

$$I_\gamma \leq 1$$

Where C_U, C_{Th} and C_K are the radioactivity concentrations of ^{238}U, ^{232}Th and ^{40}K in water samples.

An increase in I_γ greater than 1 often results to radiation risk which may result to the modification of human cells thereby causing cancer.

Value of I_γ = 1 corresponds to an annual effective dose of less than or equal to 1 mSv.

Value of I_γ= 0.5 corresponds to annual effective dose less or equal to 0.3 mSv (Avwri et al., 2014).

Annual Gonadal Equivalent Dose (AGED):The gonads, bone cells and bone marrow are centre of interest by UNSCEAR (2000) because of their sensitivity to radiation. As the AGED increases, the bone marrows are affected, causing damage of the red blood cells and are then substituted with white blood cells. This results in a blood cancer known as leukemia.

AGED can be evaluated using:

$$AGED\ (\mu Sv.yr^{-1}) = 3.09C_U + 4.18C_{Th} + 0.314C_K \quad (viii)$$

Where C_U, C_{Th} and C_K are the radioactivity concentration of ^{238}U, ^{232}Th and ^{40}K in water samples.

2.3.2 Chemical Toxicity Risk:

The chemical toxicity risk was calculated using the lifetime average daily dose (LADD) of uranium through drinking water intake, and related it with the reference dose (RFD) of 0.6 $\mu g.kg^{-1}.day^{-1}$ (Ye-shin et al., 2004) produced a hazard quotient usingas standards for uranium in several foreign organizations.

$$\text{Hazard quotient} = \frac{LADD}{RFD} \quad (ix)$$

$$\text{Ingestion LADD of drinking water} = \frac{EPC \times IR \times EF \times ED}{AT \times BW} \quad (x)$$

Where LADD = Lifetime Average Daily Dose ($\mu g.kg^{-1}.day^{-1}$)

EPC = Exposure Point Concentration ($\mu g.L^{-1}$); IR = Water Ingestion Rate ($L.day^{-1}$)

EF = Exposure Frequency ($days.year^{-1}$); ED = Total Exposure Duration (years)

AT = Average Time (days); BW = Body Weight (kg)

Using IR = $2L.day^{-1}$; EF = 350 days; ED = 45.5 years; AT = 16,607.5 (from 45.5 x 365); BW = 70kg (standard man)

Conversion: 1 $Bq.L^{-1}$ = 27.0 $pCi.L^{-1}$; 1$\mu g.L^{-1}$=1$pCi.L^{-1}$/0.67 *(xi)*

3.0 RESULTS AND DISCUSSION

Activity Concentrations of Radionuclides in the Samples: The radionuclides activity concentrationsin the water samplestaken from the study areasare presented in Tables 1 below. The activity concentration of ^{40}K in the borehole water samples ranged from 7.25±0.60 to 62.15±4.48$Bq.L^{-1}$ with an average value of 23.12±1.59 $Bq.L^{-1}$ and that of ^{238}U ranged between 3.08±0.45 and 15.24±3.07$Bq.L^{-1}$with an average value of 6.27±2.01 $Bq.L^{-1}$ while that of ^{232}Th in the samples ranged from 1.08±1.10 to 17.75±1.59$Bq.L^{-1}$ with an average of 7.01±0.89 $Bq.L^{-1}$.

Hence, ODF sample had the lowest concentration of ^{40}K while ISA had the highest concentration. The highest and lowest activity concentration values of ^{238}U were found in ISA and ODF respectively. While the highest concentration of ^{232}Th was found in ODF, the lowest was found in ESW. These variations are attributed to the different sources of water samples. Thus, ^{40}K contributed the largest activity concentration while ^{238}U contributed the least activity in the samples.

Researcher's evaluation of potential radiological hazards to humans is made possible by knowledge regarding the distribution of these radionuclide activities found in natural materials. However, they will be utilized to calculate all radiological impact hazard parameters so that we can knowto what extent the local population are exposed.

Table 1: Activity concentration of ^{40}K, ^{238}U and ^{232}Th in the borehole water samples

Water Samples	^{40}K (Bq.L^{-1})	^{238}U (Bq.L^{-1})	^{232}Th (Bq.L^{-1})
BAB	9.92±0.61	4.52±1.89	1.79±0.13
BAP	13.44±1.05	6.66±1.37	5.92±0.51
COM	29.70±1.81	3.09±1.03	1.17±0.75
ESW	16.24±0.98	5.01±4.23	1.08±1.10
ISA	62.15±4.48	15.24±3.07	14.34±1.26
ODF	7.25±0.60	3.08±0.45	17.75±1.59
MEAN±S.D.	23.12±1.59	6.27±2.01	7.01±0.89
WHO	**10.00**	**10.00**	**1.00**

BAB – Babalaje; BAP – Baptist; COM – Community; ESW – Essawe; ISA – Isao; ODF - Odofin

*Radiological Hazard Parameter in Water: Absorbed Dose Rate (*D*):*The absorbed dose rate (nGy.h^{-1}) inthe samples were evaluated withequation (ii), and the results presented in Table 2.The absorbed dose rate values ranged from 3.3727 to 18.2939 nGy.h^{-1}with an average value of 8.0922 nGy.h^{-1}for the study areas. From this study, the estimated average value was lesser than the world average value of 57 nGy.h^{-1} (UNSCEAR, 2000) and hence, poses nosevere health risk.

Annual Effective Dose (μSv.yr^{-1}): The annual effective dose resulting from the ingestion ofwater samples were calculated using equation (iii). The values ranged from 0.4194 to 3.1896μSv.yr^{-1}with an average value of 1.4872μSv.y^{-1} for the study areas.

It was eminent that the values assessed for all the water samples were lower than the world average value of 1000 μSv.yr^{-1} hence it is within the safe limit.

*Radium Equivalent activity Index (Ra_{eq}):*The Radium Equivalent Activity Index (Ra_{eq}) from water samples were estimated using equation (iv).The result ranged between 13.2435 to 87.4418 Bq.Kg^{-1}with an average value of 34.9186 Bq.Kg^{-1}for the study areas. The obtained average value was below the world average value of 370 Bq.Kg^{-1}and hence poses no significant health hazard (UNSCEAR, 2000).

Excess Lifetime Cancer Risk (ELCR): The excess life time cancer risk for the analyzed water samples were evaluated using equation (v). The ELCR ranged from 1467.95 x 10^{-6} to 11163.64 x10^{-6} with an average of 5205.15 x 10^{-6}. It is worthy of note that the obtained average value was greater than the world average value of 0.2 x 10^{-3} (200 x 10^{-6}) (UNSCEAR, 2000). This suggests that there may be a significant risk of cancer associated with spending a70 yearaverage in this environment without associating with other environment for food and shelter.

The high value of the ELCR index for water samples is due to high AED caused by high activity concentration of ^{40}K radionuclide in the samples.

RadiationHazard Indices: The radiation hazard indices in water samples, both the external and the internal were evaluated using equation (via) and (vib) respectively. The external radiation hazard index (H_{ext}) ranged between 0.0274 to 0.1507 with an average of 0.0657.~~while~~ While the internal radiation hazard index (H_{int}) ranged from 0.0190 to 0.1094 with an average of 0.0488 for the study. The two values were found to be lesser than the world average value of unity, therefore poses no significant health hazard (UNSCEAR, 2000).

*Gamma Index (I_γ):*The gamma indices for the samples were calculated using equation (vii). The average value estimated for the water samples was 0.1273 $mSv.yr^{-1}$ for values ranging between 0.0521 and 0.2864. This value is within the safe limit of less than unity, the universal standard.

*Annual Gonadal Equivalent Dose (AGED):*The AGEDsof the water samples were calculated using equation (viii). The average value estimated was 55.9175 $\mu Sv.yr^{-1}$ forvalues ranging between 23.7645 and 126.5479. This value is within the safe limit of less than the universal standard.

Table 2: Radiation hazard parameters for the water samples

Sample Code	D ($nGy.h^{-1}$)	H_{int}	H_{ext}	Ra_{eq} ($Bq.Kg^{-1}$)	I_γ ($mSv.yr^{-1}$)	AGED ($\mu Sv.yr^{-1}$)	AED ($\mu Sv.yr^{-1}$) Adults	ELCR (x 10^{-6})
BAB	3.5831	0.0212	0.0334	13.2435	0.0547	24.5639	0.4939	1728.72
BAP	7.2130	0.0436	0.0617	22.9405	0.1126	49.5452	1.2735	4457.53
COM	3.3727	0.0190	0.0274	33.6600	0.0521	23.7645	0.4324	1513.30
ESW	3.6441	0.0211	0.0346	19.0349	0.0550	25.0947	0.4194	1467.95
ISA	18.2939	0.1094	0.1507	87.4418	0.2864	126.5479	3.1896	11163.64
ODF	12.4463	0.0783	0.0867	33.1908	0.2029	85.9887	3.1142	10899.76
MEAN	**8.0922**	**0.0488**	**0.0657**	**34.9186**	**0.1273**	**55.9175**	**1.4872**	**5205.15**
UNSCEAR	**57.00**	**1.00**	**1.00**	**370.00**	**1.00**	**300**	**1000.00**	**290.00**

The result presented in table 3 showed that the exposure dose ranged from 3.40 – 16.83 $\mu g.kg^{-1}.day^{-1}$. The LADD value was observed highest in ISA sample. This might beas theresult of the depth of the water source and the geochemistry.

By comparing the lifetime average daily dose (LADD) obtained in this study and the reference dose (RFD) (0.6 $\mu g.kg^{-1}.day^{-1}$), the acceptable level, the chemical toxicity hazard

because of the uranium in the water samples were all above the RFD. This suggests that there are health hazards associated with uranium in the water samples which are basically due to the chemical toxicity risk.

Table 3: The activity concentrations, mass concentrations and estimated Lifetime Average Daily Dose of Uranium in the borehole (BH) water samples in the study area

Sample	^{238}U (Bq.L^{-1})	^{238}U (pCi.L^{-1})	^{238}U (µg.L^{-1})	LADD (µg.kg^{-1}.day^{-1})
BAB	4.52	122.04	182.16	4.99
BAP	6.66	179.82	268.40	7.35
COM	3.09	83.43	124.53	3.41
ESW	5.01	135.27	201.90	5.53
ISA	15.24	411.48	614.17	16.83
ODF	3.08	83.16	124.12	3.40

4.0 CONCLUSION

The activity concentrations of ^{40}K, ^{238}U and ^{232}Th in borehole water samples in Igbajo town, Boluwaduro Local Government, Osun State, Nigeria have been examined and the possible radiotoxicity has been documented in this study. The average values of the activity concentrations of ^{40}K, ^{238}U and ^{232}Th exceeded the recommended value by the WHO but a slight difference in the value of ^{238}U.

Similarly, the activity concentrations were related to the mass concentrations of uranium in the samples and these were found to vary from 124.12 to 614.17 µg.L^{-1}. This indicates that the measured mass concentrations of uranium in the borehole water in the area were relatively high when compared with the recommended safe limits by some various international organizations. It was inferred that the human risk due to uranium content in water may likely be to the chemical toxicity of uranium as a heavy metal, however, this study represents a valuable radiometric data that are vital tools in radio-epidemiological assessment, diagnosis and prognosis of radionuclide-induced diseases to the population of the studied area.

5.0 RECOMMENNDATION

- Effective management of mining and milling activities should be practiced
- Promotion of organic fertilizer (green chemistry is safer and cheaper) instead of synthetic ones.
- Treatment of water should be encouraged and practiced

- Adequate socialization on the risk associated with radionuclides and ways to remediate it in the society should be encouraged

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