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Radiological Hazard of Limestone at Tse-Kucha Mining and Processing Site, Gboko, Nigeria

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ABSTRACT

Background and Objective: Mining activities expose buried unstable radionuclides to the subsurface which releases harmful energy in form of radiation to the environment during decay. This study examined the radiological risk from radionuclides in limestone samples at the Tse-Kucha mining and processing site, Gboko, Nigeria and proffer radiation safety advice. **Materials and Methods:** The radiological assessment was carried out using radiation alert inspector Exp$^+$ for ambient radiation measurement, a Global Positioning System (GPS) for mapping sampled points and Sodium Iodide Thallium Activated [NaI(Tl)] detector for measuring activity concentrations and distribution patterns of the radioisotopes (^{40}K, ^{238}U and ^{232}Th). The study used Microsoft Excel and SPSS for radiological and Pearson correlation analysis. **Results:** Despite the high mean background radiation measurement of 2.445 mSv yr^{-1}, accepted limits of 339.34±18.01 Bq kg^{-1} for ^{40}K, 8.41±1.02 Bq kg^{-1} for ^{238}U and 10.99±0.69 Bq kg^{-1} for ^{232}Th were recorded. Similarly, the estimated radiation hazard parameters recorded mean concentrations within the UNSCEAR recommended values except excess lifetime cancer risk with 73.1E-5 against 29E-5. **Conclusion:** The study shows that the radionuclides are not evenly distributed in the limestone. The work also shows that continuous radiation exposure will enhance the tendency of suffering from cancer. As a result, the study recommends regular radiological studies of the area and the mandatory use of personal protective equipment when accessing the environment.

KEYWORDS

Specific activity concentration, background radiation, radium equivalent, outdoor radiation dose, annual effective dose equivalent, annual gonadal dose equivalent, excess lifetime cancer risk

INTRODUCTION

The activities of mining and processing of limestone uncover the subsurface, encapsulated naturally occurring radionuclides (^{238}U, ^{232}Th and ^{40}K) which are significant sources of radiation[1]. These associated natural gammas, beta and alpha-emitting radionuclides find their way into the air and other environmental components, thereby elevating the natural background radiation of the environment. Depending on the level of human exposure, radiation from these radionuclides can cause respiratory diseases, skin diseases[2], cataracts, cancer and other health challenges. As a result of these effects, a radiological study of environments with such activities is highly recommended, hence, this study.

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The West African Portland Cement Plc Ewekoro, located in Southwestern Nigeria where mining and processing of limestone is common, was evaluated unsafe with high activity concentrations of 904.42±39, 296.86±12 and 171.14±6 Bq kg^{-1} in limestone against 500, 50 and 50 Bq kg^{-1} for ^{40}K, ^{238}U and ^{232}Th, respectively[3]. The corresponding mean absorbed dose rate of 264.96 nGy h^{-1} or 2.444 against 1 mSv yr^{-1} in the air at 1 m above the ground was estimated from the study. In Kogi state, another high activity concentration of ^{40}K and ^{238}U radionuclides in limestone was reported from evaluating natural radionuclide contents in raw materials[4]. The study reported a mean activity concentration of 4694.0±366.0, 547.0±242.0 and 0.0±32.0 Bq kg^{-1} for ^{40}K, ^{238}U and ^{232}Th in limestone, respectively and a high absorbed dose rate of 3.797 mSv yr^{-1}. In 2010, a study was conducted on radionuclide pollutants in bedrocks (limestone and shale) and soils from the Ewekoro cement factory in Southwest Nigeria[5]. The assessment reported average specific activity concentrations of 35.86±7.06, 91.30±2.33 and 5.75±2.57 Bq kg^{-1} for ^{40}K, ^{238}U and ^{232}Th from the limestone bedrock type, respectively and an absorbed dose rate of 0.39 mSv yr^{-1}. In this study, only the activity concentration of ^{238}U was found to exceed the safe limit of 50 Bq kg^{-1} recommended by UNSCEAR[6]. Similarly, a radiological assessment of limestone samples from Sinai and Eastern desert in Egypt estimated high activity concentration of 212.41±0.64 Bq kg^{-1} for ^{238}U radionuclide[7].The ^{40}K and ^{232}Th activity concentrations in this study were observed to be within the UNSCEAR accepted limit, with activity concentration values of 151.16±0.10 and 22.97±0.20 Bq kg^{-1}, respectively. Furthermore, Malczewski and Żaba[8] evaluated the natural radioactivity in rocks of the Modane-Aussois (SE France) and reported safe activity concentrations of 18 Bq kg^{-1} for ^{40}K, 26 Bq kg^{-1} for ^{238}U and 0.7 Bq kg^{-1} for ^{232}Th in limestone. Kehinde *et al.*[9], also assessed the radionuclides in limestone at Ewekoro, South Western Nigeria in 2019 and reported high mean concentrations for ^{238}U and ^{232}Th. The study reported mean activity concentrations of 158.47±5.86, 121.30±14.80 and 112.25±6.73 Bq kg^{-1} for ^{40}K, ^{238}U and ^{232}Th, respectively. Annual effective dose equivalent from outdoor terrestrial gamma radiation of 0.166 mSv yr^{-1} was estimated from this study. Najam *et al.*[10] evaluated natural radioactivity levels of limestone rocks at eleven different locations in Northern Iraq. The study revealed high mean activity concentrations of 578.43 and 51.94 Bq kg^{-1} for ^{40}K and ^{238}U, respectively from Sadbakhma and a corresponding annual effective dose equivalent from outdoor gamma radiation of 0.244 mSv yr^{-1}. Natural radionuclides in rock and radiation exposure index from uranium mine sites in parts of Northern Nigeria were studied[11]. The study which did not report any significant radiation exposure to the workers and dwellers did not include Benue, the North Central part of Nigeria where the Tse-Kucha mining and processing site is situated.

At Yandev (near Tse-Kucha Mining and processing site), Gboko, Nigeria, a study of limestone elemental constituents and concentration revealed a harsh relationship between the limestone deposit and health[12]. Though the authors suggested proper management of wastes and particulate emissions by the company to attain environmental safety, the radiation hazard analysis of radionuclides (which act as a significant source of radiation) in limestone at the mining site was not evaluated. Another environmental study conducted around the mining site (study area) is soil analysis. It indicated that the soils are polluted with some oxides and heavy metals originating from limestone mining and cement production[13]. This study only reported the chemical pollution of the soil, the radiological risk from the radionuclides in the environment was not reported. At the same location, an appraisal of the social and health impact of the Dangote cement plant was carried out and severe health-related impacts of limestone mining and cement production on the populace were confirmed[14]. The study recommended measures to curb these effects but did not evaluate the radiation profile of the study area. Both Jibiri and Temaugee[15] and Olanrewaju and Avwiri[16] had previously assessed the radiological hazard from limestone at various locations. Jibiri and Temaugee[15] reported mean activity concentrations of 124.81 and 12.30 Bq kg^{-1} for ^{40}K and ^{232}Th, respectively, with 61.90 nGy h^{-1} absorbed dose rate and 0.0761 mSv yr^{-1} outdoor annual effective dose rate from limestone samples, while Olanrewaju and Avwiri[16] reported mean activity concentrations of 155.44, 11.81 and 12.43 Bq kg^{-1} for ^{40}K, ^{238}U and ^{232}Th, respectively, 15.59 nGy h^{-1} absorbed dose rate and 0.019 mSv yr^{-1} outdoor annual effective dose rate. Both assessments were carried out with only two samples each within the study area. It is also observed that the correlation analyses which reveal the relationship between the radionuclides and hazard indices were not reported in the studies.

In this current study, the radioactivity content of radionuclides in limestone and radiation hazard indices as well as the relationship between the radionuclides and hazard indices at the Tse-kucha mining and processing site, Gboko, Nigeria, has been determined along with appropriate recommendations.

MATERIALS AND METHODS

Study area: The study area is the Tse-kucha village limestone quarry site of Dangote Cement Company Gboko plant B. It is situated at kilometre 72 Makurdi, Gboko Road, on latitude 7°20 N and 7°30 N and longitude 8°56 E and 9°00 E within the Benue trough.

***In situ* measurements:** A radiation alert inspector Exp$^+$ (Serial No: 24650), a Global Positioning System (GPS) and a measuring tape were used to carry out the *in situ* radiation measurements.

The ambient radiations in mR/hr and geographical location at 10 different points within the study area were carefully taken using the radiation alert inspector Exp$^+$ and GPS, respectively.

Laboratory measurement

Sampling: From the administrative office, ten samples of limestone were collected weighing 500-1000 g in intervals of 100 m in the study area and stored in thoroughly rinsed black polythene bags to avoid contamination.

Sample preparation for laboratory analysis: The samples were dried and crushed separately into fine particles and stored in beakers for at least 30 days to attain secular equilibrium[1].

Laboratory analysis: Using Sodium Iodide Thallium activated [NaI(Tl)] detector, the activity concentrations of ^{40}K, ^{238}U and ^{232}Th in Bq kg^{-1} were carefully measured.

Computation of radiological risk parameters

Radium equivalent index: The radium equivalent index (Ra_{eq}) is an index for assessing activity concentrations and accounting for the radiation effects of radionuclide materials containing ^{238}U, ^{232}Th and ^{40}K. It is given by the Eq. 1[17,18]:

$$Ra_{eq} = A_u + 1.430A_{Th} + 0.0774A_K \quad (1)$$

External hazard index: It is denoted by (H_{ex}) and used to evaluate supplemental radiation hazards of natural gamma radiation. The H_{ex} is given by Tawfik *et al.*[18], in the Eq. 2:

$$H_{ex} = \frac{A_U}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad (2)$$

Internal hazard index: It is denoted by (H_{in}) and used to estimate radiation-based internal dangers such as asthma and other respiratory diseases. H_{in} is given by:

$$H_{in} = \frac{A_U}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad (3)$$

Outdoor absorbed dose rate (D_{out}): Outdoor absorbed dose rate (D_{out}) is a measure of the energy deposited in a medium by ionizing radiation per unit mass. D_{out}, measured in nGy h^{-1} is given by Qureshi *et al.*[19], in the Eq. 4:

$$D_{out} = 0.436A_u + 0.599A_{Th} + 0.042A_K \quad (4)$$

Indoor absorbed dose rate (D_{in}): D_{in} measured in nGy h^{-1}, is given by[19]:

$$D_{in} = 0.92A_u + 1.1A_{Th} + 0.081A_K \quad (5)$$

Annual effective dose equivalent of radiation based on absorbed dose rate of radiation in air (E): It estimates the average effective dose equivalent received by a person. The annual effective dose is divided into annual outdoor and annual indoor effective doses given as[20]:

$$E_{out} = D_{out} \times OF \times CF \times 10^{-6} \quad (6)$$

$$E_{in} = D_{in} \times OF \times CF \times 10^{-6} \quad (7)$$

Where, D_{out}= Outdoor absorbed dose rate, D_{in} = Indoor absorbed dose rate, OF = Occupancy factor/time of stay in the outdoor (20% of 8760 hrs = 1752 hrs) while the occupancy factor under the annual indoor effective dose is 80% of 8760 hrs = 7008 hrs and CF= Conversion factor (0.7 Sv Gy^{-1}).

Annual Gonadal Dose Equivalent (AGDE): Annual Gonadal Dose equivalent is a parameter used to monitor the radiation sensitivity of the reproductive organs such as the testis and ovaries. It also indicates the radiation dose level absorbed by the bone marrow. AGDE can be estimated using the Eq. 8[21]:

$$AGDE = 3.09A_U + 4.18A_{Th} + 0.314A_K \quad (8)$$

Excess Lifetime Cancer Risk (ELCR): The excess lifetime cancer risk from the ionizing gamma rays is computed using the equation by Sayed *et al.*[18]:

$$ELCR = Etotal \times DL \times RF \quad (9)$$

Where, E_{total} ($E_{in}+E_{out}$) is the annual effective Dose Equivalence, DL is the average duration of life estimated to be 70 yrs and RF is the risk factor (i.e. fatal cancer risk) given as 0.04 Sv^{-1} [$4\times10^{-5}(mSv)^{-1}$].

Pearson correlation coefficient analysis: The correlation coefficient between variables ranges from -1 to +1. A positive correlation indicates a direct relationship between variables, while a negative correlation indicates an inverse relationship between variables. A correlation coefficient of 0-0.49 indicates a weak relationship between the variables while a correlation coefficient of 0.5-1 indicates a strong relationship between the variables.

Note: Where, A_U, A_{Th} and A_K represent the specific activity concentrations of ^{238}U, ^{232}Th and ^{40}K, respectively.

RESULTS AND DISCUSSION

Figure 1 shows the area contour map which identifies the hilly/elevated areas with the higher radiation levels in the field represented by the closed loops which spread out through the low land areas with fewer radiation levels compared to the hilly areas. The sky blue colour represents the hilly/elevated areas with the highest radiation levels between 0.039 and 0.042 mR hr^{-1}, while the deep blue represents low land areas with the lowest radiation levels between 0.019 and 0.023 mR hr^{-1}. All radiation levels exceeded the 0.01 mR hr^{-1} UNSCEAR standard.

Table 1 presents the activity concentration, respective mean values and standard deviations of the radionuclides. The overall activity concentration measured in this study ranges from BDL (Below Detection Limit) to 813.05±42.27 Bq kg^{-1}. It ranges from 48.27±2.65-813.05±42.27 Bq kg^{-1} for ^{40}K, BDL to 38.09±4.29 Bq kg^{-1} for ^{238}U and 0.99±0.06-1.37±0.08 Bq kg^{-1} for ^{232}Th. The average activity concentration

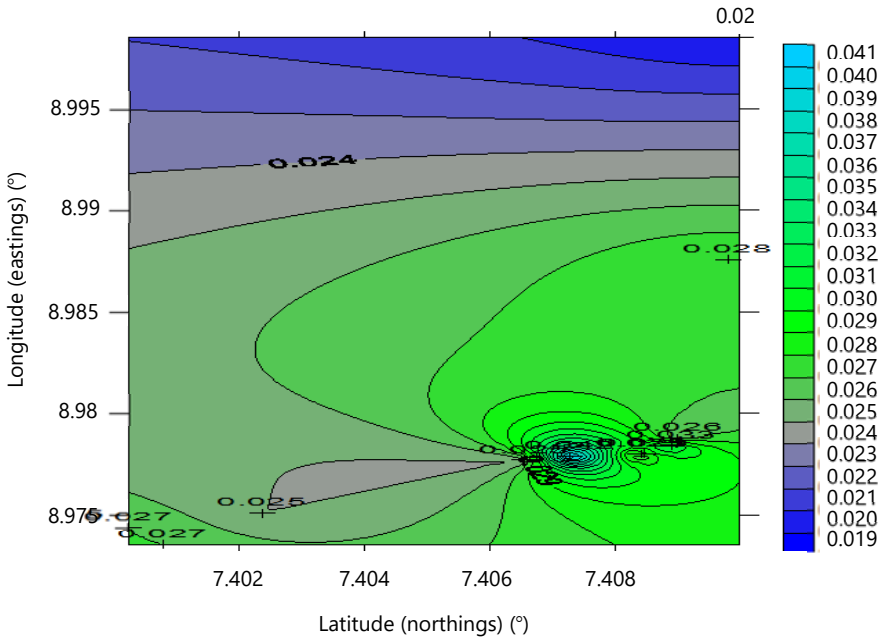

Fig. 1: Contour map of the study area

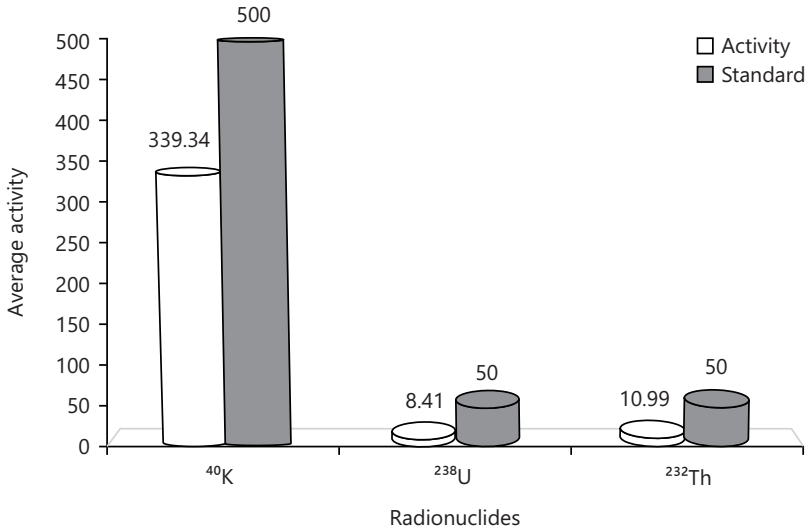

Fig. 2: Average activity concentration of the radionuclides with standard values

of ^{40}K, ^{238}U and ^{232}Th in The limestone samples were estimated to be 339.34±18.01, 8.41±1.02 and 10.99±0.69 Bq kg^{-1}, respectively. ^{40}K with the highest average concentration value is therefore the dominant radionuclide in the mineral. Figure 2 compares the average activity concentrations of ^{40}K, ^{238}U and ^{232}Th with their standard values. This analysis reveals that this present result is within the UNSCEAR recommended values.

Table 2 compares the mean activity concentrations of ^{40}K, ^{238}U and ^{232}Th of the present study with other studies around the world. The values in this study are observed to be less than the 904.42±39, 296.86±12 and 171.14±6 Bq kg^{-1} mean concentrations for ^{40}K, ^{238}U and ^{232}Th, respectively reported by Awodugba *et al.*[3] and also less than the 4694.0±366.0 and 547.0±242.0 Bq kg^{-1} mean concentrations for ^{40}K and ^{238}U, respectively but higher than 0.0±32.0 mean concentration for ^{232}Th obtained by Ajayi *et al.*[4]. The values obtained in this study are also seen to be less than the 212.41±0.64 and 22.97±0.20 Bq kg^{-1} mean concentration for ^{238}U and ^{232}Th, respectively but higher than 151.16±0.10 Bq kg^{-1} mean concentration for ^{40}K obtained by Fakeha *et al.*[7]. Furthermore, from the table, the mean concentration

Table 1: Activity concentration and average activity concentration (Bq kg^{-1}) of radionuclides

Samples	K^{-40}	U^{-238}	Th^{-232}
A-1000 m	266.74±14.41	3.68±0.50	19.36±1.21
B-900 m	351.25±19.13	8.86±1.25	9.57±0.62
C-800 m	48.27±2.65	BDL	0.99±0.06
D-700 m	60.77±3.27	0.70±0.09	1.37±0.08
E-600 m	813.50±42.27	38.09±4.29	22.96±1.42
F-500 m	71.31±3.81	3.58±0.34	1.83±0.11
G-400 m	804.08±42.50	12.30±1.65	19.74±1.24
H-300 m	368.85±19.94	BDL	3.45±0.23
I-200 m	110.09±5.78	1.08±0.13	3.47±0.21
J-100 m	498.55±26.35	15.85±1.99	27.20±1.68
Mean	339.34±18.01	8.41±1.02	10.99±0.69
S. deviation	2.80E+02	11.19	9.72

BDL: Below detection limit

Table 2: Comparison of mean activity concentration (Bq kg^{-1}) of Radionuclides in the present study and other studies around the world

Region	^{40}K	^{238}U	^{232}Th	References
Tse-Kucha, Nigeria	339.34±18.01	8.41±1.02	10.99±0.69	Present study
Kogi state, Nigeria	4694.0±366.0	547.0±242.0	0.0±32.0	Ajayi *et al.*[4]
Ewekoro, Nigeria	35.86±7.06	91.30±2.33	5.75±2.57	Gbadebo and Amos[5]
West African Portland Cement Plc Nigeria	904.42±39	296.86±12	171.14±6	Awodugba *et al.*[3]
Sinai and Eastern Desert, Egypt	151.16±0.10	212.41±0.64	22.97±0.20	Fakeha and Hamidalddin[7]
Modane-Aussois, France	18.00	26.00	0.7	Malczewski and Zaba[8]
Ewekoro SW Nigeria	158.47±5.86	121.30±14.80	112.25±6.73	Oyeyemi *et al.*[9]
Sadbakhma Northern Iraq	578.43	51.94	-	Najam *et al.*[10]
Tse-Kucha Gboko (2013)	124.81	-	12.30	Jibiri and Temaugee[15]
Tse-Kucha Gboko (2017)	155.44	11.81	12.43	Olanrewaju and Avwiri[16]
UNSCEAR standard	500.00	50.00	50.00	Mbonu and Ben[6] Qureshi *et al.*[19]

values of this study are higher than the 35.86±7.06 and 5.75±2.57 Bq kg^{-1} for ^{40}K and ^{232}Th mean concentrations, respectively but less than 91.30±2.33 Bq kg^{-1} for ^{238}U from a study of radionuclide pollutants in bedrocks from Southwest Nigeria[5] and also higher than 18 and 0.7 Bq kg^{-1} for ^{40}K and ^{232}Th mean activity concentrations, respectively but less than 26 Bq kg^{-1} for ^{238}U from a study of natural radioactivity in rocks of the Modane-Aussois (SE France)[8]. The table also shows the comparison of the mean concentration of the present study to be much lower than 121.30±14.80 and 112.25±6.73 Bq kg^{-1} values for ^{238}U and ^{232}Th, respectively estimated by Oyeyemi *et al.*[9], but higher than 158.47±5.86 Bq kg^{-1} for ^{40}K radionuclide. The mean concentration values from this study are also seen to be much lower than 578.43 and 51.94 Bq kg^{-1} for ^{40}K and ^{238}U concentration values from Najam *et al.*[10]. Similarly, the mean concentrations from this study are seen to be lower than the mean concentration value of 12.30 Bq kg^{-1} for ^{232}Th but higher than the value of 124.81 Bq kg^{-1} for ^{40}K obtained by Jibiri and Tamaugee[15] and lower than the 155.44, 11.81 and 12.43 Bq kg^{-1} for ^{40}K, ^{238}U and ^{232}Th, respectively reported by Olanrewaju and Avwiri[16]. The table also shows that the values are higher than the recommended values for ^{238}U (except Malczewski and Żaba[8] and Olanrewaju and Avwiri[16]), but within acceptable limits for ^{232}Th (except Awodugba *et al.*[3] and Oyeyemi *et al.*[9]) and ^{40}K (except Awodugba *et al.*[3], Ajayi *et al.*[4] and Najam *et al.*[10]).

Table 3 presents the estimated radiation hazard parameters with their mean concentrations, standard deviations, world average and UNSCEAR[6] recommended values. It shows that the evaluated hazard indices range from 5.13±0.29-133.56±9.58 Bq kg^{-1} with a mean value of 50.26±3.39 Bq kg^{-1} for radium equivalent activity, 0.01±0.00-0.36±0.03 Bq kg^{-1} with a mean value of 0.14±0.01 Bq kg^{-1} for external hazard indices and 0.01±0.00-0.46±0.04 Bq kg^{-1} with a mean value of 0.16±0.01 Bq kg^{-1} for internal hazard indices. These values are much lower than the standard value of 370 Bq kg^{-1} for radium equivalent activity and less than unity (1) for both external and internal hazard indices. The Table also shows the estimated values ranging from 2.62±0.15-64.53±4.50 nGy h^{-1} with average value of 24.51±1.62 nGy h^{-1} for outdoor absorbed dose rate, 5.00±0.28-126.19±8.93 nGy h^{-1} with an average value of 47.32±3.16 nGy h^{-1} for indoor absorbed dose rate, 0.00±0.00-0.08±0.01 mSv yr^{-1} with an average value

Table 3: Estimated radiation hazard parameters in limestone and their worldwide average limits and recommended values

Samples	Raeq	H-ex	H-in	D-out	D-in	E-out	E-in	E-Total	AGDE	ELCRXE-5
A-1000 m	51.90±3.34	0.14±0.01	0.15±0.01	24.40±1.55	46.29±2.96	0.03±0.00	0.23±0.00	0.26±0.02	0.176	72.8
B-900 m	49.59±3.61	0.13±0.01	0.16±0.01	24.35±1.72	47.13±3.38	0.03±0.00	0.23±0.02	0.26±0.02	0.178	72.8
C-800 m	5.13±0.29	0.01±0.00	0.01±0.00	2.62±0.15	5.00±0.28	0.00±0.00	0.02±0.00	0.02±0.00	0.019	5.6
D-700 m	7.34±0.46	0.02±0.00	0.02±0.00	3.68±0.22	7.07±0.44	0.00±0.00	0.03±0.00	0.03±0.00	0.027	8.4
E-600 m	133.56±9.58	0.36±0.03	0.46±0.04	64.53±4.50	126.19±8.93	0.08±0.01	0.62±0.00	0.71±0.01	0.469	198.8
F-500 m	11.69±0.79	0.03±0.00	0.04±0.00	5.65±0.37	11.08±0.74	0.01±0.00	0.05±0.00	0.05±0.00	0.041	14
G-400 m	102.44±6.70	0.28±0.02	0.31±0.02	50.96±3.25	98.16±6.32	0.06±0.00	0.48±0.03	0.54±0.03	0.373	151.2
H-300 m	33.33±1.86	0.09±0.01	0.09±0.01	17.56±0.98	33.67±1.87	0.02±0.00	0.17±0.01	0.19±0.01	0.13	53.2
I-200 m	14.52±0.88	0.04±0.00	0.04±0.03	7.17±0.43	13.73±0.82	0.01±0.00	0.07±0.00	0.08±0.00	0.052	22.4
J-100 m	93.13±6.42	0.25±0.02	0.29±0.02	44.14±2.98	84.88±5.81	0.05±0.00	0.42±0.03	0.47±0.03	0.319	131.6
Mean	50.26±3.39	0.14±0.01	0.16±0.01	24.51±1.62	47.32±3.16	0.029±0.00	0.231±0.01	0.251±0.01	0.032	73.1
SD	42.93	0.12	0.14	20.75	40.32			0.228	0.21	9.69E+11
UNSCEAR Standard	370.00	1.00	1.00	59.00	84.00	0.07	0.41	1.00	0.30	29.0

SD: Standard derivation

Table 4: Comparing mean annual effective dose equivalent (mSv yr^{-1}) of radiation of the present study with other studies around the world

Region	Mean AEDE	References
Tse-Kucha, Nigeria	0.251±0.01	Present study
West African Portland Cement Plc Nigeria	2.444	Awodugba *et al.*[3]
Obajana, Kogi State, Nigeria	3.797	Ajayi *et al.*[4]
Southwest Nigeria	0.390	Gbadebo and Amos[5]
Ewekoro SW Nigeria	0.166	Oyeyemi *et al.*[9]
Sadbakhma, Northern Iraq	0.240	Najam *et al.*[10]
Baathra, Northern Iraq	0.270	Najam *et al.*[10]
Tse-Kucha, Nigeria (2013)	0.076	Jibiri and Temaugee[15]
Tse-Kucha, Nigeria (2017)	0.019	Olanrewaju and Avwiri[16]
Minia, Egypt	0.290	Elsaman *et al.*[20]

of 0.029±0.00 mSv yr^{-1} for outdoor annual effective dose equivalent, 0.02±0.00-0.62±0.00 mSv yr^{-1} with an average value of 0.231±0.01 mSv yr^{-1} for indoor annual effective dose equivalent and 0.02±0.00-0.71±0.01 mSv yr^{-1} with an average value of 0.251±0.01 mSv yr^{-1} for total annual effective dose equivalent (E_{out}+E_{in}). Similarly, these average values are within the worldwide average limit of 59 and 84 nGy h^{-1} for outdoor and indoor absorbed dose rates, respectively, 0.07 and 0.41 mSv yr^{-1} for outdoor and indoor annual effective dose equivalent and 0.48 mSv yr^{-1} for total annual effective dose equivalent[3]. Annual Gonadal Dose Equivalent (AGDE) and Excess Lifetime Cancer Risk (ELCR) are also represented in the Table with values ranging from 0.019-0.469 mSv yr^{-1} with an average value of 0.032 mSv yr^{-1} for annual gonadal dose equivalent and 5.6E-5-198.8E-5 with an average value of 73.1E-5 for excess lifetime cancer risk. The Table shows that the average annual gonadal dose equivalent from this study is within the worldwide average limit of 0.3 mSv yr^{-1} while the average value of excess lifetime cancer risk exceeds the accepted limit of 29E-5[6], implying that long-term exposure to radiation from the study area will enhance the danger of suffering cancer. The mean excess lifetime cancer risk from the present study was compared with the reported mean value from Olanrewaju and Avwiri[16], who assessed the hazard index in limestone using only the outdoor annual effective dose equivalent from the same study area. The analysis shows that excess lifetime cancer risk from this study exceeds the previous study (6.7E-5) by over 90%.

Table 4 presents the comparison of the mean annual effective dose equivalent of the present study with other studies around the world. It is seen that the present study reports the least dose values except for Ewekoro, SW Nigeria[9], Sadbakhma, Northern Iraq[10], Tse-Kucha, Nigeria (2013)[15] and Tse-Kucha, Nigeria (2017)[16]. It is far less than 2.444 and 3.797 mSv yr^{-1} measured at West African Portland Cement Plc, Nigeria[3] and Obajana, Kogi State, Nigeria[4], respectively but slightly less than 0.270, 0.290 and 0.390 mSv yr^{-1}, measured at Baathra, Northern Iraq[10], Minia, Egypt[20] and Southwest Nigeria[5], respectively.

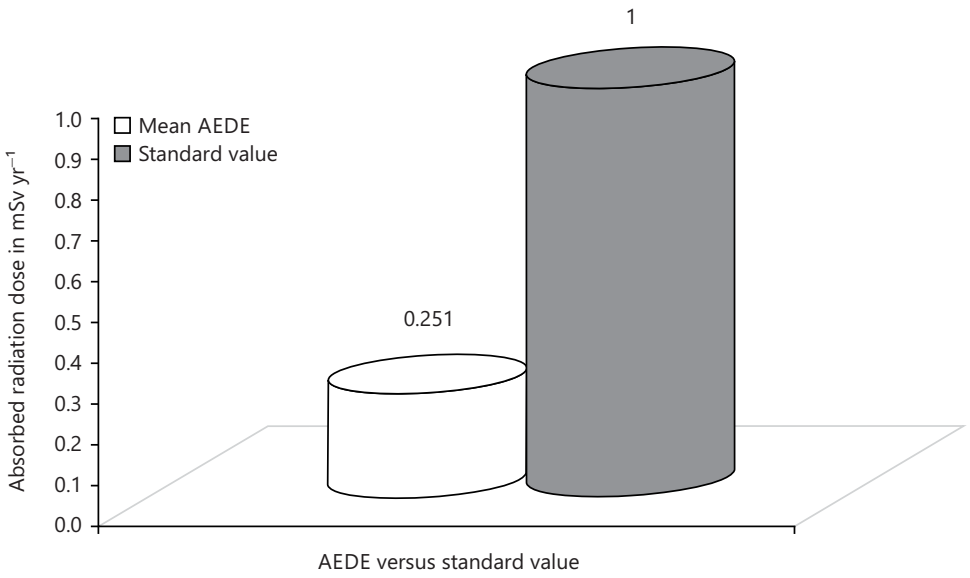

Fig. 3: Comparing mean AEDE with the standard value

Table 5: Pearson correlation coefficient among the estimated quantities

	^{40}K	^{238}U	^{232}Th	R_{aeq}	H_{ex}	H_{in}	D_{out}	D_{in}	E_{out}	E_{in}	E_{total}	AGDE	ELCR
^{40}K	1.000												
^{238}U	0.802	1.000											
^{232}Th	0.793	0.727	1.000										
R_{aeq}	0.960	0.892	0.905	1.000									
H_{ex}	0.962	0.889	0.905	1.000	1.000								
H_{in}	0.947	0.929	0.884	0.996	0.995	1.000							
D_{out}	0.969	0.887	0.894	0.999	1.000	0.994	1.000						
D_{in}	0.969	0.892	0.890	0.999	0.999	0.996	1.000	1.000					
E_{out}	0.963	0.898	0.882	0.995	0.995	0.994	0.996	0.996	1.000				
E_{in}	0.969	0.890	0.892	0.999	0.999	0.995	1.000	1.000	0.996	1.000			
E_{total}	0.968	0.893	0.890	0.999	0.999	0.996	1.000	1.000	0.996	1.000	1.000		
AGDE	0.971	0.885	0.891	0.999	0.999	0.994	1.000	1.000	0.996	1.000	1.000	1.000	
ELCR	0.968	0.893	0.890	0.999	0.999	0.996	1.000	1.000	0.996	1.000	1.000	1.000	1.000

However, the mean total annual effective dose equivalent from this study is above the 0.24 and 0.166 mSv yr^{-1} values from Sadbakhma, Northern Iraq[10] and Ewekoro, SW Nigeria[9], respectively but far greater than the 0.076 and 0.019 mSv yr^{-1} obtained previously from Tse-Kucha, Nigeria (2013)[15] and Tse-Kucha, Nigeria (2017)[16], respectively. This variation of annual effective dose equivalent results from Tse-Kucha, Nigeria, is because these previous studies evaluated only the outdoor annual effective dose equivalent while the present study evaluated the total annual effective dose equivalent (i.e., sum of outdoor and indoor annual effective dose equivalent). This change is also associated with the difference in the number of samples considered for the analysis. Figure 3 compares the total Annual Effective Dose Equivalent (AEDE) from the present study with the UNSCEAR accepted limit of 1 mSv yr^{-1} for the general populace. It shows that the mean value of the total annual effective dose equivalent estimated from this study is within safe limits.

Table 5 presents the pearson correlation coefficients among the estimated quantities. The Table shows a strong, positive correlation between any two quantities estimated with the least coefficient of +0.727, a value obtained after the correlation analysis between ^{232}Th and ^{238}U. The highest correlation coefficient from this analysis has the maximum value of +1. This value is commonly seen from the analysis of the same parameter. From the Table, the outdoor absorbed dose rate (D_{out}) with the highest correlation coefficient of +1 followed by indoor absorbed dose rate (D_{in}), indoor annual effective dose equivalent (E_{in}),

total annual effective dose equivalent (E_{total}), Annual Gonadal Dose Equivalent (AGDE) and Excess Lifetime Cancer Risk (ELCR), shows that it has a better relationship among estimated parameters than any other hazard parameter. Figure 4-6 show the correlation plot between the radionuclides. From the correlation plots, a closer interrelationship between ^{40}K and ^{238}U radionuclides was revealed in this study.

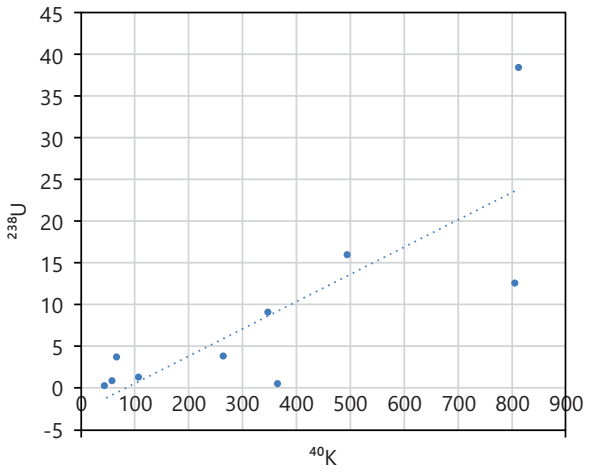

Fig. 4: Correlation plot between ^{40}K and ^{238}U

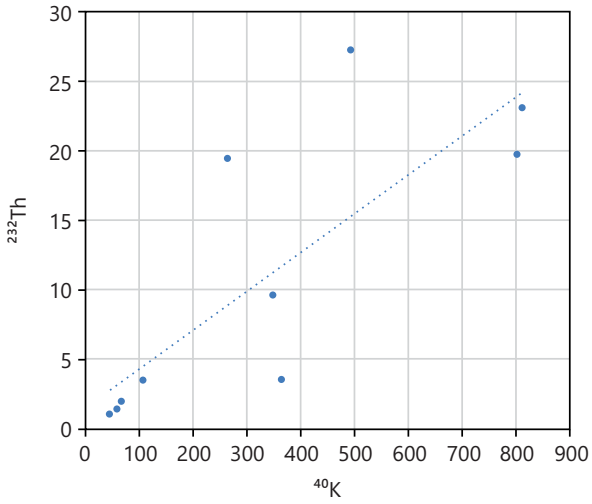

Fig. 5: Correlation plot between ^{40}K and ^{232}Th

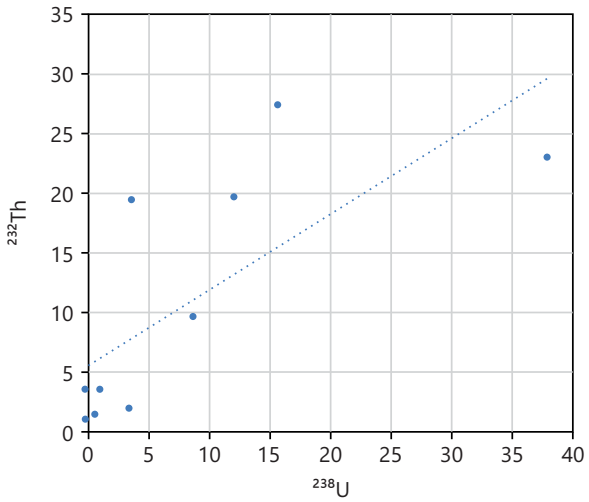

Fig. 6: Correlation plot between ^{238}U and ^{232}Th

CONCLUSION

The assessment of the radiological hazard of limestone at the Tse-Kucha mining and processing site in Gboko, Benue state, shows that the radionuclides in limestone at the study area are not evenly distributed. The average activity concentrations of the radionuclides and radiation hazard indices are all within the world's average limits except for the Excess Lifetime Cancer Risk (ELCR), implying that long-term exposure to radiation from the environment will enhance the tendency of suffering from cancer. Furthermore, Pearson correlation analysis of the study revealed a strong positive correlation between any two parameters. We, therefore, recommend the use of Personal Protective Equipment (PPE) for the occupational workers and the general populace assessing the quarry site as well as regular radiological evaluation of the environment.

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