Open Access Maced J Med Sci electronic publication ahead of print,
published on October 10, 2019 as https://doi.org/10.3889/oamjms.2019.661

ID Design Press, Skopje, Republic of Macedonia
Open Access Macedonian Journal of Medical Sciences.
https://doi.org/10.3889/oamjms.2019.661
eISSN: 1857-9655
Public Health

Radiogenic Components of Limestone Samples Collected from Ewekoro SW Nigeria: Implications for Public Radiological Health Risks Assessment and Monitoring

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Citation: Oyeyemi KD, Aizebeokhai AP, Ekhaguere OA, Chinwuba DE, Onumejor CA. Radiogenic Components of Limestone Samples Collected from Ewekoro SW Nigeria: Implications for Public Radiological Health Risks Assessment and Monitoring. Open Access Maced J Med Sci. https://doi.org/10.3889/oamjms.2019.661

Keywords: Medical geology; Public health assessment; Radiogenic composition; Natural radioactivity; Ewekoro limestone; Cancer risks

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Received: 05-May-2019; **Revised:** 01-Sep-2019; **Accepted:** 02-Sep-2019; **Online first:** 10-Oct-2019

Funding: This research did not receive any financial support

Competing Interests: The authors have declared that no competing interests exist

Abstract

AIM: This research presents the radiogenic components in thirteen limestone samples from a quarry site in Ewekoro, southwestern Nigeria.

METHODS: The distributions of natural radionuclides (^{238}U, ^{232}Th and ^{40}K) in the limestone samples were determined by gamma spectroscopy using a well-type thallium-doped sodium iodide detector. Also, estimated associated radiological hazards are presented and compared with the standard threshold values.

RESULTS: The activity concentrations for ^{238}U, ^{232}Th and ^{40}K radionuclides range 18.09 ± 3.43-239.50 ± 25.74 Bqkg-1, 8.33 ± 0.83 - 360.01 ± 21.33 Bqkg-1 and 11.28 ± 0.81-735.26 ± 0.95 Bqkg-1 respectively. The radium equivalent activity concentration in the samples ranges 58.857-758.832 Bqkg-1 with samples S3, S4 and S11 values higher than the threshold limit of 370 Bqkg-1. Estimated dose rate and annual effective dose rate (AEDE) from the samples have ranges 28.754-330.917 nGyh-1 and 35.26-405.84 μSvy−1 respectively greater than the standard limit of 59 nGyh-1 and 70 μSvy−1 respectively for all samples except S9. The estimated external and internal indices are ranging 0.16 – 2.05 and 0.21 – 2.68 respectively, greater than permissible unity in some limestone samples such as S3, S4, S8, S11 and S13. Excess lifetime cancer risk was also computed using a life expectancy of 54.5 years. The results of higher radiological parameters in the limestone samples revealed that the miners have a high probability of contracting induced cancer.

CONCLUSION: A regular check-up is recommended for the miners and staffs within the quarry site. Also, the residents within the environs should be relocated far away from the quarry site, as the particulates from the limestone rock blasting could contaminate the air in the study area.

Introduction

The exploration of the mineral is of economic importance to any nation as it contributes greatly to the nation's wealth. Despite the great and remarkable contributions of mineral prospecting towards the economic growth and advancement, environmental pollution impacts of these exploration activities are of serious concern all over the world. An aspect of geology that is concerned with the detailed understanding of several health implications of geological factors such as the occurrence of mineral deposit (including its exploration and exploitation) on humans, animals and plants is term medical geology [1], [2], [3]. The utmost significance of this area of geology is from the fact that rocks and minerals constitute fundamental building blocks of the planet, and as such, they contain several natural occurring chemical elements that are essential to plant, animals and human in considerable small doses. Hazardous contaminations of these elements in nature are also inimical to human health, since these elements are taken into human body through food, water and air [4], [5], [6], [7], [8]. Weathering of rocks form the soils on which crops and animals are raised. Drinking water percolates through soils and rocks as part of the hydrological cycle. The elements can also be inhaled through atmospheric dusts and gases.

Exposure to mineral dusts in quarry sites can affect the health of the miners and the inhabitants of the community where the minerals are being exploited or utilized [9]. Several human carcinogens have been reportedly associated with mine workers [10], [11], [12]. Huang et al. (2006) reviewed several pneumoconiosis cases among coal worker.

Pulmonary talcosis, silicosis and siderosis associated with inhalation of talc (from kaolinite), silicates and iron oxides respectively. Davies (2010) identified some potential harmful elements (PHEs) from mining operations in Africa, among which are arsenic associated with African Precambrian greenstone belts, mercury associated with the gold mining and Radiation (and radon gas) from radioactive gas formed naturally by the radioactive decay of uranium that occurs in all rocks and soils. Although, there is no place in the world that is free from radiation, natural environmental radioactivity largely depends on the geological rock types and mineral deposits in an area. Several investigations have been carried out on the natural radioactivity levels and their consequent radiological hazards associated with different crystalline rock types and soils [13], [14], [15], [16], [17], [18], [19], [20], [21], [22], tar sand and bitumen deposits [23], [24], [25], [26], [20] as well as phosphate mineral deposits [27], [28], [29], [30], [6].

Quarrying mineral deposits that contain naturally occurring radionuclides that are above the maximum permitted exposure limit can be very dangerous and can pose serious health risks for people residing within the locality where it is being mined for commercial purposes. Gene pool damage is one of the side effects of radiation exposure [31], [32], [33], [19]. Survived victims of an acute radiation exposure disease or any other radiation sickness are posed to high risk of developing cancer later in life. This study, therefore, focuses on the determination of the radiogenic composition of the limestone rock type of the Ewekoro Formation southwestern Nigeria and evaluation of the consequent radiological hazards associated with their commercial exploration and exploitation. The area of study is a limestone quarry site situated between the easting of 3°05' to 3°15' and northing 6°40' to 6°55' located in Ewekoro L.G.A, Ogun State, SW Nigeria (Figure 1).

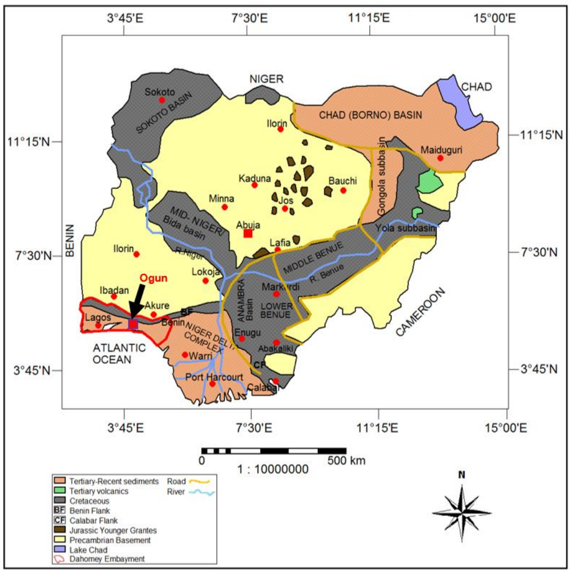

Figure 1: Geological Sketch Map of Nigeria Showing the Major Geological Components: Basement, Younger Granites, and Sedimentary Basins (Modified After Obaje, 2009) [34]

Ogun state is bounded to the North, South, West and East by Oyo state, Lagos state, Benin republic and Ondo state. Figure 2 shows that the study area lies geologically within the Eastern Dahomey Basin with east-westward trend sediments deposition and six lithostratigraphic units comprising Benin, Ilaro, Oshosun, Akinbo, Ewekoro and Abeokuta Formations from youngest to the oldest geological formation. Ewekoro Formation is known to be a Paleocene shallow marine deposit of non-crystalline and non-fossiliferous limestone strata.

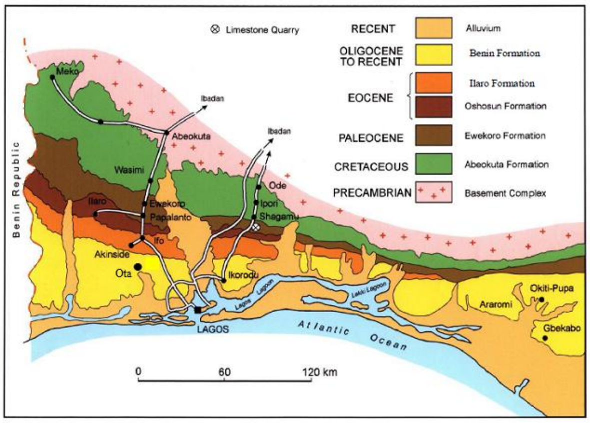

Figure 2: Geological Map of the Nigerian Part of the Dahomey Embayment Showing the Ewekoro Limestone (Modified After Gebhardt et al., 2010) [35]

Methods

Sample Collection, Preparation and Radioactivity measurements

A total of 13 samples were collected from the study area. The limestone samples were picked down to a depth of about 30 cm at each location point using hammer and hand trowel into a sealed polythene bags to prevent the samples from mixing up. In the laboratory, each sample was air-dried, pulverised, homogenised and then sieved using a 2 mm mesh. A 0.2 kg weight of the sieved sample was poured into a standard container (plastic), tightened and sealed to prevent ^{220}Rn and ^{222}Rn gasses from escaping. The sealed samples were left for thirty days to allow for secular equilibrium between parent and daughter nuclei.

The radionuclides activity concentrations were measured using NaI (TI) detector-based gamma spectrometric system where the digiBASE system that combines a miniaturised preamplifier and detector with a powerful digital multichannel analyser and special features for fine time-resolution measurements. The digiBASE incorporates into the NaI (TI) detector provides a gain stabiliser to significantly reduce the sensitivity of the detector to changes in ambient temperature and magnetic fields.

Three gamma-ray lines of interest were 1460 keV, 1764 keV and 2615 k eV which were resolved without much interference. The cylindrical plastic containers of radiation source were of diameters 7 cm. The seven soil samples each of mass 0.2 kg were dried, grinded and kept for more than thirty days in standard plastic containers to reach secular equilibrium were kept above the detector for the counting process.

About 10800 seconds (3 hours) was set as the counting time, which is considered enough for the detector to be able to show clearly and be able to distinguish the desired peak from a spectrum of signals. Multichannel analyser algorithm was used to compute the areas under each peak which represent the count number for a radionuclide in a particular sample. Uranium reference material termed RGU-1 from the International Atomic Energy Agency (IAEA) was used to calibrate the energy of the gamma spectrometer. The reference material was weighed into a standard cylindrical plastic sample container and placed on a NaI detector surface enclosed inside a lead shield of the spectrometer. This was counted for a lifetime of 10800 seconds.

A spectrum was captured, and specifically, three of the energy peaks identified on the spectrum were used in the energy calibration. Corresponding to the locations (channel numbers), the peaks of interest were: 295keV, 1120keV and 1765keV. To convert the count rate (cps) response of the spectrometer to desirable activity (Bq) for each of the three radionuclides (^{40}K, ^{226}Ra and ^{232}Th), the three reference materials RGK-1, RGU-1 and RGTh-1 from International Atomic Energy Agency (IAEA) were used. The γ-ray lines of ^{214}Bi at 1764 keV, ^{208} Tl at 2014 keV and 1460.8 keV were used to determine the specific activity of ^{226}Ra, ^{232}Th and ^{40}K respectively.

Results

Radionuclides Activity Concentration

The measured activity concentrations of ^{238}U, ^{232}Th and ^{40}K natural radionuclides in limestone samples within the quarry site along with the geographic coordinates of the points are presented in Table 1. Activity concentrations for the radionuclides were estimated in Bq / Kg based on the dry weight. The gamma-ray spectra for samples S10 and S12 are presented in Figure 3.

The activity concentrations of ^{238}U range from 18.09 ± 3.43 Bqkg^{-1} to 239.50 ± 25.74 Bqkg^{-1} with a mean of 121.30 Bqkg^{-1}, ^{232}Th range from 8.33 ± 0.83 Bqkg^{-1} to 360.01 ± 21.33 Bqkg^{-1} with a mean of 112.25 Bqkg^{-1} while ^{40}K ranges from 11.28 ± 0.81 Bqkg^{-1} to 735.26 ± 0.95 Bqkg^{-1} with a mean of 158.47 Bqkg^{-1}.

These results were then compared with the worldwide average activity concentration of 35 Bqkg^{-1}, 30 Bqkg^{-1} and 400 Bqkg^{-1} for ^{238}U, ^{232}Th and ^{40}K, respectively [36], [14], [15].

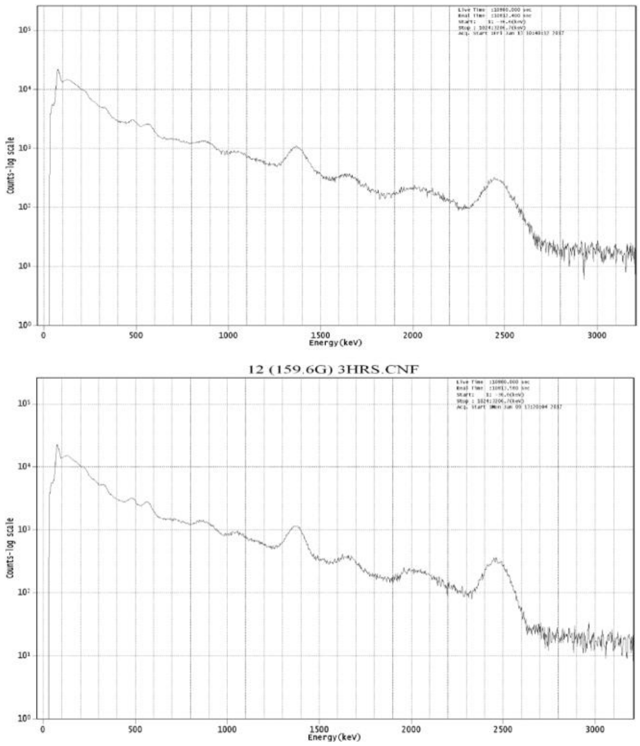

Figure 3: Representative Gamma-Ray Spectra for Samples S10 and S12

The measured activity concentration of ^{238}U was more than the worldwide average value in all of the limestone samples except for samples S2 and S9; ^{232}Th activity concentration levels in the samples were also higher than the worldwide average value except for sample S9. In contrast, ^{40}K activity concentration was below the worldwide average values for all samples considered except S12 (Table 1). Figure 4 shows the highest radionuclides ^{238}U, ^{232}Th and ^{40}K for S3, S4, and S12 respectively.

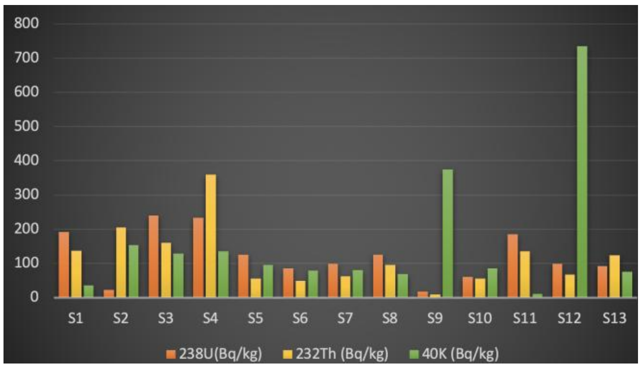

Figure 4: Measured Activity Concentrations at Each Sample Location

Discussion

The radiological maps (Figure 5A, 5B, and 5C) further reveal the higher concentration of ^{238}U localising around the northeastern (NE) and southeastern (SE) regions of the study area, while that of ^{232}Th localised around the SE part of the study area. A good positive correlation with relatively high coefficient is observed between ^{238}U and ^{232}Th radionuclides in Table 2 which imply that they are of the same source since their decay series occur together in nature. However, ^{40}K has weak negative correlation coefficients with both ^{238}U and ^{232}Th radionuclides confirming that K-40 originates from a different decay series. Table 2 also confirms that ^{40}K radionuclide has little contribution to the radioactivity of the limestone samples and consequently, the estimated radiological attributes due to its low activity concentration levels. There are strong positive correlation coefficients of ^{238}U and ^{232}Th with all estimated radiological attributes which imply that the high activity concentration of both ^{238}U and ^{232}Th are the major causative factors of the gamma radiation emission from the Ewekoro limestone. Several radiological parameters are needed to be evaluated to assess various health risks prone to by both the miners and the entire people residing around the quarry area. They include gamma-ray hazard indices, annual gonadal dose equivalence, dose rate and excess lifetime cancer risks.

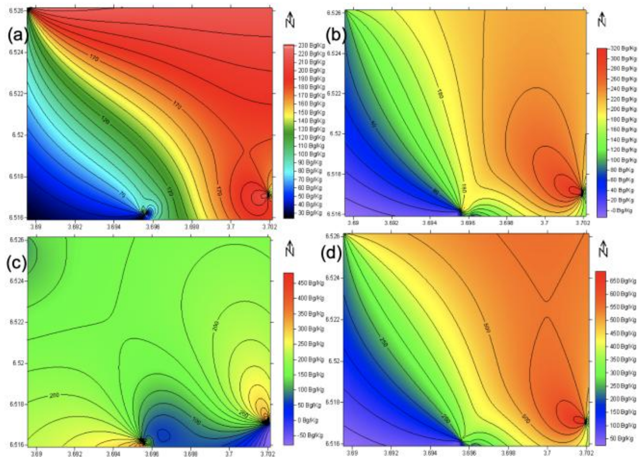

Figure 5: Radiological Map of the Study Area Showing; A) ^{238}U Activity Concentration (Bq/Kg); B) ^{232}Th Activity Concentration (Bq/Kg); C) ^{40}K Activity Concentration (Bq/Kg); and D) Absorbed Gamma Dose Rate (Ngyh^{-1})

X-Ray Radiation Hazard Indices

There is a great need to evaluate the threats of the gamma-ray radiation to both miners and residents of the communities where the limestone deposit is situated. Radium equivalent activity (Ra_{eq}) is used to compare the activity concentration of samples that contain different amounts of ^{238}U, ^{232}Th and ^{40}K natural radionuclides. This radiation index is calculated using equation (1). Where AC_U, AC_{Th} and AC_K are the limestone activity concentration in Bq / kg of ^{238}U, ^{232}Th and ^{40}K radionuclides respectively. The expression of the equation has a assumption that 370 $Bqkg^{-1}$ of ^{238}U, 259 $Bqkg^{-1}$ of ^{232}Th, and 4810 $Bqkg^{-1}$ of ^{40}K will produce the same radiation dose rate (Farai and Ademola, 2005; Usikalu et al., 2015a; Usikalu et al., 2015b)[37], [16], [17].

Table 3 shows the range of radium equivalent activity values in the limestone samples, which varies from 58.857 $Bqkg^{-1}$ in S9 to 758.832 $Bqkg^{-1}$ in S4 with a mean value of 299.96 $Bqkg^{-1}$. Ra_{eq} values corresponding to samples (S3-4 and S11) are well above the maximum permissible limit of 370 $Bqkg^{-1}$ [36], [14], [15]. These anomalously high radium equivalent values are concentrated around the southeastern region of the study area (Figure 5D). Major contributions to this hazard index are from the measured ^{238}U and ^{232}Th activity concentrations within the samples.

$$Raeq\ (Bqkg^{-1}) = AC_U + 1.43AC_{Th} + 0.077AC_K \quad (1)$$

Table 1: The Measured Radionuclides Activity Concentrations for all the Limestone Samples

SAMPLE NO	EASTING	NORTHING	^{238}U(Bq/kg)	^{232}Th (Bq/kg)	^{40}K (Bq/kg)
S1	3.69561	6.51619	191.92 ± 35.22	137.42 ± 13.67	35.94 ± 2.56
S2	3.69569	6.51621	21.99 ± 2.46	204.36 ± 12.26	153.73 ± 2.07
S3	3.68957	6.52616	239.50 ± 25.74	160.13 ± 9.55	128.39 ± 6.64
S4	3.70195	6.51708	233.59 ± 24.10	360.01 ± 21.33	135.43 ± 7.04
S5	3.70201	6.51703	125.43 ± 11.04	55.43 ± 5.04	95.43 ± 4.04
S6	3.70206	6.51712	85.43 ± 7.04	48.43 ± 3.04	79.36 ± 4.12
S7	3.69612	6.51591	98.43 ± 4.04	61.47 ± 6.04	80.28 ± 4.18
S8	3.69554	6.5162	126.10 ± 13.50	95.37 ± 1.04	69.43 ± 3.04
S9	3.69552	6.51613	18.09 ± 3.43	8.33 ± 0.83	374.75 ± 26.20
S10	3.68955	6.52615	61.04 ± 11.31	55.92 ± 5.57	85.43 ± 7.04
S11	3.70213	6.51707	184.38 ± 34.23	135.43 ± 4.04	11.28 ± 0.81
S12	3.70203	6.51712	98.59 ± 18.28	66.573 ± 3.07	735.26 ± 0.95
S13	3.69606	6.51594	92.43 ± 2.04	124.37 ± 2.07	75.43 ± 7.04
Minimum			18.09 ± 3.43	8.33 ± 0.83	11.28 ± 0.81
Maximum			239.50 ± 25.74	360.01 ± 21.33	735.26 ± 0.95
Mean			121.30 ± 14.80	112.25 ± 6.73	158.47 ± 5.86
*UNSCEAR (2000)			35	30	400

The external and internal hazard indices (H_{ex} and H_{in}) are being used to assess both the external exposure of the limestone miners and the inhabitants within the locality to γ-radiation in the outdoor air and the internal exposure to radon respectively. These indices have to be below unity (1) to have insignificant radiation [36], [16], [17]. They are estimated using equations (2 and 3) respectively. Representative level index I_r is another radiation index that is used to compute γ-radiation level about the measured activity concentrations of ^{238}U, ^{232}Th and ^{40}K radionuclides [38]. It is computed by using equation (4) [39] and its values when less than or equal to unity is the same as the annual effective dose less than or equal to 1 mSv.

$$H_{ex} = \frac{AC_U}{370(Bq/kg)} + \frac{AC_{Th}}{259(Bq/kg)} + \frac{AC_K}{4810(Bq/kg)} \quad (2)$$

$$H_{in} = \frac{AC_U}{185(Bq/kg)} + \frac{AC_{Th}}{259(Bq/kg)} + \frac{AC_K}{4810(Bq/kg)} \quad (3)$$

$$I_r = \frac{AC_U}{150} + \frac{AC_{Th}}{100} + \frac{AC_K}{1500} \quad (4)$$

The estimated H_{ex} values range from 0.16 to 2.05 (Table 3) with an average value of 0.81, while the computed H_{in} values range from 0.21 to 2.68 with a mean of 1.14. Samples (S3-4 and 11) had H_{ex} values greater than unity while Samples (S3-4, 8, 11 and 13) had H_{in} values greater than unity. The calculated values for $\mathbf{I_r}$ in the limestone samples range from 0.45 to 5.25 with an average of 2.08. These values are high (except S9) and exceed the upper threshold limit for I_r which is unity (UNSCEAR, 2000) [36]. Results of the γ-Ray radiation hazard indices revealed that exploration of Ewekoro limestone is radiologically hazardous for both the miners and the inhabitants of the area.

Table 2: Correlation Coefficient (r^2) of the Naturally Occurring Radionuclide Concentrations and all the Estimated Radiological Parameters from the Limestone Samples

*Variables	^{40}K	^{238}U	^{232}Th	Ra_{eq}	D	AEDE	H_{ex}	H_{in}	I_r	ELCR	AGED
^{40}K	1										
^{238}U	-0.286	1									
^{232}Th	-0.209	0.5	1								
Ra_{eq}	-0.189	0.802	0.905	1							
D	-0.188	0.797	0.899	0.994	1						
AEDE	-0.188	0.797	0.899	0.994	1	1					
H_{ex}	-0.189	0.802	0.905	1	0.994	0.994	1				
H_{in}	-0.226	0.897	0.822	0.984	0.978	0.978	0.984	1			
RLI	-0.167	0.792	0.909	1	0.994	0.994	1	0.98	1		
ELCR	-0.188	0.797	0.899	0.994	1	1	0.994	0.978	0.994	1	
AGED	-0.106	0.715	0.875	0.953	0.951	0.951	0.953	0.923	0.955	0.951	1

Annual Gonadal Dose Equivalent (AGDE)

The gonads are part of the vital organs in the body that are of interest because they are highly sensitive to radiation. An increase in AGDE has been known to affect the bone marrow and red blood cells. It is calculated using equation (5).

$$AGDE\ (\mu Svy^{-1}) = 3.09AC_U + 4.18AC_{Th} + 0.314AC_K \quad (5)$$

The estimated AGDE values are presented in Table 3. The annual gonad equivalent dose ranged from 208.39 μSvy^{-1} to 2269.16 μSvy^{-1} with a mean of 870.31 μSvy^{-1}. The average value obtained is almost thrice that of the average world value for exposure limit of 300 μSvy^{-1} [36]. Therefore, the radiations emitted from the limestone endanger bone marrow and the bone surface of the miners and residents of the area.

Dose Rate

The absorbed gamma dose rate (D) refers to the amount of radiation energy absorbed or deposited per unit mass of the substance. This radiological parameter is used to characterise the external primordial gamma radiation in the air at about 1 m above the surface of the ground, and it was calculated using equation (6) as proposed by (UNSCEAR, 2000) [36]. The annual effective dose equivalent (AEDE) in μSvy^{-1} resulting from the calculated absorbed dose values (D) was determined using equation (7), where O_C represents the occupancy factor taken as 0.2 and conversion coefficient (F_C) of 0.7 is used to convert absorbed gamma dose rate (D) to AEDE.

$$D\ (nGyh^{-1}) = 0.462AC_U + 0.604AC_{Th} + 0.041AC_K \quad (6)$$

$$AEDE\ (\mu Svy^{-1}) = D\ (nGyh^{-1}) \times O_C \times F_C \times 8760 \times 10^{-3} \quad (7)$$

The absorbed gamma dose rate value ranged from 28.754 $nGyh^{-1}$ in Sample S9 to 330.917 $nGyh^{-1}$ in Sample S4 with a mean of 135.289 $nGyh^{-1}$ (Table 3). All the limestone samples except Sample S9 were above the world average (populated-weighted) adsorbed gamma dose rate of 59 $nGyh^{-1}$ according to UNSCEAR (2000) [36] and Taskin et al., (2009) [40]. The AEDE values in the limestone samples ranged from 35.26 μSvy^{-1} in Sample 9 to 405.84 μSvy^{-1} in Sample S4, with an average of 165.92 μSvy^{-1}. All samples except Sample S9 had relatively high AEDE values greater than the average world value of 70 μSvy^{-1} (UNSCEAR, 1988) [41]. Therefore, based on radiation dose evaluation, the limestone in the study area is unsafe.

Excess Lifetime Cancer Risk (ELCR)

ELCR is the tendency to develop cancer over a lifetime at a given *γ*-radiation exposure limit. It is estimated using the equation (8), where DL is the life expectancy in Nigeria taken to be 54.5 years according to world health organisation report (2015) and the risk factor (RF in Sv^{-1}) of the general public estimated to be 0.05.

Table 3: The Estimated Radiological Parameters from the Limestone Samples Including Annual Gonadal Dose Equivalent AGDE, Absorbed Dose D, Annual Effective Dose AEDE, Excess Lifetime Cancer Risk ELCR, Ra_{eq} Radium Equivalent, I_r Representative Level Index, H_{ex} External Hazard Index, and H_{in} Internal Hazard Index

SAMPLE NO	Ra_{eq} (Bq/kg)	D ($nGyh^{-1}$)	AEDE (μSvy^{-1})	H_{ex}	H_{in}	I_r	ELCR	AGED (μSvy^{-1})
S1	391.2	173.14	212.34	1.057	1.58	2.68	0.58	605.63
S2	326.06	139.9	171.57	0.88	0.94	2.29	0.47	970.45
S3	478.37	212.63	260.77	1.29	1.94	3.28	0.71	1449.71
S4	758.83	330.92	405.84	2.05	2.68	5.25	1.11	2269.16
S5	212.04	95.34	116.93	0.57	0.91	1.45	0.32	649.24
S6	160.8	71.97	88.27	0.43	0.67	1.11	0.24	491.34
S7	192.51	85.89	105.34	0.52	0.79	1.32	0.29	586.3
S8	267.83	118.71	145.58	0.72	1.06	1.84	0.4	810.1
S9	58.86	28.75	35.26	0.16	0.21	0.45	0.1	208.39
S10	147.58	65.48	80.3	0.4	0.56	1.02	0.22	449.18
S11	378.91	167.45	205.36	1.02	1.52	2.59	0.56	1139.37
S12	250.4	115.9	142.15	0.68	0.94	1.81	0.39	813.79
S13	276.09	152.67	187.24	0.75	1	1.91	0.51	871.31
Minimum	58.86	28.75	35.26	0.16	0.21	0.45	0.1	208.39
Maximum	758.83	330.92	405.84	2.05	2.68	5.25	1.11	2269.16
Mean	299.96	135.29	165.92	0.81	1.14	2.08	0.45	870.31
**Limits	370	59	70	1	1	1	0.29	300

The estimated excess lifetime cancer risk range 0.10 to 1.11, with a mean value of 0.45 (Table 3). Only four out of the thirteen samples considered were safe and below the world permissible value of

0.29 (Taskin et al., 2009) [41]. A high level of ELCR within the study area implies a higher probability of induced cancer that a miner or a resident within the study area would be exposed to.

$$ELCR = AEDE \times DL \times RF \qquad (8)$$

In conclusion, the assessment of radiological parameters is important to evaluate the corresponding health hazards. The specific activity concentration of ^{238}U, ^{232}Th and ^{40}K in the limestone samples collected from Ewekoro, Ogun State had been determined using gamma-ray spectroscopy method. The activity concentration of ^{238}U and ^{232}Th in the limestone samples were higher than the safe limit except for samples S2 and S9 for ^{238}U and S9 for ^{232}Th. The computed radium equivalent activity values were higher than the global standard limit of 370 $Bqkg^{-1}$ in samples S3, S4, and S11. All the investigated limestone samples except S9 had gamma dose rate values higher than the global average, the estimated radiological hazard parameters than the permissible exposure limits. Hence, the health of the miners and inhabitants of the Communities where the limestone is being mined, processed and utilises is endangered due to the exposure to the radiation. To ensure good quality health, there is a need to protect people from the harmful effects of exposure to ionising radiation. To guarantee a high level of protection for miners, it is recommended that they should wear protective clothing to shield themselves from the radiation and reduce the time of exposure. Also, a routine health check-up should be conducted for the quarry workers and management. It is also recommended that the people should reside far away from the quarry.

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