



Natural Radionuclides Distribution in Quarry Products from Quarries in Osun and Oyo States, Southwestern Nigeria

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ABSTRACT

Granites are one of the major quarry products that are widely used in building materials; thus, it is imperative to investigate their radiological safety with regard to human protection. This study, therefore, determines the concentrations of natural radionuclides of ^{40}K , ^{238}U , and ^{232}Th in quarry products of three different sizes: Stone-Dust ($< 2\text{ mm}$), 3/4-Down ($\approx 10\text{ mm}$) and 3/4-Up ($\geq 15\text{ mm}$), collected from quarries in Osun and Oyo States, Southwestern Nigeria, using the gamma-ray spectrometry technique. The average radioactivity concentrations of ^{232}Th , ^{238}U , and ^{40}K for Stone-Dust samples are 8.92 ± 0.81 , 15.70 ± 1.19 , and $374.26 \pm 12.74\text{ Bq/kg}$, respectively; 2.78 ± 0.39 , 3.99 ± 0.65 , and $146.99 \pm 2.55\text{ Bq/kg}$ respectively for 3/4-Up samples and 5.68 ± 0.64 , 6.61 ± 0.57 , and $250.78 \pm 4.02\text{ Bq/kg}$, respectively for 3/4-Down samples. The results of one-way analysis of variance (ANOVA-1) and Scheffe pair-wise comparisons tests showed that there exists a significant difference between the activity concentration of the quarry products ($F_{cal} > F_{va}$) at $\alpha = 0.05$, but all the analyzed samples have lower activity concentrations when compared with the recommended limits. It can therefore be concluded that the activity concentrations of the radionuclides decrease as the sample size increases and that quarry processes play a significant role in the activity concentrations of quarry products. Thus, adequate ventilation is needed to ensure diffusion of natural radionuclides whenever these quarry products are used as part of building materials.

INTRODUCTION

Natural radionuclides and their progenies are generally spread on the earth's crust with a considerable amount of these radionuclides existing in many mining compositions, such as quarry soil and other environmental matrices, such as water bodies surrounding the quarry site (Nduka *et al.*,

2022; Amusat *et al.*, 2025). These radionuclides are not evenly distributed within the quarry ecosystem; rather, they decay to release toxic ionizing radiations that have detrimental health effects (Akinyose *et al.*, 2018; Isola *et al.*, 2019; Amodu *et al.*, 2023). Quarry products comprise different natural rocks with different mineral contents, crushed into various sizes at quarries (Gbenu *et al.*,

2016). These products, also known as aggregates, are heavily used in the building and construction industries in the construction of houses, roads, hospitals, schools, and other ornamental purposes. Radiologically, naturally occurring radioactive materials (NORMs) such as ^{238}U , ^{232}Th , and ^{40}K are found in trace amounts in building materials (Isola *et al.*, 2021; Lawal *et al.*, 2021; Aremu *et al.*, 2023). Hence, there is a growing concern about the use of materials that are less radioactive for building purposes (Ayanlola *et al.*, 2025). This is because ionizing radiation (IR) possesses sufficient energy to liberate electrons from atoms, altering the chemical arrangement of the materials with which it traverses. In living tissues, this can cause damage to the deoxyribonucleic acid (DNA), potentially resulting in cell death or mutations, such as cancer (UNSCEAR, 2000; ICRP, 2007). Thus, while buildings serve as a fundamental source of shelter, the materials used during their construction should be free of toxic or harmful substances.

Studies on radionuclide distribution in the environment provide vital radiological baseline information, which is essential in understanding human exposure from natural and man-made sources of radiation (Saleh *et al.*, 2007) and also necessary in establishing rules and regulations relating to radiation protection (Quindos *et al.*, 1994). Research has also shown that quarrying processes increase the activity concentration of radionuclides in the production environment (Gbadebo, 2011; Akinloye *et al.*, 2019). Similarly, Lawal (2019) investigated the impact of granite size on the activity concentrations of the natural radionuclides in the granite samples and concluded that the higher the granite size, the higher the activity concentrations of the natural radionuclides. Thus, this present study, apart from measuring the specific activities of ^{40}K , ^{238}U , and ^{232}Th in the three different sizes of quarry products, also investigated

the effects of quarry products' different sizes and finally compared the radionuclides' mean activity concentrations with the World Average Values.

METHODOLOGY

Study Areas

Osun and Oyo States are both located in the Southwestern geopolitical zone of Nigeria. Osun State lies between longitudes 4° and 5° E. of the Greenwich Meridian and latitudes 6° and 8° N of the Equator (Osun State Government, 2014), while Oyo State lies between longitudes 3° and 5° E of the Greenwich Meridian, latitudes 6.5° and 9° N of the Equator. The two States fall entirely in the tropics. Both states are characterized by a Precambrian crystalline basement complex and the dominant geological features include migmatite-gneiss complex, meta-sediments (like quartz and mica schist), and older granites. These rocks are typically found within a tropical rainforest zone. In view of these endowed natural resources, the quarry processes in both states have increased, contributing significantly to the economic growth of both states. In all, a total of 8 quarry sites (3 from Osun and 5 from Oyo) were assayed.

Collection and Preparation of Samples

A total of 52 quarry product samples of three different sizes: Stone-Dust (< 2 mm), 3/4-Down (≈ 10 mm) and 3/4-Up (≥ 15 mm) were collected from quarries in Osun and Oyo States, Southwestern Nigeria. The samples were air-dried at room temperature until a constant weight was achieved. The dried samples were then crushed into powder. The powdered samples of the same size from the same source were each packed into a plastic container that matches the geometry of the detector and tightly sealed with the aid of polyvinyl chloride (PVC) tape. All samples were weighed and kept for a period of 28 days before measurement in order to attain radioactive secular equilibrium (Gbenu *et al.*, 2016).

Samples Coding

The samples were coded as follows, in order to prevent identification errors. The sample codes consist of two letters and a number. The alphabets connote the study area, that is, the State from which the products were collected and the number stands for the serial number of the sample. For instance, OY 2 stands for sample number 2 from Oyo State, while OS 4 stands for sample number 4 from Osun State and so on.

Gamma Spectrometry Analysis

The gamma spectrometry system used in analyzing the radionuclide contents of the quarry products consists of a 3 x 3-inch NaI (TI) detector, a product of Princeton Gamma Tech, USA. The detector is housed in a cylindrical lead shield to attenuate the influx of background radiation. The detector was coupled to a Gamma Spectacular (model GS-2000 Pro) multichannel analyzer and further linked to a computer for display. Data acquisition and analysis of gamma-ray spectra were achieved using Theremino software. The spectrometry system was calibrated for its qualitative and quantitative relationship between the gamma peak position and the corresponding gamma-ray energy using the RSS8 gamma source set traceable to Spectrum Techniques LLC, USA and a reference mixed gamma source consisting of known radionuclide activities.

Counting Procedure

The sealed samples after attaining a state of secular equilibrium were each placed on the detector one after the other for analysis and were counted for 18000 s. An empty container was also counted for the same period of time so as to determine the background gamma-ray distribution count. The activity concentration A (Bq/kg) of each identified radionuclide in the sample was estimated using Equation 1:

$$A = \frac{C_{net}}{P_{\gamma} \times \epsilon \times m \times t} \quad (1)$$

where C_{net} is the net peak count (count/seconds) for each radionuclide present in the sample after subtracting the background count from the gross count, P_{γ} is the absolute gamma ray emission probability of the identified radionuclide, ϵ is the obtained full energy peak efficiency for each identified radionuclide, m is the mass (kg) of the sample and t is the counting time (s).

In addition, the Minimum Detectable Activity (MDA) for each radionuclide was also calculated using Equation 2:

$$MDA = \frac{2.71 + 4.66 (\sigma)}{P_{\gamma} \times \epsilon \times m \times t} \quad (2)$$

where σ is the standard deviation of the background collected during time t over the energy range of interest.

Statistical Analysis

The mean activity concentrations of each radionuclide for each category of sample size were subjected to One-Way Analysis of Variance (ANOVA-1) and Scheffe Pair-Wise Comparisons tests to investigate any significant differences in their mean values.

RESULTS AND DISCUSSION

The measured activity concentrations of the natural radionuclides ^{40}K , ^{238}U , and ^{232}Th in the three different sizes (stone-dust, $\frac{3}{4}$ -down, and $\frac{3}{4}$ -up) of the quarry products assayed are presented in Tables 1a - c. The results of activity concentrations of stone-dust (Table 1a) samples size showed specific activities in the range 440.48 ± 8.47 to 301.08 ± 21.94 Bq/kg for ^{40}K , 23.89 ± 1.17 to 11.16 ± 0.80 Bq/kg for ^{238}U and 10.96 ± 0.90 to 6.76 ± 0.71 Bq/kg for ^{232}Th , respectively. The $\frac{3}{4}$ -down sample size (Table 1b) has activity concentrations ranging from 304.05 ± 2.41 to 196.89 ± 1.18 Bq/kg for ^{40}K , 9.95 ± 0.46 to 4.65 ± 0.47 Bq/kg for ^{238}U , and $7.74 \pm$

0.75 to 2.92±0.26 Bq/kg for 232Th. Similarly, the concentration of ¾ -up samples size (Table 1c) ranged between 18.15±4.32 to 113.55± 1.48 Bq/kg for 40K, 5.27± 1.05 to 2.78±0.16 Bq/kg for 238U,

and 4.74± 0.59 to 0.77±0.14 Bq/kg for 232Th. The corresponding average radionuclide concentrations of 40K, 238U and 232Th for stone-dust samples are 374.26±12.74 Bq/kg, 15.7±1.19 Bq/kg, and

Table 1a: Radionuclides and their activity concentrations (Bq/kg) in stone dust samples

SAMPLE ID	K-40	U-238	Th-232
OY 1	440.15 ± 8.49	18.38 ± 1.13	8.93 ± 0.81
OY 2	438.94 ± 8.55	18.21 ± 1.03	8.88 ± 0.81
OY 3	440.48 ± 8.47	18.30 ± 1.03	10.04 ± 0.86
OY 4	398.58 ± 10.88	13.75 ± 1.89	10.19 ± 0.87
OY 5	374.31 ± 12.73	11.89 ± 1.83	8.35 ± 0.78
OY 6	356.77 ± 14.35	16.21 ± 1.97	8.47 ± 0.79
OY 7	394.71 ± 11.15	16.28 ± 1.97	8.50 ± 0.79
OY 8	414.23 ± 9.88	18.27 ± 1.03	8.80 ± 0.80
OY 9	425.26 ± 9.25	17.46 ± 1.00	8.57 ± 0.79
OY 10	329.25 ± 17.54	23.89 ± 1.17	10.96 ± 0.90
OY 11	324.59 ± 18.18	11.59 ± 0.82	10.33 ± 0.87
OS 12	301.08 ± 21.94	11.16 ± 0.80	10.87 ± 0.89
OS 13	351.59 ± 14.89	15.26 ± 0.94	8.77 ± 0.80
OS 14	373.69 ± 12.78	15.50 ± 0.95	8.31 ± 0.78
OS 15	370.75 ± 13.04	15.93 ± 0.96	8.03 ± 0.77
OS 16	317.84 ± 11.16	13.43 ± 0.88	6.76 ± 0.71
OS 17	310.12 ± 13.28	11.47 ± 0.81	6.94 ± 0.71
Range	440.48 - 301.08	23.89 - 11.16	10.96 - 6.76
Mean	374.26 ± 12.74	15.70 ± 1.19	8.92 ± 0.81
Variance	2123.31 ± 13.39	10.40 ± 0.17	1.39 ± 0.00

Table 1b: Radionuclides and their activity concentrations (Bq/kg) in ¾ –down samples

SAMPLE ID	K-40	U-238	Th-232
OY 18	288.72 ± 3.36	7.66 ± 0.50	6.85 ± 0.71
OY 19	286.30 ± 4.88	7.59 ± 0.46	6.72 ± 0.70
OY 20	239.13 ± 3.03	7.63 ± 0.56	7.74 ± 0.75
OY 21	235.56 ± 4.52	5.73 ± 0.59	6.79 ± 0.71
OY 22	232.87 ± 4.70	4.95 ± 0.56	7.44 ± 0.74
OY 23	264.31 ± 3.38	6.76 ± 0.57	7.10 ± 0.72
OY 24	229.17 ± 4.41	6.78 ± 0.44	5.77 ± 0.65
OY 25	230.16 ± 4.94	7.61 ± 0.45	5.88 ± 0.66
OY 26	230.71 ± 4.68	7.28 ± 0.41	5.92 ± 0.66
OY 27	259.06 ± 3.95	9.95 ± 0.46	6.44 ± 0.69
OY 28	292.14 ± 2.66	4.83 ± 0.45	6.65 ± 0.70
OY 29	304.05 ± 2.41	4.65 ± 0.47	7.24 ± 0.73
OY 30	290.03 ± 2.09	6.36 ± 0.50	6.78 ± 0.71
OY 31	248.69 ± 4.97	6.46 ± 0.31	4.47 ± 0.36
OY 32	230.68 ± 4.16	6.64 ± 0.34	3.96 ± 0.29
OY 33	196.89 ± 1.18	5.59 ± 0.61	5.77 ± 1.30
OY 34	199.03 ± 4.60	4.78 ± 0.78	3.70 ± 0.34
OY 35	208.43 ± 4.12	8.00 ± 0.26	2.92 ± 0.26
OS 36	275.38 ± 4.45	5.98 ± 0.98	2.92 ± 1.15
OS 37	260.31 ± 5.64	5.14 ± 1.20	3.88 ± 0.37
OS 38	264.71 ± 6.22	8.41 ± 1.11	4.24 ± 0.26
Range	304.05 - 196.89	9.95 - 4.65	7.74 - 2.92
Mean	250.78 ± 4.02	6.61 ± 0.57	5.68 ± 0.64
Variance	938.04 ± 1.42	8.87 ± 0.06	10.62 ± 0.07

Table 1c: Radionuclides and their activity concentrations (Bq/kg) in 3/4 –up sample

SAMPLE ID	K-40	U-238	Th-232
OY 39	153.62 ± 4.84	3.42 ± 0.38	4.74 ± 0.59
OY 40	154.58 ± 3.66	3.70 ± 0.39	4.23 ± 0.56
OY 41	158.03 ± 4.92	3.58 ± 0.39	4.68 ± 0.59
OS 42	113.55 ± 1.48	4.21 ± 1.95	3.81 ± 0.87
OS 43	118.66 ± 1.60	3.62 ± 0.17	3.84 ± 0.88
OS 44	188.15 ± 4.32	4.98 ± 0.72	2.03 ± 0.17
OY 45	184.96 ± 4.15	4.92 ± 1.14	0.77 ± 0.14
OY 46	129.94 ± 3.26	4.92 ± 0.72	1.32 ± 0.16
OS 47	152.45 ± 3.47	3.63 ± 0.24	1.94 ± 0.21
OS 48	154.57 ± 0.62	3.92 ± 1.13	1.99 ± 0.42
OS 49	143.29 ± 0.75	2.78 ± 0.16	2.58 ± 0.23
OY 50	141.97 ± 0.76	5.27 ± 1.05	1.78 ± 0.25
OY 51	142.57 ± 0.76	3.52 ± 0.17	2.72 ± 0.23
OY 52	121.51 ± 1.13	3.38 ± 0.51	2.52 ± 0.14
Range	188.15 - 113.55	5.27 - 2.78	4.74 - 0.77
Mean	146.99 ± 2.55	3.99 ± 0.65	2.78 ± 0.39
Variance	454.70 ± 3.06	11.739 ± 0.25	1.494 ± 0.07

8.92±0.81 Bq/kg, respectively, while for ¾-down samples are 250.78± 4.02Bq/kg, 6.61±0.57 Bq/kg, and 5.68±0.064 Bq/kg, respectively. The average activity concentrations of 40K, 238U, and 232Th for ¾-Up samples are 146.99±2.55 Bq/kg, 3.99±0.65 Bq/kg, and 2.78±0.39 Bq/kg, respectively. Generally, high values of 40K were observed in all the samples. The observed high concentration of 40K in all the sample types may be a result of the geological formation underlying the studied areas. As illustrated in Figure 2, activity concentrations of

the radionuclides in the stone-dust samples were higher than those in the ¾ -down and ¾ - up samples. The order is Stone dust > ¾ - down > ¾ - up. This result is in agreement with earlier studies by Gbadebo (2011), Akinloye *et al.* (2019), that quarrying or quarry processes, which involve blasting, crushing, and processing of rocks into different aggregates (sizes), increase the activity concentrations of radionuclides in the production environment (Gbadebo, 2011; Akinloye *et al.*, 2019).

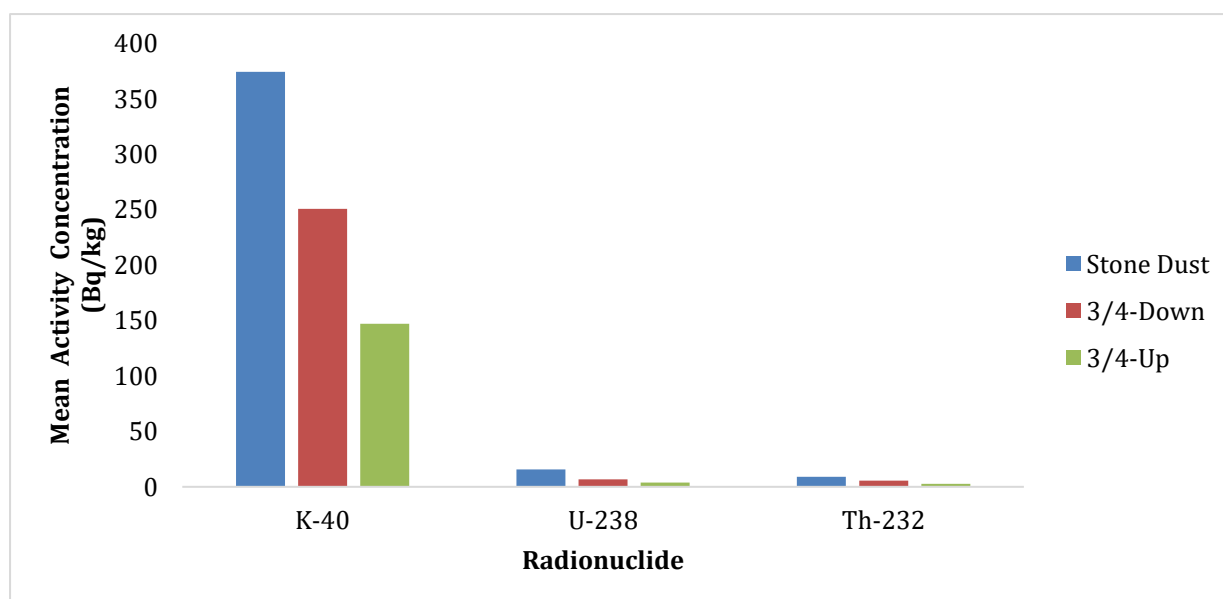


Figure 2: Mean activity concentrations of each radionuclide according to sample types

However, this present study disagrees with Lawal (2019), who claimed that the higher the granite size, the higher the activity concentrations of the radionuclides. The results from this study showed clearly that the smaller the size, the higher the activity concentrations of the natural radionuclides in the samples, as revealed in Figure 2. Table 2 shows the results of one-way Analysis of Variance conducted on the mean activity concentrations of each radionuclide in the different sizes at a 0.05 level of significance. Table 3 also showed the results of the Scheffe Pair-Wise Comparisons tests

conducted. The results actually confirmed that stone-dust samples have the highest mean activity concentrations of each radionuclide, followed by 3/4 - down samples, while the 3/4 - up samples have the least mean values of activity concentrations of radionuclides, as earlier presented in Tables 1a - c. Hence, there is a significant difference between the activity concentration of the quarry products ($F_{cal} > F_{val}$) for each radionuclide. Thus, quarrying processes increase the activity concentrations of natural radionuclides.

Table 2: Results of one-way analysis of variance

Radionuclide	Ho	Fcal	Fval (0.05,2,49)
⁴⁰ K	$\mu_1=\mu_2=\mu_3$	167.94	3.23
²³⁸ U	$\mu_1=\mu_2=\mu_3$	60.881	3.23
²³² Th	$\mu_1=\mu_2=\mu_3$	28.166	3.23

where: μ_1, μ_2, μ_3 are the mean activity concentrations (Bq/kg) of each nuclide in stone-dust, 3/4-down, and 3/4-up sample types, respectively. (Ho is the null hypothesis, Fcal is the calculated F-statistic, Fval is the critical F-statistic at $\alpha = 0.05$. Ho is rejected as long as $F_{val} \leq F_{cal}$, which means there are significant differences between the means.

Table 3: Results of the Scheffe Pair-Wise Comparisons of the Mean Activity Concentrations (Bq/kg) of each Radionuclide According to Size

Radionuclide	Sample Pair	Fcal	Fval	Conclusion
⁴⁰ K	μ_1 and μ_2	331.39	6.46	Difference exist
	μ_1 and μ_3	119.71	6.46	Difference exist
	μ_2 and μ_3	75.61	6.46	Difference exist
²³⁸ U	μ_1 and μ_2	103.93	6.46	Difference exist
	μ_1 and μ_3	76.64	6.46	Difference exist
	μ_2 and μ_3	5.69	6.46	No Difference
²³² Th	μ_1 and μ_2	55.813	6.46	Difference exist
	μ_1 and μ_3	19.019	6.46	Difference exist
	μ_2 and μ_3	13.62	6.46	Difference exist

where: $\mu_1, \mu_2,$ and μ_3 are the mean activity concentrations of each radionuclide for stone-dust, 3/4 -down, and 3/4-up sample types, respectively (Fcal is the calculated value for Scheffe's test, and Fval is the critical value for Scheffe's test)

average values. The results revealed that the mean activity concentrations of each radionuclide are well below the world average values, which implies that the materials pose no radiological risk to the environment. Table 4 shows the comparisons of the

mean activity concentration of each radionuclide with the world

Table 4: Comparison of the Mean Activity Concentrations of the Radionuclides (Bq/kg) in the Different Sizes with World Average Values.

Sample Type	K-40	U-238	Th-232
Stone-Dust	374.26 ± 12.74	15.70 ± 1.19	8.92 ± 0.81
3/4 – Down	250.78 ± 4.02	6.61 ± 0.57	5.68 ± 0.64
3/4 – Up	146.99 ± 2.55	3.99 ± 0.65	2.78 ± 0.39
World Average Values (UNSCEAR, 2010)	400	35	35

CONCLUSION

This study has presented the results of the measurement of gamma radioactivity levels in quarry products collected from quarries in Osun and Oyo States, Southwestern Nigeria, using the gamma spectrometry method. The study confirmed the presence of ⁴⁰K, ²³⁸U, and ²³²Th in the products and has shown that ⁴⁰K is the major naturally occurring source of radiation in the products. All the natural radionuclide activity concentration values were lower than the world average values. The highest concentrations of the three radionuclides measured were obtained in the stone-dust samples size followed by ¾ - down samples size while the least concentrations were in the ¾ - Up samples. Thus, it can be concluded that the activity concentrations of the radionuclides decrease as the sample size increases. In other words, the higher the size, the smaller the activity concentrations. In corollary, the smaller the size, the higher the activity concentrations of the radionuclides. Therefore, quarry processes actually increase the activity concentrations in the products.

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