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Assessment of Radioactivity Concentration Levels in Soil Samples Around Some Quarry Sites in Akamkpa, Cross River State, Nigeria

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Abstract

The presence of natural radionuclides in soil from quarry sites may present health risk potentials to the quarry workers and occupants of the quarry areas. This study is aimed to determine the activity concentration levels in soil around some quarry sites in Akamkpa, Cross River State, Nigeria. The study covers only Akamkpa, Cross River State and three quarry sites were selected at different locations. An estimation of radioactivity concentration levels in soil samples collected from the study area were carried out using Sodium Iodide (NaI) detector system at the National Institute of Radiation Protection and research, Ibadan, Nigeria thereby estimating the Absorbed Dose, Annual Effective Dose Rate, Gonadal Dose Equivalent, and Excessive Life Cancer Risk. The radioactivity concentration of ^{40}K, ^{232}Th, and ^{238}U at Fuaha Nigeria Limited (QMS –A) ranged from 480.00 ± 23.88 Bq/kg to 1575.09 ± 77.21 Bq/kgwith mean value of 1167.115 Bq/kg, 8.81 ± 1.10 Bq/kg to 47.83 ± 4.79 Bq/kg with mean value of 30.642 Bq/kg, and 3.96 ± 0.23 Bq/kg to 19.84 ± 1.14 Bq/kg with mean value of 12.237 Bq/kg respectively. At Xin-Xin Quarry Company (QMS-B) activity concentration ranged from 180.08 ± 9.05 Bq/kg to 1977.39 ± 96.40 Bq/kgwith mean value of 1194.113 Bq/kg, 20.36 ± 2.43 Bq/kg (B6) to 93.46 ± 8.50 Bq/kg with mean value of 49.475 Bq/kg, and 4.03 ± 0.23 Bq/kg to 17.95 ± 1.04 Bq/kg with mean value of 11.707 Bq/kg respectively. The radioactivity concentration in soil samples from the study area of Wings of Heaven Quarry Ltd. (QMS-C) were 492.29 ± 23.68 Bq/kg to 1632.49 ± 79.83 Bq/kg with mean value of 912.402 Bq/kg, 16.12 ± 1.93 Bq/kg to 48.23 ± 3.60Bq/kg with mean value of 35.965 Bq/kg, and 6.56 ± 0.37 Bq/kg to 22.32 ± 1.28 Bq/kg with mean value of 13.688 Bq/kg respectively. The absorbed doses for QMS –A, QMS-B and QMS-C were 72.830, 85.086 and 66.094nGy/hr which are respectively higher than the world average value of 50nGy/hr. The evaluated annual effective dose equivalents were 0.081mSv/y to 0.104mSv/y with a mean value of 0.091mSv/y and gonadal dose equivalents ranged from 479.125 µSv/y to 617.931 µSv/y, with a mean value of 543.141 µSv/y were found to be lower than the permissible level of 1 mSv/yr for public and 20 mSv/yr for radiation workers respectively. The estimated Excess Life Cancer Risk ranged from 0.284 x $10^{-3}$to 0.365 x $10^{-3}$with a mean value of 0.321x 10^{-3}. Residents and workers in and around Fuaha Nigeria Limited and Xin-Xin Quarry Company have higher chances of contracting cancer due to long term exposure to background radiation than those in Wings of Heaven Quarry Ltd.

Keywords: Mining, Quarrying, radiological hazard Indices, Public Health

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INTRODUCTION

Naturally occurring radionuclides in food, building supplies, air, and the ground, outer space and even our physical forms play a substantial role in the continuous and unavoidable exposure of human to ionizing radiation. These radionuclides are known as Naturally Occurring Radioactive Material, 'NORM'. The release of these radionuclides into the environment results in human exposure to external and internal radiation (EPA, 2019).

The commonest are the radioactive isotope of potassium 40K and the radionuclides originated from the decay of ^{238}U and ^{232}Th series, both widely distributed in soil and rocks of the earth's crust. Natural environmental radioactivity concentrations and the associated external exposure due to gamma radiation depend mainly on geological and geographical conditions and appear at different levels in soils of each region in the world (UNSCEAR, 2000b; Shittu *et al*., 2015).

The radioactivity concentration in soil give information on both natural and man – made sources which is important in radiological monitoring, assessment of radiation dose for public and also their ability to act as excellent biochemical and geochemical traces in the environment (Odeleye *et al*., 2019). Human actions can alter some exposures to natural radiation sources. For example, the release of natural radionuclides into the environment during mineral processing, the use of phosphate fertilizer processors, the combustion of fossil fuels, and quarry operations can increase exposure to natural radiation. Radiation exposure can result in harm and clinical manifestations, such as chromosomal changes, cancer induction, the production of free radicals, and bone necrosis (ICRP, 2002).

The human body is exposed to terrestrial radioisotopes such as thorium, uranium, and potassium through the food chain, mostly through food consumption. Through their roots, plants absorb these radionuclides, which then build up in their edible sections.
When these plants are processed and consumed, the accumulated radionuclides constitute internal radiation dose to humans (Ilemona *et al.,* 2016).

The two main naturally occurring radionuclides that should be of concern are ^{40}K and the decay products of ^{232}Th and ^{226}Ra. The main forms of decay for uranium and thorium are alpha and beta decay, which are difficult to find. Nonetheless, a large number of their daughter products have high gamma emissions. Since they can penetrate deeper than alpha or beta particles, gamma rays are most frequently employed to describe the natural radiation environment's terrestrial component. Therefore, the amounts of ^{232}Th and ^{238}U radioactive daughter products are estimated from their gamma ray emissions (Ibrahim *et al.,* 2013).

When radionuclides are released into the atmosphere, plants are the ones that are most likely to be contaminated by radiation and enter the food chain. Plants can become contaminated through two primary mechanisms: either by root uptake or direct aerial deposition of radioactive fallout on plants (Ezekiel, 2017). Also, the amount of radioactivity in soil is transferred in small quantities into plants (EPA (2010). It is necessary to carry out an accurate evaluation of these radionuclides in the soil in order to predict the degree of risk and harmful effects to public health (Mohannad & Khalil, 2014).

Absorbed dose is the amount of energy that is delivered from the mass per unit volume of ionizing radiation. 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, 2000b).

METHOD

Study Area

Akamkpa is a Local Government Area of Cross River State, Nigeria. , it lies between latitudes 4° 58' 11" N and 5° 46' 48" N, longitudes 8° 7' 11" E and 8° 54' 25" E (www.maphill.com), covering a land mass of 4586 square kilometers with a population of 149,705 at the 2006 Nigerian Census (https://citypopulation.de/en/nigeria/admin/cross_river/NGA009002__akamkpa/). It was divided into 260 villages arranged under 30 clans for political and administrative purposes. It is bordered to the west and south by the Odukpani and Akpabuyo Local Government Areas and to the north and west by the Biase and Yakurr Local Government Areas. To the north are the Local Government Areas of Obubra, Ikom, and Etung; to the east is the Republic of Cameroon.

Speaking Iyong-Iyong and Ejagham, there are two major ethnic groups in Akamkpa Local Government Area: the Dusauga Iyong-Iyong people and the Ejagham people. Christians make up the majority of the population, and both English and Efik are frequently utilised in social and business contexts.

Numerous agricultural estates have been created as a result of its rich agricultural past. In addition, there are abundant reserves of crude oil, and fishing is a well-liked economic activity. In addition to the availability of granite rocks, which have given rise to numerous quarrying firms, other solid minerals such as limestone, kaolin, and others are reportedly abundant but have not yet been fully utilized.
https://www.manpower.com.ng/places/lga/207/akamkpa

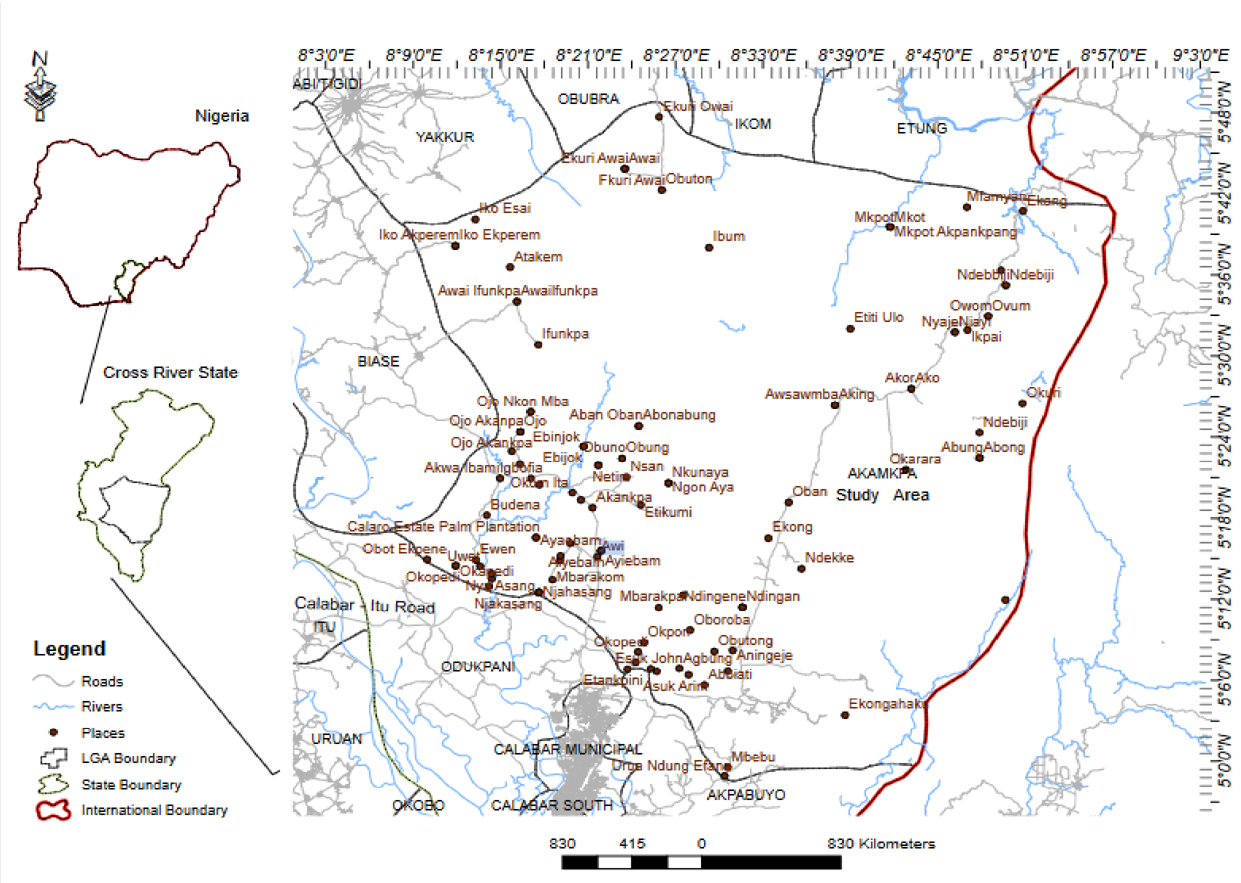

Figure 1: The geographical map of Akamkpa.

Sampling Techniques
Systematic random sampling approach was adopted for the sample gathering for this study. A defined random beginning point with periodic interval was chosen using this probability sampling technique.

Sample Collection
A total of eighteen (18) samples of soil were collected at selected points from three selected quarry sites in Akamkpa, Cross River State. These samples were collected systematically by mapping the respective coordinates from each sampling point in each quarry sites. Soil samples were collected at 500m apart from each sampling points using systematic sampling techniques to achieve statistical sensitivity of sampling and for accurate reference of result. A shovel was used to collect soil samples from a depth of about 10 cm. Each composite soil sample weighing about 500g of mass was collected and placed in a well labeled polythene bag and sealed to avoid cross contamination of the samples during transportation.

Sample Preparation
To eliminate moisture, the soil samples were allowed to air dry for seven days at room temperature. To achieve homogeneity of sample size, stony samples were first ground into a powder using a mortar and pestle and then sieved using a wire mesh with holes of 0.3 mm in diameter. The 400g soil samples were sealed in clearly labelled zip lock polythene bags and stored for 28 days in order to achieve secular equilibrium between ^{234}U, ^{232}Th, and ^{40}K and their offspring before taking it to the Secondary Standard Dosimetry Laboratory at the National Institute of Radiation Protection and Research, Ibadan for analysis.

Method of Data Analysis

Measurement of Absorbed Dose Rate
Absorbed Dose Rate (D) was determined using the equation:

$$\mathrm{D}\ (\mathrm{nGy}.hr^{-1}) = 0.462\mathrm{A_U} + 0.604\mathrm{A_{Th}} + 0.0417\mathrm{A_K} \quad (1)$$

Measurement of Annual Effective Dose Rate

Annual Effective Dose Rate (AEDR) was evaluated using the equation:

$$\mathrm{AEDR}(mSv.y^{-1}) = \mathrm{D}\ \times 8760\ \times 0.2 \times 0.7\ \times 10^{-6} \quad (2)$$

Measurement of Annual Gonadal Dose Equivalent
Annual Gonadal Dose Rate (AGDR) was calculated using the equation:

$$AGDR\left(\mu\frac{Sv}{y}\right) = 3.09\,AU + 4.18ATh + 0.314\,AK \quad (3)$$

Measurement of Excessive Life Cancer Risk
Excess Life Cancer Risk (ELCR) was determined using the equation:

$$ELCR = AEDR\ \times RF \times DL \quad (4)$$

RESULT AND DISCUSSION
The activity concentration of ^{40}K, ^{232}Th and ^{238}U for FUAHA Nigeria Limited (QMS –A), Xin-Xin Quarry Company (QMS-B) and Wings of Heaven Quarry Ltd. (QMS-C) were presented in Table 1, 2 and 3 respectively.

The estimated Radiological Hazards parameters of Soil Samples Collected from FUAHA Nigeria Limited (QMS –A), Xin-Xin Quarry Company (QMS-B) and Wings of Heaven

Quarry Ltd. (QMS-C) were presented in Table 4, 5 and 6 respectively. While the Comparison of the Present study and Previous Studies is presented in Table7 below.

Table 1: Activity concentration of ^{40}K, ^{232}Th and ^{238}U of the soil sample from FUAHA Nigeria Limited (QMS –A)

		Activity concentration (Bq/kg)		
S/N	CODE	K-40	Th-232	U-238
1	A1	949.50 ± 46.83	47.83 ± 4.79	11.43 ± 0.66
2	A2	1575.09 ± 77.21	12.37 ± 1.34	19.84 ± 1.14
3	A3	1374.54 ± 67.47	38.84 ± 4.18	3.96 ± 0.23
4	A4	1448.80 ± 70.98	35.35 ± 4.92	14.87 ± 0.86
5	A5	1174.76 ± 57.75	40.65 ± 4.67	8.84 ± 0.51
6	A6	480.00 ± 23.88	8.81 ± 1.10	14.48 ± 0.84
	Mean	**1167.115**	**30.642**	**12.237**

Table 2: Activity concentration of ^{40}K, ^{232}Th and ^{238}U of the soil sample from Xin-Xin Quarry Company (QMS-B)

		Activity concentration (Bq/kg)		
S/N	CODE	K-40	Th-232	U-238
1	B1	180.08 ± 9.05	46.71 ± 4.77	7.28 ± 0.42
2	B2	1859.20 ± 90.88	79.79 ± 7.55	17.95 ± 1.04
3	B3	1025.85 ± 50.77	27.29 ± 3.24	11.11 ± 0.64
4	B4	1977.39 ± 96.40	93.46 ± 8.50	13.72 ± 0.79
5	B5	1814.87 ± 88.63	29.24 ± 3.36	16.15 ± 0.93
6	B6	307.29 ± 15.34	20.36 ± 2.43	4.03 ± 0.23
	Mean	**1194.113**	**49.475**	**11.707**

Table 3: Activity concentration of ^{40}K, ^{232}Th and ^{238}U of the soil sample from Wings of Heaven Quarry Ltd. (QMS-C)

		Activity concentration (Bq/kg)		
S/N	CODE	K-40	Th-232	U-238
1	C1	533.09 ± 26.55	23.61 ± 2.77	11.87 ± 0.69
2	C2	492.29 ± 23.68	48.23 ± 3.60	13.77 ± 0.77
3	C3	1420.17 ± 69.65	38.22 ± 4.00	6.56 ± 0.38
4	C4	564.46 ± 28.06	42.09 ± 4.51	22.32 ± 1.28
5	C5	1632.49 ± 79.83	16.12 ± 1.93	20.93 ± 1.20
6	C6	831.91 ± 39.93	47.52 ± 3.53	6.68 ± 0.37
	Mean	**912.402**	**35.965**	**13.688**

Table 4: Calculated Radiological Hazards parameters of Soil Samples Collected from FUAHA Nigeria Limited (QMS –A)

S/N	D(nGy/h)	AEDE(mSv/yr)	AGDE(µSv/yr)	ELCR x 10^{-3}
1	73.764	0.090	533.391	0.317
2	82.319	0.101	607.590	0.353
3	82.607	0.101	606.193	0.355
4	88.636	0.109	648.635	0.380
5	77.624	0.095	566.107	0.333
6	32.027	0.039	232.289	0.137
Mean	**72.830**	**0.089**	**532.368**	**0.313**

Table 5: Estimated Radiological Hazards parameters of Soil Samples Collected from Xin-Xin Quarry Company (QMS-B)

S/N	D(nGy/h)	AEDE(mSv/yr)	AGDE(μSv/yr)	ELCR x 10^{-3}
1	39.086	0.048	274.288	0.168
2	134.015	0.164	972.777	0.575
3	64.394	0.079	470.519	0.276
4	145.246	0.178	1053.958	0.623
5	100.802	0.124	741.996	0.433
6	26.973	0.033	194.047	0.116
Mean	**85.086**	**0.104**	**617.931**	**0.365**

Table 6: Calculated Radiological Hazards parameters of Soil Samples Collected from Wings of Heaven Quarry Ltd. (QMS-C)

S/N	D(nGy/h)	AEDE(mSv/yr)	AGDE(μSv/yr)	ELCR x 10^{-3}
1	41.974	0.051	302.758	0.180
2	56.021	0.069	398.730	0.240
3	85.337	0.105	625.963	0.366
4	59.272	0.073	422.145	0.254
5	87.481	0.107	644.657	0.376
6	66.479	0.082	480.495	0.285
Mean	**66.094**	**0.081**	**479.125**	**0.284**

Table 7: Comparison of the Present study and Previous Studies

Reference	Activity Concentration (Bq/kg)			D(nGy/Hr)	AEDE(mSv/y)	ELCR x 10^{-3}
	K-40	Th-232	U-238			
Present study						
Fuaha (QMS –A)	1167.115	30.642	12.237	72.830	0.089	0.313
Xin-Xin (QMS-B)	1194.113	49.475	11.707	85.086	0.104	0.365
Wings (QMS-C)	912.402	35.964	13.688	66.094	0.081	0.284
Yusuf *et al.* (2022)	475.340	84.890	107.600	120.31	0.148	0.369
Mbet *et al.* (2019)	216.020	75.970	47.060	76.64	0.939	NA
Ogundele *et al.* (2020)	146.200	19.800	171.800	97.4000	0.500	0.004
UNSCEAR 2000b ICRP 103	420.00	45.000	33.000	60.000	1.000	0.290

The activity concentration of^{40}K, ^{232}Th, and ^{238}U in soil samples collected from the study area of Fuaha (QMS –A) ranged from 480.00 ± 23.88 Bq/kg to 1575.09 ± 77.21 Bq/kg with mean value of 1167.115 Bq/kg, 8.81 ± 1.10 Bq/kg to 47.83 ± 4.79 Bq/kg with mean value of 30.642 Bq/kg, and 3.96 ± 0.23 Bq/kg to 19.84 ± 1.14 Bq/kg with mean value of 12.237 Bq/kg respectively. The result obtained in the present study shows that the activity concentration of^{40}K is higher compare to similar study such as that of Ekeziel (2017), Mbet *et al.* (2019), Ogundele *et al*. (2020) and world average value of 412 Bq/kg, the activity concentration of ^{232}Th is lower compare to similar study such as that of Ekeziel (2017), Mbet *et al.* (2019) and world average value of 45Bq/kg but higher than that of Ogundele *et al* (2020), the activity concentration of ^{238}U is lower than that of Yusuf *et al.* (2022), Mbet*et al.* (2019), Ogundele *et al.* (2020) and world average value of 33Bq/Kg. At Xin-Xin (QMS-B) activity concentration ranged from 180.08 ± 9.05 Bq/kg to 1977.39 ± 96.40 Bq/kgwith mean value of 1194.113 Bq/kg, 20.36 ± 2.43 Bq/kg (B6) to 93.46 ± 8.50 Bq/kg with mean value of 49.475 Bq/kg, and 4.03 ± 0.23 Bq/kg to 17.95 ± 1.04 Bq/kg with mean value of 11.707 Bq/kg respectively. The result obtained in Xin-Xin (QMS-B) shows that the activity concentration of^{40}K is higher compare to similar study such as that of Yusuf *et al.* (2022), Mbet *et al.* (2019), Ogundele *et al.* (2020) and world average value of 412 Bq/kg. The activity concentration of ^{232}Th is lower compare to similar study such as that of Ezekiel (2017), Mbet *et al.* (2019) but higher than that of Ogundele *et al.* (2020) and world average value of 45Bq/kg. The activity concentration of ^{238}U is lower than that of Ezekiel (2017), Mbet *et al.* (2019), Ogundele *et al.* (2020) and world average value of 33Bq/Kg.The activity concentration of ^{40}K, ^{232}Th, and ^{238}U in soil samples collected from the Wings (QMS-C) ranged from 492.29 ± 23.68 Bq/kg to 1632.49 ± 79.83 Bq/kg with mean value of 912.402 Bq/kg, 16.12 ± 1.93 Bq/kg to 48.23 ± 3.60Bq/kg with mean value of 35.965 Bq/kg, and 6.56 ± 0.37 Bq/kg to 22.32 ± 1.28 Bq/kg with mean value of 13.688 Bq/kg respectively. Comparison of the results obtained in Wings (QMS-C) with published data from similar investigations in Nigeria shows that the activity concentration of ^{40}K is higher than that of Ezekiel (2017), Mbet *et al.* (2019), Ogundele *et al.* (2020) and world average value of 412 Bq/kg Shittu *et al.* (2015). The activity concentration of ^{232}Th is lower compare to that of Ezekiel (2017), Mbet *et al.* (2019) and world average value of 45Bq/kg but higher than that of Ogundele *et al.* (2020). The activity concentration of ^{238}U is lower than that of Ezekiel (2017), Mbet *et al.* (2019), Ogundele *et al.* (2020) and world average value of 33Bq/Kg Shittu *et al.* (2015).

Absorbed dose rate, Annual effective dose rate, Annual gonadal dose rate, and Excess life cancer risk were calculated from the activity concentrations of ^{238}U, ^{232}Th and ^{40}K measured from soil samples of Fuaha (QMS –A). The value of absorbed dose rate ranged from 32.027 nGy/h to 88.636 nGy/h, with a mean value of 72.830 nGy/h higher than world average of 60.00nGy/h (UNSCEAR 2020, Annex B). The value of Annual Effective Dose Rate ranged from 0.039 mSv/y to 0.109 mSv/y, with a mean value of 0.089 mSv/y lower than that of the International Commission on Radiation Protection (ICRP) which recommends the AEDE limit of 1 mSv/y for individual members of the public and 20 mSv/y for radiation workers. The value of annual gonadal dose equivalent ranged from 232.289 µSv/y to 648.635 µSv/y, with a mean value of 532.368 µSv/y. The value of excess life cancer risk ranged from 0.137 x 10^{-3} to 0.380 x 10^{-3}, with a mean value of 0.313 x 10^{-3} higher than the ICRP 103 recommended value of 0.290 x 10^{-3}. At the study area of Xin-Xin (QMS-B) the absorbed dose rate, Annual effective dose rate, Annual gonadal dose rate, and Excess life cancer risk were calculated from the activity concentrations of ^{238}U, ^{232}Th and ^{40}K measured from soil samples. The value of absorbed dose rate ranged from 26.973 nGy/h to 145.246nGy/h, with a mean value of 85.086 nGy/h higher than world average of 60.00nGy/h. The value of Annual Effective Dose Rate were 0.033 mSv/y to 0.178 mSv/y, with a mean value of 0.104 mSv/y lower than that of the International Commission on Radiation Protection (ICRP) which recommends the AEDE limit of 1 mSv/y for individual members of the public and 20 mSv/y for radiation workers. The value of Annual Gonadal Dose Equivalent ranged from 194.047 µSv/y to 1053.958 µSv/y, with a mean value of 617.931 µSv/y. The value of excess life cancer risk ranged from 0.116 x 10^{-3} to 0.623 x 10^{-3}, with a mean value of 0.365 x 10^{-3} higher than the ICRP 103 recommended value of 0.290 x 10^{-3}.Absorbed dose rate, Annual effective dose rate, Annual gonadal dose rate, and Excess life cancer risk were calculated from the activity concentrations of ^{238}U, ^{232}Th and ^{40}K measured from soil sample of Wings of Heaven Quarry Ltd. (QMS-C). The value of absorbed dose rate ranged from 41.974 nGy/h to 87.480 nGy/h, with a mean value of 66.094 nGy/h slightly higher than world average of 60.00nGy/h. The value Annual Effective Dose Rate ranged from 0.051mSv/y to 0.107 mSv/y, with a mean value of 0.081 mSv/y lower than that of the International Commission on Radiation Protection (ICRP) which recommends the AEDE limit of 1 mSv/y for individual members of the public and 20 mSv/y for radiation workers. The value of annual gonadal dose equivalent ranged from 302.758 µSv/y to 644.657 µSv/y, with a mean value of 479.125 µSv/y. The value of excess life cancer risk were 0.180 x 10^{-3}.to 0.376 x 10^{-3}., with a mean value of 0.284 x 10^{-3}.slightly lower than the ICRP 103 recommended value of 0.290 x 10^{-3}.

CONCLUSION

The study employed NaI(Tl) gamma ray spectrometry to analyse the activity concentrations of 238U, 232Th, and 40K in soil samples obtained from specific quarry sites in Akamkpa, Cross River State. The obtained results demonstrated that there was variation in the natural radionuclide distribution within the soil samples. With the exception of Xin-Xin (QMS-B), the study area's 40K activity concentration was higher than the global average, while the 238U activity concentration was lower than the global average. Since the assessed absorbed dose rate above the global average, the research area is often categorized as a High Background Radiation Area (HBRA) and is not appropriate for habitation. The yearly effective dosage equivalent and gonadal dose equivalent that were assessed were determined to be less than the annual maximum dose of 1 mSv for the general population and 20 mSv for radiation professionals. Because of the quarry operations, residents and workers in the study area are less likely to experience severe radiation deterministic consequences. Long-term background radiation exposure near Fuaha (QMS –A) and Xin-Xin (QMS-B) increases the risk of cancer in workers and the general public compared to workers at Wings (QMS-C).

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