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Measurement of Activity Concentrations of ^{40}K, ^{226}RA and ^{232}TH For the Assessment of Radiation Hazards from Dangora Surface Soils.

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Abstract

Activity concentrations of natural radionuclides ^{40}K, ^{226}Ra and ^{232}Th in surface soils from Dangora town, Kiru Local Government Area of Kano State of Nigeria were determined using gamma spectroscopy with NaI(TL) detector. The specific activities of the radionuclides range from 294.7 – 403.4Bq/Kg for ^{40}K, 24.2 – 48.9Bq/kg for ^{226}Ra and 43.9 – 78.7Bq/kg for ^{232}Th. ^{40}K (348.2±3.1Bq/kg) and ^{226}Ra (34.7±1.5Bq/kg) are lower than their respective world background averages of 420Bq/kg and 50Bq/kg. Average concentration of ^{232}Th is however, higher than the world mean. Result obtained for radium equivalent activities show that they range from 141.8 – 183.7Bq/kg with a mean of 137.3Bq/kg. Since these values are lower than the established upper limit (370Bq/kg), the soil from within the area of study is considered safe for use as building materials. Absorbed dose rate due to gamma irradiation at 1m above the ground surface ranges from 52.8 – 83.5nGy/h with an average of 69.3nGy/h both of which are comparable with 18-93nGy/h and 58.0nGy/h - the global range and mean respectively. Values of external hazard indices range from 0.31 – 0.50 with a mean of 0.45. Since these values are less than unity, it follows that radiation hazard due to exposure to natural background radiation is neglible. The estimated annual effective dose ranges from 0.07 – 0.10mSv with mean of 0.77mSv that is comparable with the recommended limit (0.07mSv) for normal background. Finally, it is concluded that both ^{226}Ra and ^{232}Th and their decay products are the contributors to the reportedly high level of alpha radioactivity in Dangora's water bodies but elevated by the condition of the aquifer. Due to the high level of ^{40}K concentration recorded in this work, its abundance in the environment, and also the aquifer's state, it is concluded that ^{40}K is the main contributor to the high level of beta activities recorded earlier in a water screening test.

Keywords: Absorbed Dose Rate, Activity Concentration, Annual Effective Dose, External Hazard Index, Radium Equivalent Activity

1.0 Introduction

Radionuclides in their attempt to achieve structural stability, emit one or more of alpha (α), beta (β) and gamma (γ) radiation. The fact that radioactivity has been occurring for geological time scales account for the reasons why radionuclides are omnipresent in the environment [1]. Radioactive materials are introduced into the environment from both natural and man-made sources. Over 80% of these sources are contributed by the natural sources out of which 63% is contributed by terrestrial products. U-238, Th-232 and their decay products are the main contributors to terrestrial radioactivity [2]. These radioelements have half lives comparable to the age of the earth and undergo secular equilibra with gamma emitting radionuclides in their respective decay series. Radionuclide of interest among the relatively low element is k-40[3,4].

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Terrestrial background radiation results from the gamma ray emitted by the natural occurring radioactive materials (NORMs) and constitute the main external source of human irradiation [3,4,5]. Research reports have attributed the high natural background radiation recorded in some regions of the world to the abnormal occurrence of uranium and its decay products in rocks and soils and thorium in monazite sands [6].It follows that knowledge of natural radioactivity present in soils does enable scientists to assess any possible radiation hazard, following interaction with soils via the computation of radiation hazard indices [7]. However, natural radioactivity under certain conditions can reach hazardous levels. Exposure to background radiation has been linked to the increase in cancer risk, mental retardation in children and genetic effects [3].The knowledge of specific activities and distributions of radionuclides in the environment is therefore of interest as it provides useful information in the monitoring of environment. Worldwide survey on environmental radioactivity for risk assessment began in the past few decades [8]. This research work is therefore, undertaken to determine the radioactivity concentrations of K-40, Ra-226 and Th-232 in top soils of Dangora town of Kiru Local Government area of Kano State, Nigeria. The result shall then be used to estimate hazard indices. This is considered of importance as research of this type has never been conducted in the area despite the fact that Dangora drinking water sources have been proven to carry abnormally high level of gross α and β activities following a water screening test [1]. Furthermore, reports on global external background radiation reveal that the level of the background varies from one location to the other. It is hoped that the study will complement the few other studies that had been conducted elsewhere in the country[9].

2.0 The Study Area

Dangora is located at Kiru Local Government Area of Kano state, Nigeria. It is located at Latitude $11^0$31`N and longitude $80^0$00`E. It is one of the 15 political wards of the LGA which bounded by the North, South, East and West by Bauda, Dansoshiya, Kogo and Kafin Maiyaki wards respectively. Majority of the people are farmers while a few are traders and civil servants.

3.0 Materials and Methodology

3.1 Sample Collection and Preparation

Twelve soil samples of about 1kg each were collected at a depth of about 10cm using a steel hand geological auger, which was cleaned with acid, detergent and rinsed with tap water. After removing the stones and vegetation; all the samples were dried up at room temperature for slightly over 24 hours in the laboratory. Following thorough drying the soil was pulverized to tin powder and packed to fill cylindrical containers of height 7cm by 6cm in diameter which is the same as the geometry of the counting detector. Each container accommodates approximately 300g of sample. The filled container is then carefully sealed and kept for over 30days so that secular equilibrium can be attained between Ra-226 and Th-232 and their respective short lived daughters and ground-daughters [5].

4.0 Activity Determination

The activity concentration of Th-232 and Ra – 226 were determined by a 7.62cm by 762cm NaI (TL) detector housed in a 6cm thick lead shield (to assist in the reduction of the background radiation) and lined with cadium and copper sheets (CERT Manual, 1999). The samples were placed on the detector surface and each counter for 29,000 seconds in reproducible sample geometry. The configuration and geometry was maintained throughout the analysis, as previously characterized based on well established protocol of the laboratory (at the centre for Energy Research and Training, Zaria, Nigeria). A computer based Multichannel Analyzer (MCA) MAESTRO programme from ORTEC was used for data acquisition and analysis of gamma spectra. The 1764 keV gamma-line of Bi – 214 for U-238 used in the assessment of the activity concentration of Ra-226, while 2614.5KeV Gamma – line of Tl-208 was used for Th-232. The single 1460keV gamma-line of K-40 was used in its content evaluation. The activity of each radionuclide in the samples was determined using the relation:

$$As = \frac{C_{net}}{\varepsilon_\gamma Y_\gamma m_s} \quad - \quad - \quad - \quad - \quad - \quad - \quad - \quad (1)$$

Where ε_γ is the detector efficiency at γ energy of interest. C_{net} is the count per second of the sample, m_s is the mass of the sample Y_γ is the intensity of gamma ray at the particular energy being counted.

All the obtained raw data were converted to convenient units using calibration factors to determine the activity concentrations of k-40, Ra-226 and Th-232 respectively. In order to determine the specific activity concentrations in the samples, the IAEA mixed standard consisting of k-40, Ra – 226 and Th-232 of the same dimension as the samples were subjected to the same experimental procedures. After subtraction of background counts, concentration in Bq/kg was performed using the conversion factors which are different for each nuclide such that k-40, Ra-226 and Th-232 are 6.431, 8.632 and 8.768 respectively.

5.0 Radiation Hazard Indices Calculation

To assess the hazard indices due exposure to the environmental background radiation, four quantities are considered in this work:

5.1 Radium Equivalent Activity (Ra_{eq})

This quantity is defined mathematically as [10]:

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.07C_k \quad - \quad - \quad - \quad - \quad - \quad (2)$$

Where C_{Ra}, C_{Th} and C_k are the activity concentrations of Ra-226, Th – 232 and K – 40 in Bq/kg respectively. The expression for Ra_{eq} above assumes that 370Bq/kg of Ra – 226 or 259Bq/kg of Th – 232 or 4610Bq/kg of K-40 produce the same gamma dose rate. Ra_{eq} describes the uniformity of exposure to Ra-226, Th-232 and K-40 in a environment though these radionuclides are usually not uniformly distributed. Ra_{eq}, therefore, is a common index used to compare the specific activities of ^{226}Ra, ^{232}Th and ^{40}K in materials by a single quantity, which takes into account the radiation hazards associated with them. The activity index provides a useful guideline in regulating the safety standard dwellings. Therefore, the use of materials whose Ra_{eq} concentration exceeds 370Bq/kg is discouraged to avoid radiation hazards[10].

5.2 External Gamma Absorbed Dose Rate (D)

D is mathematically defined as [5]:

$$D = 0.462\ C_{Ra} + 0.604C_{Th} + 0.042C_K \quad - \quad - \quad - \quad - \quad - \quad (3)$$

Where D is the absorbed dose rate in the air at 1m above the ground level, C_{Ra}, C_{Th}, and C_K are the measured activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in soil respectively. In deriving equation (3) it is assumed that radioactivity from radionuclides of negligible abundance as $^{137}C_s$, ^{90}Sr and the ^{235}U series can be neglected as they contribute very little to the total dose from environmental background. It is also assumed that all the decay products of ^{226}Ra and ^{232}Th are in radioactive equilibrium with their precursurs [10].

External Hazard Index (H_{ex})

H_{ex} is defined as [5]

$$H_{ex} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_k}{4810} \quad - \quad - \quad - \quad - \quad - \quad (4)$$

Where C_{Ra}, C_{Th} and C_K are the respective activity concentrations ^{226}Ra, ^{232}Th and ^{40}K. The value of this index must be less than unity for the hazard to be negligible. H_{ex} equal to unity corresponds to the upper of Ra_{eq} (370Bq/kg) [10].

Annual Effective Dose Equivalent (AEDE)

The annual effective dose equivalent received outdoor by a member is calculated from the absorbed dose rate by apply dose conversion factor of 0.7Sv/Gy and the occupancy factor for outdoor was 0.2(5/24). AEDE is determined using the following[5]:

AEDE (outdoor) ($(\mu Sv/y)$ = Absorbed dose (nGy/h) x 8760h x 0.7Sv/Gy x 0.2 x 10^{-3} - (5)

6.0 Results and Discussion

The results of radionuclide concentrations for 12 soil samples collected at different locations at Dangora town are presented in table 1. The specific activities of the radionuclides ranged between 294.7 and 403.4Bq/kg for ^{40}K, 24.2Bq/kg and 48.9Bq/kg for ^{226}Ra and 43.9Bq/kg and 78.7Bq/kg for ^{232}Th. The respective mean values for K-40, Ra – 226 and Th – 232 are 348.2 ±3.1Bq/kg, 34.7±1.5Bq/kg and 64.9Bq/kg.

While the mean activity concentrations due to K-40 and Ra-226 are lower than their respective world background averages of 420Bq/kg and 50Bq/kg, the mean value for Th – 232 is higher than the world mean (50Bq/kg). The table further reveals that none of the soil samples carry Th-232 concentration below the world average. Though the mean activity of Th-232 is less than that of K-40, it is more hazardous than the later as its permissible level is much lower than that of K-40 and Ra – 232[9].The relatively high concentration of ^{40}K recorded in this work could be attributed to its abundance in the earth crust [2]. A typical decay chain for ^{238}U to which ^{226}Ra belongs is [4]: U-238 (α) → Th-234 (β) → Pa-234 (β) → U-234 (α) → Th-230(α) → Ra-226 (α) → Ra-222 (α) → Po-218 (α) → Pb – 214 (β) → Bi-214 (β) → Po – 214 (α) → Pb – 210 (β) → Bi-210 (β) → Po - 210 (α) → Pb-206 stable.

A typical decay chain of Th-232 series is [11]. Th-232(α) → Ra-226(β) → AC – 228 (β) → Th-228(α) → Ra-224 (α) → Rn -220 (α) → Po – 216 (α) → Pb – 212 (β) → Bi – 212 (β) → Tl-208 (β) → Pb – 208 stable. Though there is beta emission in both chains, being a good beta emitter, with a relatively high abundance, ^{40}K could be regarded as the main contributor to the high levels of beta activities reported in the water bodies in the area [1].

The high values of the total alpha and beta activities reported earlier in the borehole could be attributed to the possibility of the bedrock being metamorphic which usually carries high content of ^{238}U and ^{232}Th series [12].

High level of natural radioactivity has also been linked to different solubility of thorium and radium and partly due to upward diffusion from the surrounding sediments [13].

Since the concentrations of ^{226}Ra, ^{232}Th and ^{40}K recorded in this work are relatively moderate, high levels of gross alpha and beta activities in Dangora ground water sources is therefore attributed to a number of factors which are summarized as follows [12]

1. The uranium content of the source rocks, sediments or soils, and the processes through which uranium may be leached.
2. The degree of hydraulic isolation of the water by fresher surface or groundwater.
3. The oxidation of the water and
4. The concentration of suitable complexing agents, which can increase the solubility of uranium.

In essence Dangora ground water sources may be carrying substantial amount of dissolved oxygen and relatively high dissolved solids to which nuclides as radium are adsorbed to increase their concentrations [13]. Furthermore, the bedrock could be metamorphic which usually carries high content of ^{238}U and ^{232}Th series. Hence the need for further research into these factors.

When the results of this work is compared with world average and other reports from Nigeria and other parts of the world [5,6], it was realized that the respective specific activities K-40, Ra-226 and Th-232 obtained fell within the ranges of the world mean values. The ranges of Ra – 226 and Th-232 concentrations are however comparable with those of Rajasthan, Indian[6]. The concentration range of K-40 is comparable with that of Bagega, Zamfara, Nigeria[9].

Table 1: Specific Activities of radionuclides in Dangora Soil Samples

Samples	**Specific Activity of Radionuclides (Bq/kg)**		
	K-40	Ra – 226	Th – 232
Soil 1	393.9 ± 8.6	36.7 ± 5.8	54.2 ± 2.6
Soil 2	317.8 ± 9.8	48.9 ± 7.9	78.7 ± 4.9
Soil 3	403.4 ± 9.0	24.2 ± 3.9	69.1 ± 3.9
Soil 4	394.6 ± 10.4	43.7 ± 3.6	59.0 ± 1.3
Soil 5	327.4 ± 7.6	27.1 ± 3.4	43.9 ± 1.9
Soil 6	390.7 ± 1.8	45.1 ± 3.0	61.9 ± 2.6
Soil 7	332.5 ± 6.8	24.6 ± 2.4	75.6 ± 5.1
Soil 8	321.2 ± 2.0	27.0 ± 3.1	61.9 ± 1.1
Soil 9	336.9± 8.6	31.7 ± 4.1	84.4 ± 3.6
Soil 10	343.4 ± 4.4	36.2 ± 2.7	51.1 ± 5.0
Soil 11	294.7 ± 2.5	33.4 ± 2.9	71.2 ± 3.8
Soil 12	321.5 ± 5.8	37.8 ± 3.5	57.4 ± 1.0
Range	294.7 – 403.4	24.2 – 48.9	43.9 – 78.7
Mean	348.2 ± 3.1	34.7 ± 1.5	64.1 ± 1.5

Calibration factors: K-40 = 0.000643, Ra-226 = 0.000863, Th – 232 = 0.000877

Conversion Factor: $X10^{-4}$ CPS/Bq/Kg^{-1}

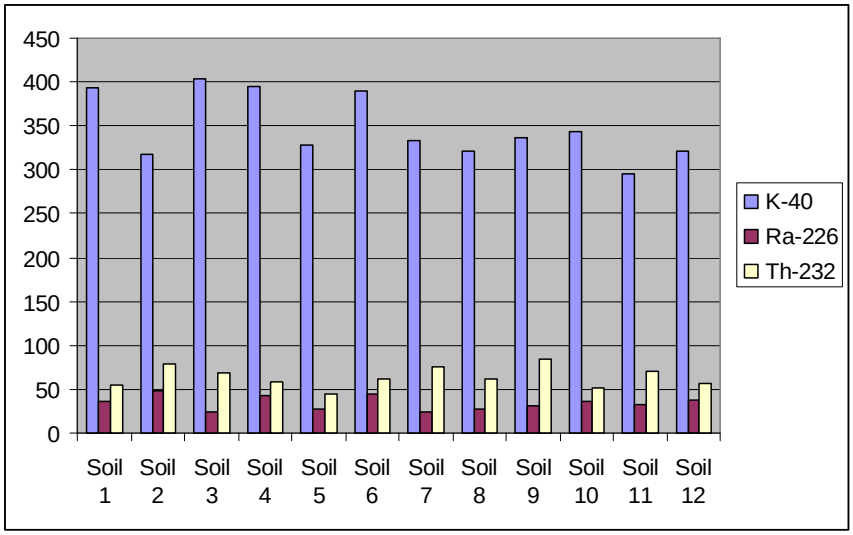

Figure 1: Relative Concentration of K-40, Ra-226 and Th-232

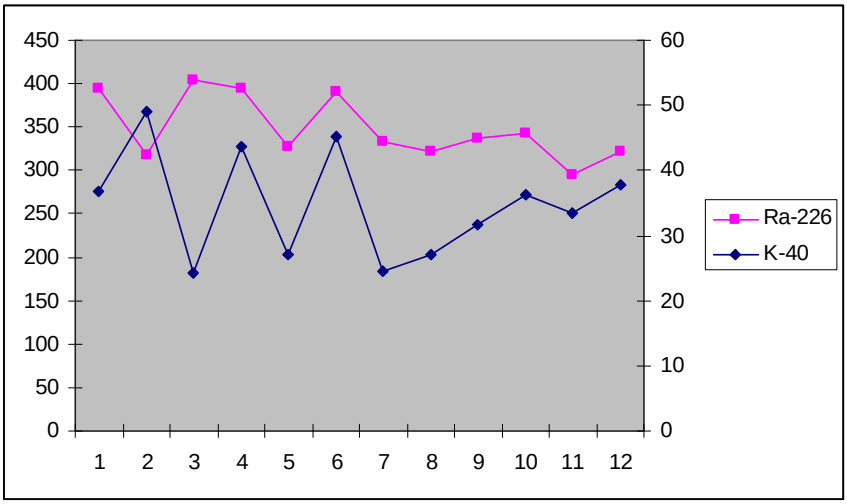

Figure 2: Correlation between K-40 and Ra-226 Concentrations

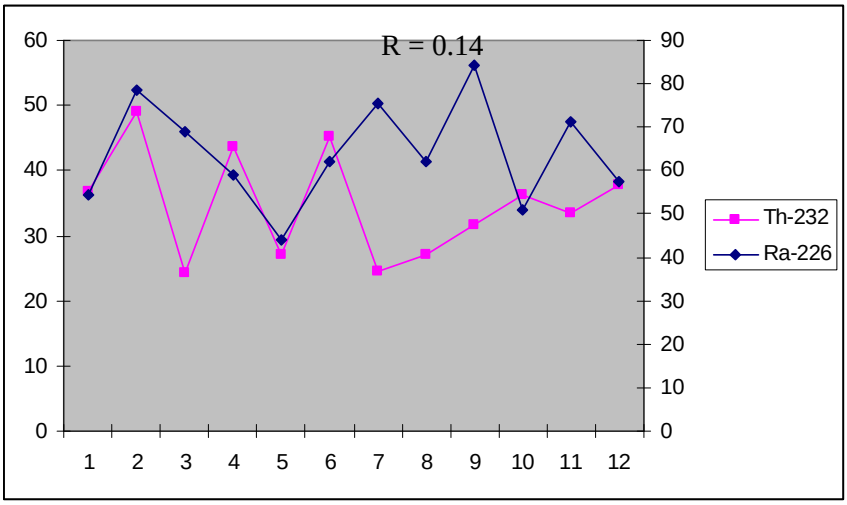

Figure 3: Correlation between Ra-226 and Th-232 Concentrations

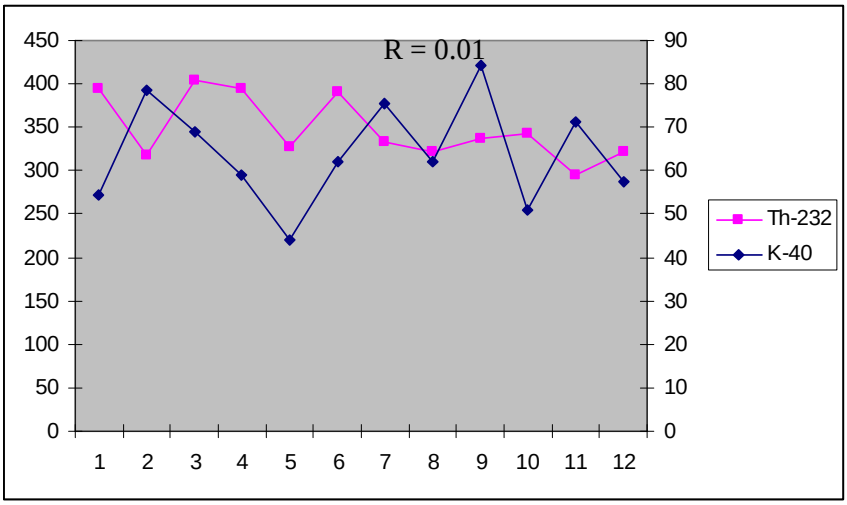

Figure 4: Correlation between K-40 and Th-232 Concentrations

Correlation coefficients of 0.14, 0.01 and -0.21 were obtained using the Spearman's formula between C_k/C_{Ra}, C_{Ra}/C_{Th} and C_k/C_{Th} respectively. Hence, the activities in samples are weakly correlated for both the positive and negative values.

Table 2: Radium equivalent activity, dose rate, annual effective dose and external hazard index at each point of measurement

Sample	Radium equivalent activity (Bq/Kg)	Dose rate (nGy/h	Annual effective dose (10^{-6}Sv)	External Hazard index
Soil 1	141.8	66.2	81.2	0.39
Soil 2	183.7	83.5	102.4	0.50
Soil 3	151.3	69.9	85.7	0.42
Soil 4	155.7	72.4	88.8	0.43
Soil 5	112.8	52.8	64.8	0.31
Soil 6	161.0	74.6	91.5	0.44
Soil 7	156.0	71.0	87.0	0.43
Soil 8	138.0	63.3	77.6	0.38
Soil 9	176.3	79.9	98.0	0.48
Soil 10	133.3	62.0	76.0	0.37
Soil 11	155.8	70.8	86.8	0.43
Soil 12	152.4	65.6	80.5	0.39
Range	141.8 – 183.7	52.8 – 83.5	64.8 – 102.4	0.31 – 0.50
Average	137.3	69.3	76.6	0.43

The results obtained for Ra_{eq} show that none of the values is higher than the established upper limit (370Bq/kg). they ranges from 141.8 – 183.7Bq/kg with a mean of 137.3Bq/kg and the soil is safe for use as constructing material.

The absorbed dose rates (D) due to gamma radiation in air at 1m above the ground surface for the uniform distribution of the naturally occurring radionuclides (^{226}Ra, ^{232}Th and ^{40}K) were obtained base on the guidelines provided by [10]. Using these guidelines, it is assumed that the contributions for other naturally occurring radionuclides were insignificant. Table 2 shows that the dose rate ranges from 52.8 – 83.5nGy/h with a mean of 69.3nGy/h. Both the range and the mean values are comparable with the global range of 18-93nGy/h and 59nGy/h respectively [5]. The range falls within those reported from other parts of Nigeria though differences exist for the means[5,9].

The values of external hazard indices ranges from 0.31 – 0.50 with a mean of 0.43. Since these values are less than unity, it follows that radiation hazard due to exposure to external radiation field from NORMs is negligible. Hence, Dangora populace are not likely to be affected by external radiation induced diseases as erythema, cataract and skin cancer due to the exposure to radiation field from NORMs. Since all concentrations and estimated risk factors are relatively moderate, the high concentration of gross activities reported earlier in Dangora water bodies, could be attributed only to the condition of aquifer - the oxidation state, dissolved solid etc[13]

Finally, the estimated annual effective dose, D ranges from 0.07 – 0.10mSv, with a mean value of 0.77mSv which is higher than the world average of 0.48mSv [5] but comparable with the recommended limit (0.07mSv) for normal background[5].

7.0 Conclusion

Activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th in surface soils from 12 different locations at Dangora town have been evaluated using gamma spectroscopy with NaI(Tl) detector. Mean concentrations of the radionuclides except that of ^{232}Th (which is higher) were found to be below their respective world background averages. When the concentrations were used to compute the risks for the assessment of radiation hazard indices, it was realized that the mean value for radium equivalent activities fell below the established upper limit, it was therefore concluded that the soil from the area of study is safe for use as building material. Similarly, calculation of absorbed dose rates at 1m above the ground revealed that the values obtained were comparable with the world mean and range. Computed values for external hazard indices fall below unity and it was concluded that radiation hazard due to exposure to background radiation is negligible. The estimated annual effective dose was found to have a range and mean that are comparable with the recommended limits for normal background. Finally the decay chains of both ^{226}Ra and ^{232}Th showed that both are alpha emitters and contribute to the reportedly high level of alpha radioactivity in the water bodies of the study area via factors as the oxidation state, dissolved solid etc. Due to the high level of ^{40}K recorded in this work, and its abundance in the environment, it is considered as the main contributor to the reported high level of beta activities in water screening test.

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