Measurement of Natural Radionuclides and Radon Gas Concentration in Surface Soil samples within Jalingo Metropolis, North East Nigeria.

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Abstract
Activity concentrations of radionuclides (\(^{226}\)Ra, \(^{232}\)Th, \(^{40}\)K) and radon gas in soil samples collected within Jalingo Metropolis were assessed by gamma spectrometric techniques using Na (TI) scintillation detector. The result showed an average activity concentration of \(^{226}\)Ra, \(^{232}\)Th and \(^{40}\)K to be 18.626±7.31 Bq/kg, 16.709±10.96 Bq/kg and 167.935±389.33 Bq/kg. The concentrations of \(^{226}\)Ra, \(^{232}\)Th were lower than the world average value while \(^{40}\)K was far higher that the recommended value. Most people in the study area use soil for building construction therefore, it was necessary to assess if there are any radiological hazards associated with the soil. This was achieved by determining Radium equivalent activity (Raeq), internal hazard index (Hin) and Annual effective dose rate. The result indicates that the indices are within normal limit. The Radon concentration in soil varies 11.126±1.315 kBq/kg to 30.374±3.331 kBq/kg with a mean value of 17.881±7.019 kBq/kg which is within the safety limits. Generally, the result showed that the soil in the study area might not pose major hazard to the members of the public.

Keywords: Radionuclides, Radon, Soil, Radioactivity, Environment.
Introduction

Radiation is the energy which is transported either in the form of particles or electromagnetic waves through space or material medium. The natural sources of radiation exposure to humans are artificial (medical applications, fertility application, mining etc) and natural (terrestrial and cosmogenic radionuclides) both radiation sources lead to internal and external radiation exposure. External exposure occurs due to gamma decay of primordial radionuclides while internal exposure results from the inhalation of airborne contaminants or ingestion of food and water contaminated by radionuclides [1].

The earth is naturally radioactive and about 90% of human radiation exposure arise from sources such as cosmic radiation, radon gas and terrestrial radionuclides. Their concentrations is dictated to a good degree by the underlying geological features of an area, its geographical location and anthropogenic activities [2] and [3]. Radon (\(^{226}\text{Ra}\)), a radioactive gas produced by the decay of naturally occurring radionuclide is a by-product in the uranium (\(^{238}\text{U}\)) series. Its three naturally existing isotopes namely \(^{222}\text{Rn}\), \(^{220}\text{Rn}\) and \(^{219}\text{Rn}\) are distinctively known as Radon, Thoron and Actinon respectively [4].

Radon and natural radionuclides have become subject of interest in recent times because of their detrimental effects on human health [5]. Acute and chronic exposure to radiation can cause adverse health effects such as cataract, lesions and stochastic effects such as cancer induction and hereditary diseases[6] and [7].

Jalingo’s natural environment (soil, water, air) have not been subjected to radiological regulatory control just like other regions in Northern Nigeria. Thus, Data on radionuclides and radon concentrations insurface soils and public exposure are scanty. Consequently, there is lack of awareness and knowledge of the radiological hazards emanating from surface soil and other components of environment. Hence, the urgent need to ascertain the radiological safety level of surface soils in Jalingo Metropolis

Materials and Method

Study Area.

Jalingo, the state capital of Taraba State has an estimated population of 11800(National Population Census,2006). Inhabitants of Jalingo are civil servants and farmers, fishermen and traders. Figure 1 is the map of Jalingo metropolis.

Figure 1: Map of Jalingo showing the sampling areas.
**Materials**

Measurement of Natural radionuclide and Radon gas concentration in Jalingo surface soils was performed using Gamma spectrometry system NaI (TI) detector (model: 802), 2mm sieve, Polyethylene sample bags, Nitric acid, and Multichannel etrex GPS meter.

**Surface Soil Samples Collection and Preservation**

The ten (10) soil samples were collected in a label polyethylene bag from the following areas Nukkai, Mile six, Mayogwai, Sabongari, NTA and along the bank of river Nukkai and Mayogwoi at a depth of 30cm, each sampling locations were carefully chosen to represent areas where human population is involve in various activities such as fishing, vegetable farming, extraction of building materials (sharp sand, gravel etc) and others. The samples were sun dried to constant weight and sieved using 2mm mesh to obtain a fine-powdered texture that will give an equilibrium level with the detector. The samples were then send to the laboratory at University of Ibadan for Radionuclides analysis.

**Activity measurement**

The activity concentrations of $^{40}$K, $^{226}$Ra, and $^{232}$Th in the prepared soil samples were measured using the gamma spectrometric technique. The gamma spectrometric system consists of a 7.62 × 7.62 cm Na (TI) scintillation detector (Bicron Corp model 3M/3), encapsulated in a 5.5-cm-thick lead shield to reduce environmental background radiation. The detector was coupled to a preamplifier (Bicron Corp Model PA-14), an amplifier (Canberra Model, 2022), and an analogue-to-digital converter (ADC) (Canberra Model 8075), which supplied an output to a Canberra S100 multi-channel analyzer (MCA). The activity concentration of $^{40}$K was measured from its gamma-ray energy of 1460 keV, and the transition lines of 1120.3 keV for $^{214}$Bi and 911 keV for $^{228}$Ac were applied for $^{226}$Ra ($^{238}$U series) and $^{228}$Ra ($^{232}$Th series), respectively.

A standard soil sample supplied by the International Atomic Energy Agency (IAEA), Vienna, Austria (Reference Material IAEA-375 for radionuclides and trace elements in soil), was used for the calibrations of the detector. The background radiation was considered as an empty container having the same geometry as the container of the standard sample and was counted for 25,200 s (7 h). Each of the prepared soil samples was counted for 7 h to determine the activity concentration of the radionuclides in them. The activity concentrations of the radionuclides in the samples were obtained using the comparative method according to Equation 1 [8]:

$$\frac{A_s}{A_{SD}} = \frac{N_s}{N_{SD}}$$

(1)

Where $A_s$ and $A_{SD}$ are the activity concentration (Bq/kg) of the sample and the reference sample, respectively, and $N_s$ and $N_{SD}$ are the net count rates under the region of interest for the sample and the reference (Standard) sample, respectively. The counting was performed in the Radiological Laboratory of Centre for Energy, Research and Development (CERD) University of Ibadan.

**Data Evaluation**

**The activity concentration of NORMs in soil samples**

The activity concentration of $^{226}$Ra, $^{232}$Th and $^{40}$K were calculated using the following equation 2 [9].

$$A_{sp} = \frac{N_{sam}\times\exp(\Delta T d)}{P\varepsilon TC \times \varepsilon \times M_{sam}}$$

(2)

$N_{sam}$ – Net counts of radionuclides in the sample

$P\varepsilon$ – Gamma ray emission probability (gamma yield)

$\varepsilon$ – Total counting efficiency of the detector system

$\Delta T d$ – delay time between sampling and counting

exp($\Delta T d$) – correlation factor between sampling and counting

$Tc$ – sampling counting time

$M_{sam}$ – mass of sampling (ka) or volume (L)
Assessment of radiological hazards in soil

The radiation hazards due to the natural nuclides $^{226}\text{Ra}$, $^{232}\text{Th}$ and $^{40}\text{K}$ were assessed by various radiation hazards indices.

i. **Radium equivalent activity** ($\text{Ra}_{\text{eq}}$): In the present study, the radium equivalent activity ($\text{Ra}_{\text{eq}}$) is given by the equation [3] [10].

$$\text{Ra}_{\text{eq}}(\text{Bq/Kg}) = 0.077C_k + C_{\text{Ra}} + 1.43C_{\text{Th}}$$ (3)

Where $C_k$, $C_{\text{Ra}}$ and $C_{\text{Th}}$ are the activity of Potassium, Radium and Thorium.

ii. **Internal hazards index** ($H_{\text{in}}$): Internal radiation hazards index is given by the equation (4)

$$H_{\text{in}} = \frac{C_{\text{Ra}}}{185} + \frac{C_{\text{Th}}}{259} + \frac{C_{\text{K}}}{4810}$$ (4)

Estimation of annual effective dose (AED) of NORMs in soil

To evaluate the year – long effective dose rates, the conversion coefficient from absorbed dose in the air to effective dose $(0.75\text{vGy}\cdot\text{Gy}^{-1})$ and outdoor occupancy factor $(0.25\text{vGy}\cdot\text{Gy}^{-1})$ suggested by [1] was applied.

- Indoor Effective Dose Rate (msv$^{-1}$):
  $$\text{Dose Rate } (\text{nGy}\cdot\text{h}^{-1} \times 8760) \times 0.8 \times 0.7\text{svGy}^{-1} \times 10^6$$ (5)

- Outdoor Effective Dose rate (msv$^{-1}$):
  $$\text{Dose rate } (\text{nGy}\cdot\text{h}^{-1} \times 8760\text{h}) \times 0.2 \times 0.7\text{svGy}^{-1} \times 10^6$$ (6)

**Radon in the soil**

The activity concentration of radon gas in the soil samples were calculated based on the radium (R-226) concentration using equation (7) [9].

$$C_{\text{Rn}} = C_{\text{Ra}} - 226 \times f \times \rho_s \times \varepsilon^{-1}(1 - \varepsilon) \times [\text{K}_T - 1] + 1)^{-1}$$ (7)

Where
- $C_{\text{Rn}}$ – is the concentration of radon-222 in (Bq/m$^3$)
- $C_{\text{Ra}}$ – activity concentration of dry mass of 226Ra in soil (Bq/Kg)
- $f$ – soil emanation factor for Rn222 = 0.2
- $\rho_s$ – density of soil (1600Kg/m$^3$)
- $\varepsilon$ – is the soil porosity (0.25)
- $K_T$ – is the radon partition coefficient between water and air phases and if the soil samples are dried before measurement, then, $m = 0$, the last term in equation (7) disappears.
- $m$ – is the porosity fraction, $m = 0$ for dry soil.

**Results and Discussion**

The result of activity concentration of radionuclide (Ra-226, Th-232 and K-40) and Radon gas; Radium equivalent index, internal hazard index, Annual effective dose and the correlation between Ra-226 and Rn-222 in the surface soil samples are presented in Table 1 – 5 and figure 2 below.

**Table 1:** Concentration of Natural Radionuclides In Soil Samples (Bq/Kg)

| Sample Location | Ra-226 | Th-232 | K-40      |
|-----------------|--------|--------|-----------|
| NUK-1           | 11.59±1.37 | 2.39±0.14 | 2152.68±109.95 |
| NUK-2           | 04±1.79   | 21.36±1.28 | 1804.19±92.39  |
| MLS-1           | 61±1.50   | 10.39±0.62 | 1035.79±53.39  |
| MLS-2           | 34±1.60   | 11.45±0.63 | 1020.89±52.24  |
| SBG-1           | 54±3.70   | 13.44±0.81 | 1669.47±85.49  |
| SBG-2           | 46±2.60   | 13.55±0.26 | 1517.59±80.59  |
| MYG-1           | 96±1.32   | 4.43±0.56  | 2101.53±89.26  |
| MYG-2           | 46±1.23   | 27.21±1.23 | 1889.23±80.34  |
| NTA-1           | 64±3.47   | 37.45±2.80 | 1848.46±82.45  |
| NTA-2           | 62±2.73   | 25.42±1.33 | 1689.52±80.32  |
| MIN             | 59±1.30   | 2.39±0.14  | 1020.89±52.24  |
| MAX             | 64±3.47   | 37.45±2.80 | 2152.68±109.95 |
| AVERAGE±STDV    | 626±7.31  | 16.709±10.96 | 1672.935±389.33 |
Table 2: Radium Equivalent Index \((Ra_{eq})\) and Internal Hazard Index \((H_{in})\)

| Sample Location | \(Ra_{eq}(Bq/Kg)\)     | \(H_{in}(Bq/Kg)\)     |
|-----------------|-------------------------|------------------------|
| NUK-1           | 180.764±10.036          | 0.519±0.031            |
| NUK-2           | 184.507±10.334          | 0.539±0.033            |
| MLS-1           | 107.224±6.498           | 0.324±0.216            |
| MLS-2           | 109.322±6.523           | 0.334±0.022            |
| SBG-1           | 169.308±11.441          | 0.515±0.041            |
| SBG-2           | 161.69±8.613            | 0.505±0.032            |
| MYG-1           | 181.113±8.994           | 0.524±0.028            |
| MYG-2           | 197.841±9.175           | 0.571±0.028            |
| NTA-1           | 227.525±22.403          | 0.700±0.070            |
| NTA-2           | 194.064±10.162          | 0.599±0.036            |
| MIN             | 107.224±6.498           | 0.324±0.216            |
| MAX             | 227.525±22.403          | 0.700±0.070            |
| AVERAGE±STDV    | 171.3358±37.693         | 0.513±0.112            |

Table 3: Annual Effective Dose Due to Ingestion of Norms \((Ra_{226}, Th_{232} \text{ and } K_{40})\) in Soil Samples

| Sample location | Indoors     | Outdoors    | \(\sum AED\)-in, out (msv/\text{y}) |
|-----------------|-------------|-------------|-------------------------------------|
| NUK-1           | 0.481±0.182 | 0.120±0.046 | 0.601±0.228                        |
| NUK-2           | 0.468±0.156 | 0.117±0.045 | 0.585±0.201                        |
| MLS-1           | 0.272±0.090 | 0.068±0.023 | 0.340±0.113                        |
| MLS-2           | 0.276±0.089 | 0.069±0.025 | 0.645±0.114                        |
| SBG-1           | 0.431±0.147 | 0.108±0.037 | 0.539±0.151                        |
| SBG-2           | 0.409±0.136 | 0.102±0.034 | 0.511±0.170                        |
| MYG-1           | 0.430±0.149 | 0.118±0.037 | 0.548±0.186                        |
| MYG-2           | 0.500±0.150 | 0.125±0.034 | 0.625±0.184                        |
| NTA-1           | 1.004±0.159 | 0.161±0.039 | 1.165±0.198                        |
| NTA-2           | 0.486±0.115 | 0.121±0.034 | 0.607±0.149                        |
| MIN             | 0.272±0.09  | 0.068±0.023 | 0.340±0.113                        |
| MAX             | 1.004±0.159 | 0.161±0.039 | 1.165±0.198                        |
| AVERAGE±STDV    | 0.476±0.203 | 0.111±0.027 | 0.591±0.211                        |

Table 4: Activity Concentration of Radon Gas \((Rn_{222})\) Calculated from the Soil Samples

| Sample location | Ra-226 Bq/Kg | Rn222(KBq/m) |
|-----------------|-------------|--------------|
| NUK-1           | 11.59±1.370 | 11.126±1.315 |
| NUK-2           | 15.04±1.790 | 14.438±1.718 |
| MLS-1           | 12.61±1.500 | 12.106±1.440 |
| MLS-2           | 14.34±1.600 | 13.766±1.536 |
| SBG-1           | 21.54±3.700 | 20.678±3.552 |
| SBG-2           | 25.46±2.600 | 24.442±2.496 |
| MYG-1           | 12.96±1.320 | 12.442±1.267 |
| MYG-2           | 13.46±1.230 | 12.923±1.181 |
| NTA-1           | 31.64±3.470 | 30.374±3.331 |
| NTA-2           | 27.62±2.730 | 26.515±2.621 |
| MIN             | 11.59±1.370 | 11.126±1.315 |
| MAX             | 31.64±3.470 | 30.374±3.331 |
| AVERAGE±STDV    | 18.626±7.311 | 17.881±7.019 |
Table 5: Comparison of Soil Gas Radon-222 with other Studies around the World

| Country | Sample Location | Measurement techniques | Concentration (R-22) Bq/m³ | Reference |
|---------|-----------------|------------------------|---------------------------|-----------|
| Nigeria | Jalingo         | NaI(Tl)                | 11,126-30,374             | This study|
| Ghana   | Mine            | HPGe                   | 12,500-41,300             | Faanu, (2011)|
| Ghana   | Fault           | Alpha Guard            | 9,910-42,010              | Amponsah P, (2008)|
| Sudan   | Soil            | SSNTD                  | 5,500-15,100              | Elmonien, (2015)|
| India   | -               | RAD7 Radon meters      | 3,200-17,200              | Mehra et al, (2015)|
| Russia  | -               | -                      | 17,000-24,000             | Iakovleva et al, (2003)|
| Italy   | Volc/mountain   | RAD7 meters            | 232-104,300               | Giammanco et al, (2007)|
| Turkey  | Geotherm area   | SSNTD                  | 98-8594                   | Tabar et al, (2013)|
| India   | Upper siwalik   |                        | 11500-78470               | Singh et al, (2010)|

Figure 2: Correlation between Ra-226 and Rn-222 in soil sample

Table 1, shows that $^{226}$Ra activity concentration ranges from 11.59±1.30 to 31.64±3.47Bq/Kg with an average value of 18.62±7.31Bq/Kg; $^{232}$Th concentration ranges from 2.39±0.14 to 37.45±2.80Bq/Kg with an average value of 16.70±10.96Bq/Kg while $^{40}$K concentration ranges from 1020.89±52.24 to 2152.68±109.95Bq/Kg with an average value of 1672.93±389.33Bq/Kg. In comparison, $^{226}$Ra and $^{232}$Th concentrations were found to be below the world average value of 35 and 30Bq/Kg. The low concentration might be attributed to low geochemical composition of uranium associated with sedimentary rock forming soils in the study area. The concentration of $^{40}$K were found to be far higher than the world average value of 400Bq/Kg. Its high concentration may be due to their relative abundance in the environment and the application of fertilizer rich in $^{40}$K on farm lands within the metropolis.

The calculated Radium equivalent activity (Ra$_{eq}$) values ranged from 107.22±6.498 to 227.52±22.403Bq/Kg with average value of 171.33±37.693Bq/Kg while the Internal Hazard Index (H$_{in}$) was found to range from 0.34±0.216 to 0.70±0.070Bq/Kg with an average 0.51±0.112Bq/Kg. This shows that the estimated value of Ra$_{eq}$ is less than the global limit of 370Bq/Kg and that of H$_{in}$ is within the acceptable limit of 1(Table 2).

Table 3 shows the total annual effective dose due to external and internal gamma dose which
ranged from 0.340±0.113 to 1.165±0.198 mSv/y with an average of 0.591±0.211 mSv/y and is also below the world value of 1 mSv [1].

The radon (Rn-222) concentration shown in Table 4 varies from 11.126±1.315 KBq/Kg to 30.374±3.331 KBq/Kg with a mean value of 17.881±7.019 KBq/Kg. A comparison of radon concentration in soil samples from different location around the world presented in table 5 shows clearly that the radon concentration in soil samples in Jalingo metropolis is within the safety limit. Figure 2, the plot of Ra-226 against Rn-222 shows a strong positive correlation between Ra-226 and Rn-222. This implies that the radon gas in the soil comes from Ra-226.

CONCLUSION

The result obtained indicates that the average activity concentration of $^{266}$Ra and $^{232}$Th is less than world average value of 35 and 30 Bq/kg while that of $^{40}$K is higher than the average world value for soil samples [1]. Findings further revealed that all the radiological indices are within the safety limits [1]. Though, the activity concentration and radon gas concentration in surface soils within the region are relatively low but continues exposure to these radiations may cause severe health hazards to the inhabitants of Jalingo metropolis.

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