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Assessment Of Radionuclides In Selected Granite Quarry Sites Within Ohimini And Gwer-East Local Government Areas Of Benue State Nigeria

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***Abstract:** The assessment of radionuclides in selected granite quarry sites in Ohimini and Gwer-East Local Government Areas of Benue State was carried out using a 76x76mm NaI (TI) detector crystal optically coupled to a photomultiplier tube (PMT). The detector is enclosed in a 6cm lead shield with cadmium and copper sheet and data acquisition was done by Canberra Nuclear Products (Meastro) at Center for Energy Research Training, Ahmadu Bello University, Zaria. These sites include Anmuda, Awulema and Ikpayongo quarry sites with five (5) samples from each site making a total of fifteen (15) samples. From the preliminary investigation, the mean absorbed dose in Anmuda, Awulema and Ikpayongo are 12.3nGy/h, 17.27nGy/h and 18.87nGy/h respectively and the corresponding Mean Annual Effective Dose equivalent are 0.15mSv/y, 0.21mSv/y and 0.25mSv/y which are actually below the recommended safe values. The result of the assessment of radionuclides in granite samples include Radium-226 (^{226}Ra), Thorium-232 (^{232}Th) and Potassium-40 (^{40}K). The mean activity concentration in Anmuda for ^{40}K is 86.717±4.656Bq/kg, that of ^{226}Ra is 7.862±3.728 Bq/kg and ^{232}Th is 5.280±1.505Bg/kg, in Awulema, ^{40}K is 155.656±5.074Bq/kg, ^{226}Ra is 3.614±4.220Bq/kg and ^{232}Th is 8.478±3.842Bq/kg and in Ikpayongo^{40}K is 329.737±9.405Bq/kg, ^{226}Ra is 5.394±5.032Bq/kg and ^{232}Th is 7.067±3.820Bq/kg. The mean absorbed dose in Anmuda, Awulema and Ikpayongo are 10.6094nGy/h, 13.845nGy/h and 21.212nGy/h. The mean annual effective dose equivalent in Anmuda is 0.13mSvy^{-1}, Awulema is 0.16mSvy^{-1} and Ikpayongo is 0.25mSvy^{-1}. The average value falls within the global range of outdoor radiation exposure given by UNSCEAR-2000 publication.*

***Keywords:** Radionuclides, granite quarry, Activity concentration, Absorbed Dose, Annual Effective Dose and Radiation Hazards.*

I. INTRODUCTION

Radiation is defined as the transfer of energy through space and matter in the form of wave and particle. Natural and artificial sources of ionizing radiation are present in the environment in which we live. Naturally occurring radionuclides are present in air we breathe, the food we eat and the water we drink and have resulted in adverse health consequences on the public. Human can be exposed to radionuclide from natural and artificial sources through human activities such as mining and drilling (Aregunjo et al, 2004). The artificial sources also include human activities such as nuclear and atomic bomb testing, nuclear reactor explosions, mining activities, industrial waste and effluent from factories (Agba et al 2006). International Atomic Energy Agency (IAEA) estimation shows that over 85% of radiation dose received by man are derived from the naturally occurring radionuclides while the remaining 15% is from cosmic rays

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and nuclear processes all around the globe (IAEA, 1986). Over exposure to radiation could cause adverse health effects such as leukemia, chromosomal breakage, bone necrosis, bone cancer, mutation of genes, cataracts of eye lens etc The mining of mineral resources such as granite, limestone, marbles etc can facilitate the release of radioactive materials from the host material (ores) into the environment since most minerals (ores) co-exist with naturally occurring radionuclides (Paul et al; 1986).

On the earth, natural radioactive mineral deposits are available in many suitable geological environments (Bhaumik et al., 2004). The high exposure level of these radionuclides in such an area may be harmful to people residing in the region. The greatest contribution to human exposure comes from natural background radiation (UNSCEAR, 2000).

The energy transferred by ionizing radiation to the mass per unit volume, is called absorbed dose. The probability of affecting the human health is directly related to the absorbed dose. The worldwide average natural dose to humans is about 2.4 mSv per year (UNSCEAR, 2000).

The major natural radionuclides of concern are Thorium-232 (^{232}Th) and Radium-226 (^{226}Ra) and their decay products as well as Potassium-40 (^{40}K). Thorium and Uranium primarily undergo alpha and beta decay and are not easily detectable. However, many of their daughter products are strong gamma emitters. Gamma rays are more penetrating than alpha or beta particles and are most often used to characterize the terrestrial component of the natural radiation environment. Thus, the gamma ray emissions from ^{232}Th and ^{238}U radioactive daughter products are used to estimate their concentrations (Ibrahim et al., 2013).

Granite is the best-known igneous rock which has extensive applications. Granite is used to make many objects that we encounter in life daily. These include counter tops, floor tiles, paving stone, stair, building veneer, and cemetery monuments (Geology and Health Science News, 2016). At the same time granite is a natural source of radioactivity like most natural rocks. The main part of the radioactivity in this kind of samples is attributed to the potassium-40 radioisotope. ^{40}K exists in a standard relation with total potassium content, with a natural abundance of 0.012%. Granites contain also some dozens of parts per million (ppm) of Uranium and Thorium as well (IAEA, 1986).

Granite quarries produce massive amount of dust particles and such dust particles increase the level of Radon gas. (USGAS, 2016). The main way of saving the humans from radiation injuries is to protect it from exposure to radiation and this is why radiation monitoring and measurement are very essential in our society today.

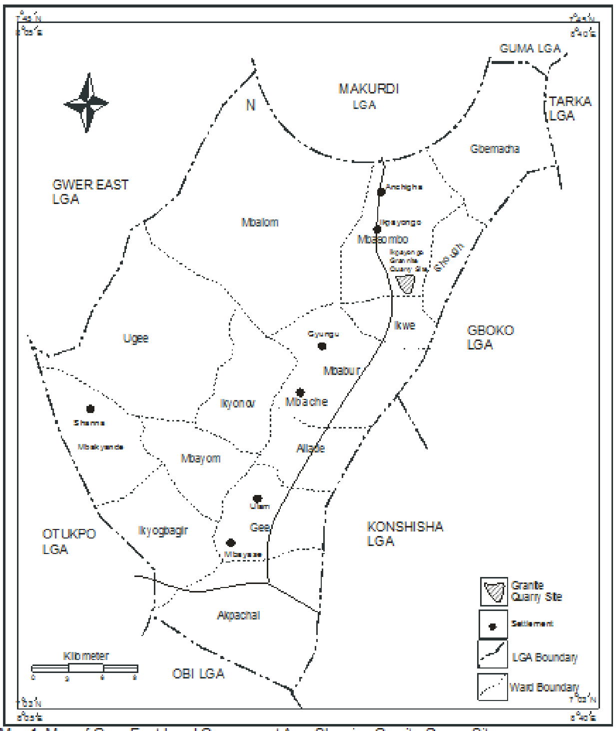
II. EXPERIMENTAL PROCEDURE (METHODOLOGY)

DESCRIPTION OF STUDY AREA

S/No	Location	Bearing	Latitude	Longitude
1	Anmuda	249°	7.193°	8.09166°
2	Awulema	262°	7.22315°	8.09166°
3	Ikpayongo	288°	7.58157°	9.69039°

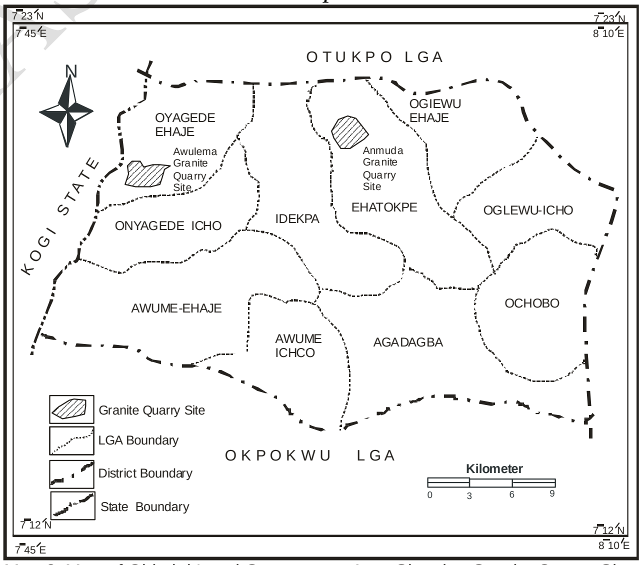
(Google map, 2016), (Geody Lab, 2016), (Amali, 2002).

Table 1.1: Sample location and their coordinate in Gwer-East and Ohimini Local Government



Map 1: Map of Gwer East Local Government Area Showing Granite Quarry Site
Source: Ministry of Lands and Survey Makurdi

Map 1



Map 2: Map of Ohimini Local Government Area Showing Granite Quarry Sites
Source: Ministry of Lands and Survey, Makurdi

Map 2

The sample analysis was done at Center for Energy Research, Ahmedu Bello University, Samaru Main Campus Zaria Nigeria.

SAMPLE COLLECTION

Five (5) samples of granite were collected each from Anmuda and Awulema quarry sites both in Ohimini local Government Area, similarly five (5) samples of granite were collected from Ikpayongo quarry site in Gwer-East Local Government area making a total of fifteen (15) samples

PRELIMINARY SURVEY AND SAMPLE PREPARATION

The assessment of the present of radiation in the sites was carried out using radiation meter (radiation alert inspector). The background radiation within the environment was measured. A small hand microprocessor attached to monitor was held about 4-5cm above the sea level and the radiation emitted was recorded within an interval of one minute (60 seconds) each. The average dose rate was measured in count per minute and converted to exposure dose rate using 1200(CPM) = 1μSv/h as conversion factor. Also a conversion factor of 1μSv/h = 1000nGy/h for absorbed dose rate enabled the estimation of annual effective dose rate. (UNSCEA, 1998)

The collected samples were sun-dried and pulverized to $1mm^2$ mesh sizes with a jaw crusher and ceramic mortar, and then homogenized to obtain a fine texture powder that gave a uniform matrix to the detector. The mass of each of the final powdered samples that was used for the analysis was 350g. The packaging of the samples into radon-impermeable cylindrical plastic containers which were selected based on the space allocation of the detector vessel which measures 7.6cm by 7.6cm in dimension (geometry) was also carried out. To prevent radon-222 escaping, the packaging in each case was triple sealed. The sealing process included smearing the inner rim of each container lid with Vaseline jelly, filling the lid assembly gap with candle wax to block the gap between lid and container, and tight-sealing lid-container with masking adhesive tape. Radon and its short-lived progenies were allowed to reach secular radioactive equilibrium between precursor and progeny radionuclides by storing the samples for 30 days prior to gamma spectroscopy measurement (Kaharan; et al. 2000).

EVALUATION OF RADIOACTIVITY OF SAMPLE AND THE DOSE

The analysis was carried out using a 76x76mm NaI (TI) detector crystal optically coupled to a photomultiplier tube (PMT). The assembly has a preamplifier incorporated into it and a 1kilovolt external source. The detector is enclosed in a 6cm leadshield with cadmium and copper sheet. This arrangement is aimed at minimizing the effects of background and scattered radiation.

The calibration of the system for energy and efficiency were done with two calibration point sources, Cs-137 and Co-60. These were done with the amplifier gain that gives 72% energy resolution for the 66.16KeV of Cs-137 and counted for 30 minutes.

The data acquisition was done by Canberra Nuclear Products (Meastro). The samples were measured for a period of 29000 seconds each. The peak area of each energy in the spectrum was used to compute the activity concentration in each sample by using the following equation:

$$C\ (Bq/kg) = \frac{C_n}{C_{fk}} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots \text{(1)}$$

Where:

C = activity concentration of the radionuclides in the sample given in $BqKg^{-1}$

C_n = count rate (counts per seconds) count per second (cps) = Net count/ live Time

C_{fk}= Calibration factor of the detecting system which is the product of the efficiency of the detector, the intensity of the gamma line in the radionuclide and the mass of the sample.

The actual concentration is gotten after deducting the reduction factor for each radionuclide given as; 12.3 Bq/kg for ^{226}Ra, 15.5Bq/kg for ^{232}Th and 18.5Bq/kg for ^{40}K. (Avwiri and Ononugbo, 2012)

The absorption dose was calculated based on guidelines provided by ICRP-60 using the following relation;

$$Do = 0.427\ C_{Ra} + 0.662\ C_{Th} + 0.043\ C_k \ldots\ldots\ldots\ldots\ldots\ldots \text{(2)}$$

Where;

C_{Ra}, C_{Th} and $C_k$are the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K

0.427, 0.662 and 0.043 are dose conversion factor for ^{226}Ra, ^{232}Th and ^{40}K by the new ICRP dosimetric setting. (ICRP 2013).

The Annual Effective Dose Rate can be estimated by taking into account the conversion coefficient from the absorbed dose in air to the effective dose in air ($0.7SvGy^{-1}$) and an outdoor occupancy factor of 0.2 received by adults. The annual effective dose equivalent was calculated by the following equation (UNSCEAR 2000)

$$E\ (mSvy^{-1}) = D(nGyh^{-1}) \times 8760(h) \times 0.2 \times 0.7(mSvy^{-1}) \times 10^{-6} \quad \ldots\ldots\text{(3)}$$

III. RESULT AND DISCUSSION

PRELIMINARY SURVEY

The following tables show a comparative result of the radioactivity measured in terms of dose rate and exposure in different locations within the three quarry sites with the use of radiation meter inspection.

S/No	Sample code	Dose rate (CPM)	Actual Dose rate(CPM)	Exposure (μSv/hr)	Annual effective Dose (mSv/yr)
1	NN1	40.500	16.900	0.014	0.172
2	NW1	36.500	12.900	0.011	0.135
3	NC1	40.300	16.700	0.014	0.172
4	NE1	42.000	18.400	0.015	0.184
5	NS1	36.000	12.400	0.010	0.123
6	NN2	37.500	13.000	0.011	0.135
7	NW2	33.000	9.4000	0.008	0.098
8	NC2	41.000	17.000	0.014	0.172
9	NE2	39.500	15.900	0.013	0.160
10	NS2	38.000	14.400	0.012	0.147
11	NN3	36.800	13.200	0.011	0.135
12	NW3	40.000	16.400	0.014	0.172
13	NC3	42.800	19.200	0.016	0.196
14	NE3	35.500	11.900	0.010	0.127
15	NS3	37.000	13.400	0.011	0.135

NN=Anmuda North, NW=Anmuda West, NC=Anmuda Center, NE=Anmuda East NS=Anmuda South

Table 1.2: Doses at different locations in Anmuda quarry site with a background radiation of 23.0 Counts Per Minute (CPM)

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S/No	Sample code	Dose rate (CPM)	Actual Dose rate(CPM)	Exposure (µSv/hr)	Annual effective Dose (mSv/yr)
1	WN1	41.000	23.000	0.019	0.233
2	WW1	38.000	20.000	0.017	0.208
3	WC1	40.200	22.200	0.019	0.233
4	WE1	40.800	22.800	0.019	0.233
5	WS1	31.200	13.200	0.011	0.135
6	WN2	38.800	20.000	0.017	0.208
7	WW2	42.000	24.000	0.020	0.245
8	WC2	41.000	23.000	0.019	0.233
9	WE2	36.400	18.400	0.015	0.184
10	WS2	38.500	20.000	0.016	0.196
11	WN3	40.000	22.000	0.018	0.221
12	WW3	41.500	23.500	0.020	0.245
13	WC3	36.000	18.000	0.015	0.184
14	WE3	35.800	17.800	0.014	0.172
15	WS3	42.200	24.200	0.020	0.245

Table 1.3: Doses in different locations in Awulema quarry site with a background radiation of 18.0 Count Per Minute (CPM)

S/No	Sample code	Dose rate (CPM)	Actual Dose rate(CPM)	Exposure (µSv/hr)	Annual effective Dose (mSv/yr)
1	IN1	44.000	24.000	0.020	0.245
2	IW1	45.000	25.000	0.021	0.256
3	IC1	45.800	25.000	0.021	0.256
4	IE1	45.500	25.500	0.021	0.256
5	IS1	46.800	26.800	0.022	0.270
6	IN2	48.000	28.000	0.023	0.282
7	IW2	37.000	17.000	0.014	0.172
8	IC2	48.500	28.500	0.024	0.294
9	IE2	51.200	31.200	0.026	0.319
10	IS2	44.800	24.800	0.021	0.256
11	IN3	41.500	21.500	0.018	0.221
12	IW3	41.000	21.000	0.018	0.221
13	IC3	52.000	32.000	0.027	0.331
14	IE3	42.000	22.000	0.018	0.221
15	IS3	36.000	16.000	0.013	0.159

Table 1.4: Doses in different locations in Ikpayongo quarry site with a background radiation of 20.0 Count Per Minute (CPM)

S/No	Sample code	^{40}K (Bq/kg)	^{226}Ra (Bq/kg)	^{232}Th (Bq/kg)
1	NN	58.016±3.736	9.016±3.475	6.164±1.140
2	NW	65.787±3.287	7.712±3.455	4.534±1.772
3	NC	78.930±3.324	7.601±3.212	5.287±1.621
4	NE	103.939±4.332	7.578±3.982	5.482±1.921
5	NS	126.912±8.600	7.394±4.518	5.138±1.071
6	WN	132.977±4.417	2.297±4.286	0.577±2.052
7	WW	155.021±4.908	4.683±4.445	2.838±3.112
8	WC	160.933±4.546	3.554±4.219	6.790±2.891
9	WE	158.715±5.902	3.735±5.134	13.820±2.936
10	WS	170.635±5.599	3.803±3.012	18.365±8.233
11	IN	354.595±10.264	5.193±4.402	15.286±3.192
12	IW	336.690±9.328	5.938±4.523	2.943±2.277
13	IC	310.956±9.760	6.473±5.451	4.043±3.754
14	IE	362.780±10.673	5.342±5.920	12.043±3.841
15	IS	284.035±6.998	4.023±4.865	1.019±2.280

Table 1.5: The radionuclides and their corresponding activity concentrations

S/No	Sample Code	Absorbed dose (nGyh^{-hr})
1	NN	10.425
2	NE	9.123
3	NC	10.140
4	NW	11.333
5	NS	12.026
6	WN	7.081
7	WE	10.525
8	WC	12.933
9	WW	17.568
10	WS	21.119
11	IN	27.583
12	IE	18.961
13	IC	18.812
14	IW	25.853
15	IS	14.606

Table 1.6: The absorbed dose in the quarry sites

S/No	Sample Code	Annual Effective Dose (mSvy^{-1})
1	NN	0.128
2	NE	0.112
3	NC	0.124
4	NW	0.139
5	NS	0.147
6	WN	0.087
7	WE	0.129
8	WC	0.158
9	WW	0.215
10	WS	0.259
11	IN	0.338
12	IE	0.233
13	IC	0.231
14	IW	0.317
15	IS	0.179

Table 1.7: the annual effective dose in the quarry sites

GRAPHICAL REPRESENTATIONS

Below are figure 1 and 2 showing graphical representation of the specific activity, the absorbed dose and the annual effective dose in the various quarry sites respectively.

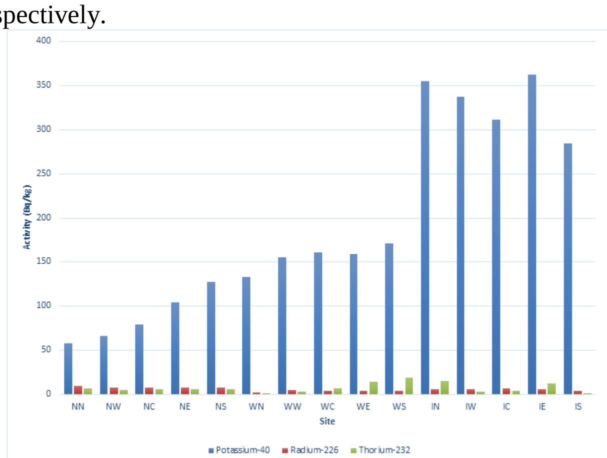

Figure 1: Specific activity of ^{40}K, ^{226}Ra and ^{232}Th in the various Quarry sites

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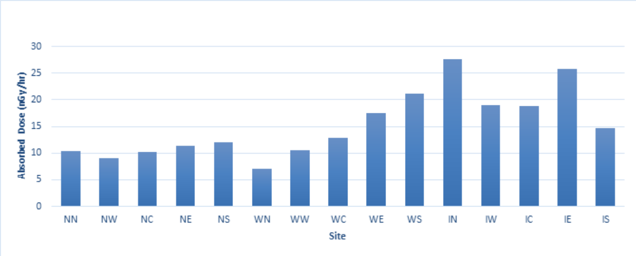

Figure 2: Variation of absorbed dose with sites

IV. CONCLUSION

The preliminary investigation recorded the mean absorbed dose (D_o) in Anmuda, Awulema and Ikpayongo are 12.3nGy/h, 17.27nGy/h and 18.87nGy/h respectively and the corresponding mean Annual Effective Dose equivalent (AED) are 0.15mSv/y, 0.21mSv/y and 0.25mSv/y which are below the recommended safe values.

The result of the assessment of radionuclides in granite samples are ^{226}Ra, ^{232}Th and ^{40}K. The mean activity concentration in Anmuda of ^{40}K is 86.717±4.656Bq/kg, ^{226}Ra is 7.862±3.728Bq/kg and ^{232}Th is 5.280±1.505Bg/kg. In Awulema ^{40}K is 155.656±5.074Bq/kg, ^{226}Ra is 3.614±4.220Bq/kg and ^{232}Th is 8.478±3.842Bq/kg. In Ikpayongo ^{40}K is 329.737±9.405Bq/kg, ^{226}Ra is 5.394±5.8.032Bq/kg and ^{232}Th is 7.067±3.820Bq/kg. The activity concentrations are lower than the worldwide average value of 420Bq/kg, 33Bq/kg and 45Bq/kg for ^{40}K, ^{226}Ra and ^{232}Th recommended by UNSCEAR. ^{40}K is a beta decay radionuclide, ^{226}Raand ^{232}Th are alpha decay radionuclides.

The mean absorbed dose in Anmuda, Awulema and Ikpayongo are 10.6094nGy/h, 13.845nGy/h and 21.212nGy/h which are all lower than 30-70nGy/h by UNSCEAR

The mean annual effective dose equivalent inAnmoda is 0.13mSvy^{-1}, Awulema is 0.16mSvy^{-1} and Ikpayongo is 0.25mSvy^{-1}. The annual effective dose rates in the three samples are lower than the recommended 0.50mSvy^{-1} by UNSCEAR (2000).

^{40}K contributes the most in all the three sites follow by ^{226}Ra in Anmuda and Ikpayongo but the contribution of ^{232}Th is more than that of ^{226}Ra in Awulema. The percentage contribution to the absorbed dose of ^{40}K, ^{226}Ra and ^{232}Th in Anmuda are 86.8%, 7.8% and 5.3%. The percentage contribution to the absorbed dose of ^{40}K, ^{226}Ra and ^{232}Th in Awulema is 92.7%, 2.2% and 5.1% and the percentage contribution to the absorbed dose of ^{40}K, ^{226}Ra and ^{232}Th in Ikpayongo is 96.3%, 1.6% and 2.1% respectively. The activity concentration shows variations from sample to sample which may be attributed to the granite type (the geological characteristics of the area where the granite was extracted). It should be noted that, the main contribution in the specific activity of each sample due to ^{40}K is high because of the high concentration in potassium in the sample and the surrounding.

The average units of all the calculated radiological indices extracted from those activities and the absorbed dose in all the investigated samples are within the level recommended by UNSCEAR 2000 report.

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