

Radionuclide contents in raw minerals and soil samples and the associated radiological risk from some mining sites in Benue State North-Central Nigeria

N.N Jibiri and S.T Temaugee*

Abstract- A total of fifty eight (58) samples in which nine (9) minerals and forty nine (49) soil samples were collected from five mining sites of Clay, Kaolin, Limestone and Baryte in Benue State, North Central, Nigeria. Radioactivity concentration was measured using NaI(Tl) γ -ray spectroscopy with an accumulating time for about 36000secs. From the measured γ -ray spectra, activity concentration of the primordial radionuclides ^{40}K , ^{226}Ra (^{238}U), and ^{232}Th for the mineral samples ranged from 74.31-441.24 Bq kg⁻¹, 15.28-111.67 Bq kg⁻¹, and 8.99-51.39 Bq kg⁻¹ respectively. The mean value of the measured activity concentration of the radionuclides in the soil samples at the mining sites were 425.92±216.06, 40.34±12.58, 33.69±4.73 Bq kg⁻¹ for ^{40}K , ^{226}Ra (^{238}U), and ^{232}Th respectively. The total Absorbed dose rate in air at the mining sites ranged from 52.25-64.00 nGy h⁻¹ with mean of 57.32±4.44 nGy h⁻¹ resulting to a corresponding annual effective dose equivalent of 63.2 μSvyr^{-1} to 78.7 μSvyr^{-1} and mean 71±6 μSvyr^{-1} . The radium equivalent activity (R_{eq}), external hazard index (H_{ex}), internal hazard index (H_{in}) and the gamma activity index (I_{yr}) were also estimated and compared with the international recommendation for radiological safety. Results of the study indicate that the minerals and soil samples from the sites will pose no health risk to the populace if used for building and construction, ceramics and pottery production in the area.

Keywords: Absorbed dose, Activity Concentration, Hazard index, Radionuclides.



1. Introduction

The natural radioactivity concentration of raw minerals depends on their mineralogy and geochemistry, and a few raw minerals are occasionally found to have comparatively high concentrations of natural radioactivity [9]. This is in line with the observation of Habshi [15] that; the specific levels of terrestrial environmental radiations are related to the composition of each lithologically separated area, and to the content of the rock from which the soils originate. Therefore, knowledge of radiation levels and basic radiological parameters in building materials is essential to assess possible radiological risk to human health.

The presence of ^{40}K , ^{238}U , and ^{232}Th , in the earth crust contributes to small but measurable amount of naturally occurring radioactivity. The environmental effect of radiation from mineral resources depends on the use of the raw material or end product of such minerals in the environment.

The end products of mineral resources have a variety of uses by human; Cement from Limestone is used extensively in building of houses and several construction purposes. Due to inherently complex physical, chemical and mineralogical characteristics, clays are used mainly in the manufacture bricks, Portland and other cements, concrete blocks and structural concrete, refractory and ceramics (electrical porcelain, dinnerware, floor and wall tiles, and pottery). Clay has been a major source for building of

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thatched houses in Benue State. Kaolin is clay that usually contains 85–95% of kaolinite, which is formed by rock weathering. In addition to kaolinite, it frequently contains quartz, mica, feldspar and illite. Kaolin is widely used as a raw material in the paper industry, ceramics, refractory, bricks, cement, textiles, rubber, medical industries and special types of plastics. Kaolin is utilized in pottery and ceramic industries for ceramic wares; Baryte is the main source of barium and its compounds. It may be used as an aggregate in “heavy” cement, for which it is crushed and screened to a uniform size. Most baryte is ground to a small, uniform size and then used as filler in the production of such goods as paper, textiles, linoleum, rubber, and plastics. In addition, it is used in paints and in a special type of mud for sealing petroleum wells in the course of drilling. In the study area people use Baryte for the construction of their buildings where they dwell.

Building and industrial materials contribute to environmental radioactivity in two ways, first by gamma radiation mainly from ^{238}U , and ^{232}Th , ^{40}K , and their progenies to whole body dose and in some cases by beta radiation to a skin dose, and secondly by releasing the noble gas radon, its radioactive daughters which are deposited in human respiratory tract. Enhanced or elevated level of natural radioactivity in such materials may cause doses in order of several mSv yr^{-1} [17].

Due to health risk associated with the exposure to indoor radiations, many governmental and international bodies such as the International Commission on Radiological protection (ICRP), the World Health Organization (WHO), etc, have adopted strong measures aimed at minimizing such exposures. In this context limits have been set on the concentrations of radionuclides in various building materials and the use of materials with abnormally high levels of radioactivity has been banned [13]. This has called for the measurement of natural radioactivity in all regions of the world. Hence, the measurement of natural radioactivity is needed to implement precautionary measures

whenever the dose rate is found to be above the recommended limits [4].

The estimation of the exposure levels in building materials has gained the interest of much environmental health Physicist in recent times. Many researches carried out gave useful information on the exposure levels in building material like; cement, brick, concrete, clay, kaolin, marble, pumice, sand, etc. in some parts of the world [5], [6], [17], [3], [2], [19], [14]. However, there is no published data existing presently on the concentration of these radionuclides in Benue state, Nigeria, especially for the raw minerals considered in this study.

2 Materials and Methods

2.1 Sample collection and preparation

Five major mining sites in Benue state were identified. These are mining sites from which the raw minerals, Limestone, Kaolin, Clay and Baryte are obtained in the state. At each site two (2) samples of the type of mineral mined were collected. Also about 10 soil samples were collected at different locations around the mining area. The soil samples were collected to a depth of 150mm at each site. A total of nine (9) mineral samples and forty-nine (49) soil samples were collected around the mining areas. A map of the study area is shown in Fig 1.

The collected samples were air-dried and pulverized to 1mm² mesh sizes with a jaw crusher and Ceramic mortar, and then homogenized. The mass of each of the final powdered sample used for the analysis was 200 g. The powdered samples were hermetically sealed in plastic containers which had been verified to be non-radioactive, so as to prevent the escape of ^{222}Rn , the daughter of ^{226}Ra . The sealed samples were stored for 30days so as to allow radium (^{226}Ra) and its short-lived progenies to reach secular equilibrium.

2.2 Radioactivity measurement

The quantitative measurement of natural radioactivity in the soil and minerals samples was done using a 7.6cm × 7.6cm NaI (TI) detector (Model No 802-series) by Canberra Inc. This is coupled

to a Canberra series 10 plus Multi-Channel Analyzer (MCA) (Model No 1104) through a preamplifier base. The detector has a resolution of about 8% at 0.662MeV energy of ¹³⁷Cs. This gave photo peaks well defined enough to distinguish the gamma-ray energies considered during these measurements. The 1.460 MeV peak was used for ⁴⁰K, 2.615 MeV for ²³²Th, and 1.750 MeV for ²³⁸U.

The energy calibration of the spectrometer was performed using certified Reference material for radiometric measurement from the International Atomic Energy Agency (IAEA), Vienna. Sources used for energy calibration were ²²Na, ¹³⁷Cs, ⁶⁰Co, ⁴⁰K, ²³⁸U, ²³²Th emitting γ -rays with energies ranging between 0.511 to 2.615MeV. The detection efficiency calibration of the detector was performed using a reference soil sample, prepared by Rockedyn Laboratories, CA, which is traceable to a mixed standard gamma source (No. 48772-356, from Analytic Inc. Atlanta, Georgia (USA)). The reference sample has a similar matrix to the soil samples. The source was counted for 36000s and count rate above the background (C_s) for each of the radionuclides was recorded. The efficiencies for each radionuclide were calculated and used to estimate the activity concentration of each of the radionuclide in the samples. The detection efficiency of the system was determined using equation 1 [12]:

$$\epsilon_p = \frac{C_s}{tA_r Y m} \quad 1$$

Where, C_s is the net count above the background after counting

known Activity A_r (Bq kg⁻¹) and mass m (kg) for a time t (s) and Y is the gamma ray emission probability.

3 Results and Discussion

3.1 Activity concentration of radionuclides in raw minerals and soil samples

The mean activity concentration of radionuclides in the raw minerals and soil samples in the study area is presented in table 1 and 2. The reported uncertainty in the mean values of the activity concentration is the standard deviation (1σ) (except where only one sample was obtained), which gives information on the spatial spread of activity concentration of radionuclides in the study area. The ranges of values for the three primordial radionuclides are wide, resulting in large values of standard deviations.

In table1 the activity concentrations of ⁴⁰K, ²²⁶Ra and ²³²Th in the raw minerals varied from 74.78 ± 0.66 Bq kg⁻¹(Kaolin) to (436.78±6.31) Bq kg⁻¹(Clay), 15.60 Bq kg⁻¹ (Baryte*) to 106.18±7.76 Bq kg⁻¹(Limestone) and 9.74±1.06 (Baryte*) to 47.49±5.52 Bq kg⁻¹(Kaolin) respectively. The mean total of the concentration of the radionuclides in the soil samples is 425.92 ± 216.06 Bq kg⁻¹, 40.34 ± 12.58 Bq kg⁻¹, 33.69 ± 4.73 Bq kg⁻¹, for ⁴⁰K, ²²⁶Ra, ²³²Th respectively as shown in table2. This can also be seen graphically in Fig.2 and 3.

the reference sample of

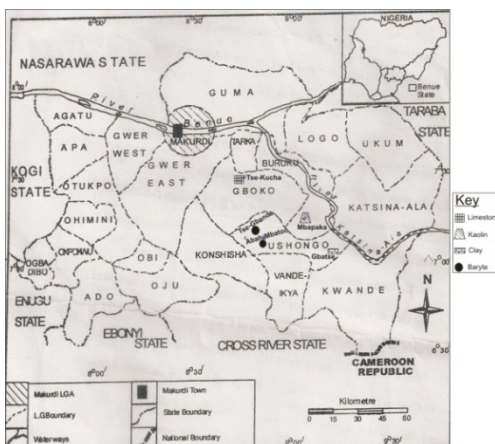


Fig 1. Map of Benue State.(locations indicated by author)

Table 1. Activity concentration of radionuclides in the raw material samples

Raw mineral (no. of samples)	Activity Concentration in (Bq kg ⁻¹)		
	⁴⁰ K	²²⁶ Ra (²³⁸ U)	²³² Th
	Range (mean±SD)	Range (mean±SD)	Range (mean±SD)
Clay (2)	432.31- 441.24 (436.78±6.31)	20.22-24.05 (22.14±2.71)	29.67-33.21 (31.44±2.50)
Kaolin (2)	74.31-75.25 (74.78±0.66)	19.24-32.18 (25.71±9.15)	43.58-51.39 (47.49±5.52)
Limestone (2)	121.97-127.65 (124.81±4.02)	100.69-111.67 (106.18±7.76)	11.19-13.41 (12.30±1.57)
Baryte* (2)	120.51-129.49 (125.00±6.35)	15.28-19.44 (17.36±2.94)	8.99-10.49 (9.74±1.06)
Baryte** (1)	135.64	15.60	11.07

Baryte* : Baryte samples from Tse-Gbande mining site in Ushongo L.G.A

Baryte** : Baryte sample from Aba-Mbatoo mining site in Ushongo L.G.A

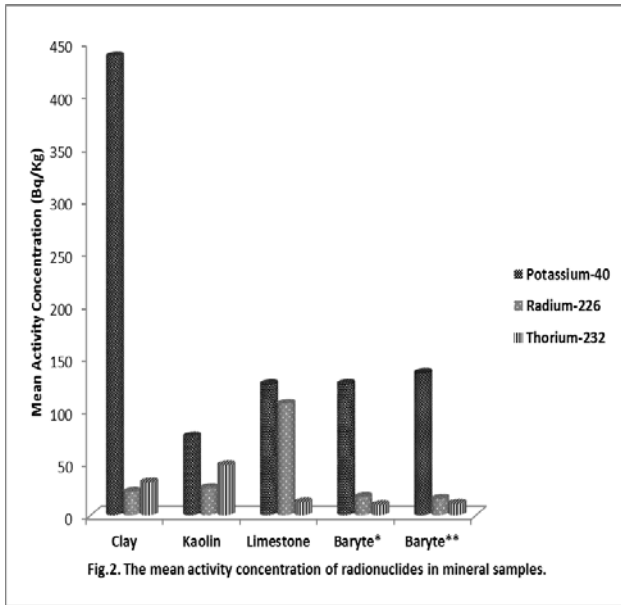


Fig.2. The mean activity concentration of radionuclides in mineral samples.

Table 2. Mean activity concentration of the radionuclides in soil samples

Site Name	Activity Concentration in (Bq kg ⁻¹)		
	⁴⁰ K	²²⁶ Ra (²³⁸ U)	²³² Th
	Range (mean±SD)	Range (mean±SD)	Range (mean±SD)
A(Clay)	433.95-1015.11 (721.55±205.05)	6.31-34.97 (19.90±8.75)	23.06-39.50 (31.75±5.51)
B(Kaolin)	66.41-598.87 (231.10±221.77)	4.10-67.02 (43.79±21.73)	25.68-65.73 (40.88±13.57)
C(Limestone)	130.28-284.30 (196.32±52.53)	34.39-67.41 (53.82±9.71)	24.69-41.00 (30.92±5.66)
D(Baryte*)	256.81-921.48 (476.87±271.92)	23.99-66.82 (39.31±13.53)	17.03-41.06 (29.02±8.28)
E(Baryte**)	329.61-753.46 (503.77±165.71)	21.71-64.16 (44.86±14.12)	21.38-45.15 (35.86±7.61)
Mean Total±SD	425.92±216.06	40.34±12.58	33.69±4.73

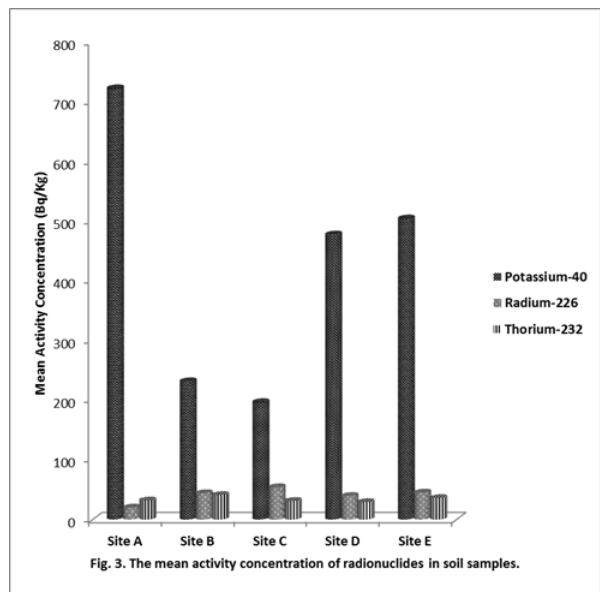


Fig. 3. The mean activity concentration of radionuclides in soil samples.

3.2 Absorbed dose rate and Annual effective dose equivalent

The absorbed dose rate and the corresponding annual effective dose equivalent (AEDE) in air at 1m above the ground were estimated for the mineral and soil samples base on the provision by UNSCEAR [21].

$$D(nGyh^{-1}) = 0.462C_{Ra} + 0.621C_{Th} + 0.0417C_K \quad 2$$

The annual effective dose rate was also estimated using the conversion coefficient from absorbed dose in air to effective dose (0.7SvGy⁻¹) and outdoor occupancy factor (0.2) proposed by UNSCEAR [21] and also used by Arafa [7]. The annual effective dose rate in (mSv yr⁻¹) was estimated as follows:

$$AEDE(mSvyr^{-1}) = D(nGyh^{-1}) \times 0.7SvGy^{-1} \times 8760h yr^{-1} \times \left(\frac{10^3mSv}{10^9}\right) \times 0.2$$

$$AEDE(mSvyr^{-1}) = D(nGyh^{-1}) \times 1.2264 \times 10^{-3} \quad 3$$

The results of the absorbed dose rates and annual effective dose equivalent for each mining site are presented in table 3 and 4 and graphically presented in figure 4 and 5. The mean total absorbed dose in air for all the five mining sites is (57.32±4.44) nGyh⁻¹.

The values of absorbed dose rate in air for the mineral samples varied from 19.28 (Baryte*) to 61.90 (Limestone) nGy h⁻¹. These values are within the range reported by Tzortzis [19] and also the maximum value obtained for Limestone is higher than the world's average value of 55 nGy h⁻¹ [21].

This value is within the average range of gamma absorbed dose rate in air across Nigerian cities which ranged between 19 ± 5 and 88 ± 44 nGy h⁻¹ obtained earlier by Farai and Jibiri [11] and later by Obed [16].

3.3 Radiation Hazard Indices

In order to ascertain the suitability of the raw minerals for use as building materials. Some basic radiological parameters such as Radium equivalent activity (Ra_{eq}), external hazard index (H_{ex}) and internal hazard index (H_{in}) and gamma activity index (I_γ) for the minerals were estimated.

3.3.1 Radium Equivalent Activity (Ra_{eq})

In order to represent the activity levels of ²²⁶R (²³⁸U), ²³²Th and ⁴⁰K by a single quantity, which takes into amount the radiation hazards associated with them, a common radiological index has been introduced. This index is called Radium equivalent (Ra_{eq}). The radium equivalent activity is mathematically defined by UNSCEAR [20]

$$Ra_{eq}(Bq kg^{-1}) = C_{Ra} + 1.43C_{Th} + 0.077C_K \quad 4$$

Where, C_{Ra} is the activity concentration of ²²⁶Ra (²³⁸U), C_{Th} is the activity concentration of ²³²Th, and C_K that of ⁴⁰K.

As could be seen from table 5 the calculated values of the radium equivalent activity (Ra_{eq}) for the four minerals ranged from 40.91 Bq kg⁻¹(Baryte**) to 133.38 Bq kg⁻¹ (Limestone).

The criterion in equation above considers the external hazard due to γ rays and it corresponds to a maximum Ra_{eq} of 370 Bq.kg⁻¹ for the material. The highest calculated values of the radium equivalent activity due to the activities of ²²⁶Ra, ²³²Th and ⁴⁰K in the minerals is 133.38 Bq kg⁻¹ for limestone samples which is less than the permissible value of 370Bq kg⁻¹ [20].

Table 3. Absorbed dose rate and annual effective dose equivalent

Raw Minerals	Absorbed Dose Rate (nGyh ⁻¹)	Annual Effective Dose rate (mSv yr ⁻¹)
Clay	47.96	0.0590
Kaolin	44.48	0.0547
Limestone	61.90	0.0761
Baryte*	19.28	0.0237
Baryte**	19.74	0.0242

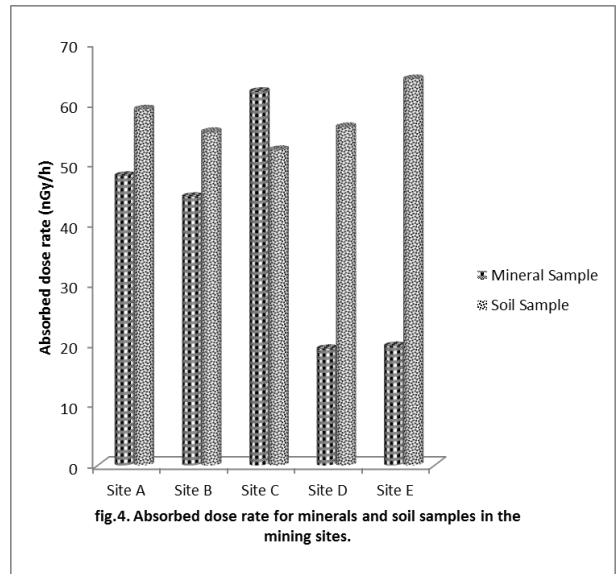


fig.4. Absorbed dose rate for minerals and soil samples in the mining sites.

Table 4. Absorbed dose rate and annual effective dose equivalent for soil samples

Name of Site	Absorbed Dose Rate (nGyh ⁻¹)	Annual Effective Dose rate(mSv yr ⁻¹)
A	58.99±11.11	0.073±0.014
B	55.25±22.24	0.068±0.027
C	52.25±6.98	0.063±0.008
D	56.07±14.33	0.069±0.018
E	64.00±11.06	0.079±0.014
Mean±SD	57.32±4.44	0.071±0.006

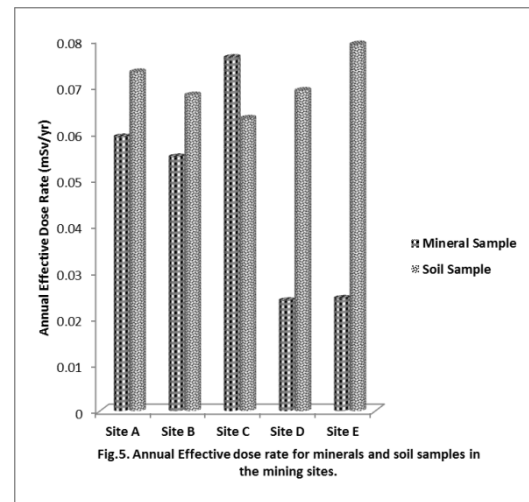


Fig.5. Annual Effective dose rate for minerals and soil samples in the mining sites.

3.3.2 External hazard index (H_{ex}), and Internal hazard index (H_{in}) and gamma activity index ($I_{\gamma r}$)

Another widely used hazard index (reflecting the external exposure) is called the external hazard index H_{ex} and is defined by equation 5.

$$H_{ex} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \quad 5$$

Beretka and Mathew [8] and also proposed by UNSCEAR [21].

More to the external hazard index, Radon and its short-lived products are also hazardous to the respiratory organ, the internal exposure to radon and its daughters is quantified by the internal hazard index H_{in} , which is given by equation 6 as used by Abel-Ghany [1] in their study.

$$H_{in} = \frac{C_{Ra}}{185} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \quad 6$$

Where, C_{Ra} , C_{Th} , and C_K are the activity concentrations of Uranium, Thorium and Potassium.

The gamma activity concentration index ($I_{\gamma r}$) representative level index is also estimated to correlate with the annual effective dose equivalent due to the excess external gamma radiation caused by superficial material. It is defined using equation 7, proposed by the European Commission [10];

$$I_{\gamma r} = \frac{C_{Ra}}{150} + \frac{C_{Th}}{100} + \frac{C_K}{1500} \quad 7$$

Values of index $I_{\gamma r} \leq 1$ corresponds to $0.3mSv yr^{-1}$, while $I_{\gamma r} \leq 3$ corresponds to $1mSv yr^{-1}$.

The results obtained in this work for the external and internal hazard indices and the gamma activity index are presented in table 5

The values of the external hazard index (H_{ex}) are also shown in table 5 for the minerals considered in this study. The highest value obtained is 0.380 for limestone while the lowest value is 0.107 for Baryte*. The values of the H_{ex} estimated in this study are less than unity, and as such, none of the minerals considered in this study are a major source of external radiation exposure. In addition to the external hazard, radon and its short-lived products also posed a hazard to the respiratory organs. The internal exposure to radon and its progenies is quantified by the internal hazard index (H_{in}). If the maximum concentration of radium is half that of the normal acceptable limit, then H_{in} will be less than 1 [13]. The calculated values of H_{in} in this study for the minerals ranged from 0.103 to 0.356. The values are less than unity.

Table 5: Radiation hazard indices for the mineral samples

Raw Mineral	Ra _{eq} (Bq kg ⁻¹)	H _{ex}	H _{in}	I _{γr}
Clay	100.45-101	0.271-0.273	0.266-0.267	0.751-0.755
	100.73±0.39	0.272±0.001	0.266±0.001	0.753±0.002
Kaolin	98.52-100.22	0.266-0.271	0.248-0.253	0.692-0.700
	99.37±1.20	0.268±0.003	0.251±0.003	0.696±0.005
Limestone	126.08-140.68	0.341-0.380	0.319-0.356	0.864-0.964
	133.38±10.32	0.360±0.02	0.336±0.026	0.914±0.07
Baryte*	39.56-42.27	0.107-0.114	0.103-0.110	0.287-0.306
	40.91±1.91	0.111±0.005	0.106±0.005	0.296±0.01
Baryte**	41.87	0.113	0.109	0.305

The gamma index ($I_{\gamma r}$) calculated as could be seen from table 5 ranges from 0.287 (Baryte*) to 0.964 (Limestone). Although the gamma activity index of sample Limestone, Clay and Kaolin has mean values which are close to unity but since the mean gamma indices of Clay, Limestone and Kaolin is less than 1, the minerals will pose no major radiological risk to the inhabitants of buildings and external exposure from materials used in the homes that are end products of these minerals.

4.0 Conclusion

Measurements of the activity concentrations of ^{40}K , ^{226}Ra (^{238}U) and ^{232}Th in minerals and soil samples around some mining sites have been carried out in this work and the resulting hazard indices and the absorbed dose in air have been estimated. No anthropogenic radionuclide was detected in any of the samples. This shows that there has not been any radioactive contaminant in the mining sites studied. The average outdoor absorbed dose rate in air for the soil samples is $57.32 \pm 4.44 \text{ nGy h}^{-1}$ which corresponds to an annual effective dose equivalent of $71 \pm 6 \mu\text{Sv yr}^{-1}$ while the values of absorbed dose rate in air for the mineral samples varied from 19.28 (Baryte*) to 61.90 (Limestone) nGy h^{-1} corresponding to an annual effective dose 24 to $76 \mu\text{Sv yr}^{-1}$.

Limestone had the highest values of the radiological hazard indices recorded. However, the values do not call for alarm, since they are still lower than the limiting levels. The maximum values are slightly higher than the world average value but lie within the range of the results obtained previously in other parts of Nigeria [11], [16]. From the estimated values of radium equivalent (R_{eq}), external hazard index (H_{ex}), internal hazard index (H_{in}) and gamma activity index (I_{gr}) for the mineral samples. The low values of radiological hazard indices show that the minerals are safe for use as building and construction materials and any other use of such in dwellings in the study area.

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