THE NATURAL RADIOACTIVITY MEASUREMENTS AND EVALUATION OF RADIOLOGICAL HEALTH HAZARD INDICES OF QUARRY PRODUCTS FROM SOUTH EASTERN NIGERIA

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Abstract

The naturally occurring radioactivity concentrations of ⁴⁰K, ²³⁸U and ²³²Th in quarry samples from different locations in some South Eastern part of Nigeria was carried out using NaI (TI) detector. The mean values obtained for 40 K, 238 U and 232 Th were 1.028.63 ± 27.48 Bg kg⁻¹, 5.88 ± 3.8 Bg kg⁻¹ and 2.65 ± 1.4 Bg kg⁻¹ respectively. These radioactivity concentrations were used to determine the indoor and outdoor values of the following ; Absorbed Dose Rate, Annual Effective Dose Equivalent, the Health Hazard Indices and Excess Lifetime Cancer Risk using standard analytical methods. The mean values of the outdoor absorbed dose rate and outdoor annual effective dose equivalent which are 49.99 nGyh⁻¹ and 60.17 µSvy⁻¹ are lower than the world permissible UNSCEAR levels of 60.00 nGyh⁻¹ and 70.00 μ Svy⁻¹ respectively. But the mean values of the indoor absorbed dose rate and the indoor annual effective dose equivalent which are 95.19 nGyh⁻¹ and 466.85 μ Svy⁻¹, are slightly higher than the world permissible levels of 84.00 nGyh⁻¹ and 410.00 μ Svy⁻¹ respectively. The mean value of the outdoor excess lifetime cancer risk is 0.21 which is lower than the world permissible level of 0.29. However the mean value for the indoor excess lifetime cancer risk is 1.64 which is also slightly higher than the world permissible UNSCEAR level of 1.16. The mean values of the outdoor and indoor health hazard index are 0.02 and 0.29 respectively. These are less than unity. The mean value of the radium equivalent activity is 82.97 Bq kg⁻¹, which is much lower than the world permissible UNSCEAR value of 370.00 Bq kg⁻¹. The mean value of the annual gonad dose equivalent is 365.94 Bq kg⁻¹, which is slightly higher than the world recommended value of 300.00 Bq kg⁻¹. This shows that there is a very low radiation and cancer risk for the people living around the surroundings of the quarry sites. However, there is a high radiation and cancer risk to people living inside buildings constructed with quarry materials.

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Keywords : Natural radionuclides, radiological health hazard indices, quarry materials, buildings, excess lifetime cancer risk, UNSCEAR values

1. Introduction

Living organisms are continuously exposed to a wide range of ionizing radiations from naturally occurring radioactive materials (NORMs) and artificial radionuclides (Isinkaye and Emelue, 2015). Natural radiation sources could be grouped into cosmic and primordial radiations. Cosmic radiation is highly penetrating and of galactic in origin. It is formed by the interactions of cosmic rays and particles in the atmosphere. They include heavily high speed particles, high energy photons and muons. Examples are ${}^{3}H$, ${}^{7}Be$, ${}^{14}C$, and ${}^{22}Na$ which are the dominant sources of ionization in the atmosphere from 1 Km to 70 Km above the sea level. The intensities of these secondary radiations at any point depend on the altitude. Primordial radionuclides are radioactive elements that have survived since the time of the formation of the earth. They have long half lives, and their decay products are mostly the isotopes of Potassium, ${}^{40}K$, Thorium ${}^{232}Th$ and Radium (Uranium) ${}^{226}Ra$ (Aznam et al., 2009). They are wide spread in the soil, water, rocks and living tissues. Artificial radionuclides are from the bye – products of nuclear power plant, nuclear accidents and medical and industrial applications.

Natural radiation from NORMs constitutes about 87% of the radiation doses received by humans (Shetty and Narayana, 2010). Moreover, they give rise to a very much larger radiological effect on the public than that caused by the artificial sources of radiation because of their wide distribution (UNSCEAR 2000). Most building materials contain a very high level of NORMS, which could constitute health hazards to human

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beings that live in them (Shoeib and Thabayneh, 2014). Hence, the measurement of activity concentrations in building materials is very important in the assessment of radiation exposure to the population, because individuals spend about 80% of their time indoors (Lust and Realo, 2012). This radiation, is caused by gamma radiation originating from the Potassium (⁴⁰K), Uranium (²³⁸U) and Thorium (²³²Th) series (Amrani and Tahtat, 2001)

A quarry site is a place from which dimension stone, rock, construction aggregate, riprap, sand, gravel, or slate has been excavated from the ground. This include different geological materials such as gneiss, granite, diorite, granodiorite and other rocks that after an industrial process are suitable for use as building material and ornamental rocks. It could also be referred to as an open – pit mine from which raw materials, stone or metal ore used for building constructions are extracted (www.thoughtco.com). Privately and organized mining operations have been on the increase in Nigeria leaving behind a numbers of abandoned mines of minerals (limestone, gemstones etc) and vast bare of degraded land (Ashiawa, 2007). The cumulative effect of these pockets of mining activities is that active and abandoned mines and quarries sites could have negative influence on the environmental. This is because these activities provides much of the materials used in traditional hard flooring, granite, limestone, marble, sandstone, slate and ceramic tiles (Oyinloye and Ajayi, 2015). Cement is produced majorly from limestone and some small quantities of other materials such as clay, shale ash and ion oxide (Nartey et al., 2012). Very high degrees of respiratory morbidity are associated with quarry deposits, which constitute most of the materials used for building. Fine rocks and mineral dust of different kinds have shown to be carcinogenic when inhaled.

The measurement of radiation concentrations in the environment is essential for the assessment of possible radiological risk to human (Aznam et al., 2009). Prolong exposure of the public to radiation is very dangerous if it is not detected and checked (UNSCEAR, 2000). The long – term human exposure to uranium and radium and their progenies, has several health effects such as chronic lung diseases, acute leucopoenia, anemia and necrosis of the mouth. Radium causes bone, cranial and nasal tumours. Thorium

exposure can cause lung, pancreas, hepatic, kidney cancer and leukaemia (Taskin et al 2009). Radiological health implications such as radium equivalent activity, absorbed dose rate, annual effective dose rate, hazard indices depend on the activity concentrations of ⁴⁰K, ²²⁶Ra and ²³²Th (Emelue et al 2014). There is therefore a great need to accurately determine the activity concentrations and the health hazard implications from the quarrying activities. This is because these quarrying products provide much of the materials used in traditional hard flooring, granite, limestone, marble, sandstone, slate and ceramic tiles (Oyinloye and Ajayi, 2015).

Nigeria is divided into six geo – political zones. Three of the zones are in the northern part, which includes North – East, North – West and North Central. While the remaining three zones are in the southern part, which are South – East, South – West and South – South. The South – Eastern part of Nigeria has five (5) states namely Abia, Anambra, Ebonyi, Enugu and Imo states. The major quarry locations of quarries in the South - East are scattered around Ishiagwu and its environments in Ebonyi state, Okigwe and its environments in Imo and Abia states. The map of the study area is given below in figure 1. Gbenu and others studied the radiological hazards of quarry products from southwestern Nigeria (Gbenu et al., 2015), while Essien and Akpan did a similar research work in Itu Akwa Ibom state (South South) (Essien and Akpan, 2016), but data is scarce about the radioactivity levels and the health hazard implications of quarry products from southeastern Nigeria. The purpose of this research therefore is to measure the activity concentrations of the natural radionuclides in selected quarry sites in the south eastern Nigeria. These activity concentrations obtained will be used to determine the radiological health hazard implications and excess lifetime cancer risks both to the people living in the surroundings of the sites, but also to the people living inside the house built by materials from the quarry products. This could serve as a baseline study for future references and research.

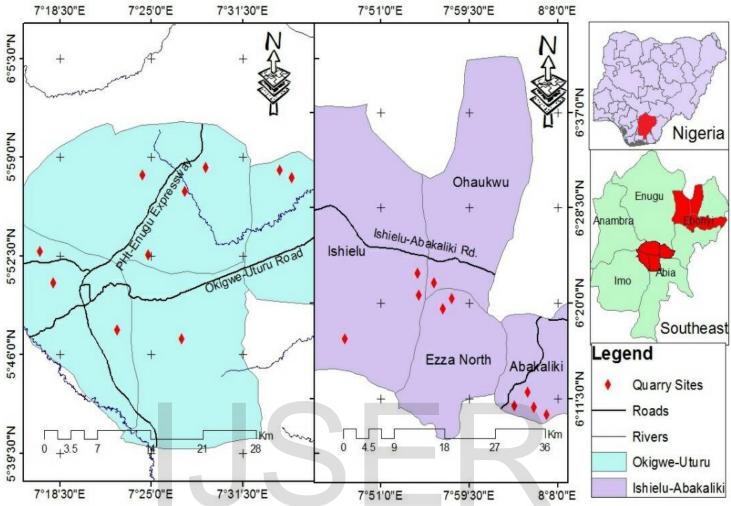


Figure 1: The map of the study area

2. Materials and Methods

The quarry samples were randomly collected in waterproof nylon bags at different locations in, around quarry sites, labeled appropriately, and transported to the radiation laboratory for analysis. The samples were air – dried and homogenized to pass through a 1mm mesh sieve. About 0.2 Kg of the each sample was weighed and transferred into a plastic container of about 250 ml which has been verified to be non radioactive. These samples were sealed for about 28 days for the short – lived members of Radon – 222 (²²²Ra) and Radon – 220 (²²⁰Ra) series to reach a secular equilibrium. Each of the samples were placed symmetrically on top of the detector and measured for 10 hours (36,000s). The net area under the corresponding photopeaks in the energy spectrum was computed by subtracting count due to Compton



scattering of the background source from the total area of the photopeaks. The radionuclides were computed using the algorithm of the multichannel analyzer (MCA).

The scintillation detector used in this work is a lead shield Canberra 76mm x 76mm NaI (TI) crystal models number 802 – series. One face of the cylindrical detector is free while the other is optically coupled to a Photomultiplier tube that detects the small visible light photons produced in the crystal and converts them into amplified electrical pulses, which is fed into analyzer systems (Canberra series 10 plus multichannel analyzer MCA) through a preamplifier base. This gamma spectrometry detector was first calibrated before it was used for the analysis of the samples. This was done to ensure that the radiation parameters in the samples could be expressed in physical radiometric units. This calibration was done in two stages. These are energy and efficiency calibrations. The energy calibration converts channel numbers to gamma ray (γ – ray) energy in Mev. This was done by placing different gamma sources of known energy in the detector. After a preset counting time of 10 hours (36,000s), the channels of the various photopeaks corresponding to the gamma energies were then identified. Then the efficiency calibration was to determine the gamma ray counting efficiencies over energy range of 0.662 - 2.615 MeV. This was done by converting the count per seconds under the photopeaks to activity concentration in Bq kg⁻¹ of certified reference standard samples. The certified reference standard samples have activity concentrations of 7.24 Bq kg⁻¹ for 137Cs (0.662 Mev), 578.40 Bq kg⁻¹ for 40K (1.460 Mev), 20.90 Bq kg⁻¹ for 238U (1.760 Mev) and 10.47 Bq kg⁻¹ for 232Th (2.615 Mev).

3. Results and Discussions.

In this study, the following parameters were determined in order to adequately assess the health hazard indices and excess lifetime cancer risk.

3.1 Activity concentrations of ⁴⁰K, ²³⁸U and ²³²Th (Bq kg⁻¹)

The activity concentrations of the primordial radionuclides of 40 K, 232 Th and 238 U from the samples of quarry sites were obtained. The mean activity concentration of Potassium (40 K) is 1,028.63 ± 27.48 Bq kg⁻¹,

Uranium (²³⁸U) in the samples is 5.88 ± 3.8 Bq kg⁻¹, while that of Thorium (²³²Th) in the samples is 2.65 ± 1.4 Bq kg⁻¹. Table 2 shows the activity concentration of the three primordial radionuclides. While Figure 2 is showing the distribution of ⁴⁰K, ²³⁸U and ²³²Th with respect to the samples in the quarry sites

3.2 Absorbed Dose rate. D (nGyh⁻¹)

The contribution of natural radionuclides to the absorbed dose rates depends on the concentrations of various radionuclides in the samples of the quarries (Erenturk et al., 2014). The indoor gamma dose (D_{in}) imparted by the emission of gamma – ray from ⁴⁰K, ²³⁸U and ²³²Th in the samples when utilized as building materials was calculated using the formula given by (Qureshi et al., 2014) for a standard room of dimensions 4 m x 5 m x 2.8 m in equation 1.

$$D_{(indoors)} (nGyh^{-1}) = 0.081C_k + 0.92C_u + 1.1C_{Th}$$

The outdoor gamma dose rate (Dout) at 1m above the ground surface is assessed from the gamma radiation originating from ⁴⁰K, ²³⁸U and ²³²Th. The formula for calculating the outdoor dose rate according to (Bede et al., 2015), is given in equation 2.

$$D_{(outdoors)} (nGyh^{-1}) = 0.0417C_K + 0.429C_u + 0.666C_{Th}$$

Where C_k , C_u and C_{Th} are the activity concentrations of ${}^{40}K$, ${}^{238}U$ and ${}^{232}Th$ respectively.

The values of the outdoor and indoor absorbed dose rates compared to the world standard values are given in table 2. It was observed that the mean outdoor dose rate for samples, which is 49.99 nGyh⁻¹, is less than the recommended standard (UNSCEAR) value of 60.00 nGyh⁻¹ (UNSCEAR, 2000). But the mean indoor dose rate of 95.19 nGyh⁻¹ is slightly higher than the world recommended (UNSCEAR) value of 84 nGyh⁻¹ (UNSCEAR, 2000). The high concentration of ⁴⁰K contributed significantly to the high value of indoor dose rates in the samples.

 Table 1: The activity concentrations of of ⁴⁰K, ²³²Th and ²³⁸U from samples of quarry sites in the

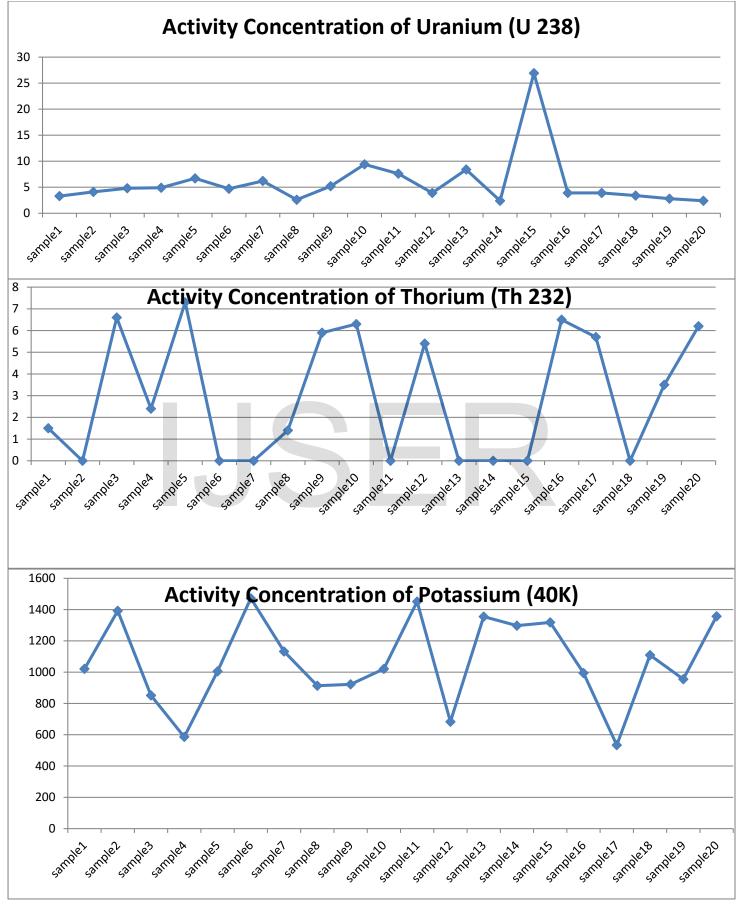
 South Eastern Nigeria

Samples	⁴⁰ K Bq kg ⁻¹	²³⁸ U Bq kg ⁻¹	²³² Th Bq kg ⁻¹



518	0 0	, , , 1		
1		1021.1 ± 5.7	3.3 ± 2.6	1.5 ± 0.05
2		1392.2 ± 6.3	4.1 ± 3.0	BDL
3		851.4 ± 6.5	4.8 ± 3.2	6.6 ± 1.6
4		586.3 ± 7.1	4.9 ± 3.8	2.4 ± 0.4
5		1005.2 ± 5.5	6.7 ± 3.7	7.3 ± 0.1
6		1472.3 ± 6.9	4.7 ± 5.3	BDL
7		1132.3 ± 5.2	6.2 ± 3.6	BDL
8		913.3 ± 6.1	2.6 ± 2.2	139 ± 0.1
9		922.3 ± 6.1	5.2 ± 3.4	5.9 ± 1.5
10		1021.0 ± 6.5	9.4 ± 3.2	6.3 ± 1.7
11		1451.3 ± 6.7	7.6 ± 2.7	BDL
12		683.1 ± 6.5	3.9 ± 3.1	5.4 ± 1.3
13		1355.1 ± 6.4	8.4 ± 3.2	BDL
14	IU	1297.5 ± 5.9	2.4 ± 2.1	BDL
15		1318.1 ± 5.8	26.9 ± 2.2	BDL
16		994.7 ± 6.4	3.9 ± 2.7	6.5 ± 1.7
17		533.7 ± 6.4	3.9 ± 3.0	5.7 ± 1.6
18		1109.3 ± 5.0	3.4 ± 2.7	BDL
19		955.3 ± 4.5	2.8 ± 2.4	3.5 ± 0.9
20		1357.1 ± 6.7	2.4 ± 2.1	6.2 ± 0.9
Mean		$1,028.63 \pm 27.48$	5.88 ± 3.8	2.65 ± 1.4

BDL = Below Detection Limit. (UNSCEAR 2000)



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3.3 Annual Effective Dose Equivalent (AEDE) (Bq kg⁻¹)

The indoor annual effective dose equivalent (AEDE indoors) to the population due to radioactivity from the quarry sites was computed from the indoors absorbed dose rate by applying a dose conversion factor of 0.7 SvGy⁻¹ and indoor occupancy factor of 0.8. This was done on the estimation that an average person spends about 19 hours indoors on daily (Veiga et al., 2006). The formula used for the calculation is given in equation 3. (Sivakumar et al., 2014).

$$AEDE_{in} (\mu Svy^{-1}) = D_{in} (nGyh^{-1}) \times 8660 \times 0.7 \times 0.8 \ 10^{-6}$$

The indoor annual effective dose is given in table 2. The mean value obtained in this research work is 466.85 μ Svy⁻¹, which is slightly higher than the world standard (UNSCEAR) value of 410.00 mSvy⁻¹ as recommended by (UNSCEAR, 2000).

The outdoor annual effective dose equivalent was computed from the outdoor absorbed dose rate by applying a conversion 0.7 SvGy^{-1} and an outdoor occupancy factor of 0.2. This was done with the estimation that an average person spends about 5 hours outdoors daily. The formula used for the calculation is given in equation 3. (Sivakumar et al., 2014).

$$AEDE_{out} (\mu Svy^{-1}) = D_{out} (nGyh^{-1}) \times 8660 \times 0.7 \times 0.2 \times 10^{-6}$$

The outdoor annual effective dose is given in table 2. The mean value obtained in this research work is $60.17 \ \mu \text{Svy}^{-1}$, which is lower than the world standard (UNSCEAR) value of $70.00 \ \mu \text{Svy}^{-1}$ as recommended by (UNSCEAR, 2000).

3.4 External and Internal Hazard Indices

The external hazard index (Hex) was calculated using the formula in equation 5. (Beretka and Matthew, 1985).

$$H_{ex} = \frac{C_U}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \le 1$$
 5

In order to assess the suitability of any material to be used for building constructions, and to determine the dose delivered internally to individuals in a house constructed with such material, a dose criterion called internal hazard index (Hin) was estimated using Equation 6 (Beretka and Matthew, 1985).

$$H_{in} = \frac{C_U}{185} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \le 1$$
 6.

where C_u , C_{Th} , and $_k$ are the activity concentrations of ²³⁸U, ²³²Th and ⁴⁰K in Bq kg⁻¹ respectively. For radiological safety precautions, both the external and internal hazard indices should individually be less than one (1) (Rati et al., 2010). The values of the external (Hex) and internal hazard indices (Hin) are shown in table 2, while the chart comparing them to the world standard (UNSCEAR) values are shown in figure 3.

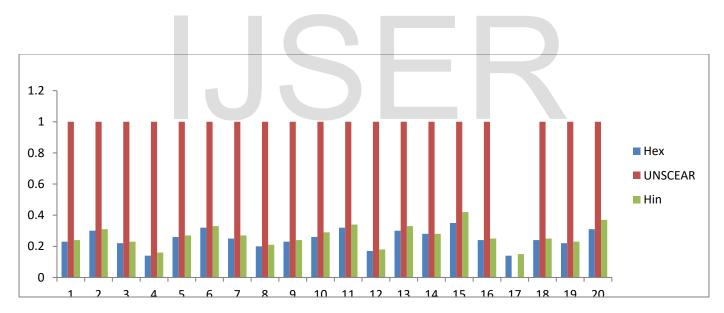


Fig.3. The external and internal health hard indices compared to the world standard values.

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Table 2: Radiological Health parameters in samples from quarry sites in Eastern Nigeria

Sam	Dose Rate	Dose Rate	AEDEout	AEDEin	Hex	Hin	Raeq	AGED	ELCRout	ELCRin
	(Out) (10 ⁻⁹)	(In) (10 ⁻⁹)	(10 ⁻⁶)	(In) (10 ⁻⁶)					(10-3)	(10-3)
1	45.00	87.40	55.19	428.75	0.23	0.24	76.92	337.09	0.193	1.501
2	59.80	116.54	73.34	571.70	0.30	0.31	101.55	449.82	0.257	2.001
3	42.00	80.64	51.51	395.59	0.22	0.23	73.84	309.76	0.180	1.385
4	28.15	54.64	34.52	268.04	0.14	0.16	49.37	209.27	0.121	0.938
5	49.65	95.62	60.89	469.07	0.26	0.27	87.50	366.85	0.213	1.642
6	63.41	123.58	77.77	606.23	0.32	0.33	68.63	476.25	0.272	2.122
7	49.88	97.42	61.17	477.90	0.25	0.27	85.46	374.70	0.214	1.673
8	40.11	77.91	49.19	379.79	0.20	0.21	68.53	300.66	0.172	1.329
9	44.62	85.98	54.72	421.78	0.23	0.24	79.20	330.33	0.192	1.476
10	50.80	98.23	62.62	481.88	0.26	0.29	89.88	375.97	0.218	1.687
11	63.78	124.55	78.22	610.99	0.32	0.34	109.19	479.19	0.274	2.139
12	33.76	64.86	41.40	318.18	0.17	0.18	59.44	249.12	0.145	1.114



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13	60.11	117.49	73.72	576.36	0.30	0.33	103.26	451.46	0.258	2.017
14	55.14	107.31	67.62	526.42	0.28	0.28	93.23	414.83	0.237	1.843
15	66.50	131.51	81.56	645.14	0.35	0.42	119.17	497.00	0.285	2.258
16	47.48	91.31	58.23	447.93	0.24	0.25	82.82	351.56	0.204	1.568
17	27.73	53.09	34.01	260.44	0.14	0.15	49.41	203.46	0.119	0.912
18	47.72	92.98	58.52	456.12	0.24	0.25	81.05	258.83	0.205	1.595
19	43.37	83.81	53.19	411.14	0.22	0.23	74.68	323.25	0.186	1.439
20	61.75	118.95	75.73	583.52	0.31	0.32	106.26	459.46	0.265	2.042
Mean	49.99	95.19	60.17	466.85	0.02	0.29	82.97	365.94	0.21	1.64
World std	60.00	84.00	70.00	410.00	1.00	1.0	370.00	300.00	0.29	1.16

3.5 Excess Life Time Cancer Risk (ELCR)

The excess lifetime cancer risk is the probability of developing cancer over a lifetime at a given exposure level. In this research work, the risk of developing cancer for the people in the surroundings of the quarry sites known as (external lifetime cancer risk) and also for the people living in a house in a building constructed with such materials (known as internal lifetime cancer risk) were calculated. The equation for both external and internal cancer risk (ELCR) is given in equation 7. (Amrani and Tahtat, 20010).

$$ELCR = AEDE \times DL \times RF$$
 7

Where AEDE ext = the external annual effective dose equivalent, is used to calculate the external cancer risk.

While AEDE (internal) is the internal annual effective dose equivalent is used to calculate the internal cancer risk.

DL = the average life duration (estimated to be 70 years) and

RF = the risk factor of contacting cancer. The International Commission for Radiological Protection (ICRP, 1999) uses 0.05 for stochastic effect for the public (Thabayneh and Jazzar, 2013). The values of both the internal (ELCRint) and external risks (ELCRext) are given in table 2. The chart comparing both the internal and external excess lifetime cancer risks with the world (UNSCEAR) values are shown in figure 4.

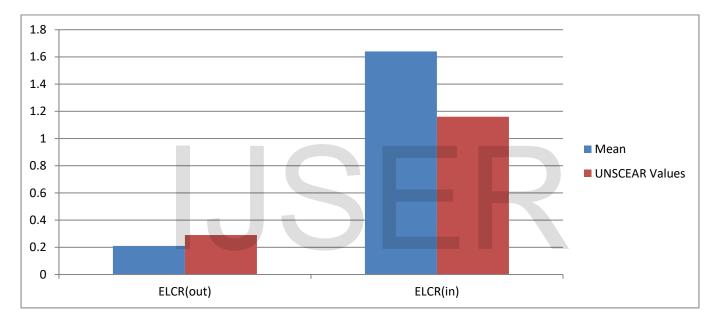


Fig 4. Showing the excess lifetime cancer risk compared to the world permissible UNSCEAR values

3.6 Annual Gonad Dose Equivalent (AGDE) (Bq kg⁻¹)

The annual gonads dose equivalent was considered as organs of interest in this research work because of its importance in cell formation. This is the measure of threat to sensitive cells from exposure to a particular level. This sensitive cells include the gonads surface cells and the bone marrow (UNSCEAR, 1993). The values of AGED was calculated using equation 8 and the values obtained are recorded in Table 2.

$$AGDE = 3.09C_{u} + 4.18C_{Th} + 0.314C_{k}$$
 8

Where C_u , C_{th} and C_k are the activity concentrations of Uranium, Thorium and Potassium respectively. The mean value is 365.94 Bq kg⁻¹, which is slightly higher than the world permissible (UNSCEAR) values

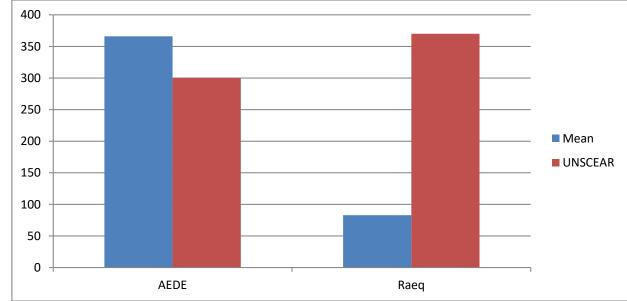
IJSER © 2019 http://www.ijser.org (UNSCEAR, 1993) value of 300.00 Bq kg⁻¹. The chat comparing the mean with the world permissible (UNSCEAR) value is shown in figure 5.

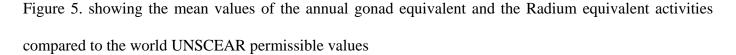
3.7 Radium Equivalent Activity (Raeq) (Bq kg⁻¹)

Due to the non – uniform distributions of radionuclides in the quarry sites, radium equivalent activity has been defined as a single radiological parameter that compares the specific activity of materials containing varying concentrations of ²³⁸U, ²³²Th and ⁴⁰K (Veiga et al., 2006). It is the sum of the weighted activities of ²³⁸U, ²³²Th and ⁴⁰K based on the estimation that 10.00 Bq kg⁻¹ of ²³⁸U, 7.00 Bq kg⁻¹ of ²³²Th and 130.00 Bq kg⁻¹ of ⁴⁰K produced the same gamma dose rate (SreshGandhi et al., 2014, Tripathi et al., 2013). In terms of the radiological health safety assessment, the maximum permissible limit of Raeq \leq 370.00 Bq kg⁻¹ had been set for materials to be used as component of building construction (Sivakumar et al., 2014, Beretka and Matthew, 2010). The formula used for the calculation is given in equation 9.

$$Ra_{eq} = \left[\frac{C_u}{370} + \frac{C_{th}}{259} + \frac{C_k}{4810} \right] \times 370$$

 C_u , C_{th} and C_k are the activity concentrations of Uranium, Thorium and Potassium respectively The values of R_{aeq} are recorded in table 2 and the chat comparing the mean with the world permissible (UNSCEAR) values (UNSCEAR, 1993) is shown in figure 5. The mean value is 82.97 Bq kg which is lower than the world permissible (UNSCEAR) values (UNSCEAR, 1993) of 370.00 Bq kg





4.0 Conclusions

The activity concentrations of naturally occurring radionuclides in samples from quarry sites in the south Eastern states of Nigeria were measured using NaI(Tl) gamma ray spectrometer. The activity concentrations of the radionuclides of ²³⁸U, ²³²Th and ⁴⁰K, were used to calculate the indoor and outdoor values of the followings: Absorbed Dose Rates, Annual Effective Dose Equivalent, Radiation Health Hazard Indices and the Excess Lifetime Cancer Risk. It was also used to calculate the values of the Annual Gonad Dose Equivalent and the Radium Equivalent Activity using standard analytical methods. All the outdoor values obtained in this research were lower than the world permissible (UNSCEAR) values, while the indoors values were higher than the world permissible (UNSCEAR) values. This shows that there is a very low radiation and cancer risk for the people living around the surroundings of the quarry sites. However, quarry materials are not safe for use as buildings because of the radiation and cancer risks for indoor exposures. This research could be used as a baseline data for future reference and research.

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