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Radiological Health Implications of Radiation Levels In Rock Fragments From Quarry Sites In Akamkpa, South-South Nigeria

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Abstract
The concentrations of the naturally occurring radionuclides in rock fragments from two quarry sites in Akamkpa, Cross Rivers State, South-south Nigeria have been measured, using gamma ray spectrometry (NaI (TI) detector). The photo peaks observed with reliable regularity in all the samples analysed belonged to the naturally occurring radionuclides (^{238}U, ^{232}Th and ^{40}K). No artificial radionuclides were detected in any of the samples. The mean concentration for ^{40}K, $^{238}U$and ^{232}Th were 868.70 ± 47.05 Bq.kg^{-1}, 36.51 ± 5.34 Bq.kg^{-1} and 40.46 ± 3.18 Bq.kg^{-1}, respectively. The concentrations of these radionuclides were lower than the world average concentrations of 412, 33, and 45 Bq.kg^{-1} for ^{40}K, ^{238}U, and ^{232}Th, respectively. The gamma absorbed dose rate (DR), radium equivalent activity (Ra_{eq}), annual effective dose equivalent (AEDE), activity concentration index(I), external radiation index (H_{ex}) , and internal radiation hazard index (H_{int}) associated with the radionuclides were evaluated and were found to be lower than permissible limits. The result indicates therefore, that the rock fragments may not pose a radiological hazard to both the employees and the general public. Hence, they are safe for use as geomaterials for construction and beautification purposes.
Key Words: *Gamma radiation, natural radionuclide, Gamma spectroscopy, radioactivity, quarry sites*

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I. Introduction

The human environment, when exposed to deleterious substances, can become polluted, thus having a harmful effect on man and other biotic organisms in the environment. Radioactive elements are one of the sources of pollution in the environment, if allowed to build up due to natural occurrence or anthropogenic activities. Radionuclides which emit radiation are found naturally in air, water and soil. Natural radioactivity is common in the rocks and soil that make up our planet, in waters and oceans, and in building materials [1].

Human beings have always been exposed to ionizing radiation from the earth. The natural radioactivity in rock comes from thorium (^{232}Th) and uranium (^{238}U) series and potassium (^{40}K). Quarry activities which involve blasting, crushing and processing of rocks into different aggregates releases radionuclides into the atmosphere and the radiological implication of these radionuclides is gamma exposure of the body and irradiation of the lung from inhalation and ingestion of radon and its daughters [2]. Therefore, the assessment of gamma radiation dose from natural sources is of particular importance as natural radiation is the largest contributor to the external dose of the world population [3].

Mining has been identified as one of the potential sources of exposure to naturally occurring radioactive materials [3]. The activities associated with mining of rock fragments are prevalent in the study area. As a result, it is necessary for baseline radiological studies to be carried out prior to the commencement of mining activity and subsequently similar studies (auditing) be conducted to ascertain the levels of these radionuclides as a result of the mining activities in the operational phase of the mine [4].

The town of Akamkpa in Cross River State, South-south Nigeria, hosts a significant number of quarry sites due to the presence of large outcrop of rocks mainly granites and gneisses that belong to the bedrocks of Oban Massif. Consequently, there is a concentration of rock-blasting activities within the area, more so due the fact that majority of south-south region of the country is underlain mainly by sedimentary rock formations. Hence, any outcrop of hard rock found in this part of the country is seen as a goldmine, resulting in high mining (blasting of the rocks) activities for construction and related purposes.

Therefore, it is important to carry out measurements and determine the background radiation exposure levels in the area, in order to ascertain whether the activities of quarry operational activities pose a health risk to both quarry site employees and the general public. Not only that, the collection of data on these quarries within the study area will improve our understanding of exposure levels, as well as add to the body of already existing knowledge, which will be referenced in the future by researchers.

II. Location And Local Geology Of The Study Area

The study areas are COSEL Quarry within latitudes 5° 10' N and 5° 42' N and longitudes 8° 21' E and 8° 44' E in New Netim in Akamkpa Local Government Area, Cross Rivers State (Location 1) (Fig. 1) and Alabi Brothers Quarry within latitudes 5° 12' N and 5° 32' N and longitudes 8° 20' E and 8° 34' E in Netim in Akamkpa Local Government Area of Cross Rivers State (Location 2) (Fig. 1). It has an area of 5,003km^2 and is easily accessible to other major and urban centers in the state.

Fig. 1: Map of Cross River State showing Akamkpa (The study area)

The study area is underlain by rocks of the Oban Massif. The Oban basement Massif of South-eastern (now South-south) Nigeria is composed of metamorphic rocks including phyllites, schists, gneisses and amphibolite's cut by pegmatite dykes of varying lengths and thicknesses, which intruded the metamorphic rocks [5].

There is a concentration of quarrying activities within Akamkpa because of the huge potential of basement lithology that are found within the area. Rock fragments from these quarries find extensive utility in construction and home beautification. Granites are used in road surfacing and all kinds of civil engineering works. The gems and other accessory minerals that abound within the area have been reported by several workers to be of high grade and are economically valuable.

III. Materials And Methods

3.1 Sample Collection and Preparation

A total of ten (10) samples were collected from two (2) different quarry sites (five samples from each site). These sites spread across Akamkpa LGA in Cross Rivers State where major quarry activities take place in South-South, Nigeria. Each sample was sealed in a well labelled polythene bag and taken to gamma laboratory at National Institute of Radiation Protection and Research Centre, University of Ibadan, Ibadan, for preparation and analysis. Wet samples were air dried at room temperature to constant weight and all dried samples were crushed and pulverized with Rock lab ring mill, after which they were sieved to pass 2-mm mesh. About 200g each of the samples was weighed into cylindrical polyvinylchloride containers sealed and kept for 28 days in order to attain secular equilibrium between the parent and the daughter nuclides present. The activity concentration of natural radioactivity in the samples were determined using a 7.62cm x 7.62cm NaI (Tl) detector employed with adequate lead shielding (with 1.5mm thickness) which reduced the background by a factor of about 95% [6]. Energy calibration was done using Standard sources, of known gamma-ray energies and activities, prepared by the Isotope Products Laboratories, Burbank California, USA. The calibration provided qualitative and quantitative analysis of the radionuclides present in each sample. Counting was done for 25,200s for each of the samples; the calibrated reference material and an empty container were used to determine the background. Current decay data for nuclides were obtained from literature [5, 7]. The activities of various radionuclides were determined in Bq kg-1 using the count spectra obtained from each of the samples. The gamma ray photo peaks corresponding to energy of 1120.3 keV (214Bi), 911.21 keV (228Ac) and 1460.82 keV (40K) were considered to determine the activity of 238U, 232Th and 40K. The detection limits of the NaI (Tl) detector system were calculated as 31.57, 5.73 and 0.26 Bq kg-1 for 40K, 232Th and 238U respectively for a counting time of 25,200s. The radionuclide activity concentration values for the two quarry sites are summarized in table 1 below.

Table 1: Radionuclide concentration in the samples from quarry sites

S/N	SAMPLE CODE	^{40}K(Bq.kg^{-1})	^{238}U(Bq.kg^{-1})	^{232}Th(Bq.kg^{-1})
1	CS1	971.9 ± 51.7	35.5 ± 5.9	49.1 ± 3.7
2	CS2	875.9 ± 46.8	37.5 ± 1.7	38.7 ± 3.0
3	CS3	855.9 ± 45.7	41.7 ± 7.0	35.0 ± 3.0
4	CS4	815.3 ± 47.06	37.5 ± 6.7	42.2 ± 3.2
5	CS5	824.6 ± 44.1	30.3 ± 5.3	37.4 ± 3.0
6	AB1	732.1 ± 39.4	40.4 ± 6.7	41.6 ± 3.2
7	AB2	867.5 ± 38.7	34.5 ± 2.4	37.6 ± 5.3
8	AB3	795.6 ± 24.2	28.8 ± 3.9	34.3 ± 3.7
9	AB4	689.7 ± 37.2	20.2 ± 3.8	25.9 ± 2.2
10	AB5	803.2 ± 51.8	39.7 ± 2.5	43.4 ± 4.8
	$Mean \pm SD$	823.2 ± 42.7	35.0 ± 5.0	38.5 ± 3.5

IV. Results

4.1 Absorbed Dose Rate in Air (D)

In order to assess any radiological hazard, the exposure to radiation arising from radionuclides present in rock can be determined in terms of many parameters. A direct connection between radioactivity concentrations of natural radionuclides and their exposure is known as the absorbed dose rate in the air at 1 meter above the ground surface. The mean activity concentrations of ^{238}U, ^{232}Th and ^{40}K (Bq.kg^{-1}) in the granite samples are used to calculate the absorbed dose rate (ADR) given by the following formula [8].

$$ADR = 1.60 \times 10^{-10} A\bar{E} Gys^{-1} \qquad (4.1)$$

The average absorbed dose rates (ADR) were used to calculate the annual effective dose equivalent (AEDE) in mSv/yr received by workers, visitors and people living around the area. The AEDE was computed using the relation [9, 10]:

$$AEDE\left(\frac{mSv}{yr}\right) = ADR \times T \times OF \times 0.001 \qquad (4.2)$$

Where

ADR is Absorbed Dose Rate, T is Time of Exposure, and OF is Occupancy Factor

4.2 Annual Effective Dose Equivalent (AEDE)

The absorbed dose rate in air at 1metre above the ground surface does not directly provide the radiological risk to which an individual is exposed. Using an outdoor occupancy factor of 0.2 and the conversion factor of 0.7SvGy^{-1}[9], the annual effective dose equivalent (AEDE) from the calculated outdoor terrestrial gamma radiation at 1m above the ground in the various quarry sites in Akamkpa in unit of mSvy^{-1} was obtained by using the following formula [10].

$$AEDE(mSv.y^{-1}) = Dose\ rate\ (nGyh - 1) \times 8760h \times 0.2 \times 0.7SyGy^{-1} \times 10^{-6} \qquad (4.3)$$

4.3 Radium Equivalent (Ra_{eq})

To compare the specific activities of the samples, the radium equivalent activity Ra_{eq} can be used as a common index, Radium equivalent provides a useful guideline in regulating the safety standards on radiation protection for the general public. It is the sum of the weighted activities of ^{226}Ra, ^{232}Th and ^{40}K based on the estimation that 10 Bq kg^{-1} of ^{26}Ra, 7 Bq kg^{-1} of ^{232}Th and 130 Bq kg^{-1} of ^{40}K will deliver equal or the same gamma dose rate (Jibiri and Farai,1999). The radium equivalent was calculated through the use of the following equation

$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_k$ (4.4)

Where $A_{Ra,}$ $A_{Th}$and A_k are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in $Bqkg^{-1}$, respectively. From radiological point of view and safety limits, the maximum value of Ra_{eq} for a material to be used in building construction is that; $Ra_{eq} \leq 370 Bq.kg^{-1}$ [11, 12]. Radium equivalent of 370 $Bq.kg^{-1}$ corresponds to the dose limit of 1mSv for the general population. The use of materials whose Ra_{eq} concentrations exceeds 370$Bqkg^{-1}$ is discouraged in order to avoid radiation hazards. Using equation (4.4) and activity concentrations of the radionuclides in Table 1 above, the Ra_{eq} concentrations calculated in Bq/kg varied from 147.3 to 180.5 with an average of 161.3 for Cossel Quarry, and ranged from 110.4 to 163.7 with an average of 144.8 for Alabi Brothers Quarry. The results are presented in Table 2.

4.4 External Hazard Index (H_{ex})

The external hazard index is an important criterion used to assess the radiological suitability of a material for building purposes. The external hazard index is an evaluation of the hazard of the natural gamma radiation. The prime objective of this index is to limit the radiation dose to the admissible permissible dose equivalent limit of $1mSv.y^{-1}$. The external hazard index should be below unity. The external hazard index due to natural gamma radiation was calculated using the relation [13].

$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_k}{4810}$ (4.5)

Where A_{Ra}, A_{Th} and A_k are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in $Bq.kg^{-1}$, respectively. The external hazard index is obtained from the expression for Ra_{eq} through the supposition that its maximum allowed value corresponds to the upper limit of $Ra_{eq}(370Bq.kg^{-1})\left(H_{ex} = \frac{Ra_{eq}}{370}\right)$ so that the annual external dose rate does not exceed 1.5mGy. To limit the external gamma dose for materials to 1.5mGy y^{-1} for radiation hazard to be negligible, the external hazard index should conform to the criterion: $H_{ex} \leq 1$. The external Hazard Index $H_{ex}$for COSSEL Quarry varied from 0.398 to 0.489 with an average of 0.4342, and ranged from 0.298 to 0.442 with a mean of 0.3914 for Alabi Brothers Quarry. The H_{ex} values for the two sites under consideration are also presented in Table 2.

4.5 Internal Hazard Index (H_{int})

In addition to the external hazard, there is also a threat to respiratory organs due to the ^{222}Rn, the decay product of ^{226}Ra and its short-lived decay products. The internal hazard index is defined generally so as to reduce the maximum permissible concentration of ^{226}Ra to half the value appropriate to external exposure alone [14]. The internal hazard index (H_{int}) gives the internal exposure to carcinogenic radon and the value of this index should be less than unity in order for the radiation hazard to have negligible hazardous effects to the respiratory organs of the individuals living in the dwellings. The internal exposure to radon and its progeny products is quantified by estimating internal hazard index through the equation [12]:

$H_{int} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_k}{4810}$ (4.6)

Where A_{Ra}, $A_{Th}$and A_k are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in $Bq.kg^{-1}$, respectively. If the maximum concentration of ^{226}Ra is half that of the normal acceptable limit then H_{int} will be less than unity. For safety precautions in the use of materials in construction of dwellings; the criterion demands that $H_{int} \leq 1$. The estimated values of $H_{int}$for Cossel Quarry ranged from 0.413 to 0.538 with a mean of 0.500, and varied from 0.353 to 0.550 with an average of 0.480 for Alabi Brothers Quarry. The results along with other parameters are also presented in Table 2

Table 2: Radiological hazard indices in the samples

Sample ID	Dose (nGy/h)	Dose Error	AEDE (mSv/y)	External Hazard Index H_{ex}	Internal Hazard Index H_{int}	Gamma Index (I)	Ra (Bq/kg)
CS1	60.330	5.774	0.074	0.489	0.413	0.689	180.52
CS2	53.263	2.283	0.065	0.433	0.534	0.617	160.24
CS3	52.974	5.692	0.065	0.426	0.538	0.599	157.62
CS4	54.775	5.735	0.067	0.434	0.535	0.608	160.56
CS5	48.727	4.909	0.059	0.398	0.480	0.563	147.32
AB1	54.558	5.626	0.067	0.422	0.531	0.587	157.27
AB2	51.402	4.889	0.063	0.419	0.512	0.592	155.07
AB3	45.694	4.388	0.056	0.376	0.453	0.533	137.65
AB4	35.128	3.656	0.043	0.298	0.353	0.427	110.37

AB5	56.384	4.851	0.069	0.442	0.550	0.617	163.66

4.6 Gamma - index (I_γ)

The gamma index or representative index is a hazard parameter used to correlate the annual dose rate due to the excess external gamma radiation caused by superficial materials. It is used only as screening tool for identifying materials that might be of health concern when used as construction materials [15]. The gamma index (I_γ) was calculated as proposed by European commission [16].

$$I_\gamma = \frac{A_{Ra}}{300} + \frac{A_{Th}}{200} + \frac{A_k}{3000} \quad (4.7)$$

Where A_{Ra},$A_{Th}$and $A_k$are the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in Bq kg$^{-1,}$ respectively. Values of $I_\gamma \leq 1$ corresponds to an annual effective dose of less than or equal to 1mSv while $I_\gamma \leq 0.5$ corresponds to an annual effective dose less than or equal 0.3 mSv [17]. The estimated value of Gamma Index (I_γ) for Cossel Quarry ranged from 0.563 to 0.689 with an average of 0.689, and varied from 0.427 to 0.617 with a mean value of 0.550. The result is also presented in Table2

4.7 Excess Life-time Cancer Risk (ELCR)

This estimates the probability of developing cancer over a life time at a given level of exposure using 70 years as the average life duration of humans. The equation is given by [18] as:

$$ELCR = AEDE \times ALD \times CRF \quad (4.8)$$

Where AEDE is the Annual Effective Dose Equivalent, ALD is the Average Life Duration estimated at 70 years, later modified to 54.5 years for an average Nigerian by [19] and CRF is the Cancer Risk Factor which is 0.05 for the public.

V. Discussion

Radioactivity measurements were carried out using gamma-ray spectroscopy to determine the radionuclide contents and concentrations in the soil samples collected. The results of the analysis of the samples as could be seen from Table 1 showed that concentration of ^{40}K within the Cossel quarry site ranged from 732.14 ± 39.35 to $971.85 \pm 5172 Bq.kg^{-1}$, ^{238}U ranged from 30.33 ± 5.30 to 41.71 ± 7.01Bq.kg^{-1} and ^{232}Th varied from 34.97 ± 2.95 to 49.09 ± 2.95Bq.kg^{-1}. The mean concentration for ^{40}K is $868.70 \pm 47.05 Bq.kg -^{1}$, ^{238}U is $36.51 \pm 5.34 Bq.kg^{-1}$ and ^{232}Th is $40.46 \pm 3.18 Bq.kg^{-1}$. The average values of both ^{40}K and ^{238}U are higher while that of ^{232}Th is slightly lower than the world wide average activity concentrations of ^{40}K, ^{238}U and ^{232}Th which are 412, 33, and 45 Bq.kg^{-1} respectively [20].

Potassium (^{40}K) has the highest concentration in all areas of the study due to its relative abundance. The higher concentrations of radionuclides in some of the samples collected may be attributed to geological areas consisting of different granite rocks mainly feldspar, biotite, quartz, plagioclase and gneisses, which contain higher concentrations of potassium. From Table 2, the calculated value for the absorbed dose rate ranged from 35.128 to 60.330nGyh^{-1}with an average mean of 51.323nGyh^{-1}, which is 1.13times lower than the world wide mean of 58nGyh^{-1} [20]. The highest dose rate was recorded at location A1 with absorbed dose rate of 60.330 nGyh-1which also revealed the overall highest contribution in activity concentration for ^{232}Th. Potassium-40and ^{232}Th are the main contributor to the absorbed dose rate in most of the rock samples measured in this work due to relative abundance of ^{40}K in the areas and high presence of ^{232}Th.The annual effective dose rate as presented in Table 2 for the selected quarry sites in this study varied from 0.043 to 0.074mSv.y^{-1}, with the mean of 0.063mSvy^{-1} but when compared with the worldwide effective dose of 0.07mSv.y^{-1} [20] the results in this work was lower. The highest and lowest AEDE was found in Cossel quarry site.

The radiation hazard parameters in terms of the external hazard (H_{ext}), internal hazard (H_{int}) index and radium equivalent activity (Ra_{eq}) were also evaluated. The results for the calculated Ra_{eq} in this work are given in table 2. The values of Ra_{eq} ranged from 110.37 to 180.52 Bq.kg^{-1} with an average mean of 152.95 Bq.kg^{-1}. It can be seen that the (Ra_{eq}) values for all the rock samples in this work are lower than the safe limit of 370 Bq.kg^{-1} as recommended by the Organization for Economic Cooperation and Development (OECD). Therefore, the use of these granite rocks as raw materials for building could not pose a health risk to an individual as all the calculated values of the external hazard and internal hazard indices and for all rock samples studied in this work were all lower than the acceptable limit value of unity [21]. These values suggest that the rocks could not pose a risk to the health of the inhabitants residing in the areas.

VI. Conclusion

The activity concentrations of ^{40}K, ^{232}Th, ^{238}U and radiation exposure levels and their possible radiological effects inthe rock fragments from the selected quarry sites in Akamkpa, Nigeria, have been determined using the gamma-ray spectroscopyThe results showed that the activity concentration values varied due to the variation in geological structure of each area of study. The results also showed that the average value of radium equivalent (Ra_{eq}), absorbed dose rate in air from most of the samples were lower than the worldwide value. Furthermore, other radiological hazard indices determined showed a clear threshold within conventionally recommended safety limits. The findings in this research indicate that workers and inhabitants in the study areas are radiologically safe. It is recommended that relevant authorities in the industry should step up their monitoring efforts, to ensure that employees and residents of the area are not exposed to environmental hazards resulting from the activities of the quarry sites.

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