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EVALUATION OF NATURAL RADIONUCLIDES AND RADIATION HAZARDS OF WATER AND FLOODPLAIN SOILS OF ALATO RIVER USING GAMMA RAY SPECTROMETRY

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ABSTRACT

Natural radioactive elements can pose health risks if present in elevated concentrations. These elements can adversely affect aquatic and terrestrial ecosystems. This study evaluates the specific activity of radionuclides in floodplain soil and water samples collected from Alato river in South western Nigeria. The samples were analysed for activity concentration of ^{40}K, ^{235}U and ^{232}Th using NaI (TI) detector gamma ray spectrometry system. The mean activity concentration of radionuclides in soil samples were: ^{235}U: 8.65±01.85 to 9.74±0.45 Bq/kg, ^{232}Th: 35.58±1.79 to 48.53±2.28Bq/kg, ^{40}K: 471.84±5.33 to 593.59±5.47 Bq/kg, while that of water samples; ^{235}U: 8.71±1.02 to 9.57±0.07 Bq/l, ^{232}Th: 29.60±1.71 to 35.58±1.79 Bq/l and ^{40}K: 433.86±8.86 to 593.82±7.22 Bq/l. The average activity concentration of ^{235}U in both matrixes were lower than the global average, while those of ^{40}K were higher than guidance level of the World Health Organization (WHO). The average activity concentration of ^{232}Th in the soil samples differ at different regions which were be higher or lower than the global average, while the level in water was within tolerable range. The study revealed that the mean value of absorbed dose rate is lower than the world average limit. It is clear that the external and internal hazard associated with the soil usage in this location do not pose some radiological risk.

Keywords: Floodplain, Radionuclides, Gamma Spectrometry, Radiological Risk, Specific Activity

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1. INTRODUCTION

The environment occupied are naturally radioactive, and individuals are frequently presented to radiation from the inestimable beams, characteristic radionuclides in water, air, soil and man-made radioactivity (Ademola *et al*., 2014). The radiation to which the human population is exposed comes from many diverse sources. Some are natural, while others are the result of anthropogenic activities. Natural sources include cosmic radiation, external radiation from radionuclides in earth's crust and internal radiation from radionuclides inhaled or ingested and retained in the body. The level of the exposures depends on environment and as well as human activities. Height above sea level affects the dose rate from cosmic radiation; radiation from the ground depends on geology; and the dose from radon, which seeps from the ground (Gür *et al*., 2001).

Soils are the actual store house of radionuclide naturally occurring on the Earth's crust responsible for transferring these elements into biological systems (Garba *et al*., 2012). Radioactivity is an observable fact that is connected with energetic atomic nuclei that are automatically decomposed releasing beta, alpha, and neutron particles, or electromagnetic radiation in the form of gamma rays (Ayeni and Adebiyi, 2022). Soil radioactivity is gaining great scholar attention widely as it one of the main factors in public doses and may help predict changes in the environment. Soil characteristics, geological formations and human practices are important factors for potentially elevating the concentrations of background natural radiation. The level of natural radioactivity in an area depends on the soil types, rocks types and its geology (Ademola, 2021). However, the distribution of radionuclides in surface soil relies on the composition and distribution of radioelements in the bedrock, their physical and mechanical properties such as porosity, permeability (Alashrah *et al*., 2018). Soil, rocks and minerals commonly contains naturally occurring radioactive materials (NORM), with half-lives as longer as the Earth's age, known as primordial radionuclides (Ayeni and Adebiyi, 2022). An elevated level of radioactivity in aquatic could be due to possible radionuclides migration to surface and groundwater from human activities such as mining, milling, intensive use of fertilizers in farming, abandoned industrial sites, nuclear power plants, waste dump areas (Nkwankwo, 2013).

Humans and other living organisms are continuously exposed to ionizing radiation (Hamzah *et al*., 2011). Naturally Occurring Radioactive Material (NORM) includes long-lived radioactive elements (e.g., uranium, thorium, and potassium) found in the environment (Mansour *et al*., 2012). Wastes from NORM accumulate in tubing and surface equipment in the form of scale and sludge. In the exploration and extraction processes of oil and gas, the natural radionuclides ^{238}U, ^{235}U, and ^{232}Th, as well as the radium-radionuclides ^{223}Ra, ^{224}Ra, ^{226}Ra and ^{228}Ra, and ^{210}Pb, etc., are brought to slurry surfaces and may contain levels of radioactivity above the surface background (Gazineu *et al*., 2005). Several studies have been conducted on the concentrations of naturally radionuclides in the environment in different regions throughout the world (Al-Attar *et al*., 2015).

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Natural radioactive elements, such as uranium, thorium, and radium, can pose health risks if present in elevated concentrations. Radioactive elements can adversely affect aquatic and terrestrial ecosystems. Assessing the levels of these elements and understanding their distribution in flood plain soil enables authorities to assess potential exposure risks to nearby communities and implement measures to mitigate health hazards. This study aims to determine the specific activity of natural radionuclides in floodplain soil and water from Alato River in attempt to evaluate the occurrence, distribution and potential health hazards of the natural radionuclides in the area under investigation.

2. MATERIALS AND METHODS

2.1 Sample collection

Floodplain soil and water samples were collected from different locations along the course of Alato river, Ado Ekiti, Southwestern, Nigeria. Three samples each were randomly selected from each region, as separated or divided by a bridge. In this study, a total of six water samples were collected within the study area. The samples were collected into 1-liter pre-cleaned polyethylene/plastic bottles which were carefully labelled and rinsed thoroughly with water sample to be collected, prior collection. For gamma spectrometry, samples were taken to the laboratory, each sample was sealed in previously purified and labelled plastics containers and kept for a period of 28 days to allow parent and daughter radionuclide to attain secular equilibrium, after which counting began.

Each soil sample were collected with the use of improvised auger from the surroundings of the river. All collected samples were kept separately in non-radioactive cellophane bags, labelled and properly secured. Samples for radionuclide (gamma spectrometry) were taken to the laboratory and properly sealed. The sealed samples were kept in the laboratory for about 28 days before counting commenced.

2.2 Sample preparation

2ml HNO_3 was used to stabilize the water sample prior to counting from where 200ml each of the samples were measured into seven clean sample bottles and properly sealed. Sealed containers, of both water and sediment samples, which has been washed with 0.1M HCl were kept incubated for 28days in order to reach secular equilibrium.

2.3 Sample analysis

Using a counting time of 25,200s (7hrs), gamma counting of the samples are done using a 7.62 cm x 7.62 cm NaI (TI) detector surrounded with adequate lead shielding that reduces the background by a factor of approximately 95% in a Canberra MP2-2U version T35240K gamma spectrometer with a Canberra Genie 2000 software. The activity concentrations of various radionuclide will then

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be determined in Bq/kg from the count spectra obtained from each of the samples using the gamma ray photo peaks corresponding to energy of 484.5keV (^{214}Bi), 608KeV (^{208}Te) and 1460.82 keV (^{40}K) for ^{238}U, ^{232}Th and ^{40}K, respectively.

2.4 Energy calibration

The Sodium Iodide detector will be calibrated in terms of absolute gamma ray energy. The energy calibration process involved using known radioactive isotopes as calibration sources to establish a relationship between channel numbers and corresponding gamma-ray energies. The system will be calibrated with standard calibration sources of ^{137}Cs and ^{60}Co. In order to achieve a clearly established photo peak, the calibration sources will be counted for 25,200s. The channel number corresponding to the epicenter of each full energy peak on the MCA will be recorded and used to plot the slope and interception.

2.5 Evaluation of radiological parameters

In order to evaluate the potential risk associated with the use of water and soil in the studied area, it's necessary to evaluate and assess some radiological hazard parameters to derive a more reasonable and safer conclusion. The parameters are computed from known value of activity concentration of radionuclides in the environmental matrices.

2.5.1 Absorbed dose rate

The dose conversion factors for converting the activity concentrations of ^{238}U, ^{232}Th and ^{40}K into doses (nGy/h per Bq/kg) are 0.462, 0.621 and 0.0417 respectively. The absorbed gamma dose rate was computed using equation below:

$$D\ (nGy/h) = 0.462H_u + 0.621\ H_{Th} + 0.0417H_k$$

where D is the dose rate in nGy/h and H_u, H_{Th} and H_k are the concentrations of ^{40}K, ^{238}U and ^{232}Th, respectively

2.5.2 External and internal hazard index

The external hazard index (H_{ex}) is computed using equation below:

$$H_{ex} = \frac{H_{Ra}}{370} + \frac{H_{Th}}{259} + \frac{H_K}{4810}$$

The internal hazard index (H_{in}) is computed using equation below:

$$H_{in} = \frac{H_{Ra}}{185} + \frac{H_{Th}}{259} + \frac{H_K}{4810}$$

H_u, H_{Th} and H_k are the activity concentrations of ^{40}K, ^{238}U and ^{232}Th, respectively

2.5.3 Radium equivalent activity (Ra_{eq})

The radium equivalent activity (Ra_{eq}) is expressed mathematically by equation (UNSCEAR, 2000) below:

$$Ra_{eq}\,(Bqkg^{-1}) = Hu + 1.43\,H_{Th} + 0.0077H_k$$

H_u, H_{Th} and H_k are the activity concentrations of ^{40}K, ^{238}U and ^{232}Th, respectively.

2.5.4 Ingestion dose from water consumption

The ingestion dose due to consumption of water is measured using the annual effective dose equivalent (AEDE) due to ingestion of ^{235}U, ^{232}Th and ^{40}K in water. The AEDE is a function of the rate of consumption of water. The equation below was used to estimate the AEDE due to ingestion of a radionuclide in water (UNSCEAR, 2008; Akpanowo *et al.*, 2021).

$$AEDE_{ing,w} = \sum^{3}_{i=1} (A_i \text{ x } RC_w \text{ x } DF_i)$$

Where Ai is the activity concentration of radionuclide i(^{235}U, ^{232}Th and ^{40}K in Bq/l), RC_w is the rate of consumption of water in $1y^{-1}$ taken as 730 $1y^{-1}$ or 2 $1y^{-1}$ for adult, while DF*i* is the dose conversion factor for ingestion of i radionuclide by an adult. The dose conversion factor for ^{235}U is given as 4.5 x 10^{-5} $mSvBq^{-1}$ (Akpanowo *et al.*, 2021), for ^{232}Th is given as 2.3 x 10^{-4} $mSvBq^{-1}$, while the DF*i* for ^{40}K is given as 6.2 x 10^{-6} $mSvBq^{-1}$ (Orosun *et al.*, 2018; Akpanowo *et al.*, 2021).

2.5.5 Life time risk (LR) from ingestion of water

The risk levels from direct ingestion of natural radionuclides in water is estimated using the lifetime risk assessment (LR). This is estimated as

$$LR = D_w * D_L * R_F$$

Where Dw is the annual effective dose equivalent (Svy^{-1}). D_L is the duration of life which is taken as 55 years for Nigerian (Akpanowo *et al.*, 2021). R_F is the risk factor (Sv^{-1}) and is taken as 7.3 x 10^{-2} Sv^{-1} (ICRP, 1996).

3. RESULTS AND DISCUSSION

3.1 Specific activity and distribution pattern of radionuclides

Results of specific activities of ^{235}U, ^{232}Th and ^{40}K in soil and water samples collected in region A, B and control are shown in Tables 1 and 2. The specific activity of radionuclides in the soil samples in region A were measured as; ^{235}U: 9.81±0.03 to 11.08±1.14 Bq/kg with an average of 9.74±0.45 Bq/kg, ^{232}Th: 41.83±2.62 to 52.11±2.49 Bq/kg with an average of 48.53±2.28 Bq/kg,

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^{40}K: 417.11±3.91 to 500.23±4.91 Bq/kg with an average of 471.84±5.33 Bq/kg, while that of water samples; ^{235}U: 4.21±0.06 to 12.28±0.07 Bq/l with an average of 9.57±0.07 Bq/l, ^{232}Th: 24.58±1.09 to 32.59±3.17 Bq/l with an average of 29.60±1.71 Bq/l, ^{40}K: 388.37±1241 to 493.26±5.41 Bq/l with an average of 433.86±8.86 Bq/l. It can be deduced that the concentration of radionuclides in the soil sediments was greater than that of the water samples.

The mean values obtained were higher in ^{232}Th than the guidance levels of the World Health Organization (WHO), which are 10 Bq/l for both ^{235}U and ^{232}Th for drinking water. The mean values of ^{40}K in the water samples were several orders higher than the values of ^{235}U and ^{232}Th. This could be attributed to the relative abundance of ^{40}K terrestrially in nature compared to ^{235}U and ^{232}Th. It has also been in report that ^{235}U and ^{232}Th have poor migrating ability compared to ^{40}K (Syabaini and Iskander, 2014).

Table 1: Specific activities of ^{226}Ra and ^{232}Th, ^{40}K in soil and water samples in region A

Sample	Soil			Water		
	K-40 (Bq/kg)	U-235 (Bq/kg)	Th-232 (Bq/kg)	K-40 (Bq/l)	U-235 (Bq/l)	Th-232 (Bq/l)
A_1	417.11±3.91	9.81±0.03	52.11±2.49	493.26±7.40	12.22±0.09	24.58±1.09
A_2	500.23±4.91	11.08±1.14	41.83±2.62	388.37±12.41	12.28±0.07	32.59±3.17
A_3	498.19±7.16	8.32±0.18	51.64±1.74	414.94±6.78	4.21±0.06	31.63±0.86
mean	471.84	9.74	48.53	433.86	9.57	29.60
SD	5.33	0.45	2.28	8.86	0.07	1.71
C.V%	1.13	4.62	4.49	2.04	0.73	5.78

The specific activity of radionuclides in the soil samples in region B were measured as; ^{235}U: 6.04±1.01 to 13.11±3.43 Bq/kg with an average of 8.65±01.85 Bq/kg, ^{232}Th: 31.62±0.97 to 43.05±2.44 Bq/kg with an average of 35.58±1.79 Bq/kg, ^{40}K: 577.18±3.49 to 603.31±7:51 Bq/kg with an average of 593.59±5.47 Bq/kg, while that of water samples; ^{235}U: 8.33±0.86 to 9.04±1.02 Bq/l with an average of 8.71±1.02 Bq/l, ^{232}Th: 21.48±1.07 to 41.66±1.94 Bq/l with an average of 28.42±1.75 Bq/l, ^{40}K: 577.18±3.49 to 633.81±7:84 Bq/l with an average of 593.82±7.22 Bq/l. Some radionuclides are highly soluble in water and, therefore, more mobile in aquatic systems. When radionuclides are soluble, they are less likely to bind to sediment particles and instead remain suspended in the water column. This mobility can result in higher concentrations in water compared to soil sediment (IAEA, 2010).

The global average specific activity of these natural radionuclides is 35, 40, and 400 Bqkg^{-1} for ^{235}U, ^{232}Th and ^{40}K, respectively (UNSCEAR, 2008). The average specific activity of ^{235}U at

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region A and B were lower than the global average. At region A, the average specific activity of ^{232}Th in the soils were higher than the global average, while that of region B is lower. ^{232}Th level in water from point A and B are within tolerable range. ^{40}K average specific activity is higher than the standard at both regions. This indicates that the level of ^{40}K is above tolerable limits which can increase the risk of cancer and other health issues. The concentration of ^{40}K at region A increases as we move toward B, while that of ^{232}Th decreases. The physical processes of erosion, transport, and deposition affect how radionuclides are distributed in sediments (Syed *et al*., 2020). Lake Baikal sediments reveal historical deposition patterns of radionuclides, reflecting both natural processes and human activities (Appleby *et al*., 1998).

Table 2: Specific activities of ^{226}Ra and ^{232}Th, ^{40}K in soil and water samples in region B

Sample	Soil			Water		
	K-40 (Bq/kg)	U-235 (Bq/kg)	Th-232 (Bq/kg)	K-40 (Bq/l)	U-235 (Bq/l)	Th-232 (Bq/l)
B_1	603.31±7.51	6.80±1.12	43.05±2.44	588.21±5.41	8.75±1.17	41.66±1.94
B_2	600.22±5.41	13.11±3.43	32.09±1.98	633.81±7.84	8.33±0.86	22.13±2.25
B_3	577.18±3.49	6.04±1.01	31.62±0.97	577.18±3.49	9.04±1.02	21.48±1.07
Mean	593.59	8.65	35.58	593.82	8.71	28.42
SD	5.47	1.85	1.79	7.22	1.02	1.75
C.V%	0.92	21.39	5.03	1.23	11.71	6.16

3.2 Absorbed dose rate of radionuclides

The absorbed gamma dose rate (nGy/h) are presented in Table 3. The Absorbed gamma dose rate (D) is the quantity of absorbed ionizing radiations energy gotten per unit mass per unit time from materials. Clearly, the study showed that the mean value of D, in nGy/h is lower than the world average limit of 84 nGy/h (UNACEAR, 2000). The computed values also did compare favorably with those reported elsewhere and some part of Nigeria (Jegede *et al*., 2017).

Table 3: Absorbed gamma dose of the samples

Locations	Soil	Water
Region A	54.31	40.89
Region B	50.16	46.44
Mean	52.24	43.67

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Gamma radiation can alter the microbial composition and activity in soil, impacting nutrient cycling and soil health (Seybold *et al*., 2011). Radioactive isotopes from gamma radiation can be absorbed by aquatic organisms and biomagnified through the food chain, posing health risks to both aquatic life and humans consuming contaminated water or organisms (Smith and Beresford, 2019). Increased level of absorbed dose rate can cause the radiolysis of water, breaking down water molecules into reactive species like hydrogen peroxide and free radicals. These reactive species can further react with other compounds in the water, potentially leading to the formation of toxic substances.

3.3 External and internal hazard index of radionuclides

The internal hazard is a parameter used to estimate the internal radiation burden from terrestrial radionuclides via inhalation and ingestion pathways. For radiation hazard due to exposure to soil to be insignificant, the relation $H_{in} \leq 1$ must be satisfied. The external hazard index (H_{ex}) is used to evaluate the radiation burden due to exposure to gamma radiation from natural radionuclides. The external and internal hazard index are evaluated and shown in Table 4.

Table 4: External and internal hazard index of radionuclides of the samples

	Soil		Water	
Locations	**H_{ex}**	**H_{in}**	**H_{ex}**	**H_{in}**
Region A	0.31	0.34	0.23	0.26
Region B	0.28	0.31	0.26	0.28
Mean	0.30	0.33	0.25	0.27

This index is defined by Beretka and Matthew (Bavarneign et al., 2013). The mean value obtained is less than unity and obeys the relations $H_{ex} \leq 1$ for hazard or risk level to be insignificant. Radiation index of less than unity revealed insignificant hazard (UNSCEAR, 2000). The prime objective of this index is to limit the radiation dose to dose equivalent limit of 1 mSv/y (ICRP, 1993). It is clear that the external and internal hazard associated with the samples in these locations do not pose some radiological risk since their H_{ex} and H_{in} values didn't exceeded unity. The external value implies that dwellers are relatively safe from radiation exposure risk. This result implies that there is no significant radiation hazards associated with the soil and water bodies of the area studied. These results are comparable to similar coastal environment of other countries and part of Nigeria (Putra *et al*., 2021).

3.4 Radium equivalent activity (Ra_{eq})

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The distribution of ^{40}K, ^{235}U and ^{232}Th showed variation in the environment. Ra_{eq} is used to achieve uniformity in terms of exposure in radiation by a single quantity or index which takes into account the radiation risk associated with each radionuclide (Isinkaye, 2013). Table 5 showed the radium equivalent activity (Ra_{eq}) for samples collected from the studied area.

Table 5: Radium equivalent activity

Locations	Soil	Water
Region A	115.5	86.3
Region B	105.2	95.1
Mean	110.4	90.7

The average evaluated (Ra_{eq}), for soil samples was 110.4Bq/kg, while the water showed 90.7Bq/kg. The mean values obtained was less than the specified limit of 370Bq/kg (UNSCEAR, 2000), Indicating that the soil will not pose a significant risk if used.

3.5 Ingestion dose from water consumption

The ingestion dose due to consumption of water is measured using the annual effective dose equivalent (AEDE) due to ingestion of ^{235}U, ^{232}Th and ^{40}K in water. The AEDE is a function of the rate of consumption of water. The AEDE due to ingestion of ^{235}U, ^{232}Th and ^{40}K of the water are presented in Table 6. The estimated AEDE ranged from 7.24 to 7.75 $mSvy^{-1}$ with mean value of 7.50 $mSvy^{-1}$. The mean value obtained was in excess of the WHO limit of 0.1 $mSvy^{-1}$ (Orosun *et al*., 2018). The results revealed that the sampled water had AEDE values exceeding the WHO limit.

Table 6: AEDE and risk associated with the water samples

Locations	AEDE	Risk
Region A	7.24	29.1
Region B	7.75	31.1
Mean	7.50	30.1

3.6 Life time risk (LR) from ingestion of water

The estimated level of risk (life time risk (LR)) due to ingestion of ^{235}U, ^{232}Th and ^{40}K in the sampled water are presented in Table 6. The life time risk ranged from 29.1 31.1 with average value of 30.1. This revealed that the LR for the water sample exceeded the limit of 8.4×10^{-3} for drinking water (Al-Ghamdi, 2019).

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4. CONCLUSION

In this research work, the specific activities concentration and distribution of ^{235}U, ^{232}Th and ^{40}K of floodplain soil and water of Alato river were evaluated. The study showed that the mean value of absorbed dose rates was lower than the world average limit. It is clear that the external and internal hazard associated with the floodplain soils do not pose some radiological risk. Evaluating the specific activity of radionuclides and understanding their distribution patterns in the environment are critical for assessing environmental contamination, guiding remediation efforts, and ensuring public health and safety. Continuous monitoring and adherence to regulatory standards are essential for maintaining a safe and healthy environment.

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