

Radionuclide Concentration and Lifetime Cancer Risk Due to Gamma Radioactivity from Quarry Stone Aggregates in Jos and Its Environs, North Central Nigeria

Abiye Olatunji Solomon[1*] Nestor Monday Chagok[2] Alexander Sunday Chup[1]
Ifeoma Aquila Ekeleme[1] Joseph Azi Bulus[1] Ambura Fube Ajol[1]
Lukman Olaleke Adigun[3]
1.Department of Geology, University of Jos, Nigeria
2.Department of Physics, University of Jos, Nigeria
3.Bore & Core Nigeria Ltd, Jos, Nigeria

Abstract
Naturally occurring radionuclides ^{238}U, ^{232}Th and ^{40}K present in crushed stone aggregates from seven (7) quarries in Jos and its environs were measured using Gamma Ray Spectrometer. This is mainly for the purpose of evaluating the radiation health hazard indices and excess lifetime cancer risk (ELCR) that may arise due to the use of such crushed stones for building construction. Mean concentration of ^{238}U, ^{232}Th and ^{40}K from the quarries were found to be 4.0-15.1 ppm, 25.7-51.7 ppm, and 3.9%-4.4%, while their activity concentration range from 49.5-186.5 Bqkg^{-1}for ^{238}U, 104.3-209.9 Bqkg^{-1} for ^{232}Th, and 1158.1-1471.1 Bqkg^{-1}for ^{40}K respectively. Absorbed dose in air (149.85–264.35 nGy h^{-1}), annual effective dose equivalents for internal and external (0.69-1.22 mSv y^{-1}and 0.23-0.41 mSv y^{-1}), hazard indices (1.00-2.05 indoor, and 0.87-1.53 outdoor), and radium equivalent activity (317.51-573.11 Bqkg^{-1}) were calculated. These values are higher than the world's averages. Total excess lifetime cancer risk (ELCR) from the quarries was found to be 3.21×10^{-3} - 5.68×10^{-3} with an average of $4.45\text{x}10^{-3}$. This is 15.34 times higher than the world average of $0.29\text{x}10^{-3}$ below which negligible risk of developing cancer has been stated. Crushed stones from quarries in Jos and its environs therefore may have contributed significantly to cancer risks in this area.
Keywords: Quarry; Crushed Stones; Uranium; Thorium; Potassium; Activity Concentration; Hazard Indices; Excess Lifetime Cancer Risk; Jos Environs..

1. Introduction

Quarry stones are construction aggregate produced by breaking removed rocks into desired sizes using a crusher or hammer. Due to their angular surface which makes it easy to roll and key into cement mix, crushed stone are used mainly for construction projects. In a study by Ilangovana *et al.* (2008), the strength of quarry rock dust concrete was reported to be 10-12% more than that of similar mix of conventional concrete. Hameed & Sekar (2009) also reported that the flexural strength increases in concrete with crushed stone dust compared with the concrete with natural sand. Quarry stone aggregates are used extensively for building construction in Jos and in many cities in Nigeria. The rocks being quarried here are mainly biotite granite and biotite hornblende granite, all belonging to the Younger Granite Complex. They are known to contain significant amount of ^{238}U, ^{232}Th and ^{40}K and therefore constitute a major source of exposure to radiation to the inhabitants of the area. This study assesses the radionuclide concentrations and lifetime cancer risk due to gamma radioactivity emanating from crushed stone aggregates in Jos and its environs in relation to safety.

2. Materials and Methods

2.1 The study area

The study area (Jos and environ) is located in north central part of Nigeria. The city is the administrative capital of Plateau State. Quarrying is on the rise here because of the need to meet the increasing demand for shelter as population is increasing. According to Kinnaird *et al.* (1981), Jos and its environ is underlain essentially by rocks of the Younger Granites, as well as the Nigerian Basement Complex (Figure 1). The Precambrian Basement Complex rocks in the study area are mainly migmatite, granite gneiss and porphyritic Older Granites. Whereas the Younger Granites comprise of non-orogenic granites, ryholites, basalts, crystal-rich ignimbrites and porphyries of the Mesozoic Era (MacLeod *et al.* 1971) which occur within a distinct metallogenic province, the Jos-Bukuru, Buji, Rukuba and Shere Complexes constitute the bulk of Younger Granites suite in and around Jos, and consist essentially of biotite granite, riebeckite biotite granite, hornblende fayalite granite, hornblende biotite granite, rhyolite, syenite, gabbro, dolerites and basalts, with significant amount of natural concentration of uranium, thorium and potassium (Solomon, 2005). The alluvial deposits of cassiterite (tin oxide, SnO_2) and columbite (oxide of tantalum-niobium, iron and manganese, $(Fe,Mn)(Ta,Nb)_2O_6$), as well as radioactive mineral such as thorite ($ThSiO_4$), zircon ($ZiSiO_4$) and monazite ($(Ce,La,Yt)PO_4$) on the Jos Plateau are believed to have been derived from these Complexes.

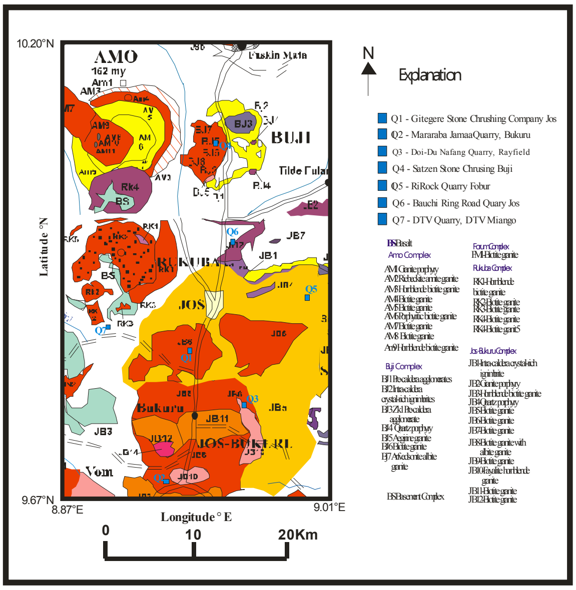

Figure 1. Geological map of Jos and environs and location of the quarries.

2.2 Determination of concentration of naturally radioactive elements

Measurement of gamma radiation emanating from the naturally radioactive constituents of the crushed rocks was carried out using RS-230 Gamma Spectrometer (Figure 2). The spectrometer is a handheld auto-stabilized 1024 channel piece of equipment which uses a large (103 cm^3) BGO (Bismuth Germanate Oxide) detector for improved level of system sensitivity and accuracy. The use of a BGO gives very significant increase in performance over the normal NaI detector. For a better accuracy, a preset time of 120 seconds was used for the measurement per point while the assay mode provided the concentrations of Potassium (K %), equivalent Thorium (eTh) in ppm, and equivalent Uranium (eU) in ppm for each point of measurement. The energy response of the equipment is 30–3000 kev. Between fifty (50) and one hundred and twenty (120) measurements were recorded from different sizes of crushed stone aggregates for each of the quarry, and the average calculated. The values were then converted into activity concentrations in $Bqkg^{-1}$ with which the lifetime cancer risk values were determined.

Figure 2. RS-230 BGO Handheld Gamma-Ray Spectrometer.

3. Results and Discussion

Radionuclide concentration, activity concentration, absorbed dose rate in air, annual effective dose equivalent (AEDE), radium equivalent activity (Ra_{eq}), hazard indices, and excess lifetime cancer risk (ELCR) were determined in this study.

3.1 Radioelement concentration and Specific Activity.

Concentration of radionuclides (Table 1) varies from one quarry to another owing to differences in the mineral composition of the rocks. Average concentration of ^{238}U, ^{232}Th and ^{40}K in the crushed stones and their activity concentrations are 15.1 ppm, 47.9 ppm, 4.4%, and 186.5, 194.5, 1377.2 Bqkg^{-1} respectively for Getegere Stone Crushing Company Zaramaganda, 14.1 ppm, 51.7 ppm, 4.1%, and 174.1, 209.9, 1283.3 Bqkg^{-1} for Mararaba Jamma Quarry Bukuru, 12.2 ppm, 39.4 ppm, 4.7%, and 150.7, 160.0, 1471.1 Bqkg^{-1} for Doi-Du Nafang Quarry Rayfield, 7.0 ppm, 25.7 ppm, 4.1%, and 86.9, 104.3, 1283.3 Bqkg^{-1} for Satzen Company Quarry Buji. They also include 9.0 ppm, 31.7 ppm, 4.3%, and 111.5, 128.7 1333.4 Bqkg^{-1} for RicRock Quarry Fobur, 5.5 ppm, 26.8 ppm, 3.7%, and 67.9, 108.8, 1158.1 Bqkg^{-1} for Bauchi Ring Road Quarry, as well as 4.0 ppm, 30.7 ppm, 3.9%, and 67.9, 108.8, 1158.1 Bqkg^{-1} for DTV Quarry, DTV-Jebu. From the analytical results, Gintegere Stone Crushing Company Zaramaganda Jos, and Mararaba Jamma Quarry Bukuru recorded the highest values of excess lifetime cancer risk (ELCR) compared to the other quarries.

Table 1. Radiation parameters for quarries in Jos and environs.

S/No	QUARRY NAME	COORDINATE		CONCENTRATION OF RADIONUCLIDES			DATA SUMMARRY
				^{238}U	^{232}Th	^{40}K	
1	Getegere Stone Crushing Company Bukuru, Jos	LAT (^{0}N) 9.85	LON (^{0}E) 8.86	15.1	47.9	4.4	Average
				20.9	56.7	5.7	Maximum
				10.8	36.9	3.3	Minimum
				186.5	194.5	1377.2	Activity Concentration (Bqkg^{-1})
2	Mararaba Jamaa Quarry Bukuru, Jos	LAT (^{0}N) 9.72	LON (^{0}E) 8.86	14.1	51.7	4.1	Average
				31.9	87.7	5.3	Maximum
				6.5	1.6	2.1	Minimum
				174.1	209.9	1283.3	Activity Concentration (Bqkg^{-1})
3	Doi-Du Nafang Quarry Rayfield, Jos	LAT (^{0}N) 9.80	LON (^{0}E) 8.91	12.2	39.4	4.7	Average
				15.8	49.8	6.1	Maximum
				9.4	29.5	3.5	Minimum
				150.7	160.0	1471.1	Activity Concentration (Bqkg^{-1})
4	Satzen Company Quarry Buji Near Jos	LAT (^{0}N) 10.03	LON (^{0}E) 8.51	7.0	25.7	4.1	Average
				11.3	35.3	5.5	Maximum
				3.6	19.1	2.7	Minimum
				86.9	104.3	1283.3	Activity Concentration (Bqkg^{-1})
5	RicRock Quarry Fobur Near Jos	LAT (^{0}N) 9.85	LON (^{0}E) 9.01	9.0	31.7	4.3	Average
				14.1	44	5.1	Maximum
				5.2	1.9	1.2	Minimum
				111.5	128.7	1333.4	Activity Concentration (Bqkg^{-1})
6	Bauchi Ring Road Quarry, Jos	LAT (^{0}N) 9.95	LON (^{0}E) 8.91	5.5	26.8	3.9	Average
				8.6	38.5	6.1	Maximum
				1.7	19.8	2.7	Minimum
				67.9	108.8	1158.1	Activity Concentration (Bqkg^{-1})
7	DTV Quarry, DTV-Jebu Near Jos	LAT (^{0}N) 9.53	LON (^{0}E) 8.46	4.0	30.7	3.9	Average
				6.4	48	5.9	Maximum
				1.4	17.1	0.8	Minimum
				49.4	124.8	1205.1	Activity Concentration (Bqkg^{-1})

These quarries feed from rocks of the Jos-Bukuru Complex known to contain significantly high amount of natural concentration of ^{238}U, ^{232}Th and ^{40}K, and as such, high values of radiation hazard indices (Solomon *et al.* 2018). Generally, granitic rocks are known to be enriched in Thorium (Th) and Uranium (U), with an average of 15 µgg^{-1} of Th and 5 µgg^{-1} of U (Faure 1986; Mènager *et al*. 1993).

3.2 Absorbed Dose Rate in Air (D)

The absorbed dose rate in the air due to gamma rays 1 metre above the ground from the quarries was calculated according to UNSCEAR (2000), using Equation 1.

$$D\ (\text{nGy h}^{-1}) = 0.462A_U + 0.621A_{Th} + 0.0417A_K \qquad (1)$$

Where 0.462, 0.621 and 0.0417 are the conversion factors for ^{238}U, ^{232}Th and ^{40}K respectively. The assumption here is that other natural radionuclides contributes negligible amount to the absorbed dose rate. The absorbed dose rate in air from the quarries investigated (Table 2) varies from 149.85–264.35 nGy h^{-1}. The Gintegere Stone Crushing Company Bukuru, Jos (Q1), and the Mararaba Jamma Quarry Bukuru, Jos (Q2) recorded the highest values. The two quarries source rock fragments for crushing from the Jos-Bukuru Complex.

3.3 Annual Effective Dose Equivalent (AEDE).

Annual effective dose equivalents (indoor and outdoor) were calculated for the seven (7) quarries under study using Equation 2 and 3.

$AEDE_{Indoor}\ (mSvy^{-1}) = D\ (nGyh^{-1})\ x\ 8760\ hy^{-1}\ x\ 0.75\ x\ 0.7SvGy^{-1} x\ 10^{-6}$ (2)

$AEDE_{Outdoor}\ (mSvy^{-1}) = D\ (nGyh^{-1})\ x\ 8760\ hy^{-1}\ x\ 0.75\ x\ 0.7SvGy^{-1} x\ 10^{-6}$ (3)

Where D is the absorbed dose in air, 8760 (*24x365*) is the number of hours in a year, 0.75 is the indoor occupancy factor, which represents the fraction of time spent indoor, and $0.7SvGy^{-1}$ is the conversion coefficient from absorbed dose in air to effective dose received by adults. The values of AEDE ranges from 0.69-1.22 mSv $y^{-1}$for internal and 0.23-0.41 mSv $y^{-1}$for external (Table 2).

3.4 Hazard Indices (HI)

Hazard Indices (internal and external) are reflections of exposure to radiation. The hazard indices were calculated using Equation 5 and 6 according to Shoeib & Thabayneh (2014).

$H_{In} = A_U/185 + A_{Th}/259 + A_K/4810$ (*4*)

$H_{Ex} = A_U/370 + A_{Th}/259 + A_K/4810$ (*5*)

Where A_U, A_{Th} and A_K are the specific activity concentrations of ^{238}U, ^{232}Th and ^{40}K respectively. The mean internal and external hazard index should be less than 1 to provide acceptable level of personal safety. The values of radiation hazard indices in the studied quarries are 1.00-2.05 for indoor, and 0.86-1.55 for outdoor. The mean internal and external hazard index are less than 1 in some of the quarries but higher in others.

Table 2. Excess Lifetime Cancer Risk and other hazard parameters for the Quarries

S/No	QUARRY ID	D (nGy h^{-1})	AEDE Int. (mSv y^{-1})	AEDE Ext. (mSv y^{-1})	RaEq. (Bqkg^{-1})	HI indoor	HI outdoor	ELCR (In)	ELCR Out	ELCR (Total)
1	Q1	264.35	1.22	0.41	570.63	2.05	1.54	4.26	1.42	5.68
2	Q2	264.31	1.22	0.41	573.11	2.02	1.55	4.25	1.42	5.67
3	Q3	230.29	1.06	0.35	492.69	1.74	1.33	3.71	1.24	4.95
4	Q4	158.25	0.73	0.24	334.47	1.14	0.90	2.55	0.85	3.4
5	Q5	187.40	0.86	0.29	398.83	1.38	1.08	3.02	1.01	4.03
6	Q6	149.85	0.69	0.23	317.51	1.04	0.86	2.41	0.80	3.21
7	Q7	151.13	0.70	0.23	321.63	1.00	0.87	2.43	0.81	3.24

Quarry Index.

Q1- Getegere Stone Crushing Company Bukuru, Jos
Q2- Mararaba Jamma Quarry Bukuru, Jos
Q3- Doi-Du Nafang Quarry Rayfield, Jos
Q4- Satzen Company Quarry Buji Near Jos
Q- Ricrock Quarry Fobur Near Jos
Q6- Bauchi Ring Road Quarry, Jos

3.5 Radium Equivalent Activity (Ra_{eq})

Radium equivalent activity (Ra_{eq}) is a single index which sums up the gamma output from different mixture of ^{238}U, ^{232}Th and ^{40}K emanating from a rock. For safety, radium equivalent activity (Ra_{eq}) is set at 370 $Bqkg^{-1}$. This is equivalent to a maximum permissible dose of 1.5 mSv y^{-1} to human from exposures to natural radiation. Radium equivalent activity was calculated according to Beretka & Matthew (1985); UNSCEAR (2000) as follows:

$Ra_{eq}\ (Bqkg^{-1}) = A_{Ra} + 1.43A_{Th} + 0.077A_K$ (*6*)

Where A_{Ra}, A_{Th} and A_K are the specific activity concentrations of uranium (^{238}U), thorium (^{232}Th) and potassium (^{40}K) respectively. The value of Ra_{eq} ranges from 317.51-573.11 $Bqkg^{-1}$ (Table 2). With a tolerable Ra_{eq} value of 370 $Bqkg^{-1}$, radiation safety may become a concern if crushed stones from quarries with higher values as identified in this study are used in buildings construction.

3.6 Excess Lifetime Cancer Risk (ELCR)

Exposure to radiation on a long-term basis is believed to have some risks of causing cancer. It means, therefore that all human to a large extent are at a risk of getting cancer because we are all exposed to many sources of radiation within our living environment. According to the National Cancer Institute (2009), American men have a 44% lifetime risk of cancer, while women have a 38% lifetime risk. It implies therefore that there is chance of about 33% (or 0.33) that a person will get some type of cancer at some stage of life. Excess lifetime cancer risk" (ELCR) refers to the additional risk of getting cancer that a person might have if that person is exposed to cancer-causing materials over a long period of time. The allowable value is additional or excess of 1 in 100,000 chances (1×10^{-5}). In line with the position of Ramasamy *et al.* (2009), Emelue *et al.* (2014), the excess lifetime cancer risk (ELCR) was calculated based on the calculated values of annual effective dose equivalents using the following equation:-

$ELCR_{(indoor)} = AEDE(indoor) \times DL \times RF$ (*7)*
$ELCR_{(outdoor)} = AEDE(outdoor) \times DL \times RF$ (*8)*

Where AEDE is the annual effective dose equivalent, DL is the average duration of life (70 years), and RF is the risk factor or fatal cancer risk per Sievert. The International Committee on Radiation Protection ICRP (1991) peg RF at 0.05 for the public.

The excess lifetime cancer risk (ELCR) of the study area ranges from 2.43 x10^{-3}- 4.26 x10^{-3}, (indoor) while for outdoor; it ranges from 0.80 x10^{-3}-1.42 x10^{-3} (Table 2). These values are higher than the reference value of 0.29×10^{-3} below which negligible risk of developing cancer has been fixed (Aziz *et al.* 2014). The total (ELCR) ranges from 3.21-5.68×10^{-3} with an average value of 4.45×10^{-3}. This is 15.34 times higher than the reference value of 0.29x10^{-3}. The quarries with the highest (ELCR) are Getegere Stone Crushing Company Bukuru, Jos and Mararaba Jamaa Quarry Bukuru, Jos.

Similar studies carried out by Solomon *et al.* (2018) on hollow aggregate filled stone-dust blocks and sandcrete blocks produced in Jos Plateau State, Nigeria, gave a range of 1.26 x10^{-3}-1.74 x10^{-3} for indoor, and a range of 0.43 x10^{-3}- 0.58 x$10^{-3}$for outdoor. For aggregate-filled stone-dust blocks, ELCR (indoor) varies from 2.46 x10^{-3}- 2.99 x10^{-3} while for the outdoor, it ranges from 0.82 x10^{-3}-1.00 x10^{-3}. The report concluded that the risk of developing cancer (ELCR) is much higher for dwellers of buildings constructed with aggregate filled stone-dust blocks compared to 'ordinary' sandcrete blocks. A study on the determination of (ELCR) in and around Warri Refining and Petrochemical Company in Niger Delta, Nigeria was carried out by Emelue *et al.* (2014). The result revealed that the risk of developing cancer is below the standard.

In a similar study carried out in high background radiation area (Kerala), India by Ramasamy *et al.* (2013), an average (ELCR*)* value of 1.7×10^{-3} was recorded, amounting to six times the world average (0.29×10^{-3}). According to Taskin *et al.* (2009), excess lifetime cancer risk (ELCR) in Kirklareli Turkey is 0.50×10^{-3}. Ramasamy *et al.* (2009) equally carried out the evaluation of (ELCR) in river sediments of Karnataka and Tamilnadu, India, and reported an average ELCR value of 0.20×10^{-3}. This is low, and less than the world average. ELCR in natural environment therefore is a function of the local geology.

4. Conclusion

The concentration and specific activity of ^{238}U, ^{232}Th and ^{40}K from crushed stones have been determined across major quarries in Jos, north central Nigeria using the RS-230 Gamma Spectrometer. Radiological parameters from the various quarries which include absorbed dose rate in air, annual effective dose equivalent, radium equivalent activity, internal hazard index and excess lifetime cancer risk were found to be generally higher within the Getegere Stone Crushing Company Bukuru, Jos and the Mararaba Jamma Quarry Bukuru, Jos. Both quarries source their aggregate product from biotite granite belonging to the Jos-Bukuru Complex. The average of total Excess Lifetime Cancer Risk from the quarries (4.45x10^{-3}) is 15.34 times higher than the world average of 0.29x10^{-3} below which negligible risk of developing cancer has been stated. Crushed stones from quarries in Jos and its environs therefore may have contributed significantly to cancer risks in this area.

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