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Natural Radioactivity and Environmental Risk Assessment of Rock Samples at Gidan Tagwaye Granite Quarry Site in Dutse Area of Jigawa State using Gamma-Ray Spectrometry

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ABSTRACT

The toxicity risks of being over exposed to ionizing radiation in the environments are of great concern to environmental and health scientists. The levels of natural radioactivity of ^{40}K, ^{226}Ra and ^{232}Th were investigated at Gidan Tagwaye Granite Quarry Site located at 15 km away from Dutse Local Government Area of Jigawa State, in the North-Western Region of Nigeria using Gamma Spectroscopy with NaI(TI) detector. Radium equivalent activity (Ra_{eq}), absorbed dose rate, annual effective dose rates, hazard indices (Hex and Hin), gamma and alpha index were calculated and compared with the world recommended value. The results obtained show the measured activity concentration due to ^{40}K ranges from 347.52 ± 31.20 Bq/kg to 460.80 ± 93.60 Bq/kg with an average of 397.82 ± 71.24 Bq/kg. ^{226}Ra ranges from 92.12 ± 13.65 Bq/kg to 153.8 ± 18.85 Bq/kg with an average of 120.15 ± 16.12 Bq/kg. ^{232}Th ranges from 216.8 ± 14.45 Bq/kg to 343.98 ± 33.71 Bq/kg with an average of 289.10 ± 30.71 Bq/kg respectively. The radium equivalent activity (Ra_{eq}) was found to be 549.398 Bq/kg which is higher than world average 370 Bq/kg, the mean absorbed dose rate obtained is 280.02 nGy/h and is higher than the world average of 55 nGy/h. The measured average indoor annual effective dose rate is 1.168 $mSvy^{-1}$ and is higher than the world average value of 1 mSv/y while the outdoor annual effective dose rate is 0.349 $mSvy^{-1}$ which is lower than the world average value of 1 $mSvy^{-1}$. The measured average values of external and internal hazard index are 1.521 and 1.957, which is higher than the world permissible limit of 1, the measured gamma and alpha indices are 3.17 and 0.839, in which the gamma index is higher than the world average value, while alpha is less than the world average value. This indicates that the sediments in all the sampling sites pose a significant radiological effect to the workers and inhabitants of the area, therefore public awareness on the health implication of these radionuclides is recommended.

Keywords: Gidan Tagwaye, Radionuclides, Granite, Gamma-ray Spectrometry, Quarry

INTRODUCTION

Man and his environment are constantly exposed to natural radiation, the primary sources of this radiation are naturally occurring radioactive materials (NORM) (UNSCEAR, 2000). They can be found in rocks, soil, and underground water. These NORM are retained in variable amounts in materials originating from these geological matrices. Materials derived from rock and soil contains mainly the natural radionuclides of the radium (^{226}Ra), thorium (^{232}Th) series and potassium (^{40}K). Available literature reveal that these radionuclides are important for dose assessment in human because we are constantly taking risk of accumulating radionuclides through direct exposure and ingestion of food and water (Fares, 2017). Ionizing radiation is continuously emitted from two main sources, natural and manmade sources. All living organism receives about 85% of the exposure from naturally occurring radiation (Dołhańczuk, 2012), which is from both cosmic and terrestrial radiation. The upper atmosphere protects the earth and blocks most of the cosmic rays that approach the earth. However, a number of

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radionuclides are produced by the interactions of cosmic rays with the earth's upper atmosphere. The radioactive elements and their radiations are rather an indispensable part of nature, their impact on living organisms is crucial and very vital to study (Mishev & Hristov, 2011). Geological formations are additional sources of environmental risk (UNSCEAR, 2000).

Abdullahi *et al*. (2013) reported that radionuclides being transferred to the soil by rain and water movement, which subsequently causes living organisms to be repeatedly exposed to background radiations with or without their understanding. By measuring the activity concentration of the radionuclides present in the samples, it is possible to calculate the concentrations of these radionuclides in soil. Gamma radiation from these radionuclides serves as the body's primary external source of radiation. Therefore the population's baseline radiation exposure is significantly influenced by radionuclides in the soil near quarry sites

Natural radionuclides and their progenies are widely distributed throughout the crust of the earth, and various mining compositions, such as quarry soil, they include significant amounts of these radionuclides (Degerlier *et al*., 2008). These radionuclides are not distributed equally throughout the quarry ecosystem instead when they degrade, they emit harmful ionizing radiation that can result in cancer and other negative health effects. The existence of naturally occurring radionuclides (^{226}Ra, ^{232}Th and ^{40}K) in building materials arising from quarry products provides radiation vulnerability both inside and outside the building environments predominantly due to gamma radiation of potassium-40 and members of the Thorium and Uranium decay series. The term "quarry products" refers to a wide range of distinct natural rocks with various mineral compositions that have been crushed into different sizes at quarries. This involves a variety of rocks utilized in industrial processes, building construction, and as ornamental rocks, including granite, gneiss, diorite, and granodiorite (Shittu *et al*., 2015). Therefore, a great interest expressed worldwide for the study of naturally occurring radiation and Environmental radioactivity has led to the performance of extensive surveys in many countries (UNSCEAR, 2000), with few works existing close to the current study area (Baba-Kutigi *et al*., 2016; Joseph *et al*., 2023).

The radioactive contamination of the environment has increased the natural background radiation caused by naturally occurring radioactive material which are brought to the earth surface through human activities such as oil and gas exploration, quarry activities and artificially produced radioactive material which make the workers of the quarry site and resident of the area to be repeatedly exposed to the gamma radiation. The three most common radioactive elements found in most quarry sites are potassium ^{40}K, ^{226}Ra and ^{232}Th. A primary health concern associated with naturally occurring radioactive material (NORMs) such as Radon-222 and Radon-220, the daughter product of Radium-226 and Thorium-232 decay chains respectively, because they result in harmful effect including anemia, cataracts and cause lung cancer upon sufficient exposure (William *et al.*, 2000; Baba-Kutigi *et al*., 2016; Joseph *et al*., 2018). It is on this basis that this work is aimed at evaluating the natural radioactivity and environmental risk assessment of rock samples at Gidan Tagwaye granite Quarry Site in Dutse Area of Jigawa State, North-Western Nigeria.

MATERIALS AND METHOD

The Study Area

The study area is Gidan Tagwaye which is about 15km away from Dutse Local Government Area of Jigawa State, in the North-Western Region of Nigeria. Dutse is on Geological coordinates of Latitude 11° 11' 20" N and Longitude 9° 54' 34" E with the total land area of about 23,154 km^2 And estimated population of about 153,000 (NPC, 2015). The environment enjoys a tropic type climate which is generally characterized by two temperatures, the hot and cold temperature, and this is largely controlled by two air masses: namely tropical maritime and tropical continental blowing from Atlantic, the air masses determine the two dominant seasons which are wet and dry season (NPC, 2015). The plate 1 shows the Location of Kaci from a Satellite Map.

Plate 1: Satellite Image of Kaci Granite Quarry Site

Sample Collection and Preparation

The samples were collected randomly within some selected areas of the study area. These samples were analyzed using gamma spectrometry to determine the activity concentration of radionuclides. The Global Positioning System (GPS) was used to record the location of all the sampling points in Table 1, with samples of about 1 *kg* each to a depth of about 10 cm was collected, initially filled in to polyethylene bags separately and labelled accordingly and then transported to the environmental laboratory at Centre for Energy Research and Training (CERT). In the laboratory, the rock samples were air-dried and then oven-dried at a temperature of 105℃ until all moisture is completely lost. The samples were crushed into a fine powder using a pulverizer and sieved through a 2-mm pore size mesh into a plastic beaker. The powder was then sealed inside plastic beaker using Vaseline, candle wax and masking tape in order to prevent trap radon gas from escape and stored for the period of 28 days for the short-lived daughters of ^{226}Ra (in the ^{238}U decay series), ^{40}K and ^{232}Th decay series to attain secular equilibrium with their long-lived parent radionuclides.

Table 1: GPS Coordinates of the rock samples

No	Sample ID	Latitude	Longitude
1	GIT 1	11°45'51.97"	9°21'49.93"
2	GIT 2	11°45'51.72"	9°21'46.84"
3	GIT 3	11°45'53.86"	9°21'47.01"
4	GIT 4	11°45'55.40"	9°21'51.39"
5	GIT 5	11°45'57.13	9°21'50.62"

Sample Analysis

The gamma-ray spectrometry set-up is made up of a 7.62 cm x 7.62 cm NaI (Tl) detector housed in a 6 cm thick lead shield (to assist in the reduction of the background radiation) and lined with cadmium and copper sheets. The samples were placed on the detector surface and each counted for about 29,000 seconds in reproducible sample detector geometry. The configuration and geometry were maintained throughout the analysis, as previously characterized based on well-established protocol of the laboratory (at the Centre for Energy Research and Training, Zaria). The specific activity of ^{238}R and ^{232}Th was measured using property of secular equilibrium with their decay products such as transition lines of ^{214}Bi (1765 KeV) and transition lines of ^{208}Tl (2614 KeV) respectively. Potassium-40 was measured directly from the photo peak at 1460 KeV. The measuring time for all samples under study were 24/h (Iqbal *et al*., 2010).

The specific activity for each detected photo peak was calculated using the equation below

$$A_r = \frac{N - N_o}{I_\gamma \varepsilon m t} \tag{1}$$

A_r is the specific activity of the radionuclide in the sample, N is the net counts of a given peak for a sample, N_o is the background of the given peak, I_γ is the number of gamma photons per disintegration, ε is the detector efficiency at the specific gamma-ray energy, m is the measured mass of the sample, and t is the measuring time.

COMPUTATION OF PARAMETERS

Radium Equivalent Activity (Ra_{eq})

The gamma–ray radiation hazards due to the specified radionuclides ^{226}Ra, ^{232}Th and ^{40}K were assessed by radiation hazard index R_{aeq}. The Radium equivalent was calculated based on the estimation that 370Bq/kg of ^{226}Ra, 259 Bq/kg of ^{232}Th and 4810 Bq/kg of ^{40}K produce the same radiation rate (UNSCEAR, 2000).

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \tag{2}$$

Calculation of Air Absorbed Dose Rate (Dab)

Radiation emitted by a radioactive substance is absorbed by any material it encounters (UNSCEAR, 2000). The absorbed dose rate was calculated from the measured activity concentrations of the ^{226}Ra, ^{232}Th and ^{40}K which are converted into doses (nGy h^{-1} per Bq/kg), by applying the conversion factors 0.462, 0.604, and 0.0412 for Radium, Thorium and Potassium, respectively using the following equation (UNSCEAR, 2000).

$$D_{ab} = 0.462_{Ra} + 0.604\ A_{Th} + 0.042\ A_k \tag{3}$$

Annual Effective Dose Rates

To measure the annual effective doses, both indoors and outdoors, considerations must be made for the conversion coefficient from absorbed dose in air to effective dose and the indoor and outdoor occupancy factors respectively. The conversion coefficient, 0.7SvGy-1 was recommended by UNSCEAR (2000). The adults spend about 80% of their time indoors, while the remaining 20% time is spent outdoors. Therefore, the indoor and outdoor occupancy factors were given as 0.8 and 0.2, respectively (UNSCEAR, 2000). Hence, the annual indoor and outdoor effective doses (mSv) are given as follows:

$$H_{E\ (indoor)} = Dab \times 8760\ h \times 0.8 \times 0.7\ SvGy\text{-}1 \times 10^{-6} \tag{4}$$

$$H_{E\ (outdoor)} = Dab \times 8760\ h \times 0.2 \times 0.7\ SvGy\text{-}1 \times 10^{-6} \tag{5}$$

External and Internal Hazard Index

These indices represent the external radiation hazards due to the inhalation of radon gas. The external hazard index is calculated using equation (5) (Innocent *et al.*, 2014)

$$H_{ex} = \frac{ARa}{370} + \frac{ATh}{259} + \frac{Ak}{4810} \tag{5}$$

The radiation risk is negligible when the maximum value of the external hazard index is less than unity (Hex ≤ 1), which is equivalent to a maximum value of the Raeq activity < 370 $Bqkg^{-1}$. Internal exposure arises from the inhalation of radon (^{222}Rn) gas and its progeny products or ingestion of other radionuclides. Hence the internal hazard index is calculated using equation (7):

$$H_{in} = \frac{ARa}{185} + \frac{ATh}{259} + \frac{Ak}{4810} \quad (7)$$

Gamma Index (Iγ)

Gamma index I_γ is used estimate the gamma ray hazard related to the natural radionuclide in specific investigated samples. The representative gamma index was estimated as follow (Avwiri & Ononugbo, 2014).

$$I\gamma = \frac{ARa}{150} + \frac{ATh}{100} + \frac{Ak}{1500} \quad (8)$$

Alpha Index (I_α)

As radon progeny decay, they emit radioactive alpha particles and attach to aerosols, dust and other particles in the air. As we inhale, radon progeny is deposited on the cells lining the airways where the alpha particles can damage DNA and potentially cause lung cancer. The excess alpha radiation due to radon inhalation originating from building materials is estimated through the alpha index (I_α), it is given by equation (9).

$$I\alpha = \frac{ARa}{150} \quad (9)$$

RESULTS AND DISCUSSION

Table 2: Activity Concentration, Radium Equivalent, Absorbed Dose Rate and Annual Effective Dose

Sample ID	Activity concentration ($Bqkg^{-1}$)					Annual effective dose	
	^{40}K (Bq/kg)	^{226}Ra (Bq/kg)	^{232}Th (Bq/kg)	Ra (eq) (Bq/kg)	Dab $nGyh^{-1}$	Effective dose (Indoor) $mSvy^{-1}$	Effective dose (Outdoor) $mSvy^{-1}$
GIT 1	408.00	153.86	319.02	641.47	280.91	1.378	0.345
GIT 2	460.80	121.52	343.98	648.89	283.26	1.389	0.347
GIT 3	413.76	101.92	280.80	535.27	234.67	1.148	0.287
GIT 4	359.04	92.12	216.84	492.85	150.331	0.737	0.184
GIT 5	347.52	131.32	284.70	565.20	247.22	1.213	0.303
Minimum	347.52	92.12	216.84	429.85	150.31	0.737	0.184
Maximum	460.80	153.86	343.98	648.89	283.26	1.389	0.347
Mean	397.82	120.15	289.10	576.74	239.28	1.173	0.300

Table 3: External and Internal Hazard Indices, Gamma Index and Alpha Index

Sample ID	Hazard Indices		Gamma index (Iγ)	Alpha index (Iα)
	H_{ex}	H_{in}		
GIT 1	1.7280	2.1505	4.4879	0.7693
GIT 2	1.7469	2.1070	4.5631	0.6076
GIT 3	1.4410	1.9650	3.7633	1.4040
GIT 4	1.5250	1.7784	3.0219	0.4606
GIT 5	1.5208	1.8810	4.3919	0.6560

Minimum	1.4410	1.7784	3.0219	0.4606
Maximum	1.7469	2.1505	4.5631	1.4040
Mean	1.5923	1.9764	4.0456	0.7790

The activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K, radium equivalent activity (Raeq), absorbed dose rate, annual effective doses, hazard index, gamma index and alpha index in rock samples collected from Gidan Tagwaye Granite Quarry Sites located at 15km away from Dutse were measured and presented in Table 2 and Table 3. From Table 2 the activity concentration of ^{40}K have been measured and ranges from 347.52 to 460.80 Bq/kg with an average of 397.82 Bq/kg. ^{226}Ra ranges from 92.12 to 153.86 Bq/kg with an average of 120.15 Bq/kg. ^{232}Th ranges from 216.84 to 343.98 Bq/kg with an average of 289.10 Bq/kg respectively. The world average concentrations are 35, 30 and 400 Bq/kg for ^{226}Ra, ^{232}Th and ^{40}K, respectively (UNSCEAR, 2000). The obtained mean activity concentration of ^{40}K is 397.82 Bq/kg, which is indeed lower than the world average of 400 Bq/kg. While the average and ranges of activity concentration of ^{226}Ra and ^{232}Th are higher than the world average value of 35, 30 respectively, reported by (UNSCEAR, 2000). The result obtained in this study were higher than the mean values reported by Joseph *et al*. (2023) in Dutsin-ma, the differences obtained in the mean values may be attributed to level of the concentration of Uranium and Thorium arise from rocks and the geological formation of the area because Dutse is underlain by formation of granite rock and (UNSCEAR, 2000) reported that higher radiation level of radionuclides are associated with granite rocks. However the activity concentration of ^{40}K in three samples (GIT 4 and GIT 5) are found to have values lower than world average value. The activity concentration for ^{226}Ra in rock samples for this area were found to be higher than the world average values in all the five (5) samples, The activity concentration for ^{232}Th in rock samples for this area are found to be higher than the world average values in all the five (5) samples. Therefore the radionuclides present in the rocks samples poses significant effect to the workers of the quarry site and also the nearby resident of the area.

Table 2 shows that the value for radium equivalent activity of all the samples varies from 492.85 to 648.89 Bq/kg minimum and maximum respectively with mean value of 576.74 Bq/kg. The result also shows that all values of the five (5) samples are higher than the 370 Bq/kg limits set by United Nation Scientific Committee. The total absorbed dose rates from the study area were calculated for all the sampling sites in Table 2. The result shows that the minimum value being 150.31 nGyh^{-1}, the maximum is 283.26 nGyh^{-1} and the mean absorbed dose rates delivered to the general public of the study area were 239.28 nGyh^{-1}. It has been observed that the calculated values of absorbed dose are found to be higher than the global average absorbed dose level which is 86 nGyh^{-1} (UNSCEAR, 2008).

Table 4: Comparison of Mean Activity Concentration of (^{226}Ra, ^{232}Th, ^{40}K) with some Reported Value

S/N	^{226}Ra (Bq/kg)	^{232}Th (Bq/kg)	^{40}K (Bq/kg)	Location	Reference
1	120.15	289.10	397.82	Dutse, Nigeria	Present Study
2	11.59	25.99	36.7	Dutsin-Ma, Nigeria	(Joseph *et al*., 2023)
3	42.3	27.6	390.4	Turkey	(Taşkın *et al*., 2009)
4	37.2	45.3	185.0	Yobe, Nigeria	(Habu *et al*., 2019)
5	130.0	352.0	412.0	Kano, Nigeria	(Oladunjoye *et al*., 2022)

The annual effective dose rate for both indoor and outdoor was calculated in Table 2 and the result for (indoors) are in the range from 0.737 to 1.389 mSvy^{-1} minimum and maximum respectively. With and average value of 1.173 $mSvy^{-1}$. For (outdoors) are ranges from 0.184 to 0.347 $mSvy^{-1}$ minimum and maximum respectively. With an average value of 0.300 $mSvy^{-1}$. For (outdoor) all the five (5) are higher than international recommended value. While for (outdoor) doses all the five (5) samples are found below the international recommended value of 1 $mSvy^{-1}$ (UNSCEAR, 2000).

The external (H_{ex}) and internal (Hi_{n}) hazard indices represent the risk associated from exposure of the radionuclides in the rocks samples. Table 3 shows the values of both external and internal hazard index of all the samples calculated. The external hazard index was found in the range of 1.26 to 1.75 minimum and maximum respectively with mean value of 1.60 while the internal hazard index ranges from 1.7 to 2.30 minimum and maximum respectively with mean value of 1.98. The values of external and internal hazard indices in the present study is higher than unity, indicating that the rocks from these granite quarry site are not safe and can pose significant harmful effects to the inhabitants if used as construction material (UNSCEAR, 2000).

The calculated values of gamma index ($I\gamma$) for the rock samples are given in Table 3. The alpha index in rock samples have been found to vary from 3.02 to 4.56 with an average value of 4.05. It is observed that five samples have values that are higher than the limit of 0.5.

The values of Alpha index ($I\alpha$) calculated from the concentrations of ^{226}Ra are presented in Table 3. However, the alpha index in rock samples have been found to vary from 0.46 to 1.40 with an average value of 0.78. It is observed that four values of $I\alpha$ are lower than maximum permissible value of $I\alpha = 1$ (Kamal, 2012).

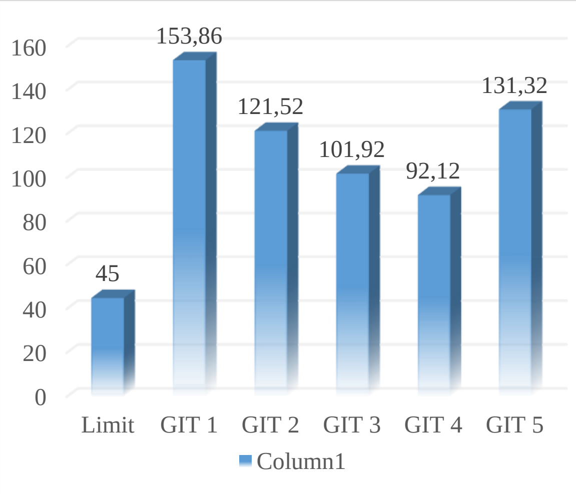

Figure 1: Chart of R-226 Concentration

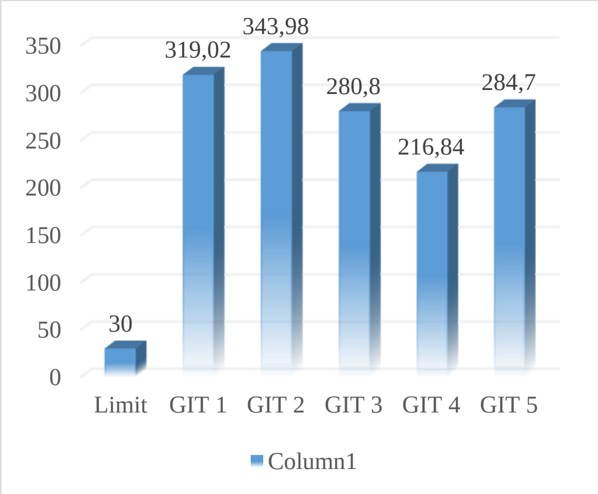

Figure 2: Chart of Th-232 Concentration

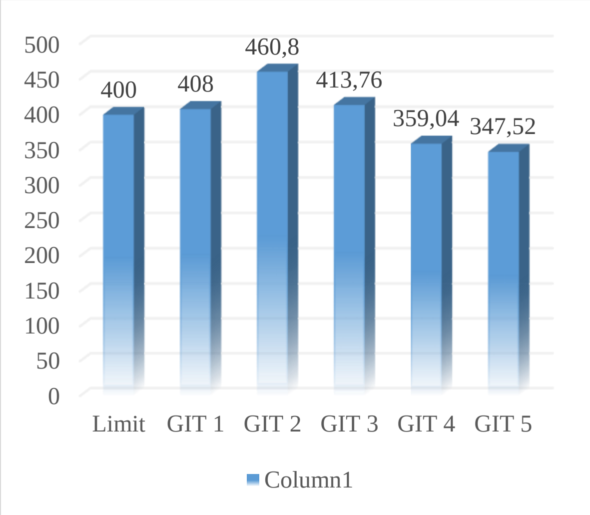

Figure 3: Chart of K-40 Concentration

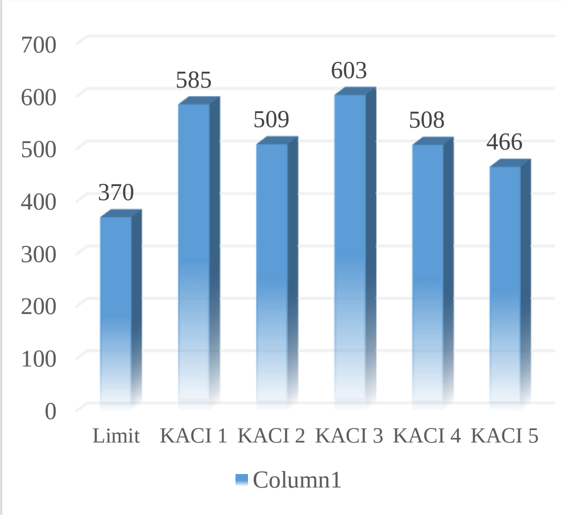

Figure 4: Chart of Radium equivalent R_{aeq}

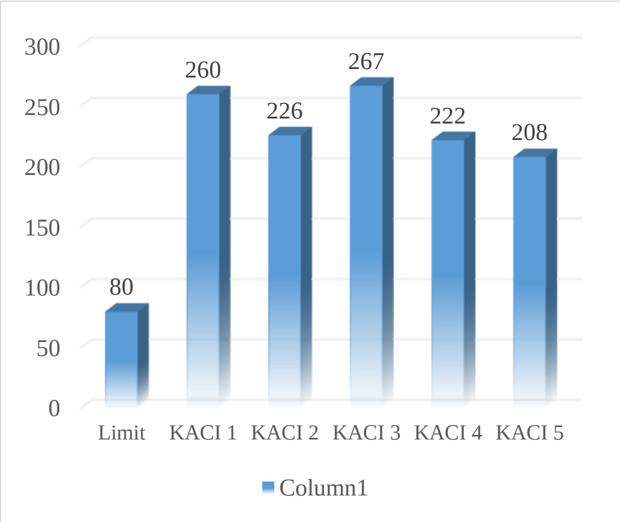

Figure 5: Chart of Absorbed Dose Rate (D_{ab})

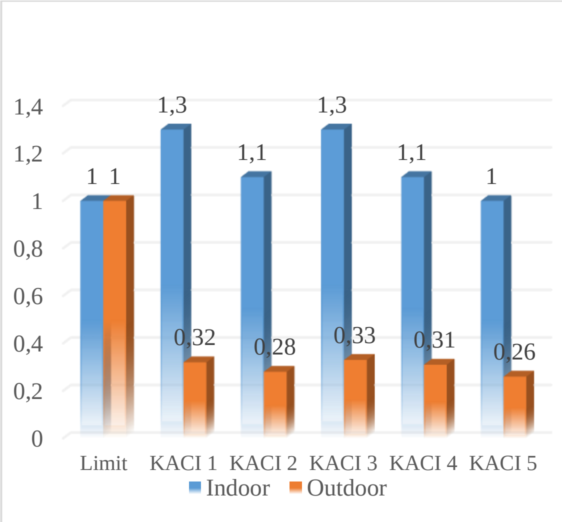

Figure 6: Chart of Outdoor and Indoor Annual Effective Dose Rate

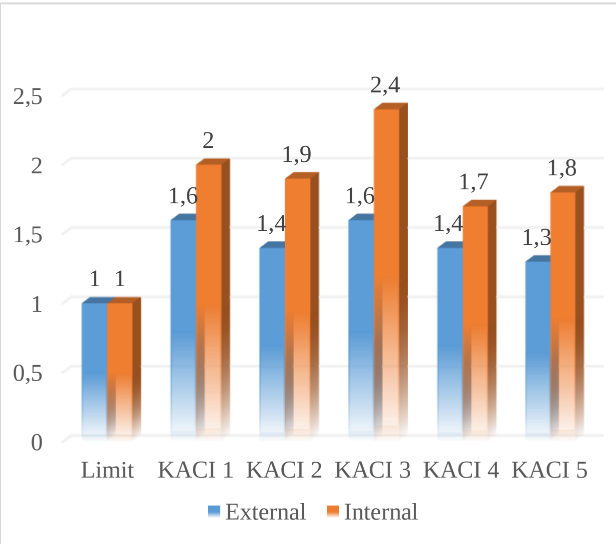

Figure 7: Chart of External and Internal Hazard Index

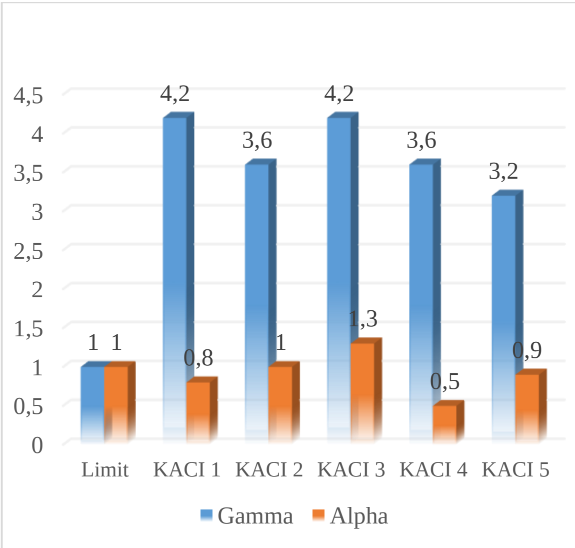

Figure 8: Chart of Gamma and Alpha index

Chart were plotted to graphically illustrate the obtained/calculated values, in each case the first bar is the limit while the remaining bars are the obtained/calculated values, so any bar that is higher than the first bar has exceeded the limit. Figure 1 gives a comparison of the measured values of the activity concentration for ^{226}Ra with recommended limits which indicates that the concentration of ^{226}Ra is greater than the recommended value at all sampling sites, which shows that the dwellings is not considered safe.

Figure 2 shows the comparison of the measured value of ^{232}Th with recommended limits which indicates that the concentration is greater than the recommended value at all sampling sites, which indicates that the dwelling is not completely safe. Similarly, a comparison of the measured value of ^{40}K activity concentration as shown in Figure 3 indicating the concentration of ^{40}K is lesser than the recommended value at some sampling site, which shows that the dwelling is considered safe. Figure 4 shows the comparison of measured value of the radium equivalent activity with recommended limit indicates the measured values is higher than the recommended value indicating the site is not safe for dwelling. Figure 5 shows the comparison of measured value of the absorbed dose rate with recommended limits indicates the measured values is higher than recommended value indicating the site is not safe for dwelling. Figure 6 shows the comparison of measured value of the annual effective dose rate with recommended limits indicates the measured values is higher than recommended value indicating the site is not safe for dwelling. Figure 7 shows the comparison of measured value of the external hazard index with recommended limits indicates the measured values is higher than recommended value indicating the site is not safe for dwelling. Figure 7 also gives a comparison of internal hazard index with its own recommended values which was found to be higher than the recommended one indicating the site is not safe for dwelling. Figure 8 shows the comparison of gamma index with its own recommended value which was found to exceed the recommended value indicating the site not good for dwelling due to its health effects. Figure 8 likewise shows the comparison of alpha index with its own recommended value which was found to fall below the recommended value.

CONCLUSION

The effect of activity concentrations of the primordial radionuclides in soil and the associated radiological hazards to the public with respect to geological formation of Gidan Tagwaye Granite Quarry. In this study, the calculated average values of activity concentration of ^{40}K, ^{226}R, and ^{232}Th are 397.82, 120.15, 289.10 Bqkg^{-1} respectively. The average values of ^{226}R, and ^{232}Th are higher than the world average value of (35 and 30 Bqkg^{-1} for ^{226}R and ^{232}Th respectively). While the average value for ^{40}K is lower than world average value of 400. And other radiological hazard parameters that were calculated include Radium Equivalent Activity (Ra_{eq}) with an average value of 576.74 Bq/kg, Absorbed Dose Rate (D_{ab}) with an average value of 239.28 nGy/h, Indoor Annual Effective Dose with an average value of 1.173 mSvy^{-1}, Outdoor Annual Effective Dose with an average of 0.300 mSvy^{-1}. Internal Hazard Index with an average value of 1.976, while External Hazard Index with an average value of 1.592. Gamma Index ($I\gamma$) with an average of 4.046 and Alpha Index (I_α) with an average of 0.780. All the radiological hazard parameters are higher than the world average value except for Effective dose (Outdoor) and alpha index. Therefore these granite quarry site are not safe because all the radiological hazard parameters are in the critical range, it also indicate that the sediment in all the sampling sites is not recommendable for construction or building.

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