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Natural Radionuclides Content in Granites from Operational Quarry Sites

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Abstract

Natural radionuclides content in granite from eight functional quarries in Osun State, Southwestern Nigeria was assessed. Eighty granite samples comprise $\frac{3}{4}$ inches. $\frac{1}{2}$ inch and stone-dust were collected from Wolid, Slava, Ayofe, Espro, Ife/Modakeke, Krystal Vountein, Clario and Omidiran quarries in the State. Measurement was done using a high purity germanium (HPGe) detector. The statistical analysis was carried out using Statistical Package for Social Sciences (SPSS) software to determine if granite size has an impact on the activity concentrations. The results revealed that the mean activity concentrations of ^{238}U (12.64 ± 1.89 Bq·kg^{-1}) and ^{232}Th (16.93 ± 2.46 Bq·kg^{-1}) were highest in $\frac{3}{4}$ inch granite and lowest in stone-dust (5.01 ± 0.77 and 8.97 ± 1.37 Bq·kg^{-1} respectively), whereas ^{40}K is highest in the $\frac{3}{4}$ inches (266.19 ± 35.53 Bq·kg^{-1}) and lowest in $\frac{1}{2}$ inches (151.85 ± 25.09 Bq·kg^{-1}) granite. Espro has the highest (23.75 ± 3.74 Bq·kg^{-1}) while Wolid has the lowest (4.11 ± 0.73 Bq·kg^{-1}) ^{238}U activity concentration and Slava has lowest for ^{232}Th (8.21 ± 1.12 Bq·kg^{-1}) and ^{40}K (109.54 ± 11.06 Bq·kg^{-1}). The radiological hazard parameters such as absorbed dose, annual effective dose radium equivalent, gamma index, external index, and internal index, were calculated to assess the radiation hazards associated with granite samples. The results obtained are lower than the recommended limits. The results were compared with the published data of other countries. Although, all the calculated radiation hazard indices were lower than the permissible limits. Therefore, people working in the quarries, granite end-users and the general public are safe from radiological health risks from the quarries, since there is no significant health hazard. The research will give reliable information on activity concentrations of natural radionuclides in granite rocks, contribute to a better understanding of radioactivity distribution in granite, and develop standards.

Keywords

Activity Concentration, Radionuclides, Granite, Quarries, Radiological Parameters

1. Introduction

Natural radionuclides are all-over in the environment; even man carries naturally occurring radioactive materials within his body [1]. Everyday inhales and ingests radionuclides with air, food, and water. Natural radioactivity is conventional in the rocks and soil that constitute our planet, in water bodies, building construction materials and homes. Radioactive elements can be subdivided into three general groups: primordial, cosmogenic and man-made [2]. The extent of terrestrial environmental radiation is a function of the geological composition of the region, and concentration of ^{226}Ra, ^{232}Th and ^{40}K in rocks [3] [4]. Rocks are used in various construction activities, which also have these natural radionuclides [5].

Radiation exposure can injure living cells, leading to death in some of them and altering others. The majority of the body organs and tissues are unaffected by the loss of even substantial numbers of cells. Nonetheless, if the number forfeited is large enough, there will be noticeable injury to organs that may result into death. Such harm occurs in individuals who are exposed to radiation above a threshold level. Other radiation injuries may also happen in cells that are not killed but altered. Such destruction is normally repaired. Provided the repair is imperfect, the resulting alteration will be transmitted to further cells and may finally cause cancer. If the cells modified are germ cells of the exposed individual, hereditary disorders may arise [4]. The existence of naturally occurring radionuclides (^{238}U, ^{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 Uranium and Thorium decay series. The phrase "quarry products" incorporates a wide number of dissimilar natural rocks with different mineral contents, crushed into various sizes at quarries. This involves varying geological materials such as granite, gneiss, diorite, granodiorite, and other rocks that are used for industrial processes, building construction and as ornamental rocks [6].

Granite is a natural and abundant resource that has great values that can be harnessed for the development of southwestern Nigeria as it has been widely used as a building construction material [7]. When it is used as cut stones or dimension stones, they are considered by many as the premium material for beauty and durability in institutional and monumental constructions. Granite as cut stones can be used in flagging, roofing slates, and mills stock slates. They can be used as curbing and paving blocks and in laboratory furniture and sinks. They have been utilized to line tube mills for grating one or other substances. However, the only most noticeable usage of these dimension stones to date is its exploitation as aggregates in small scale and monumental constructions. The overall effects of quarry activities are the sizable havoc of the environment in terms of deforestation, destruction of nearby farmlands with stone relics, gaseous pollution from the use of explosives. Other sources of environmental hazard are natural radiations from granitic bodies and other geological formations [4] [8] [9].

Researchers from different regions have carried out researches on natural radioactivity in granites unfortunately none is available for radionuclide level in granite samples of the selected quarries from the surveyed of published works. Consequently, knowledge of the level of natural radioactivity in granite from operational quarries in Osun State is of great significance. The research will give reliable information on activity concentrations of natural radionuclides in granite rocks, contribute to a better understanding of radioactivity distribution in granite and develop standards for the use of this building material. The study aims at assessing the natural radionuclides content of granite from operational quarry sites in Osun State, Southwestern Nigeria.

2. Methodology

2.1. Geology of the Study Area

Osun State is an inland state in Southwestern Nigeria with coordinates 6˚55'N 4˚06'E and 8˚07'N 5˚05'E and a total area of 9251 km^2 (3572 sq mi). Its capital is Osogbo. It has boundary with Kwara State in the North, in the east partly by Ekiti State and partly by Ondo State, in the south by Ogun State and in the west by Oyo State [10]. Osun is underlain by metamorphic rocks of the basement complex, which is visible on the surface over many parts. Rocks of the basement complex observed here are schists, associated with quartzite ridges of the type obtained in Ilesa area. The metamorphic rocks are chiefly not different, though two particular rock groups could still be pinpointed. The first group consists of the migmatite complex, including banded magmatic and augen gneisses and pegmatites with outcrops in Ilesa and Ife areas. Metasediments consisting of schists and quartzites, calsilicates, meta-conglomerates, amphibolites, and metamorphic iron beds make up the second group. They are found in Iwo and Ikire areas. Other parts of the state are underlain by undifferentiated metamorphic rocks [11] [12]. **Figure 1** is a map of Osun State Nigeria. The coloured regions are the five local governments where quarry sites under study are located.

2.2. Sample Collection and Preparation

A total of eighty granite samples, stone-dust (24 samples), a half-inch (32 samples) and twenty-four samples of a three-quarter-inch (24 samples) were collected from eight functional quarry sites in Osun State. The quarries are distributed across five Local government areas of the State. Each granite sample was packed into a polyethylene bag and clearly labeled to avoid cross-contamination.

The label contained sample location, code and collection date. The polythene bags were tightly sealed with cellophane. Table 1 shows the quarry names, sample code, sample number and GPS location. The samples were crushed with a Laboratory Jaw Crusher. The samples were transferred to the laboratory and prepared into 1 litre Marinelli beakers. The beakers were thick enough to check the imbuing of radon. The beakers were closed by screw caps and the plastic tape was wrapped over the caps and then stored for a month to allow time for ^{222}Rn to reach a state of secular equilibrium with its short-lived daughters before gamma spectroscopy. This step was necessary to ensure that radon gas was confined within the volume and that the progenies also remained in the sample.

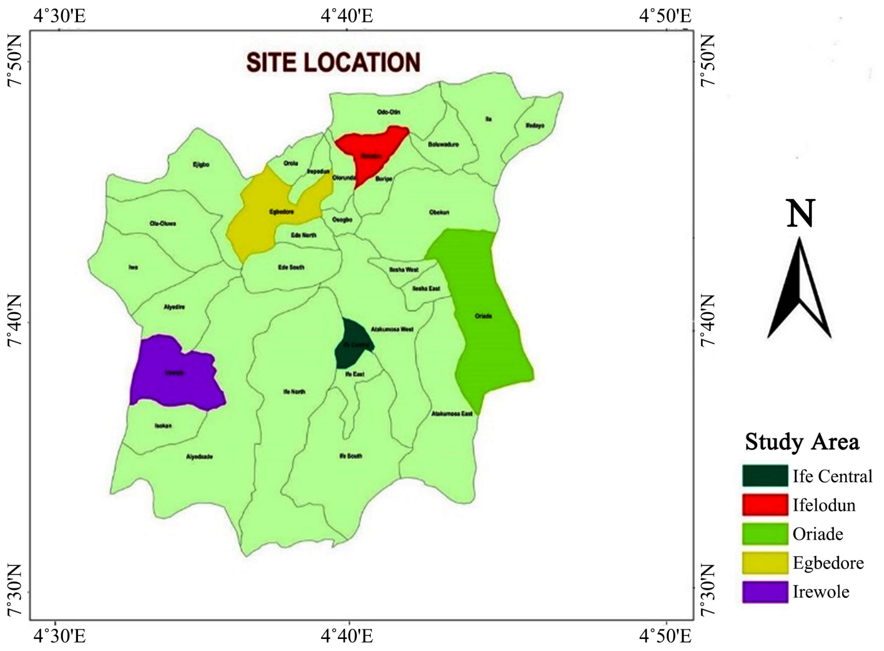

Figure 1. Local government areas in Osun State where studied quarries are located [13].

Table 1. Quarry names, sample code, sample number and GPS location.

S/N	Quarry Name	Sample Code	Number of Samples	Longitude	Latitude
1	Wolid Quarry Complex	WQ	10	4.348365	7.747812
2	SlavaYetidepe	YQ	10	4.390623	7.755893
3	Ayofe/Irepodun and Sons	AQ	10	4.392271	7.7649281
4	Espro Asphalt Prod. Co. Ltd.	EQ	10	4.259797	7.429560
5	Granite Producers Ife/Modakeke	IQ	10	4.608602	7.556943
6	Krystal Vountain	KQ	10	4.897970	7.490550
7	Clario Nig. Ltd.	CQ	10	4.667675	7.943495
8	Omidiran Nig. Ltd.	OQ	10	4.667675	7.943495

2.3. Sample Measurement and Analysis of Spectra

The activity concentrations of the samples were determined by using a computerized gamma-ray spectrometry system with high purity germanium (HPGe). The relative efficiency of the detector system was 40%, and the resolution of 1.8 keV at 1.33 MeV of ^{60}Co. The gamma spectrometer is coupled to conventional electronics connected to a multichannel analyzer card (MCA) installed in a desktop computer. A software program called MAESTRO-32 was used to accumulate and analyze the data. The detector is located inside a cylindrical lead shield of 5 cm thickness with an internal diameter of 24 cm and a height of 60 cm. The lead shield is lined with various layers of copper, cadmium, and Plexiglas, each 3 mm thick. A counting time of 36,000 seconds (10 h) was used to acquire spectral data for each sample. The activity concentrations of the uranium-series were determined using γ-ray emissions of ^{214}Pb at 351.9 keV (35.8%) and ^{214}Bi at 609.3 keV (44.8%) for ^{238}U, and for the ^{232}Th-series, the emissions of ^{228}Ac at 911 keV (26.6%), ^{212}Pb at 238.6 keV (43.3%) and ^{208}Tl at 583 keV (30.1%) were used. The ^{40}K activity concentration was determined directly from its emission line at 1460.8 keV (10.7%) (See **Figure 2**).

2.4. Estimation of Activity Concentration

The specific activity concentrations (A_{sp}) of ^{238}U, ^{232}Th, ^{40}K in Bq·kg^{-1} for the rock samples were determined using the following expression [14].

$$A_{sp} = \frac{N_{sam}}{\gamma_E \cdot \varepsilon \cdot T_C \cdot M} \tag{1}$$

where N_{sam} is net counts of the radionuclide in the sample, γ_E is gamma-ray emission probability (gamma yield), ε is total counting efficiency of the detector system, T_c is sample counting time and M is mass of sample (kg).

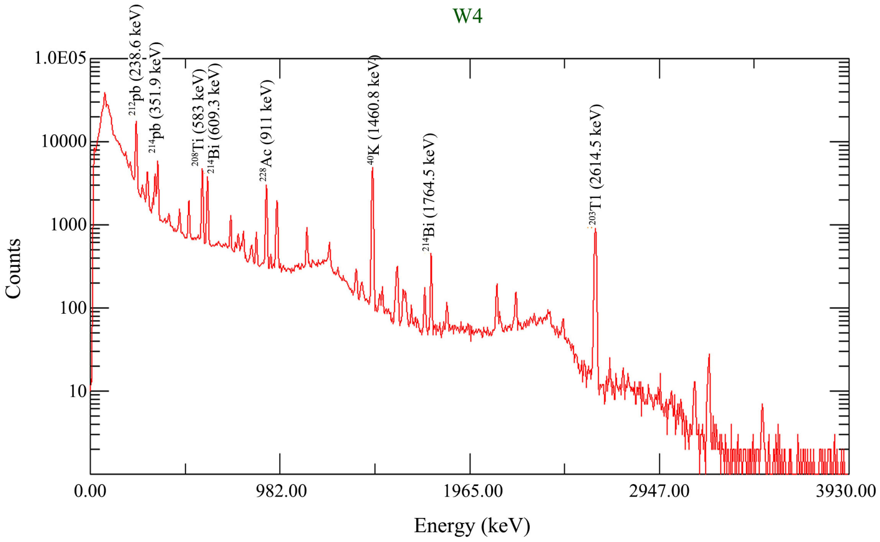

Figure 2. Typical sample spectrum of the radionuclides present in WQ.

2.5. Calculation of Radiological Parameters

2.5.1. Absorbed Dose Rate in Air (*D*)

The exposure to radiation arising from radionuclides present in the rock can be determined in terms of many parameters. A direct connection between radioactivity concentrations of ^{238}U, ^{232}Th and ^{40}K (Bq·kg^{-1}) in the granite samples are used to calculate the absorbed dose rate given by the relation [15].

$$D\left(\mathrm{nGy}\cdot\mathrm{h}^{-1}\right) = 0.462C_{\mathrm{U}} + 0.604C_{\mathrm{Th}} + 0.0417C_{\mathrm{K}} \quad (2)$$

where D is the absorbed dose rate in nG·y^{-1}, C_{U}, C_{Th} and C_{K} are the activity concentrations of ^{238}U, ^{232}Th and ^{40}K respectively. The dose coefficients in units of nG·y^{-1} per Bq·kg^{-1} were taken from [15].

2.5.2. Annual Effective Dose Equivalent (AEDE)

The absorbed rate in air at about 1metre above the ground surface does not directly provide the radiological hazard to which an individual is exposed [16]. Using an outdoor occupancy factor of 0.2 and conversion factor of 0.7 SvG·y^{-1}, the AEDE from the calculated outdoor terrestrial gamma radiation at about 1 m above the ground in a unit of mSv·y^{-1} will be obtained using the following formula [17].

$$\mathrm{AEDE}\left(\mathrm{mSv}\cdot\mathrm{y}^{-1}\right) = \text{Dose rate}\left(\mathrm{nGy}^{-1}\right)\times 8760\ \mathrm{h}\times 0.2\times 0.7\ \mathrm{Sv}\cdot\mathrm{Gy}^{-1}\times 10^{-6} \quad (3)$$

2.5.3. Radium Equivalent Activity (*Ra$_{eq}$*)

To compare the radiological effect or activity of materials that contain ^{238}U, ^{232}Th and ^{40}K by a single quantity that takes into account the radiation hazards associated with them, a common index termed Radium equivalent activity (Ra_{eq}) is used [18]. Ra_{eq} will be calculated using the relation [19].

$$Ra_{eq} = C_{\mathrm{U}} + 1.43C_{\mathrm{Th}} + 0.077C_{\mathrm{K}} \quad (4)$$

where, C_{U}, C_{Th}, and C_{K} are the radioactivity concentration of ^{238}U, ^{232}Th, and ^{40}K in the granite samples.

2.5.4. Hazard Indices

To estimate the gamma-radiation dose expected to be delivered externally from building materials, a model was suggested by various researchers to limit the radiation dose from the building materials to 1.5 mSv·y^{-1} [20]. In this model, the external hazard index (H_{ex}) is defined as [21]

$$H_{ex} = \frac{C_{\mathrm{U}}}{370} + \frac{C_{\mathrm{Th}}}{259} + \frac{C_{\mathrm{K}}}{4810} \quad (5)$$

where, C_{U}, C_{Th}, and C_{K} are the radioactivity concentration of ^{238}U, ^{232}Th, and ^{40}K in the granite samples.

Internal exposures arise from the inhalation of radon (^{222}Rn) gas and its short-lived decay products as well as from the inhalation and ingestion of other radionuclides [20]. To assess the internal exposure to ^{222}Rn gas, the internal hazard index will be determined using [21].

$$H_{in} = \frac{C_{\mathrm{U}}}{185} + \frac{C_{\mathrm{Th}}}{259} + \frac{C_{\mathrm{K}}}{4810} \tag{6}$$

where, C_{U}, C_{Th}, and C_{K} are the radioactivity concentration of ^{238}U, ^{232}Th, and ^{40}K in the granite samples.

2.5.5. Representative Gamma Index (I_γ) Will Be Determined Using Relation [13]

$$I_\gamma = \frac{C_{\mathrm{U}}}{150} + \frac{C_{\mathrm{Th}}}{100} + \frac{C_{\mathrm{K}}}{1500} \tag{7}$$

where, C_{U}, C_{Th}, and C_{K} are the radioactivity concentration of ^{238}U, ^{232}Th, and ^{40}K in the granite samples.

2.5.6. Annual Gonadal Dose Equivalent (AGED)

The gonads, the bone marrow, and the bone surface cells are considered as organs of interest [4] because they are the most sensitive parts of the human body to radiation. An increase in AGED has been known to affect the bone marrow, destroying the red blood cells that are then replaced by white blood cells. This situation results in a blood cancer called leukemia which is fatal. The annual gonadal dose equivalent (AGED) is calculated using the equation [22].

$$\mathrm{AGED}\left(\mathrm{mSv}\cdot\mathrm{y}^{-1}\right) = 3.09C_{\mathrm{U}} + 4.18C_{\mathrm{Th}} + 0.314C_{\mathrm{K}} \tag{8}$$

where, C_{U}, C_{Th}, and C_{K} are the radioactivity concentration of ^{238}U, ^{232}Th, and ^{40}K in the granite samples.

2.5.7. Excess Lifetime Cancer Risk (ELCR)

ELCR deals with the probability of developing cancer over a lifetime at a given exposure level. It is presented as a value representing the number of cancers expected in a given number of people on exposure to a carcinogen at a given dose. It is worth noting that an increase in the ELCR causes a proportionate increase in the rate at which an individual can get cancer of the breast, prostate or even blood. Excess lifetime cancer risk (ELCR) is given as [23].

$$\mathrm{ELCR} = \mathrm{AEDE} \times \mathrm{DL} \times \mathrm{RF} \tag{9}$$

where AEDE is the Annual Effective Dose Equivalent, DL is the average duration of life (estimated to 70 years), and RF is the Risk Factor (Sv^{-1}), *i.e.* fatal cancer risk per Sievert. For stochastic effects, [24] uses RF as 0.05 for the public [25].

3. Results and Discussions

3.1. Activity Concentrations of Natural Radionuclides

Quarry names, sample code, sample number and GPS location are presented in Table 1. Table 2 summarized mean activity concentrations of ^{238}U, ^{232}Th, and ^{40}K in granite samples from each quarry together with their corresponding uncertainties. Mean activity concentrations of ^{238}U, ^{232}Th, ^{40}K in the samples for different sizes in all locations are shown in Table 3. The activity concentration of ^{238}U ranged from 3.47 Bq·kg^{-1} to 58.98 Bq·kg^{-1} in three-quarter-inch granite, it ranged from 1.53 Bq·kg^{-1} to 27.51 Bq·kg^{-1} in a half-inch granite, while the range is 1.60 Bq·kg^{-1} to 9.67 Bq·kg^{-1} in stone-dust. Based on location, The mean activity concentrations of ^{238}U in three-quarter inch, a half-inch and stone-dust are 12.64 Bq·kg^{-1}, 8.76 Bq·kg^{-1}, and 5.10 Bq·kg^{-1} respectively as shown in **Table 3**. This implies that the larger the size the higher the mean activity. Aggregate size distribution likely has effects on radiation levels since the stone-dust has the least mean activity concentration. This is an indication that fine-grain aggregates allow easy escape of radiations than the coarse aggregates thereby giving a shorter period of hazard compared with other sizes [7]. The activity concentrations of ^{238}U in all locations are below the world average value of 33 Bq·kg^{-1} [4].

The activity concentration of ^{232}Th ranged from 5.23 Bq·kg^{-1} to 77.85 Bq·kg^{-1} with mean value of 16.93 Bq·kg^{-1} in three-quarter inch granite, it ranged from 5.15 Bq·kg^{-1} to 43.86 Bq·kg^{-1} with mean value of 13.69 Bq·kg^{-1} in half-inch granite, while the range of activity concentration in stone-dust is 1.62 Bq·kg^{-1} to 16.55 Bq·kg^{-1} with mean value of 8.97 Bq·kg^{-1} (see **Table 3**). The average activity concentrations of ^{238}Th in all locations and sizes are below the worldwide average activity concentrations of 45 Bq·kg^{-1} [15]. For ^{232}Th, the mean activity concentration decreases as the granite size decreases as shown in **Table 3**. Three-quarter has the highest mean activity concentration of ^{232}Th while stone-dust has the lowest.

Table 2. Mean activity concentrations of ^{238}U, ^{232}Th, ^{40}K in the samples for each sample location.

Sample location	Mean Activity Concentrations (Bq·kg^{-1})		
	^{238}U	^{232}Th	^{40}K
WQ	4.11 ± 0.73	9.58 ± 1.52	137.38 ± 25.10
YQ	6.40 ± 0.75	8.21 ± 1.12	109.54 ± 11.06
AQ	5.05 ± 0.61	7.38 ± 0.90	144.07 ± 14.76
EQ	23.75 ± 3.74	34.65 ± 4.96	315.70 ± 48.42
IQ	7.67 ± 1.15	13.16 ± 1.98	257.20 ± 38.58
KQ	7.53 ± 1.12	11.27 ± 1.68	236.47 ± 35.47
CQ	7.85 ± 1.18	11.24 ± 1.69	171.27 ± 25.69
OQ	8.07 ± 1.21	10.25 ± 1.52	152.37 ± 22.88

Table 3. Mean activity concentrations of ^{238}U, ^{232}Th, ^{40}K in the samples for different sizes in all locations.

Size (Inch)	Mean Activity Concentrations (Bq·kg^{-1})		
	^{238}U	^{232}Th	^{40}K
$\frac{3}{4}$ inch	12.64 ± 1.89	16.93 ± 2.46	266.19 ± 35.53
$\frac{1}{2}$ inch	8.76 ± 1.28	13.69 ± 1.95	151.85 ± 25.09
Stone-dust	5.10 ± 0.77	8.97 ± 1.37	164.27 ± 23.51

Of the three granite sizes, three-quarter-inch has the highest average activity of ^{40}K followed by stone-dust which is slightly greater than that of half-inch.

The range and mean activity concentration of ^{40}K for three-quarter inch, half inch granite and stone-dust respectively are (90.21 - 672.54) 266.19 Bq·kg^{-1}, (56.53 - 426.68) 151.85 Bq·kg^{-1} and (57.88 - 379.77) 164.27 Bq·kg^{-1} respectively (see Table 3).

All the mean activity concentrations values of ^{40}K are below the worldwide average value of 412 Bq·kg^{-1} [15].

3.2. Comparison of the Average Activity Concentrations of ^{238}U, ^{232}Th, and ^{40}K with Values from Other Studies Worldwide

The mean activity concentrations of ^{238}U, ^{232}Th, and ^{40}K in this work were compared with the reported values obtained by other researchers such as [4] [7] [26] [27] [28], etc.

The result is comparison in Figure 3. It is important to note that the values shown are not representative values for the countries mentioned, but only for the regions in which the samples were collected.

3.3. Radiation Hazard Index

To establish the health risk due to the exposure to natural radionuclides in granite on quarry workers, granite end-users and the general public in the study area, the radiation hazard indices were calculated (Table 4).

The mean absorbed dose rate in air in the study area ranged from 4.56 nGy·h^{-1} to 38.10 nGy·h^{-1}. This is attributed to the size of the samples and geology of the study areas. The absorbed dose rate in air in stone-dust is least in Wolid, and Slava Yetidepe (WQ and YQ) whereas highest in Ife/Modakeke and Espro (IQ and EQ). This is because the activity concentration of natural radionuclides in granite is proportional to the size of the granite. The larger the granite sizes the higher the activity concentrations of natural radionuclides in the granite of study areas, as shown in Table 3.

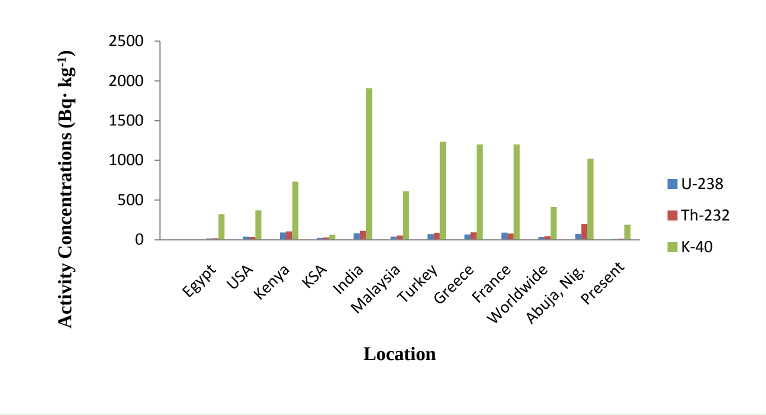

Figure 3. Comparison of the average activity concentrations of ^{238}U, ^{232}Th, and ^{40}K with values from others.

Table 4. Correlation result of ^{238}U, ^{232}Th, and ^{40}K in a three-quarter, half-inch and stone-dust.

	Mean activity concentrations (Bq·Kq^{-1})		
	^{238}U	^{232}Th	^{40}K
Pearson Correlation (r)	0.984	0.998	0.707
r^2 value	0.968	0.996	0.500

^{40}K is the highest contributor to the absorbed dose rate in air, followed by ^{232}Th and then ^{238}U. The chart showing the contributions of ^{234}U, ^{232}Th and ^{40}K to the absorbed dose rate in air in the study area is shown in **Figure 4**. ^{40}K accounts for 90% of the absorbed dose rate in air, while ^{232}Th and ^{238}U account for 6% and 4% respectively. The absorbed dose rate in air in all locations is below the world average value of 60 nGy·h^{-1} [4].

The absorbed dose rate in the air at about 1 m above the ground surface does not directly provide the radiological risk to which an individual is exposed [16].

The annual effective dose equivalent (AEDE) as presented in **Table 5** for the selected quarry sites in this study varied from 5.60 to 46.73 μSv·y^{-1}. AEDE for 3/4 inch, 1/2 inch and stone-dust ranged from 10.80 to 46.73, 6.64 to 30.69 and 5.60 to 20.18 μSv·y^{-1} respectively with mean values of 17.10, 7.45 and 7.55 μSv·y^{-1} respectively. When compared with a worldwide effective dose of 0.07 mSv·y^{-1} [15], the results in this work is lower. The highest AEDE was found in 3/4 inch granite from Espro and the least in stone dust from Wolid quarry site. According to [24], the acceptable annual effective dose rate recommended for members of the public without constraint is 1.0 mSv·y^{-1}.

The radium equivalent activity (Ra_{eq}) ranged from 28.79 Bq·kg^{-1} to 162.43 Bq·kg^{-1}, 21.64 Bq·kg^{-1} to 108 Bq·kg^{-1} and 17.13 Bq·kg^{-1} to 46.32 Bq·kg^{-1}, with mean values of 43.85 Bq·kg^{-1}, 27.44 Bq·kg^{-1} and 22.76 Bq·kg^{-1} for 3/4 inch granite, 1/2 inch granite and stone-dust respectively. This is below the permissible maximum value of 370 Bq·kg^{-1} [4] which corresponds to an effective dose of 1 mSv for the general public. The radium equivalent activity (Ra_{eq}) is highest in three-quarter inch granite from Espro (EQ). The radium equivalent activity (Ra_{eq}) is lowest in stone-dust from Wolid (WQ).

The mean annual gonadal equivalent dose (AGED) ranged from 98.32 mSv·y^{-1} to 517.90 mSv·y^{-1}, 72.15 mSv·y^{-1} to 351.12 mSv·y^{-1} and 58.40 mSv·y^{-1} to 155.02 mSv·y^{-1}, with mean values of 150.98 mSv·y^{-1}, 88.85 mSv·y^{-1} and 76.54 mSv·y^{-1} respectively for 3/4 inch granite, 1/2 inch granite and stone-dust respectively. The AGED is highest in three-quarter inch granite from Espro (EQ), and is at lowest in stone-dust from Wolid (WQ). An increase in AGED has been known to affect the bone marrow, destroying the red blood cells that are then replaced by white blood cells. This situation results in a blood cancer called leukemia which is fatal. The mean values are lower than the maximum permissible value of 300 mSv·y^{-1} [15]. Hence, the quarry workers, granite end-users and the general populace in the study area are not at risk of developing blood cancer due to the exposure to the natural radionuclides present in the study area.

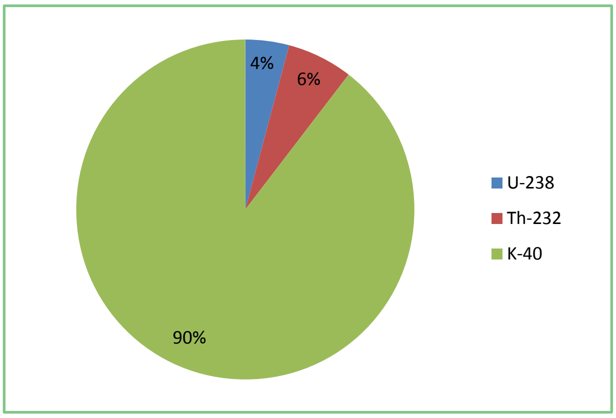

Figure 4. Contribution of ^{234}U, ^{232}Th, and ^{40}K to absorbed dose rate in air.

Table 5. Mean radiation hazard indices.

Sample Location/ Size	D (nGy·h^{-1})	AEDE (μSv·y^{-1})	Ra_{eq} (Bq·kg^{-1})	AGED (mSv·y^{-1})	ELCR × 10^{-3}	H_{ex}	H_{in}	I_{γ}
WQ	10.93	13.41	30.71	109.26	0.05	0.08	0.10	0.24
YQ	8.56	10.50	26.38	88.62	0.04	0.07	0.09	0.20
AQ	7.41	9.09	22.54	78.87	0.03	0.06	0.07	0.17
EQ	21.02	25.78	81.56	266.08	0.09	0.22	0.28	0.6
IQ	13.29	16.29	34.73	124.10	0.06	0.09	0.11	0.27
KQ	9.52	11.68	32.97	110.69	0.04	0.09	0.11	0.25
CQ	16.88	20.70	46.55	166.17	0.07	0.13	0.15	0.36
WWA	60	70	370	300	1	1	1	1
$\frac{3}{4}$ inch	3.94	17.10	43.84	150.98	0.06	0.12	0.14	0.33
$\frac{1}{2}$ inch	6.08	7.45	27.44	88.85	0.03	0.07	0.09	0.20
Stone-dust	6.16	7.55	22.76	76.54	0.03	0.06	0.07	0.17

The external hazard index (H_{ex}) ranged from 0.08 to 0.44, 0.06 to 0.29 and 0.05 to 0.13, with mean values of 0.12, 0.07 and 0.06 for 3/4 inch granite, 1/2 inch granite and stone-dust respectively. While the internal hazard index (H_{in}) ranged from 0.10 to 0.56, 0.07 to 0.35 and 0.05 to 0.15, with mean values of 0.14, 0.09 and 0.07 for 3/4 inch granite, 1/2 inch granite and stone-dust respectively. These values do not exceed the acceptable limit value of unity (ICRP, 2010), thus, suggesting that radiation hazard due to the exposure to natural radionuclides in the study area is negligible.

The external hazard index (H_{ex}) and the internal hazard index (H_{in}) are highest in three-quarter inch granite from Espro (EQ). The external hazard index (H_{ex}) is lowest in both Ayofe and Wolid (AQ and WQ) while the internal hazard index (H_{in}) is lowest in stone-dust from wolid (WQ).

The representative gamma index (I_γ) ranged from 0.22 to 1.17, 0.20 to 0.79 and 0.13 to 0.35, with mean values of 0.33, 0.20 and 0.17 for 3/4 inch granite, 1/2 inch granite and stone-dust respectively. An increase in the representative gamma index greater than the universal standard of unity may result in radiation risk leading to the deformation of epithelial and blood cells, thereby causing cancer [29]. Since the values of the representative gamma index (I_γ) is lower than 1 in all the quarries except Espro ($I_\gamma = 1.17$ for 3/4 inch granite), thus, the populace of the study area does not suffer significant health risk due to the exposure to radiation from natural radionuclides in the granite of the study area.

The excess lifetime cancer risk (ECLR) has to do with the probability of developing cancer over a lifetime at a given exposure level. The excess lifetime cancer risk (ECLR) ranged from 0.04 to 0.16 with a mean value of 0.06 for three-quarter-inch granite, ranged from 0.02 to 0.11 with a mean value of 0.03 for half-inch granite and from 0.02 to 0.07 with a mean value of 0.03 for stone-dust. These values are significantly lower than unity, so the risk that the general public of the study area would develop cancer due to the exposure to radiation emitted from natural radionuclides is very low or insignificant.

The excess lifetime cancer risk is highest in 3/4 inch granite and lowest in stone-dust (<1/2 inch granite).

The excess lifetime cancer risk (ELCR) is highest at the Espro (EQ), closely followed by the Clario quarry (CQ). The excess lifetime cancer risk is lowest at Wolid (WQ).

The values of the radiation hazard indices in all locations are below the maximum permissible limit set by ICRP. Hence, radiation emitted from natural radionuclides (^{238}U, ^{232}Th, and ^{40}K) in the granite of the study areas does not pose a serious health risk to the quarry workers, granite end-users and the general public of the study area.

3.4. Statistical Analysis

The Pearson correlation results of ^{238}U, ^{232}Th and ^{40}K in a three-quarter, half-inch granite, and stone-dust as shown in **Table 4** are 0.984 0.998 and 0.707 respectively indicating a strong positive linear relationship between the mean activity concentrations of ^{238}U, ^{232}Th and ^{40}K respectively and granite size. The r^2 values are 0.968, 0.996 and 0.500, implying that 96.8%, 99.6% 50% of the mean activity concentration of ^{238}U, ^{232}Th and ^{40}K can be explained by granite size.

4. Conclusions

The ^{238}U, ^{232}Th, and ^{40}K content in eighty granite samples from eight quarry sites in Osun State, southwest of Nigeria, was assessed using high purity germanium (HPGe) detector.

Statistical Package for Social Sciences (SPSS) software was used for statistical analysis to determine the impact of granite size on the activity concentrations of the natural radionuclides in the granite. With the results obtained, there is a strong linear relationship between granite size and the activity concentrations of the natural radionuclides in the granite of the study area. The higher the granite size, the higher the activity concentrations of ^{238}U, ^{232}Th and ^{40}K; and vice versa. In all the samples, potassium-40 contributed majorly to the absorbed dose rate in the air due to its abundance on the earth. The measured values in the present work showed a low activity concentration for ^{238}U, ^{232}Th and ^{40}K for all the granite samples analyzed, in contrast with other results found in the literature. Regarding the natural radioactivity content, the values revealed that the people working in the quarry sites, granite end-users and the general public from the study area are safe from radiological health risks from the sites since there is no significant health risk. More studies are recommended to be carried out to ascertain the relationship between granite size and activity concentrations of natural radionuclides.

Conflicts of Interest

The author declares no conflicts of interest regarding the publication of this paper.

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