Environmental Radiology of Southern Part of Zamfara State, Nigeria

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Abstract:  
The activity concentrations of the naturally occurring radionuclide for the soil samples collected from Southern Zamfara State were measured using Hyper-pure Germanium detector (HPGe). The terrestrial gamma radiation doses were measured using inspector alert, model 35448 manufactured by U.S.A from 250 locations across southern Zamfara. The mean activity concentrations of 226Ra, 232Th and 40K in the soil samples were found to be 55 Bq kg⁻¹, 35 Bq kg⁻¹ and 267 Bq kg⁻¹, respectively. The activity concentration of 232Th, and 40K obtained are within the recommended values of 45 Bq kg⁻¹ and 420 Bq kg⁻¹, while 226Ra concentration 55Bqkg⁻¹ is higher than the world average value of 32 Bq kg⁻¹. The annual effective dose (AED), external (H_e) and internal (H_i) hazard index and Gamma representative index were computed as 0.07 μSv⁻¹, 0.33, 0.98 and 0.81, respectively. The values are within the recommended safe limit of unity. Other parameters such as annual gonadal dose equivalent, radium equivalent activity (Ra_eq) and excess lifetime cancer risk were also computed as 390.61 μSv⁻¹, 123.73 Bq kg⁻¹ and 0.34. The values obtained for annual gonadal dose equivalent and excess lifetime cancer risk are higher than the recommended values of 300 μSv⁻¹ and 0.29, while radium equivalent activity (Ra_eq) is lower than the safe limit 370 Bq kg⁻¹. The result implies that the gonadal values may have some effects on the reproductive organs of the people in the study area. But other radiation hazard parameters show that it is safe to use the soil in the study area for farming and other activities.

Keywords: Natural Radioactivity, radiation hazards parameters. HPGe Detector

1. Introduction
Soils contain radioactive materials naturally [1]. The radionuclides found in soils are mostly primordial radionuclides 226Ra, 232Th and their decay products and 40K[2]. Natural environmental radioactivity and the associated external exposure due to gamma radiation depend primarily on the geological and geographical conditions and appear at different levels in the soils of each region in the world [3,4]. The higher concentrations of radium, thorium and potassium are associated with soil developed from acid intrusive rocks and clay [5,6] and probably, a majority of uranium is associated with the phosphates sands and clays of these formations [7]. Knowing the concentration levels of natural radionuclides in soil and their distribution in the environment is of great interest in several fields of science [8]. Igneous rocks such as granite have higher radiation levels, while lower levels are associated with sedimentary rocks, although some shale and phosphate rocks have a relatively high content of radionuclides[9].
2. Materials and Methods

2.1. Study Area

The study area is Southern part of Zamfara State Nigeria. The region consist of seven local government including Anka, Bukuyum, Bungudu, Gusau, Gummi, Maru and Tsafe bounded by Latitude 0°0′0″ and 12°15′0″ N and Longitude 5°0′0″ and 7°15′0″ E. These coordinates are dominated mostly by the following geological formations: Taloka formation; Gundumillo formation; Younger meta volcano sedimentary series; Older granitolds (Pan African); Older meta-sedimentary rock; Gneiss migmatite as can be seen in Figure 1 and Table 1.

![Figure 1: Geology Map of the Study Area Showing the Six Geological Formations [10]](image)

| Geological Level | Geological Name                  | Composition                                                                 |
|------------------|----------------------------------|-------------------------------------------------------------------------------|
| A1, A2 and A3    | Taloka                           | Sandstones, silt and shale, clays and shales, tuff with sandstones            |
| B1, B2 and B3    | Gundumillo                       | Clays grits and pebbles, gravels and sand, Agglomerate with sandstone         |
| C1, C2 and C3    | Younger meta volcano sedimentary series | Rhyolite                                                                        |
| G1, G2 and G3    | Older granitolds                 | Hypersthene quartz diorite, quartz syenite, fine-grained biotite granite, medium-coarse, Grained biotite, biotite-homblende granite, biotite homblende granodiorite quartz, coarse granite, granite-gneiss and migmatite. |
| M1, M2 and M3    | Older metasedimentary Rock       | banded irion formation, biotite and muscovite schists, quartzite and quartz schists, slate, phylite meta-siltstone locally homfelsic, undifferentiated muscovite, felspathic schist, metaconglomerate and quartzite |
| R1, R2 and R3    | Gneiss migmatite                 | Migmatite granite gneiss and silicified and sheared rock                      |

Table 1: The Geological Formations of Zamfara South and Their Corresponding Compositions

2.2. Sample Collection

Soils samples were collected from the study area at 18 different locations across the entire southern part of the state (Figure 2). The surface vegetation and death organic matter from the surface of each sample location were removed before taking each soil sample. The coordinate of each soil sample was located using Global Positioning system (GPS), each soil sample at the collection point was placed in a plastic bag, labeled with date and coordinate for easy identification. The 18 soil samples collected for the study area were transported back to the material science laboratory of Ahmadu Bello University for preparation.
2.2.1. Sample Preparation

Each sample was dried in an oven at a temperature of about 100°C for 24 hours in order to remove the moisture content, each dried sample was crushed to pass through a fine mesh sieve (2mm) to homogenous size. Then, each sample was packed in a plastic container selected based on the detector geometry and carefully sealed using masking tape in order to prevent Radon gas from escaping to the atmosphere. The prepared samples were allowed to attain secular radioactive equilibrium between $^{226}$Ra and its daughters by storing it for the period of a month. The activity concentrations of these samples were measured using hyper pure germanium detector (HPGe).

2.3. Experimental Set-Up

HPGe detector of serial and model number 9744 and GC 8023 was used; the detector has a relative efficiency of 80% with an energy resolution of 2.3KeV for 1.33 MeV gamma ray emission of $^{60}$Co. the length and end cap diameter of the detector is 69.8mm and 78mm. the set up was connected to a multichannel analyzer and a computer based data acquisition system. Efficiency calibration of the detector was done using $^{155}$Eu (60KeV, 86.5KeV and 105.3 KeV), $^{125}$Sb (176.3 KeV, 427.9 KeV and 600.6 KeV), $^{54}$Mn (834.8 KeV), $^{65}$Zn (1115.5 KeV), and $^{40}$K (1460.8 KeV). The energy calibration was also done using a standard source of $^{60}$Co (1173.2 and 1332.5KeV), $^{241}$Am (59.54 KeV), $^{133}$Ba (356.1 KeV) $^{137}$Cs (661.9 KeV) and $^{23}$Na (1368.6 KeV).

2.3.1. Sample Analysis

Each soil sample was set counting time of 10800s. The count rate per second for each radionuclide in each sample was analyzed and the background count was subtracted for every count. The activity concentration of $^{226}$Ra was determined from $\gamma$-ray energies 609.31 KeV, the activity concentration of $^{40}$K was determined from the $\gamma$-ray energy of 1460.80 KeV and the activity concentration of $^{232}$Th was determined from the $\gamma$-ray energy of 911.1 KeV.

2.4. Radiological Hazards Parameters

The mean activity concentrations of $^{226}$Ra, $^{232}$Th, and $^{40}$K measured in the soil samples were used to calculate the Radiological hazards parameters using the following expressions. Where in all the expressions, $A_{Ra}$, $A_{Th}$ and $A_{K}$ are the activity concentration of $^{226}$Ra $^{232}$Th and $^{40}$K, respectively.

2.4.1. Absorbed Dose Rates

The absorbed dose rate was obtained using the expression below [11]

$$D = (0.462A_{Ra} + 0.604A_{Th} + 0.0417A_{K})$$ (1)

Where D is the absorbed dose rate in mGy.h$^{-1}$.

2.4.2. Annual Effective Dose Rates

The expression used in calculating the annual effective dose rates was given below [12]:

$$AEDE = D \times 8760h \times 0.2 \times 0.7 \times 10^{-6}$$ (2)

Where AEDE is the annual effective dose equivalent in ($mSvyr^{-1}$) and D is the value of absorbed dose rate earlier calculated.
2.4.3. Radium Equivalent Activity
The Radium Equivalent Activity (Ra$_{eq}$), which is the actual activity level of $^{238}$U, $^{232}$Th and $^{40}$K in soil samples [13] was calculated using the expression below.

\[ \text{Ra}_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_{K} \]  

(3)

2.4.4. External Hazard Index
To limit the radiation exposure due to natural radionuclides in the samples to a maximum permissible limit of 1 mSv.y$^{-1}$, the External Hazard Index (Hex) was introduced [14]. It was computed by the Equation below [11]:

\[ H_{ex} = \left( \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_{K}}{4810} \right) \]  

(4)

2.4.5. Internal Hazard Index
The internal hazard index was calculated using equation (5).

\[ H_{in} = \left( \frac{A_{Ra}}{180} + \frac{A_{Th}}{259} + \frac{A_{K}}{4810} \right) \]  

(5)

2.4.6. Excess Lifetime Cancer Risk (ELCR)
Equation 6 was used in obtaining the Excess lifetime cancer risk (ELCR) of the study area.

\[ \text{ELCR} = AEDE \times (RF) \times (DL) \]  

(6)

where AEDE, DL and RF is the annual effective dose equivalent, average duration of life and risk factor.

2.4.7. Gamma Representative Index
The expression used in calculating the Gamma representative index was given in equation (7) below.

\[ I_{G} = \frac{A_{Ra}}{150} + \frac{A_{Th}}{100} + \frac{A_{K}}{1500} \]  

(7)

2.4.8. Annual Gonadal Equivalent Dose
Equation 8 was used in calculating the annual gonadal dose equivalent of study area.

\[ AGDE = 3.09 \ A_{Ra} + 4.18 \ A_{Th} + 0.314 \ A_{K} \]  

(8)

3. Results and Discussion

3.1. Activity Concentrations Based on Geological Formation
Activity Concentration for $^{226}$Ra, $^{232}$Th, and $^{40}$K in each geological formation was presented in Figure 3 and Table 2. $^{232}$Th concentration in the six geological formations ranges from 45.80 to 26.71 Bqkg$^{-1}$, with a mean value of 34.55 ± 7.99 Bqkg$^{-1}$.$^{226}$Ra ranges from 77.47 to 37.15 Bqkg$^{-1}$, with a mean value of 54.19±15.47 Bqkg$^{-1}$. While $^{40}$K concentration ranges from 75.54 to 394.72 Bqkg$^{-1}$, with a mean value of 260.00 ± 92.62 Bqkg$^{-1}$. Gneiss Migmatite formation has the highest mean concentration of $^{232}$Th, $^{226}$Ra and $^{40}$K, while Older Meta Sedimentary Rock formation has the lowest mean concentration of $^{226}$Ra and $^{40}$K, the lowest concentration of $^{232}$Th was found in Older Granitolds formation. The variation of this activity concentration across the geological formation is due to the variation of the mineral contents present in each geological formation.

\[ \text{Figure 3: Activity Concentrations Based on Geological Formation} \]
3.2. Activity Concentrations Based on Local Government

Figure 4 shows that the activity concentration for $^{232}$Th in the seven local government of the study area ranges from 26.81 to 41.39 Bq kg$^{-1}$, with a mean value of 39.31 ± 7.34 Bq kg$^{-1}$. That of $^{226}$Ra ranges from 39.40 to 63.79 Bq kg$^{-1}$, with a mean value of 51.44 ± 8.38 Bq kg$^{-1}$. $^{40}$K concentration ranges from 80.59 to 346.15 Bq kg$^{-1}$ with a mean value of 260.00 ± 70.04 Bq kg$^{-1}$. Based on the results obtained, Bukuyum local government has the highest mean concentration of $^{40}$K, 346.15 ± 8.72 Bq kg$^{-1}$ and while the lowest mean concentration of $^{40}$K and $^{232}$Th was observed in Maru local government. In terms of $^{226}$Ra, Gusau local government has the highest mean concentration of 63.79 ± 9.05 Bq kg$^{-1}$ while Tsafe local government has the lowest mean concentration of $^{226}$Ra, 39.40 ± 5.20 Bq kg$^{-1}$. The mean concentration of $^{232}$Th, was observed to be high in Gummi local government.

![Figure 4: Activity Concentrations Based On Local Government Formation](image)

3.3. Comparison of the Mean Activity Concentration with Other Published Data

The result show that, activity concentration obtained for $^{40}$K in the study area is higher than the value obtained in Abeokuta [15] but lower than the values obtained in Bagega [16], Zamfara North [17], Sri lanka [18], Egypt [19], Algeria [20] and Zambia [21]. Table 2. In terms of $^{226}$Ra, the values obtained in the study area is lower than the values obtained in Algeria but higher than those obtained in Bagega, Abeokuta, Sri lanka and Zambia. For $^{232}$Th, the concentration obtained is lower than the values obtained in Sri lanka, Egypt, Algeria and Zambia. But higher than that obtained in Zamfara North, Bagega Nigeria and Abeokuta Nigeria.

| Region         | $^{40}$K | $^{226}$Ra | $^{232}$Th | References               |
|----------------|---------|-----------|-----------|--------------------------|
| study area     | 260     | 54        | 35        | This work (2019)          |
| Zamfara North  | 269     | 23        |           | Christopher et al. (2018) |
| Bagega Nigeria | 371     | 18        | 17        | Girigisu et al. (2013)   |
| Abeokuta Nigeria | 52   | 12        | 23        | Gbadebo (2011)           |
| Sri lanka      | 584     | 35        | 72        | Hewamannyaet al. (2001)  |
| Egypt          | 276     | 54        | 63        | Uosif et al. (2011)      |
| Algeria        | 675     | 65        | 51        | Amraniet et al. (2001)   |
| Zambia         | 412     | 32        | 82        | Hayambuer et al. (1995)  |

Table 2: Comparison of the Mean Activity Concentration with Other Published Data

3.4. Radiological Indices

Table 4 and 5 present the results obtained for the hazards parameters. The annual gonadal dose equivalent obtained (390.6 μSv y$^{-1}$) was above the permissible limit of 300 μSv y$^{-1}$, the absorbed gamma dose rate 56 nGy h$^{-1}$ obtained in the study area is below the permissible limit of 59 nGy h$^{-1}$[11]. This implies that the gonadal values may pose some threat to the reproductive organs of the people in the study area. The mean radium equivalent activity found was 123.729 ± 32.166 Bq kg$^{-1}$ lower than 370 Bq kg$^{-1}$ recommended by [11]. Any radium equivalents activity (Ra$_{eq}$) concentration value
that exceeds 370 Bq kg⁻¹ may pose radiation hazards [14]. The radium equivalents activity (Ra eq) ranged from 71.825 to 185.26 Bq kg⁻¹. The annual effective dose rate ranged from 0.037 to 0.105 μSv⁻¹ with an average value of 0.069 ± 0.020 μSv⁻¹. The mean value of external and internal radiation hazard index was found to be 0.33 ± 0.09 and 0.98 ± 0.26, lower than the recommended value of 1 μSv⁻¹ [11]. The average value obtained for excess lifetime cancer risk was 0.34 higher than the world average value of 0.29 [11]. The higher values of ELCR may be attributed to the higher concentrations of radionuclide in the geological formations. Gamma representative index measured ranged from 0.16 to 1.33 with a mean value of 0.81±0.27 for the soil samples. This index must be lower than unity to keep the radiation hazard insignificant. However, a mean value obtained in this study is lower than the reference level of 1 [11].

| Sample Code | Hₑₓ | Hᵣ | AEDE | Iᵣ | ELCR (10⁻³) |
|-------------|-----|-----|------|-----|-------------|
| A1          | 0.41±0.20 | 0.56±0.23 | 0.09±0.06 | 1.06±0.31 | 0.32         |
| A2          | 0.47±0.19  | 0.51±0.25 | 0.10±0.06 | 1.24±0.30 | 0.35         |
| A3          | 0.44±0.18  | 0.58±0.24 | 0.06±0.06 | 0.16±0.28 | 0.21         |
| B1          | 0.26±0.17  | 0.35±0.10 | 0.55±0.06 | 0.69±0.27 | 1.93         |
| B2          | 0.24±0.16  | 0.33±0.11 | 0.05±0.06 | 0.64±0.25 | 0.18         |
| B3          | 0.38±0.16  | 0.50±0.21 | 0.08±0.06 | 1.02±0.29 | 0.28         |
| C1          | 0.36±0.18  | 0.47±0.21 | 0.06±0.06 | 0.74±0.28 | 0.28         |
| C2          | 0.35±0.17  | 0.46±0.21 | 0.07±0.06 | 0.93±0.27 | 0.25         |
| C3          | 0.29±0.18  | 0.41±0.22 | 0.06±0.06 | 0.74±0.29 | 0.21         |
| G1          | 0.27±0.14  | 0.39±0.17 | 0.06±0.05 | 0.72±0.23 | 0.21         |
| G2          | 0.19±0.15  | 0.27±0.17 | 0.04±0.05 | 0.52±0.23 | 0.14         |
| G3          | 0.28±0.16  | 0.37±0.18 | 0.04±0.05 | 0.73±0.24 | 0.14         |
| M1          | 0.21±0.16  | 0.29±0.18 | 0.04±0.05 | 0.54±0.25 | 0.18         |
| M2          | 0.21±0.15  | 0.30±0.18 | 0.04±0.05 | 0.56±0.24 | 0.14         |
| M3          | 0.24±0.16  | 0.44±0.23 | 0.05±0.06 | 0.62±0.26 | 0.18         |
| R1          | 0.46±0.18  | 0.82±0.27 | 0.10±0.06 | 1.20±0.30 | 0.35         |
| R2          | 0.45±0.20  | 0.77±0.28 | 0.09±0.06 | 1.19±0.31 | 0.32         |
| R3          | 0.5±0.18   | 0.86±0.27 | 0.11±0.06 | 1.33±0.28 | 0.39         |
| Range       | 0.19 to 0.5 | 0.27 to 0.86 | 0.04 to 0.11 | 0.16 to 1.33 | 0.14 to 0.39 |
| Mean        | 0.33±0.09  | 0.98±0.26 | 0.07±0.02 | 0.81±0.27 | 0.34         |
| World average | 1      | 1       | 1     | 1     | 0.29 (10⁻³) |

Table 3: Activity concentrations (in Bq kg⁻¹) in soil

Table 4: Radiological Hazards Indices
## 4. Conclusion

The results obtained show that the activity concentrations of $^{232}$Th, and $^{40}$K are within the recommended values of 45 Bq kg$^{-1}$ and 420 Bq kg$^{-1}$ [11] while the mean value of $^{226}$Ra obtained is higher than the safe limit of 32 Bq kg$^{-1}$. The estimated annual effective dose rates ranged from 0.04 to 0.1 $\mu$Sv $^{-1}$ with a mean value of 0.07 ± 0.02 $\mu$Sv $^{-1}$. The radium equivalent activity (Ra$_{eq}$) ranged from 71.83 to 185.0 Bq kg$^{-1}$, with a mean value of 123.73 ± 32.17 Bq kg$^{-1}$ which is lower than the accepted safety limit of 370 Bq kg$^{-1}$[11]. The calculated values of the external and internal hazard index for all soil samples studied varied from 0.19 to 0.5 and 0.27 to 0.86 respectively. The results show that the external and internal hazards indices are below the permissible limit of 1 mSv $^{-1}$ recommended by the International Commission on Radiological Protection (ICRP). Other parameters such as annual gonadal dose equivalent and excess lifetime cancer risk were also computed as 390.61 $\mu$Sv $^{-1}$ and 0.34. The annual gonadal dose equivalent and excess lifetime cancer risk values obtained are higher than the recommended values of 300 $\mu$Sv $^{-1}$ and 0.29 [11], the gonadal dose equivalent values may pose some threat to the reproductive organs of the people in the study area. The values of other radiation hazard parameters obtained for the current study are lower than the recommended values, and therefore confirms that it is safe to live in that area. It also shows that the soils from this area can be safely use for construction of houses and other civil engineering structures.

## 5. Acknowledgement

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### Table 5: Radiological hazards indices in the area

| Sample ID | DR (nGy h$^{-1}$) | Ra$_{eq}$ (Bq kg$^{-1}$) | AGDE (µSv y$^{-1}$) |
|-----------|------------------|----------------------|------------------|
| A1        | 69.18 ± 2.52     | 150.75 ± 3.73        | 476.38 ± 6.55    |
| A2        | 80.56 ± 2.47     | 174.22 ± 3.65        | 558.40 ± 6.43    |
| A3        | 51.81 ± 2.28     | 164.49 ± 3.39        | 514.84 ± 5.96    |
| B1        | 44.51 ± 2.18     | 96.48 ± 3.24         | 260.69 ± 5.27    |
| B2        | 41.48 ± 2.06     | 89.57 ± 3.05         | 288.90 ± 5.38    |
| B3        | 65.89 ± 2.38     | 141.19 ± 3.52        | 460.44 ± 6.20    |
| C1        | 61.40 ± 2.34     | 131.84 ± 3.46        | 428.59 ± 6.09    |
| C2        | 59.56 ± 2.22     | 128.21 ± 3.29        | 414.56 ± 5.69    |
| C3        | 48.57 ± 2.34     | 108.73 ± 3.48        | 330.62 ± 6.09    |
| G1        | 46.43 ± 1.85     | 101.54 ± 2.77        | 321.93 ± 4.85    |
| G2        | 32.91 ± 1.89     | 71.83 ± 2.80         | 227.28 ± 4.92    |
| G3        | 29.97 ± 2.01     | 102.29 ± 2.99        | 326.95 ± 5.24    |
| M         | 34.23 ± 2.01     | 76.55 ± 3.00         | 235.47 ± 5.25    |
| M         | 35.76 ± 1.92     | 79.48 ± 2.86         | 245.20 ± 5.01    |
| M         | 40.38 ± 2.11     | 89.88 ± 3.12         | 276.91 ± 5.49    |
| R1        | 77.80 ± 2.41     | 169.16 ± 3.57        | 537.85 ± 6.15    |
| R2        | 76.61 ± 2.53     | 165.88 ± 3.74        | 532.68 ± 6.59    |
| R3        | 85.34 ± 2.39     | 185.26 ± 3.52        | 593.78 ± 6.22    |
| Range     | 29.97 to 185.34  | 71.83 to 185.26      | 227.28 to 593.78 |
| Mean      | 56.02 ± 16.11    | 123.73 ± 32.17       | 390.61 ± 5.74    |

World average: 59 370 300
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