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EVALUATION OF CARCINOGENIC RISK OF GRANITE SAMPLES FROM SELECTED QUARRIES IN ONDO AND EKITI STATES, NIGERIA

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ABSTRACT: A total of twenty granite rock samples were collected from twenty quarry locations in Ondo and Ekiti States. Activity concentration measurement was done using Hyper Pure Germanium detector. The mean activity concentration is 21.01±3.15 Bq kg^{-1} for samples in ^{238}U, 51.19±6.73 Bq kg^{-1} in ^{232}Th, and 489.11±73.37 Bq kg^{-1} in ^{40}K. The mean absorbed dose rate in air is 57.21 nGy h^{-1}, the mean internal level index is 0.11 while the maximum permissible level for the four is unity and the mean excess lifetime cancer risk is 0.74 x 10^{-3}. The probability of sample from selected quarries causing carcinogenic risk on the buildings they are constructed with is very low.
KEYWORD – Activity, Detector, Germanium, Hyperpure, Gonnadal

I. INTRODUCTION

The radioactivity in rocks contributes to the external gamma dose rate that humans receive from the environment. Rocks are of three types, namely igneous, sedimentary and metamorphic rocks. Higher ionizing radiation levels are associated with igneous rocks, such as granite, as well as metamorphic rocks, whereas lowest levels with sedimentary rocks [Hareyama et al 2000]. However, there are exemptions as some shale and phosphate rocks have relatively high content of radionuclides [Abbady et al 2005]. Ionizing radiation is a form of radiation with sufficient energy to remove electrons from their atomic or molecular orbital shells in the tissues they penetrate. These ionizations, received in sufficient quantities over a period of time, can result in tissue damage and disruption of cellular function at the molecular level, most importantly their effect on deoxyribonucleic acids (DNA). In the case of carcinogens generally, whether chemical or radiological, safety standards are based on a postulated zero threshold. Increasing the size of the dose increases the probability of inducing a cancer with that carcinogen. One of the major aggregates in building construction is granite, therefore it is important to measure the radioactivity in granites because it exhibits enhanced elemental concentration of Uranium (^{238}U), Thorium (^{232}Th) and Potassium (^{40}K) compared to the very low abundance of these elements observed in the mantle and crust of the earth. Hence they may pose carcinogenic risk
The Activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in granite samples collected from five different quarry industries in Ondo State, Nigeria. From the study, it can be concluded that they do not pose any significant radiological hazard when used for construction of dwellings [Ademola et al 2010]. Evaluation of the carcinogenic risk of granite used in building materials in Zahedan using hyper pure germanium (HPGe) detector. Some of the values for internal hazard index, external hazard index and annual effective dose were higher than permissible level of unity and world average [Ahmad et al 2019]. Works concerning the evaluation of carcinogenic risk and radiological hazard of granites of granites have been studied, radioactivity of natural Uranium (^{238}U), Thorium (^{232}Th) and Potassium (^{40}K) in granites are very high in some areas. Therefore it is necessary to evaluate the carcinogenic risk of granite samples from some selected quarries that are the major source of granite for building construction in Ondo and Ekiti States. In order to know if they constitute a significant source of external exposure to radiation for the dwellers of the building they are used to construct.

II. MATERIALS AND METHODS

The Study Area

Ondo and Ekiti states are both located in south western geopolitical zone of Nigeria. Ondo State lies between longitudes 4°30' and 6°00'E of the Greenwich Meridian and latitudes 5°45' and 8°15' N of the Equator [Ondo State 2010]. While Ekiti State lies between longitudes 4°45' and 5°50'E of the Greenwich Meridian, latitudes 7°15' and 8°10' N of the Equator [Bayowa et al 2014].The two states fall entirely in the tropics
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Geology of the Study Area

There are two distinct geological regions in old Ondo State that comprises of present Ondo and Ekiti states.. First, is the region of sedimentary rocks in the south, and secondly, the region of Precambrian Basement Complex rocks in the north [Daramola et al. 2009]. Some few kilometres north of Aaye occurs the basement complex sedimentary rocks boundary. The sedimentary rocks are mainly of the post Cretaceous sediments and the Cretaceous Abeokuta Formation [Daramola et al. 2009]. The basement complex is mainly of the medium grained gneisses. These are strongly foliated rocks frequently occurring as out crops [Daramola et al. 2009]. On the surface of these outcrops, severely contorted, alternating bands of dark and light coloured minerals can be seen. These bands of light coloured minerals are essentially feldspar and quartz, while the dark coloured bands contain abundant biotic mica [Daramola et al. 2009]. Some portions of the state, especially to the northeast, have coarse grained granites and gneisses, which are poor in dark ferromagnesian minerals [Daramola et al. 2009]. Ondo State is composed of lowlands and rugged hills with granitic outcrops in several places [Daramola et al. 2009]. In general, the land rises from the coastal part of llaje/Ese-Odo (less than fifteen meters above sea level) in the south, to the rugged hills of the north eastern portion in Akoko area [Daramola et al. 2009].. Some of the more prominent hills found at Idanre and Akoko rise above 250 meters above sea level [Daramola et al. 2009]. The geomorphologic units of the creek and riverine areas include sand ridges, lagoons, swamp flats, creeks and the anatomizing distributaries of the western Niger Delta [Daramola et al. 2009].

Sample Location

The Sample location name, quarry name and numbers collected per location are shown in table 1 while map of the main studied area is shown in fig. 1.

Table 1 Sample location name, quarry name and numbers collected per location.

No.	LOCATION	QUARRY
1	ISE AKOKO	N D C
2	ITAOGBOLU	SAMCHASE
3	ITAOGBOLU	DORTMUND
4	SHASHA - AKURE	JOHNSON
5	SHASHA - AKURE	RCC
6	SHASHA - AKURE	STONEWORKS
7	AYE - IJARE	ZEBO-FM
8	AKURE	OSAC
9	ELEGBEKA	SERVETEK
10	ELEGBEKA	JAPAUL
11	ELEGBEKA	SERENA
12	ELEGBEKA	BALLESTER
13	ELEGBEKA	NIGERCAT
14	IFON	PARTIFINS
15	IFON	ROADSTONE
16	SUPARE AKOKO	STONEWORLD
17	IKERE EKITI	INLAND
18	IKERE EKITI	MAC
19	ISINBODE EKITI	ISINBODE
20	IGEDE EKITI	BCCL

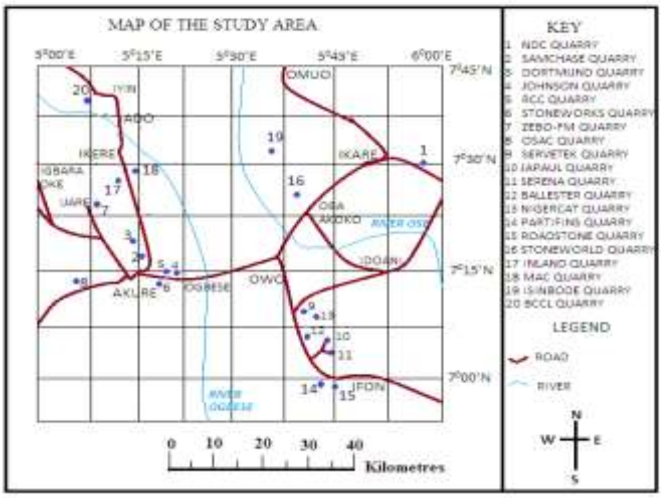

Figure 1. Locations of selected quarry sites in Ondo and Ekiti States

Sample Processing And Activity Determination

Twenty granite rock samples were collected from selected 16 quarries in Ondo State and 4 quarries in Ekiti State. Each of the twenty samples from the 20 selected quarry sites was crushed and milled to very fine particles. The prepared samples was packed in 1 litre capacity marinelli beaker that had been washed with hydrochloric acid, rinsed with distilled water and allowed to dry. The marinelli beaker lid was well

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tightened and sealed with a "cello tape". The enclosed sample was left for four weeks so that secular equilibrium could be reached. Analysis (counting) was performed on each mesh size of samples for activity concentration measurement using Hyper Pure Germanium Detector for 36000 seconds (10 hours) to acquire spectra data. 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 ^{226}Ra, 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%)

Radiological Hazard Indices Calculation
Absorbed Dose Rate in Air (D) [UNSCEAR 2000]

$$D\ (\mathrm{nGyh^{-1}}) = 0.462A_{Ra}+0.604A_{Th}+0.042A_K \quad (1)$$

where; A_{Ra}, A_{Th} and A_K are the radioactivity concentrations in Bq kg^{-1} of ^{238}U, ^{232}Th and ^{40}K respectively.

Internal (α-radioactivity) Level Index (I_α) [El-Galy et al, 2009]

$$I_\alpha = \frac{A_{Ra}}{200} \leq 1 \quad (2)$$

Excess Lifetime Cancer Risk (ELCR) [Taskin et al 2009]

$$ELCR = AED \times DL \times RF \quad (3)$$

where, AED is the Annual Equivalent Dose, DL is average Duration of Life (estimated to be 70years), and RF is the Risk Factor (Sv), i.e. fatal cancer risk per Sievert. For stochastic effects, ICRP uses RF as 0.05 for the public [Taskin et al 2009].

III. RESULT AND DISCUSSION

Activity Concentrations of Natural Radionuclides

Table 2 and fig. 2 present the three (^{238}U, ^{232}Th and ^{40}K) natural radionuclide isotopes present in the studied samples, the range of activity concentrations of ^{238}U, ^{232}Th and ^{40}K were found to be 7.44±1.12 to 46.74±7.01 Bq kg^{-1}, 17.56±2.63 to 80.69±12.10 Bq kg^{-1} and 309.84±46.48 to 777.19±116.58 Bq kg^{-1} respectively while mean values of ^{238}U, ^{232}Th and ^{40}K are 21.01±3.15 Bq kg^{-1}, 44.88±6.73 Bq kg^{-1} and 489.11±73.37 Bq kg^{-1} respectively. The mean activity concentrations in the studied granite samples were lower than permissible values of 33 Bq kg^{-1} for ^{238}U, approximately the same as 45 Bq kg^{-1} permissible value for ^{232}Th, but higher than 412 Bq kg^{-1} permissible value for ^{40}K. [UNSCEAR 2010]

Table 2 Activity concentration of natural radionuclides in the collected sample

SAMPLE	ACTIVITY CONCENTRATIONS IN Bq kg^{-1}		
	^{238}U	^{232}Th	^{40}K
NDC QUARRY	25.04±3.76	27.87±4.18	652.17±97.83
SAMCHASE QUARRY	11.60±1.74	71.97±10.80	493.74±74.06
DORTMUND QUARRY	9.12±1.37	61.18±9.18	470.13±70.52
JOHNSON QUARRY	36.04±5.41	76.42±11.46	777.19±116.58
RCC QUARRY	23.51±3.53	33.46±5.02	336.67±50.50
STONEWORKS QUARRY	10.37±1.56	21.61±3.24	434.51±65.18
ZEBO-FM QUARRY	8.65±1.30	80.69±12.10	513.54±77.03
OSAC QUARRY	32.06±4.81	44.30±6.65	590.57±88.59
SERVETEK QUARRY	14.75±2.21	59.52±8.93	610.97±91.65
JAPAUL QUARRY	29.08±4.36	38.42±5.76	776.62±116.49
SERENA QUARRY	11.97±1.80	47.62±7.14	467.60±70.14
BALLESTER QUARRY	39.47±5.92	32.96±4.94	380.39±57.06
NIGERCAT QUARRY	7.44±1.12	69.96±10.49	461.95±69.29
PARTIFIN QUARRY	46.74±7.01	52.43±7.86	385.88±57.88
ROADSTONE QUARRY	19.96±2.99	31.21±4.68	341.02±51.15
STONEWORLD QUARRY	30.65±4.60	36.72±5.51	420.98±63.15
INLAND QUARRY	8.96±1.34	35.31±5.30	505.49±75.82
MAC QUARRY	17.85±2.68	17.56±2.63	309.84±46.48
ISINBODE QUARRY	9.95±1.49	24.18±3.63	511.43±76.72
BCCL QUARRY	26.90±4.03	34.22±5.13	341.49±51.22
MEAN	**21.01±3.15**	**51.19±6.73**	**489.11±73.37**
MAX	**46.74±7.01**	**80.69±12.10**	**777.19±116.58**
MIN	**7.44±1.12**	**17.56±2.63**	**309.84±46.48**

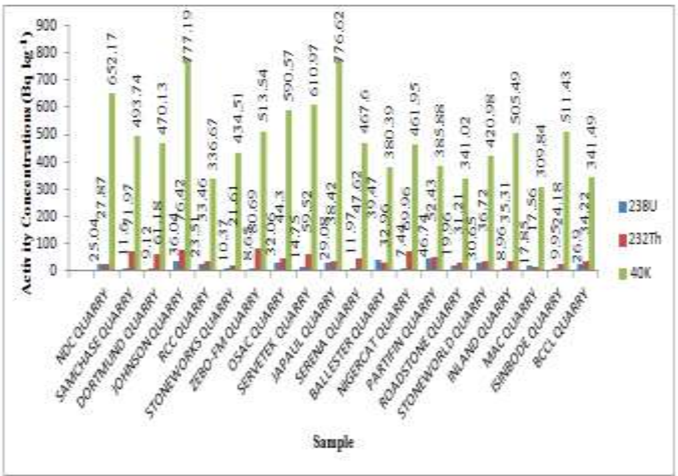

Figure 2 Activity Concentrations of Natural Radionuclides in Samples

Absorbed Dose Rate in Air (D)

The calculated absorbed dose (D) shown in table 3 shows that the absorbed dose rates due to the terrestrial gamma rays at 1 m above the ground in samples are in the range of 31.77 to 95.22 nGy $h^{-1}$with a mean value of 57.21 nGy h^{-1}. This value is higher than the world average value of 43 nGy h^{-1} in soil [ICRP 2000] as shown in fig. 3, except for samples from stoneworks, roadstone, Mac and Isinbode quarries that are lower with values 35.96, 42.29, 31.77 and 40.53 nGy h^{-1} respectively.

Table 3 Carcinogenic risk evaluation

SAMPLE	D (nGy h^{-1})	I_α	ELCR X10^{-3}
NDC QUARRY	55.59	0.13	0.71
SAMCHASE QUARRY	69.42	0.06	0.9
DORTMUND QUARRY	60.77	0.05	0.78
JOHNSON QUARRY	95.22	0.18	1.23
RCC QUARRY	45.11	0.12	0.59
STONEWORKS QUARRY	35.96	0.05	0.47
ZEBO-FM QUARRY	74.15	0.04	0.96
OSAC QUARRY	66.19	0.16	0.86
SERVETEK QUARRY	68.24	0.07	0.88
JAPAUL QUARRY	69.02	0.15	0.89
SERENA QUARRY	53.79	0.06	0.69
BALLESTER QUARRY	54.00	0.20	0.7
NIGERCAT QUARRY	64.95	0.04	0.84
PARTIFIN QUARRY	69.35	0.23	0.89
ROADSTONE QUARRY	42.29	0.10	0.55
STONEWORLD QUARRY	53.89	0.15	0.7
INLAND QUARRY	46.55	0.04	0.6
MAC QUARRY	31.77	0.09	0.41
ISINBODE QUARRY	40.53	0.05	0.53
BCCL QUARRY	47.33	0.13	0.61
MEAN	**57.21**	**0.11**	**0.74**
MAX	**95.22**	**0.23**	**1.23**
MIN	**31.77**	**0.04**	**0.41**

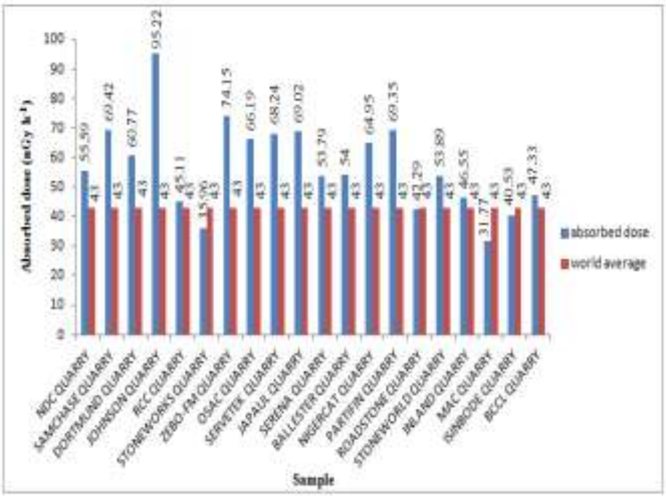

Figure 3. Calculated absorbed dose (D) compared with world average value

Internal Level Index (I_α)

Table 3 and figure 4 show that calculated internal level index (I_α) for all samples are lower than the maximum permissible value of unity [El-Galy et al 2008] [Orgun et al 2007]. The range of calculated internal level index (I_α) is 0.04 to 0.23, while the mean value is 0.11. Values calculated for internal level index (I_α) for all samples are significantly lower than the maximum permissible value of unity (the highest I_α is only 23% of the maximum permissible value of unity), therefore none of the sample can emit radon that will be concentrated enough to cause carcinogenic risk.

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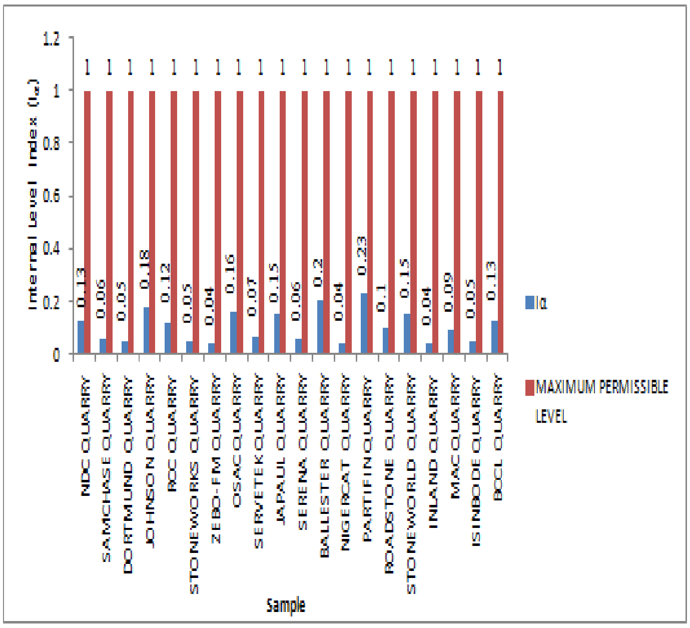

Figure 4 Internal Level Index compared with maximum permissible level

Excess Lifetime Cancer Risk (ELCR)

Table 4 shows that calculated excess lifetime cancer risk (ELCR) for all samples are higher than the world average value of 0.29 x 10^{-3} [Taskin et al 2009]. Therefore, the probability of cancer occurrence due to these samples is higher than the world average. The probability of cancer occurrence due to each sample is highlighted in table 4.

Table 4 Probability of cancer occurrence due to each sample

SAMPLE	ELCR X 10^{-3}	REMARKS (Probability of having cancer)
NDC QUARRY	0.70	7 out of 10000 people
SAMCHASE QUARRY	0.91	9 out of 10000 people
DORTMUND QUARRY	0.77	8 out of 10000 people
JOHNSON QUARRY	1.23	12 out of 10000 people
RCC QUARRY	0.60	6 out of 10000 people
STONEWORKS QUARRY	0.46	5 out of 10000 people
ZEBO-FM QUARRY	0.95	10 out of 10000 people
OSAC QUARRY	0.84	8 out of 10000 people
SERVETEK QUARRY	0.88	9 out of 10000 people
JAPAUL QUARRY	0.88	9 out of 10000 people
SERENA QUARRY	0.70	7 out of 10000 people
BALLESTER QUARRY	0.70	7 out of 10000 people
NIGERCAT QUARRY	0.84	8 out of 10000 people
PARTIFIN QUARRY	0.91	9 out of 10000 people
ROADSTONE QUARRY	0.56	6 out of 10000 people
STONEWORLD QUARRY	0.70	7 out of 10000 people
INLAND QUARRY	0.60	6 out of 10000 people
MAC QUARRY	0.42	4 out of 10000 people
ISINBODE QUARRY	0.53	5 out of 10000 people
BCCL QUARRY	0.60	6 out of 10000 people

IV. CONCLUSION

It is important to evaluate the carcinogenic effect of crushed granite samples in the selected quarries in order to evaluate the health hazard they may cause on the dwellers of the building they are used to build. In order to provide information on ionizing radiation exposure levels and carcinogenic risk associated with the use of granites from these locations and make recommendations about the need for regulations and control of exposure to radiation if carcinogenic risk is discovered. Data obtained in this study may be useful for future investigation of natural radioactivity in the selected locations and radiation impact assessment of the selected quarries on their workers or people living within the selected locations. Conclusively, though the granite samples collected have internal level index lower than the maximum permissible level, but that does not rule out carcinogenic risk. Therefore the range of probability of developing cancer in a lifetime as shown in table 4 is “4 to 12 out of 10000 people” for samples from the selected locations.

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