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Survey of Natural Radioactivity of Soil and Rock Samples and their Radiological Implications to Human Health in Ugwuele Quarry Mining Site, Uturu, Abia State, Nigeria

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ABSTRACT: This work measured the activity concentration of natural radionuclides 238U, 232Th and 40K in soil and rock samples collected in Ugwuele mining site and it surrounding communities in Uturu, Abia state, Nigeria. Fifteen different samples collected were analyzed. A NaI (Tl) spectrometry detector system was adopted for this research. The results obtained were used to estimate the radiological implication of the quarry mining in the study area. The average gamma activity obtained for 238U, 232Th and 40K in soil were 53.39 ± 3.08, 76.79 ± 9.66 and 1061.02 ± 15.72 Bq/kg respectively and 55.09 ± 5.71, 90.96 ± 16.10 and 1281.78 ± 6.38 Bq/kg respectively for rock samples. The calculated radiological hazard indices from Absorbed dose rate, Annual effective dose, Radium equivalent, internal hazard index and External hazard index gave 120.25 nGy/h, 0.147 msv/yr, 247.5 Bq/kg, 0.833 and 0.687 respectively. The average dose rate was higher than the recommended limit. Therefore, soil and rock materials found in Ugwuele could increase the radiological hazards on miners and residents of the area due to long time cumulative exposure.

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INTRODUCTION

Natural radioactivity has become a burning issue for most researchers, due to the health implication of natural occurring radioactive materials (NORMS) on man. Human are regularly exposed to natural radioactivity through divers sources as well as some special human activities. Among these human activities are mining of solid minerals and introduction of fertilizers to farmlands which have been associated with increase in natural radioactivity concentration of any location (Ibikunle *et al*, 2013). The continuous distortion of geological formation of the earth arrangement through the process of mining has in no small measure contributed to increase in radioactivity concentration in our environment especially around the mining area. (Echeweozo and Igwesi, 2021). In addition to that, most inorganic fertilizers used in farmlands, produced from Phosphate material have high concentration of naturally occurring radionuclides. Therefore, these farmland are expected to exhibit elevated level of radioactivity when compared with virgin farmlands (Fasasi *et al*., 1999). The radiological implication of elevated radioactivity concentration of natural occurring radioactive materials in mines and adjoin farmlands cannot be over emphasized due to associated risk to miners and the teaming farming population (Oladapo *et al.*, 2012). Although many studied have considered natural

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radioactivity in different mining sites and communities but for the first time, this study considered the radiological implication of the mining activities in Ugwuele mining site in Uturu, Abia State Nigeria and its impact on the adjoining farmlands and village quarters. The study also evaluated the radiological hazard indices of contiguous farmlands and village quarters with respect to the mining site.The knowledge of distribution pattern of these natural radionuclides is necessary for the determination the level of exposure of humans around these locations and estimating the associated health risk. Hence, the objective of this paper is to survey the natural radioactivity of soil and rock samples and their radiological implications to human health in Ugwuele Quarry Mining Site, Uturu, Abia State, Nigeria.

MATERIALS AND METHOD

Study Area: A cross sectional survey study approach was adopted in this work. Soil and rock samples were collected across the vicinity of Ugwuele quarry mining site and the host community in Uturu Isiukwuato L.G.A in Abia State which lies between latitude 5º 33'N and 5º 55'N to longitude 7º 22'E and 7º 42'E. The soil type in Ugwuele is characterized with false bedded sand stones of the Mastrichitan geological era with igneous outcrop (Anozie *et al* 1978). Three sites chosen for this study include Ugwuele quarry site, the village quarters (Ugwuele community) and the adjoining farmlands. The village quarters is about two kilometers from the mining site while the farmland is about one kilometer from the mining site. *Sample collection and preparation:* The choice of sampling locations were based on their proximity to the mining site and the determination to compare the rate radionuclide mobility within the mining site. Five soil samples were collected from each of the sampling area while four rock samples were collected from their bedrock in the quarry site. At each location soil samples were collected from approximately 1-2 meters apart using hand auger. Samples were obtained at depth of 5 cm from the top surface soil layer (Abaddy *et al*, 2014). Samples were collected on 13th June, 2023. 300 g of each of the soil samples were labeled and packaged in a plastic bag. Rock samples collected from different sections of the quarry pit were crushed and 300 g of each of the samples were also packaged and labeled. At the laboratory, soil and rock samples were placed in an oven at temperature of 60º for 24hours to remove residual moisture. The dried samples were pulverized into fine powder from 1mm mash size (AST M No. 18). Sample were homogenized with electric stirring device. 250 g of each of homogenized samples were then packed in a well-sealed 7 x 9 cm marinelli beaker and stored for 28days to attain radioactive equilibrium between radon 222 and radium 226 prior to spectroscopic measurement and analysis. (Avwiri, *et al*. 2013).

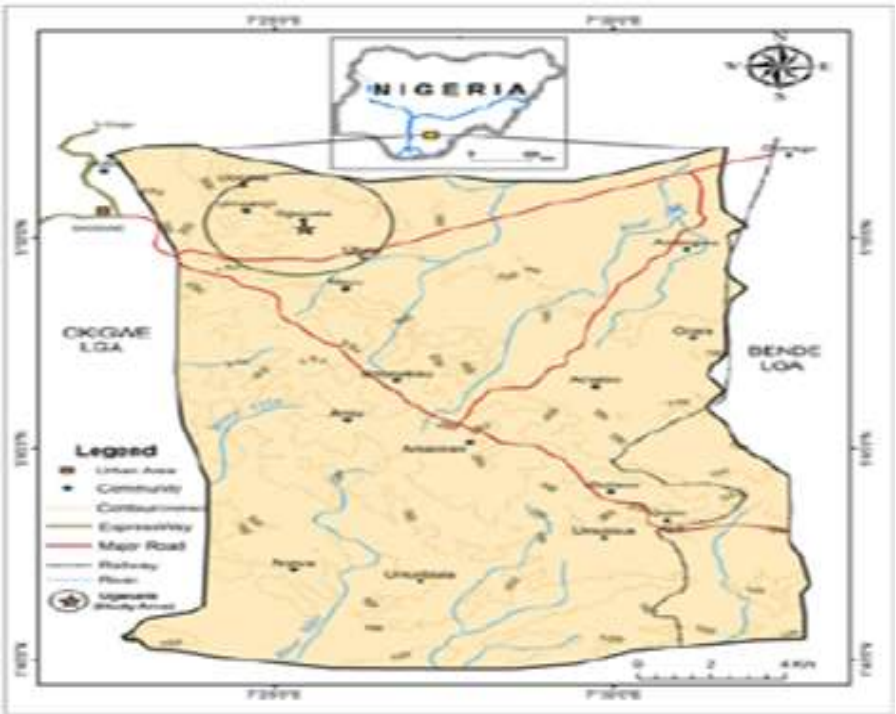

Fig 1: Map of Isiukwuato L.G.A of Abia State, Nigeria with Ugwuele community in circle (Source: Anozie *et al*, 1978)

The activity concentration of 238U, 232Th and 40K were measured with thallium scintillated 3cm x 3 cm Sodium Iodide (NaI(Ti) detector (model no 3M3/3) at the Center for Energy Research and Development, Obafemi Awolowo University , Ile-Ife. The detector was enclosed in a 5cm thick cylindrical lead shield to reduce the background radiations from possible natural radiation sources and to isolate it from other radiation sources used in the surroundings.

Energy and Efficiency calibrations: The detection energy calibration of the system was done with reference standard sources (IAEA-444) Prepared from the radiochemical center Amersham, England. The 1.460MeV photo peak was used for the measurement of 40K while the 1.120MeV photo peak from 214Bi and the 0.911MeV photo peak from 288Ac were used for the measurement of 238U and 232Th respectively. Each of the samples was counted for 36000 seconds (10 hours). The background spectrum distribution was determined by counting an empty container with the same geometry as the measured samples. Standard sources with known gamma energies and activity similar to the measured samples were used for energy calibration (see Table 2). These standard sources were prepared by Isotope product laboratories Burbank California, USA. The spectrometer system was adjusted to position the 662KeV photo peak of the 137Cs gamma line at about one third full scale and also to obtain the 0.5KeV per channel in order to obtain the highest resolution of the measuring system. The gain range and LLD of the ADC were positioned at 4K, 8K, 0.3V respectively while the amplifier coarse gain and the shaping time were positioned at 20, 0.5 and 4μs respectively. This settings were kept constant throughout the period of measurement.

Table 1: Channel for various energies of the Energy calibration sources

Standard source	Energy (KeV)	Channel
152Eu	344.27	284.28
137Cs	661.70	610.04
152Eu	778.89	734.89
	1112.02	1076.31
60C0	1173.24	1140.25
	1332.50	1305.56
152Eu	1407.95	1387.82

The absolute efficiency E_{abs} of a detector for each photo peak representing a particular photo energy of a standard source whose emission probability Pr was calculated with Equation 1 as shown in Table 2

$$E_{abs} = \frac{N(E_\gamma)}{A_t P_\gamma t_c} \qquad 1$$

Where $N(E_\gamma)$ is the peak area and t_c is the counting time. Results from the calculations are shown in Table 2

The dictation limit which is the level the true net count will be dictated above acceptable level (L_o) with a given probability when real activity is present was calculated with

$$LD = 2.706 + 4.653\delta_{NB} \qquad 2$$

Where δ_{NB} is the standard deviation of the number of counts when a blank sample is measured to determine the background count.

The minimum detectable amount of concentration is given by Equation 3

$$MDC = \frac{4.66 S_n}{\sum E_r P_r M} \qquad 3$$

Where S_n = the estimated standard error of the net count rate of the nuclide, n; Pγ = the absolute γ ray emission probability of nuclide n; $\sum E_r$ = the absolute efficiency; M = the mass of the sample in Kg

In order to determine background radiations around the detector, background measurement was counted for 10hours with no radiation source around the NaI(Ti) detector.

Measurements of radionuclide concentration in all samples: The measurement of radioactivity concentrations of all samples was carried out with the calibrated 3" x 3" NaI(Ti) detector with PMT base coupled to a Canberra series 10 plus multi-channel analyzer (MCA) which was linked to a preamplifier base. All samples were counted at a constant geometry and for a constant time of 10 hours. The counting technique applied in this work is described in (Echeweozo and Okeke 2021). The activity concentrations of radionuclides in each sample were obtained from total counts at peak emissions by subtracting background counts and divided by efficiency of the photopeak, gamma intensity of the radionuclide, mass of the sample and counting time.

The activity concentrations (C_i) of 40K, 238U and 232Th in Bq/kg were estimated using Equation 4

$$A_c = \frac{C_n}{P_\gamma M \epsilon} \qquad 4$$

Where A_c is the activity concentration of radionuclides in the samples in Bq/kg

C_n is the net count rate under the corresponding peak. P_γ is the absolute transition probability of specific ray. M is the mass of samples (kg) and $\in$ is the detector efficiency at a specific gamma energy. The total error in the determination of the specific activity depends on the error in determination of nuclide specific counting efficiency and the statistical counting errors error in determination of counting efficiency ranged from 1-4%.

Absorbed dose rate in air (D): When radiation passes through an object, part of the energy will be transferred to the material. These transferred radiations is referred to as absorbed dose. The permissible absorbed dose rate of 60 nGy/h was prescribed by UNSCEAR (2000). The mean activity concentration of 40K, 238U and 232Th in Bq/kg in the soil and rock sample were computer with Equation 5 (Abojassim *et al*, 2014).

$$D = 0.0417A_k + 0.462A_U + 0.621A_{Th} \quad 5$$

Where D is the absorbed dose rate in nGy/h and A_k, A_U and A_{Th} are the activity concentration of 40K, 238U and 232Th respectively.

Annual effective doses exposure (AEDEs): This measures the health risk to human beings due to radiation exposure. In this study the outdoor annual effective dose exposure was computer with Equation 6 (Ugbede, and Echeweozo 2017).

$$AEDE_{outdoor}(mSv) = D(nGyh^{-1}) x\ 8760\left(\frac{h}{y}\right) x\ 0.7\left(\frac{sv}{Gy}\right) 10^{-6}\ x\ 0.2 \quad 6$$

The worldwide range of the annual effective dose is 0.3 - 0.6 mSv with average of 0.48 mSv (UNSCEAR 2000).

Radium equivalent dose (R_{eq}): Radium equivalent dose gives the combined effects of specific activities of (226Ra, 232Th and 40K) in a measured sample. The published maximum admissible value is 370 Bq/Kg (UNSCEAR (2000). It was deduced with Equation 7.

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad 7$$

External and Internal Hazard Indices: External radiation exposure which is the total radiations dose due to the presence of 226Ra, 232Th and 40K is assessed by external hazard index, H_{ex}. Its level is calculated by Equation (8) (El-Taher *et al*, 2010)

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad 8$$

The internal exposure to radon and its progenies is evaluated with the internal hazard index, H_{in}, as shown in Equation (6).

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad 9$$

Internal and External hazard index which are the measure of hazardous effects of radon and its short-lived products to the respiratory organs in the body was evaluated with Equation (8 and 9).

RESULTS AND DISCUSSION

Radioactivity in samples: Table 3 represents the specific activity concentration of natural radionuclides in rock and soil samples from the studied area. The mean activity concentration of 40K, 238U and 232Th were found at 1061.02 ± 15.72, 53.39 ± 3.08 and 76.79 ± 9.61 Bq/kg respectively for all samples. These values were significantly higher than the world average values of 400, 35 and 30 for 40K, 238U and 232Th respectively. It can also be observed that the highest activity concentration of these radionuclides were observed from the quarry rock sample followed by farm land and the Village quarters. These large variation could be attributed to heavy quarry activities at the quarry site when compared to the passive farm land as shown in Figure 2. The elevated values of activity concentration observed in all sample were suspected to be influenced by geological nature of site, heavy quarrying activities and high usage of potassium fertilizer in the farmland region of the studied area.

Assessment of radiological hazard: The assessment of radiological hazard could be considered in different ways to estimate the radiation exposure dose and to assess the biological and radiological effect of these natural occurring radionuclides to humans and the environment. In this study, five radiological hazard indices considered are (i) Absorbed dose rate (D) in air, (ii) Annual effective dose equivalent (AEDE) (iii) Radium equivalent dose. (iv) External hazard index (H_{ex}) (v) Internal External hazard index (H_{in}) as shown in Table 5. The absorbed gamma dose rate in air due to the distribution of 40K, 238U and 232Th were computer and compared with recommended limit of 55nGy/h (UNSCEAR 2000). It was observed that the highest contributors to the total dose rates in the study area was from the rock samples from the quarry site. The mean absorbed dose rate in the air of the study area is 120.55nGy/h. it was also observed that this mean value was greater than acceptable dose level. The absorbed dose is a function of gamma rays (γ) emission and activity concentrations of natural occurring radionuclides. Therefore, the radiological hazard of mining activities on workers and residents of the study area is high.

Table 2: Characteristics of the energies of the nuclide in IAEA- 375 Source and Absolute efficiencies for varying photon energies of the nuclides in the sample standard source.

Nuclide	Energy (Eγ) Kev)	Emission probability (Pγ)	Half-life (T1/2) (Days)	Activity (Ao) (Bq)	N(Eγ)	A_t (Bq)	Absolute Efficiency (E_{abs})
	241.98	0.0712			517267		0.02313
	351.98	0.351			39631		0.00211
226 Ra	1120.29	0.147	584400	2000	104267	1993.97	0.00572
	1238.11	0.0578			204791		0.00034
	1764.49	0.151			142675		0.00015
232Th	969.11	0.1623	5.13×10^{12}	2500	12658	2497.54	0.00085
134Cs	795.85	0.854	754.2	4630	372152	379.37	0.00121
137Cs	661.66	0.850	109575	5280	413565	5216.61	0.00147
40K	1460.81	0.1067	4.66×10^{11}	4240	753842	4238.97	0.00023

Table 3: Specific activity concentration in rock and soil samples

S/N	Location	40K(Bq/kg)	238U(Bq/kg)	232Th(Bq/kg)
		Quarry Rocks		
1	Q_{s1}	1186.00 ± 5.86	47.27±3.11	82.90±14.93
2	Q_{s2}	1183.00 ± 6.06	75.23± 4.28	58.29±11.52
3	Q_{s3}	518.54 ± 3.80	72.20 ±4.08	16.17±6.14
4	Q_{s4}	777.29 ± 4.64	52.19± 3.31	31.07 ± 8.05
5	Q_{s5}	1189.00 ± 6.60	50.27±3.33	85.90±15.13
		Farm Land		
6	F_{s1}	820.19±4.75	34.27± 2.63	47.16± 10.25
7	F_{s2}	720.80± 14.60	45.80± 3.20	34.30±4.60
8	F_{s3}	500.00±13.90	38.50±4.00	38.70± 4.10
9	F_{s4}	749.50±15.10	31.70±4.20	43.80±4.00.
10	F_{s5}	811.30±15.11	33.80± 4.40	31.60± 5.00
		Village quarters		
11	V_{s1}	1554.52±75.25	77.06±4.35	102.77±17.68
12	V_{s2}	1445.00±20.40	51.70±1.60	121.00±3.40
13	V_{s3}	1544.10±16.80	60.60±1.30	171.60±24.50
14	V_{s4}	1470.60±17.20	79.80±1.00	147.20±6.00
15	V_{s5}	1455.40±16.30	50.60±1.40	139.40±8.90

Table 4: Average activity concentration of 40K, 238U and 232Th in the rock and soil samples

Location		40K(Bq/kg)	238U(Bq/kg)	232Th(Bq/kg)
Quarry rocks	Mean ± STD	1491.92 ±29.19	63.95 ± 1.93	136.39 ±12.09
	Range	1445- 1554	50- 79	102-171
Farm Land	Mean ± STD	720.36 ±12.69	36.81 ±3.69	39.11 ±5.59
	Range	500 - 820	31- 45	31-47
Village quarters	Mean ± STD	970.77 ± 5.28	59.43 ±3.62	54.87 ±11.15
	Range	518 -1189	47-75	16- 85
Total Mean		1061.02 ± 15.72	53.39 ± 3.08	76.79 ± 9.61
Worldwide range		140 -850	17 – 60	11-64
Worldwide Mean		400	35	30

Table 5: Radiological Hazard indices

Location	D(nGy/h)	AEDE(mSv/y)	H_{in}	H_{ex}	Ra_{eq}(Bq/kg)
Quarry rocks	174.55	0.214	1.1824	1.0096	373.86
Farm Land	70.88	0.086	0.4997	0.04003	119.85
Village	101.37	1.124	0.7349	0.4003	212.64
Mean	120.25	0..147	0.833	0.687	247.5
Permissible limit values	55	1	1	1	370

The annual effective dose equivalent from outdoor terrestrial gamma radiation for each site was estimated. The mean value of 0.147mSv/y was observed which is within the acceptable limit of less than 1mSv/y as specified by (ICRP 1994) From Fig 3The Radium equivalent dose from measured sample was estimated at a mean value of 247.50 Bq/kg. From the results, it could be seen that values for all studied area were lower than the permissible limit value of 370 Bq/kg (UNSCEAR 2000).

For internal hazard index and external hazard, elevated values were observed from the rock samples obtained from the quarry site. However, the mean values of internal hazard index and external hazard index gave 0.833 and 0.687 respectively. This mean values are below the permissible value of one or unity as recommended by ICRP (1999). It could be concluded that the study area does not constitute significant risk except at the quarry site.

Conclusion: 238U, 232Th and 40K gave 53.39 ± 3.08, 76.79 ± 9.66 and 1061.02 ± 15.72 Bq/kg respectively for soil samples and 55.09 ± 5.71, 90.96 ± 16.10 and 1281.78 ± 6.38 Bq/kg respectively for rock samples. The Absorbed dose rate, Annual effective dose, Radium equivalent, internal hazard index and External hazard index gave 120.25 nGy/h, 0.147 msv/yr, 247.5 Bq/kg, 0.833 and 0.687 respectively. The average dose rate was higher than the recommended limit. Therefore, rock materials found in Ugwuele could increase the radiological hazard on miners and residents.

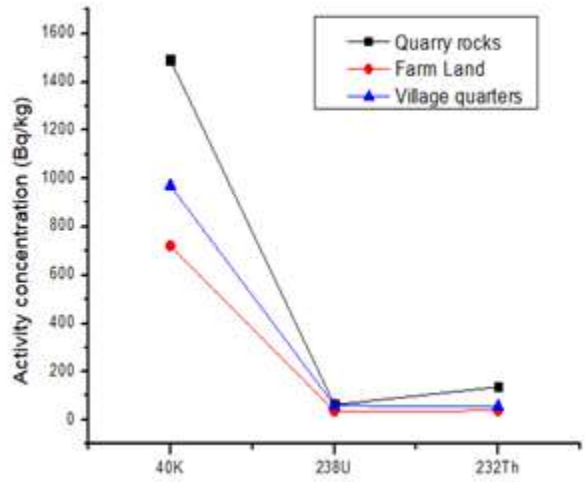

Fig. 2 Comparison Activity concentration of 40K, 238U and 232Th for all sample sites

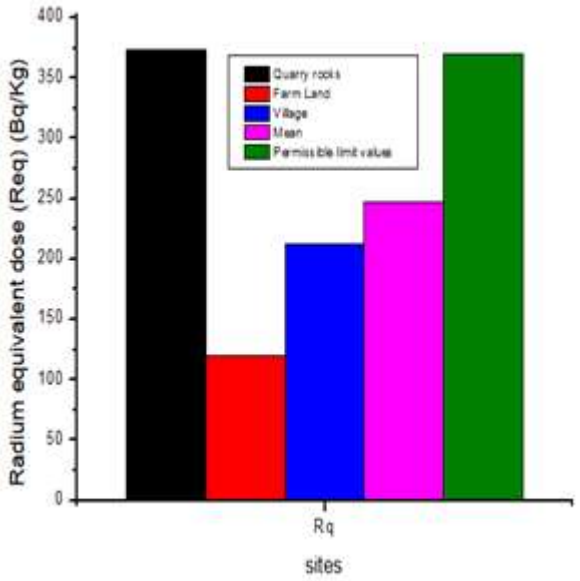

Fig. 3 Comparison of Radium equivalent dose (Ra_{eq}) for all sample sites

Conflicts of interest: There are no conflicts of interest.

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