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RADIOACTIVITY CONCENTRATION AND DOSE ASSESSMENT OF SOIL SAMPLES IN CEMENT FACTORY AND ENVIRONS IN OGUN STATE, NIGERIA

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ABSTRACT

The main natural radioactive materials that contribute externally and internally to man and his environs are radionuclides emanating from gamma series of ^{238}U, ^{232}Th, as well as ^{40}K isotope, alpha and beta sources. Also, human activities such as mining and milling of uranium and phosphate, tobacco smoking, oil exploration, air transportation, coal-fired power station, and so on can trigger The technologically Enhanced Naturally Occurring Radioactive Materials (TENORMs). It is imperative to monitor the terrestrial background radiation of radionuclides emanating from these sources in order maintain a safe environment for humans. This study measured the activity concentration of ^{238}U, ^{40}K and ^{232}Th in soil samples from cement factory and environs using the NaI(TI) detector, in three zones in Ogun State, Nigeria. The activity concentrations obtained for the three zones are in the order $^{238}U < ^{232}Th < ^{40}K$ respectively. The radiological parameters estimated from the activity measured were all within the recommended permissive limit except for the annual gonadal doses from the cement factory and its environs, which are higher than the global standard by the factors of 1.03 and 1.07 respectively. This study therefore concludes that people living and working in these areas might be exposed to high radiation burden as a result of cumulative effect of the emission from the factory.

Keywords: Radioactivity, radiation, radiological parameters, cement factory, soil

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1. INTRODUCTION

The stages involved in the production of cement include: appraisal of the location for possible prospects of limestone (a major material in cement production) shale and gypsum via geophysical approach, quarrying, grinding and blending of the clinker, as well as packaging and transportation of the finished products for sales [1 – 3]. In all these stages, the most sensitive part to human health is the quarrying stage. Although exposure to natural radiation by human being is one of the unavoidable phenomena on earth as the mechanisms of releasing radionuclides to the soil are majorly from weathering of parent-rock and other geological processes [4], previous researches have revealed that quarrying and mining processes increase the activity concentration of radionuclides in an environment [5 – 7], but a contrary opinion was given by [8, 9]. Information about the subsurface radioactivity distribution could be used for the environmental assessment and monitoring of a contaminated region [10].

The sources of radiation are both natural and artificial. The natural sources (rocks, soils, sediments, radiation emission from air, water and vegetation) take an aggregate of up to 96% of the total [10]. Naturally Occurring Radioactive Materials (NORMs) and Technologically Enhanced Naturally Occurring Radioactive Materials (TENORMs) are from the same natural source, but human activities such as mining and milling of uranium and phosphate, tobacco smoking, oil exploration, air transportation, coal-fired power station, and so on trigger the TENORMs [11]. The basic natural radioactive materials that contribute externally and internally to man and his environs are the radiation emanating from gamma (^{238}U and its series, ^{232}Th and its series, as well as ^{40}K isotope), alpha and beta sources. It is essential to monitor the terrestrial background radiation of radionuclides emanating from these sources in order maintain a safe environment for humans.

Measurement and analysis of NORMs in soil is paramount, because it enables one to visualize and project the variations in the natural background activity of an area with time. This process is not only to determine the radiological impact on dweller of an area, it could be used as indicator for estimation of geochemical and biochemical constituents in the environment [11]. The rate at which the industrial pollution affects human health and ecosystem in developing nations calls for periodic assessment and monitoring. This will assist the researchers to differentiate the contaminated regions from the uncontaminated ones, and suggest possible remedies to environment being contaminated as a result of industrial pollution.

The major raw materials used in the production of cement are shale and limestone. These materials are one of the key geological formations in the sedimentary terrain, which contain some concentrations of radioelements. It has been discovered that workers and dwellers in the vicinity of cement factory are exposed to dust arising from production processes and during different manufacturing stages. Various clinical and occupational exposures to cement dust had been reported which pose different health challenge to human health. The dust cause respiratory health problem of which lung is the most affected part observed by the occupationally workers exposed to the dust [12]. The by-products from the production of cement is envisaged to be released to the environment, soil as well as groundwater system within the production settings, thereby causing environmental toxicity [13]. Emissions to groundwater and surface water may arise from construction and various processing stages during cement manufacturing. Water quality may be affected from the effluent which can affect marine flora and fauna, and human health, most importantly when the water is used for drinking and irrigation purposes. Therefore, this Chapter is aimed to assess the radioactivity concentrations and estimate hazards in soil samples within Ewekoro Cement Factory, Ogun state, Nigeria and its environs, as well as comparing the results with the control analysis from Covenant University, Ota, Ogun state, Nigeria in order to determine the impacts of cement production on the people living around this factory. The radiological impact assessment studies from other regions within Nigeria have been reported by [14, 15].

2. MATERIALS AND METHODS

2.1. The Study Location and Geology

The study area falls within Ogun state, southwestern Nigeria. The state is bounded by Oyo state and Osun state to the north, Ondo state to the east, Benin Republic to the west, and Lagos state to the south (Figure 1). The study area is divided into three zones. The first two zones are within Ewekoro local government area (LGA), while the third zone (control) is in the headquarters of Ado-Odo/Ota local government area (Figure 1). The study area is under a tropical climate, with rainy and dry seasons respectively. The rainy season is distributed from March to November, while the dry season covered the rest of the months. The temperature distribution varied from 27.2 to 33.7°C annually. The major occupation of the dwellers in the study area is agriculture, with arable as 80% of the total landmass. The major cultivated crops are cash crops (cocoa, oil palm, cotton, and rubber) and food crops (pineapple, cassava, banana, cocoyam, plantain and citrus).

The geology of Nigeria is underlain by the Pan-African mobile belt that separates West Africa from Congo Cratons [16, 17]. Separation of this region is believed to have been as a result of thermotectonic events for years [18]. The Basement and Sedimentary terrains are the major geological settings in Nigeria [6, 7], with the two settings well represented in Ogun state. The geology of the study area is chiefly on the Nigerian Dahomey Basin. It is composed of six depositional classes, which are: Abeokuta-, Akinbo-, Ilaro-, Oshosun-, and Benin-Formations (Figure 2). The locations of this present study fall on Ewekoro and Benin Formations as revealed in Figure 2.

Three zones were chosen within the state for the study: Ewekoro cement factory, a neighbouring settlement via the factory, and Covenant University (CU), Ota as the control. The first two zones are within Ewekoro LGA (bounded by longitude 3° 13′ E and latitude 6° 56′ N), while the later is the headquarters of Ado-Odo/Ota LGA (bounded by longitude 3° 41′ E and latitude 6° 41′ N). The choice of these zones is to study the impact of cement production on the lives of factory workers and people living around the factory. The results will be juxtaposed with that of the control station. Five topsoil samples were taken at each location (central, 1.5 metres away from the central spot towards the southern; northern; eastern; and western directions respectively), with the maximum of four (4) locations per zone, which culminates to the total of sixty (60) data points. The unwanted materials and dead leaves were removed from the surface of the topsoil before taking the samples. 2 kg of topsoil sample was taken at each spot using hand trowel. The soils were mixed together in order to have a full representation of that spot. The entire samples were dried until the weight reading becomes constant. Each sample was packaged in 240 g plastic container, sealed and left for 30 days to permit the radionuclides to reach the secular equilibrium [19, 8]. After this stage, the samples were analyzed using gamma ray spectrometry technique.

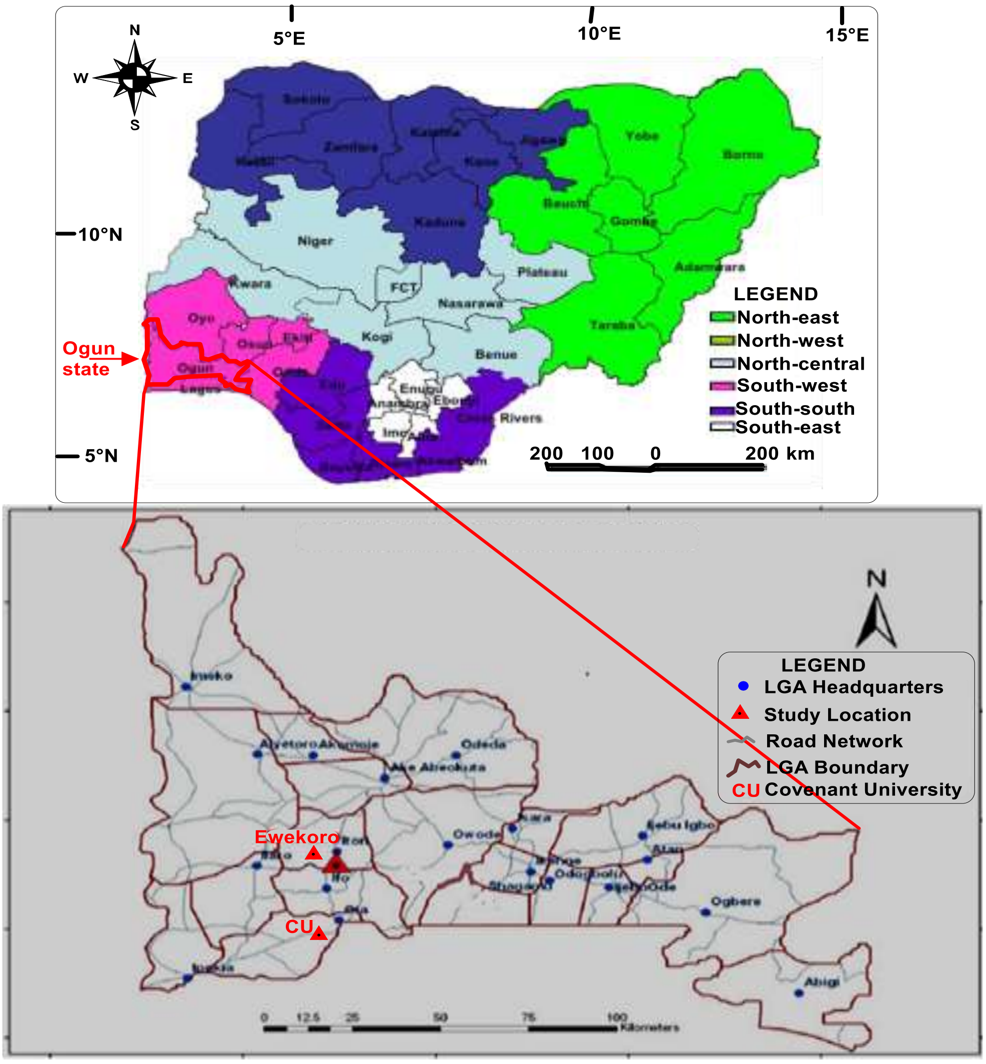

Figure 1 Zonal map of Nigeria revealing the study locations in Ogun state

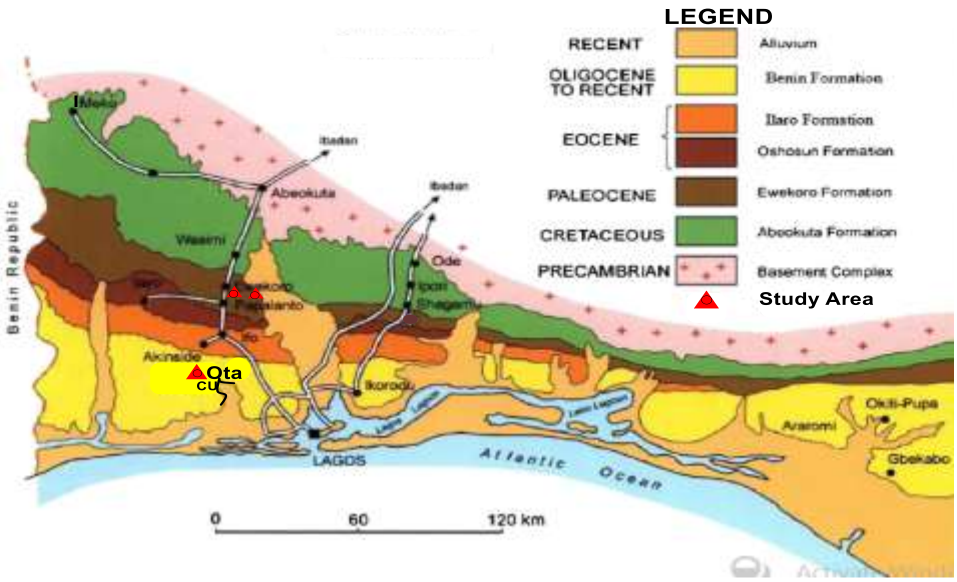

Figure 2 Geological domains in Ogun state and Nigerian Dahomey Basin

2.2. Method of Analyzing Gamma Ray Spectrometry Technique

The total time of 36, 000 seconds were used to count each sample. This will enable the processes to maintain a minimum counting error in NaI(TI) detector, which is connected to a multichannel analyzer. The resolution of the detector is approximately 8% at 0.662 MeV of ^{137}Cs, which is suitable to determine the gamma ray energies used for the collection. Its efficiency is about 25%. The measurements of ^{40}K was achieved at 1.460 MeV photopeak; ^{232}Th was achieved from ^{208}TI at 2.614 MeV photopeak; and ^{238}U was achieved from ^{214}Bi at 1.760 MeV photopeak. The detector was calibrated using IAEA-375 Reference Sample as provided by International Atomic Energy Agency. The mean specific activity concentrations (A_C) were determined by Eq. (1). The five samples taken at each location were averaged and presented as the representative of each location. The mean activity concentrations in the three (3) zones occupied in this study are presented in Figure 5 a – c.

$$A_C = \frac{C}{\varepsilon \times P_\gamma \times M_S \times t} \quad (1)$$

where ε is the efficiency of the γ-rays from the detector, C is the count rate of γ-rays, P_γ is the absolute transition probability of γ-decay, t is the counting time in secs., and M_S is the mass of the sample in kg. However, A_C was expressed in Bqkg^{-1} per dry weight.

2.3. Method of Estimating the Radiological Health Risks

In order to estimate the radiological health risks associated with the cement production workers and people living around the cement workers and people living around the cement factory, seven (7) radiological parameters were estimated. These parameters are: external absorbed dose rate ($D_{External}$), external annual effective dose ($AED_{External}$), excess life cancer risk (ELCR), external hazard (H_{Out}), radium equivalent (Ra_{Eq}), gamma index (I_γ) and annual gonadal dose (AG).

The $D_{External}$ in air at a height of 1 m above the surface of the topsoil of the three established zones based on the contributions from the activity concentrations of ^{238}U, ^{232}Th and ^{40}K were estimated using the standard as presented in Equation 2 [20].

$$D_{External} = 0.462\ C_U + 0.604\ C_{Th} + 0.0417\ C_K\ (\text{nGy h}^{-1}) \quad (2)$$

where C_U, C_{Th} and C_K are the activity concentrations of ^{238}U, ^{232}Th and ^{40}K respectively.

The foundational impact of radiological risk is determined from the assessment of annual radiation equivalent received by individual working or living in an area. The $AED_{External}$ was estimated based on Equation 3 [21, 22]. The implication of Equation 3 is that the conversion factor of 0.7 Sv Gy^{-1}, it converts the absorbed dose in air received to the effective dose received by taken human into consideration. Averagely, humans spend 20% of 24 hours in a day externally, which culminates to 20% of 365 days annually.

$$AED_{External}\ (\text{mSv y}^{-1}) = D_{External} \times 8760 \times 0.7 \times 10^{-6} \times 0.2 \quad (3)$$

Workers at cement factory as well as dwellers around the factory are at risk through the exposure of radioactive materials via inhalation of dust particles. The ELCR was estimated with the aim to assess the chances of developing cancer by the humans working and living around the cement factory. This is the estimated risk for individual that are exposed to acquired toxic materials in a lifetime. The ELCR is estimated based on Equation 4 [23, 24].

$$ELCR = AED_{External} \times DL \times RF \quad (4)$$

where DL is the duration of lifetime, which is assumed to be 70 years; RF is the risk factor, which is given as 0.05 Sv^{-1} respectively.

The H_{Out} is estimated from Equation 5. The implication of Equation 5 is that the activity concentrations of ^{238}U, ^{232}Th and ^{40}K are assumed to possess the same γ-radiation dose of 370, 259 and 4810 Bq kg^{-1} of uranium, thorium and potassium respectively.

$$H_{Out} = \frac{C_U}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \leq 1 \quad (5)$$

where C_U, C_{Th} and C_K have been defined in Equation 2.

The Ra_{Eq} were assessed using Equation 6 [21, 14]. This is one of the radiological risks that represent the uneven distribution of the activity concentration of ^{238}U, ^{232}Th and ^{40}K from the analyzed samples. The assumption of this parameter is similar to that of H_{Out} [25].

$$Ra_{Eq}\ (\text{Bq kg}^{-1}) = C_U + (C_{Th}\times 1.43) + (C_K\times 0.077) \leq 370 \quad (6)$$

The I_γ is one of the radiological indices used to assess human safety when exposed to γ-radiation (UNSCEAR, 2000). The I_γ is estimated based on Equation 7 [26].

$$I_\gamma = (C_U\times 0.0067) + (C_{Th}\times 0.01) + (C_K\times 0.00067) \leq 1 \quad (7)$$

It is essential to estimate the annual gonadal equivalent dose (AG), because it is the parameter that predicts whether the gonad, bone cells and marrow of humans are safe after exposure to γ-radiation or not [27]. The AG as a result of contributions from the activity concentrations of uranium, thorium and potassium was determined using Equation 8 [28].

$$AGED\ (\mu S_V\ y^{-1}) = (C_U\times 3.09) + (C_{Th}\times 4.18) + (C_K\times 0.314) \quad (8)$$

3. RESULTS AND DISCUSSION

The activity concentrations of ^{238}U, ^{232}Th and ^{40}K from the three zones occupied in this study were presented in Table 1. The mean activity concentrations of ^{238}U, ^{232}Th and ^{40}K from the three zones occupied in this study were presented in Figure 3 a - c. The mean activity concentration of radionuclides from the topsoil of Ewekoro Cement Factory (zone 1) measured for ^{238}U, ^{232}Th and ^{40}K were 1.95 ± 0.10 $Bqkg^{-1}$; 51.14 ± 1.87 $Bqkg^{-1}$ and 285.34 ± 13.37 $Bqkg^{-1}$ respectively. At the neighbouring settlement near the cement factory (zone 2), the activity concentration of radionuclides from the topsoil showed that the mean concentration of ^{238}U, ^{232}Th and ^{40}K were 2.21 ± 0.09 $Bqkg^{-1}$, 55.00 ± 1.92 $Bqkg^{-1}$ and 276.02 ± 14.58 $Bqkg^{-1}$ respectively. In the third zone which is the control, the mean activity concentration measured were 0.85 ± 0.06 $Bqkg^{-1}$, 34.55 ± 1.82 $Bqkg^{-1}$ and 280.19 ± 14.18 $Bqkg^{-1}$ for ^{238}U, ^{232}Th and ^{40}K respectively. For all the three zones, the radionuclides are in the order $^{238}U < ^{232}Th < ^{40}K$ respectively. The implication of this is that the soils are enriched with ^{40}K. By comparing these variables with the global standard as presented in Table 2, it was revealed that ^{40}K is higher than the global average of 250 $Bqkg^{-1}$ from the three zones by a factor of 2.05, 2.20 and 1.38 respectively [27, 21]. Over exposure to background radiation has been linked with some health challenges such as lung diseases, cancer, cataract and teeth fracture among others [7]. Thorium is available virtually everywhere. It does not evaporate from the soil, and can be transported from one surfacial region to the other. Over exposure to thorium can lead to damage of the body systems or death [29].

Comparison of the present study with the average values from literature as presented in Table 2 showed that this study is in agreement with the report obtained around a cement factory in Egypt [11]. In contrary, the report from the bedrock and soils of Ewekoro cement factory showed that the mean activity of ^{232}Th fall below the permissible limit, while the activity concentrations of ^{238}U and ^{40}K are strongly in agreement with this present study [30]. The Pearson correlation study as presented in Table 3 revealed the interactions among basic radionuclides used in this study. A significant strong positive correlation exist between ^{238}U and ^{232}Th from the three zones, such that correlation of 0.806 exist at Zone 1; correlation of 0.906 exist at Zone 2; and correlation of 0.999 exist at Zone 3 as shown in Figures 4a – c respectively. At Zone 1 and Zone 2, strong negative correlation of -0.769 and -0.578 exist between ^{40}K and ^{232}Th respectively. For other interactions, either weak positive or negative correlations were observed (Table 3). The implication of uranium-thorium interaction is that the two radionuclides originated from the same source [13]. The strong negative interaction between ^{40}K and ^{232}Th at Zone 1 and Zone 2 (Ewekoro Cement Factory and its environs) implies the contribution from different source, probably quarrying of limestone [5].

The estimated radiological health risks from Equations 2 to 8 showed varied risk parameters as presented in Table 4. The mean absorbed dose rate estimated was 43.772 $nGyh^{-1}$, 45.832 $nGyh^{-1}$ and 33.00 $nGyh^{-1}$ for zone1, 2 and 3 respectively. The values obtained from the three zones fall below the global mean of 59 $nGyh^{-1}$ [23]. The mean annual effective dose obtained for the zones were 0.054 $mSvy^{-1}$, 0.056 $mSvy^{-1}$ and 0.041 $mSvy^{-1}$ respectively. The values obtained from the three zones fall below the global mean of 0.07 $mSvy^{-1}$ according to [20]. The mean excess life cancer risk obtained in the study from the three zones fall below the global mean of 0.29×10^{-3} as reported by [20, 23].

The external hazard values obtained from the three zones fall below the global mean of unity as reported by [7]. The radium equivalent varied from 276.96 to 329.04 $Bqkg^{-1}$, 278.80 to 313.06 $Bqkg^{-1}$, and 231.23 to 279.21 $Bqkg^{-1}$ for zone 1, 2 and 3 respectively. The values obtained from the three zones fall below the global mean of 370 $Bqkg^{-1}$ as reported by [14]. The mean gamma index was 0.715, 0.749 and 0.538 for the three zones respectively. The values obtained from the three zones fall below the global mean of unity [6]. Radium equivalent activity has been linked with the external and internal hazards as well as the gamma index respectively, such that if one of these parameters falls below the global mean, the result is the same for others [25]. This is in agreement with this present study.

The annual gonadal dose varied from 299.533 to 331.314 μSvy^{-1}, 303.835 to 342.627 μSvy^{-1}, and 204.684 to 256.427 μSvy^{-1} for the three zones respectively. Virtually all the points at the cement factory (Zone 1) and its environs (Zone 2) revealed elevated signatures than the global mean of 300 μSvy^{-1} [31]. The values at CU (Zone 3), which is the control point fall below the global average. The outcome of annual gonadal doses at the cement factory and its environs showed that the workers and the dwellers in this environment are exposed to radiological risk, which might pose effect on the functionality of their organs most especially the lungs [27, 31].

Table 1 Activity concentrations of radionuclides from the three zones [8]

Stations	Zone 1 (Ewekoro Cement Factory)		
	^{238}U (Bq kg^{-1})	^{40}K (Bq kg^{-1})	^{232}Th (Bq kg^{-1})
1	2.5579±0.082	276.2125±13.252	56.6218±1.957
2	1.8268±0.082	261.5372±12.674	51.5696±1.792
3	1.5960±0.0596	342.0791±14.174	44.7819±1.833
4	1.8268±0.0827	261.5372±13.384	51.5696±1.890
Mean	1.9519±0.0964	285.3415±13.371	51.1357±1.868
Stations	Zone 2 (Neighbouring settlement)		
	^{238}U (Bq kg^{-1})	^{40}K (Bq kg^{-1})	^{232}Th (Bq kg^{-1})
1	2.6242±0.08	244.106±13.384	61.6911±1.890
2	2.6117±0.098	296.4±14.898	57.4982±2.021
3	1.7797±0.089	280.4153±14.623	50.7276±1.854

4	1.8147±0.077	283.1712±15.428	50.0746±1.925
Mean	2.20757±0.086	276.0230±14.583	54.9979±1.923
Stations	Zone 3 – Control (CU)		
	^{238}U (Bq kg^{-1})	^{40}K (Bq kg^{-1})	^{232}Th (Bq kg^{-1})
1	0.6197±0.065	301.1536±13.547	30.2785±1.812
2	0.6227±0.059	243.3481±12.571	30.2269±1.866
3	1.0749±0.063	287.1673±15.428	38.8706±1.782
4	1.0729±0.07	289.0965±15.188	38.8361±1.8
Mean	0.8476±0.064	280.1914±14.184	34.5530±1.815

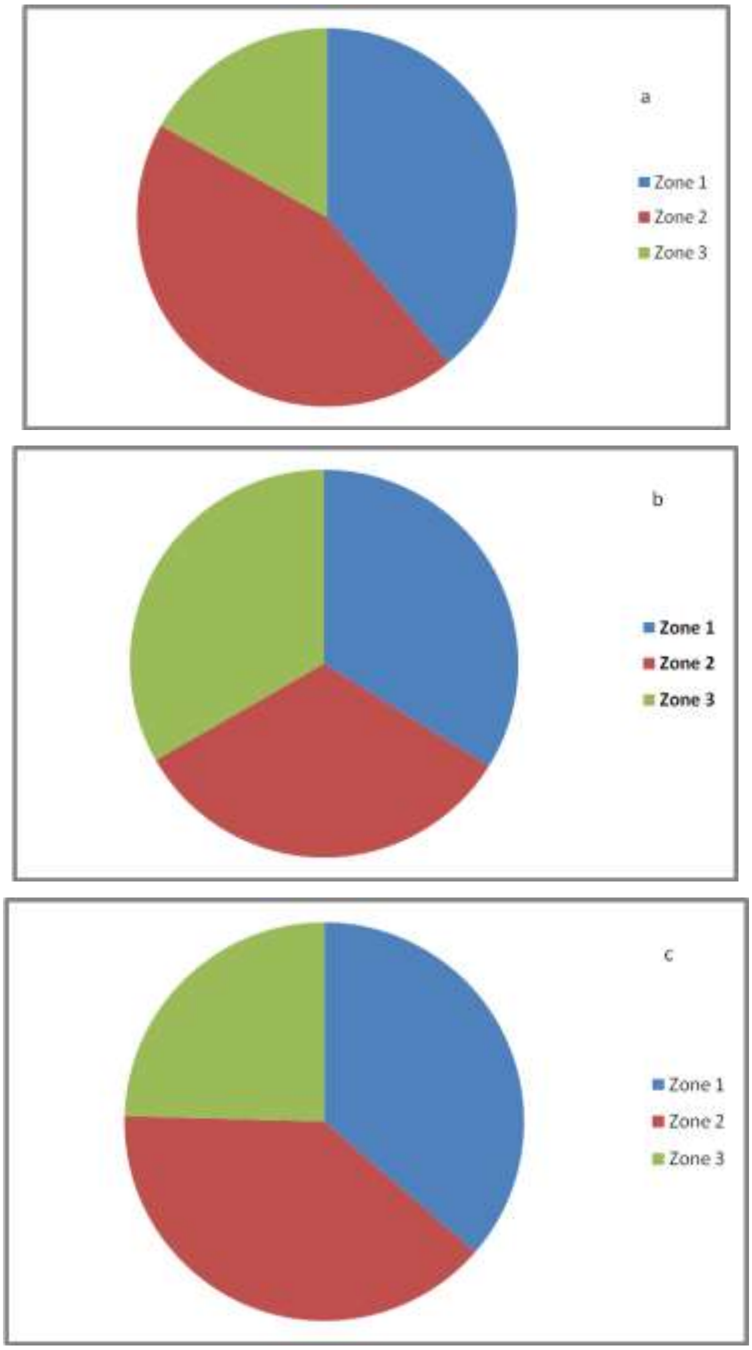

Figure 3 (a-c) Mean activity concentration of ^{238}U, ^{40}K and ^{232}Th for zone 3 respectively

Table 2 Comparison of the mean activity concentration of radionuclides in this study with the results from other locations

Location		^{238}U	^{40}K	^{232}Th	Reference
		(Bq kg^{-1})			
Pakistan	-	50.7	531.7	70.2	[21]
Egypt	Site 1	28.2	141.6	31.3	[11]
	Site 2	24.8	202.5	35.5	
Malaysia	-	19.0	243.3	16.5	[32]
Italy	-	13.1	13.5	6.0	[33]
Nigeria	-	11.3	89.6	8.0	[28]
Nigeria	-	12.8	415.6	8.8	[4]
Nigeria	-	7.8	17.6	8.9	[30]
Nigeria	Zone 1	1.9	285.3	51.1	Present study
	Zone 2	2.2	276.0	55.5	
	Zone 3	0.8	280.2	34.6	
World Mean	-	25.0	370.0	25.0	UNSCEAR (1988)

Table 3 Pearson correlation among the radionuclides

Zone 1	Parameters	^{238}U	^{232}Th	^{40}K
	^{238}U	1		
	^{232}Th	-0.410	1	
	^{40}K	0.806	-0.769	1
Zone 2	Parameters	^{238}U	^{232}Th	^{40}K
	^{238}U	1		
	^{232}Th	-0.306	1	
	^{40}K	0.906	-0.578	1
Zone 3	Parameters	^{238}U	^{232}Th	^{40}K
	^{238}U	1		
	^{232}Th	0.358	1	
	^{40}K	0.999	0.366	1

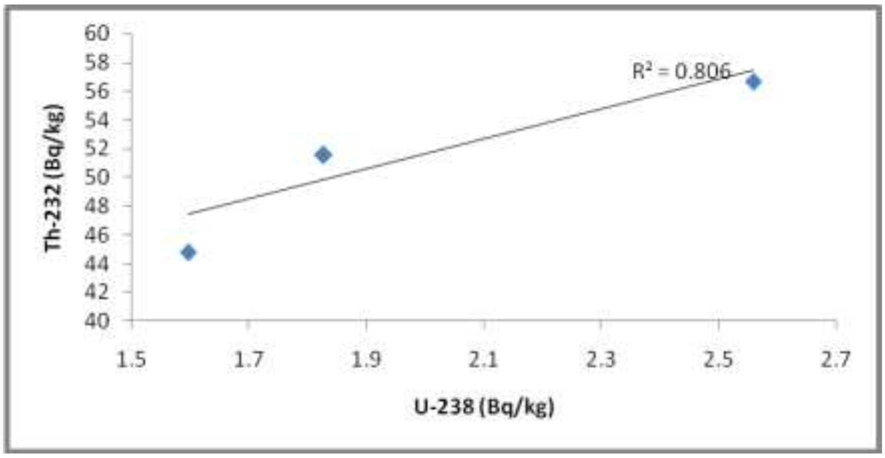

Figure 4a Uranium-thorium relationship at Zone 1

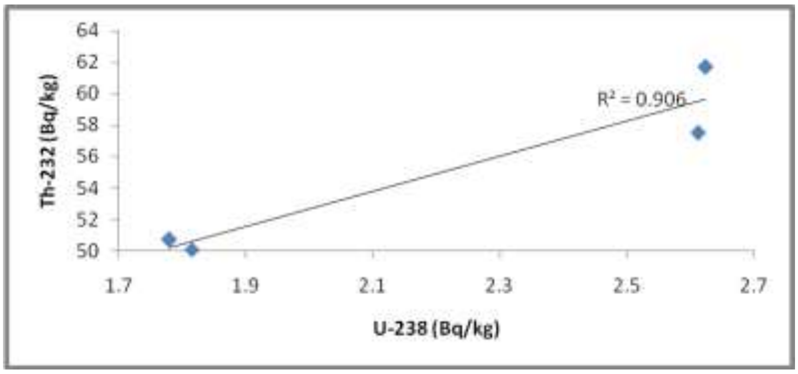

Figure 4b Uranium-thorium relationship at Zone 2

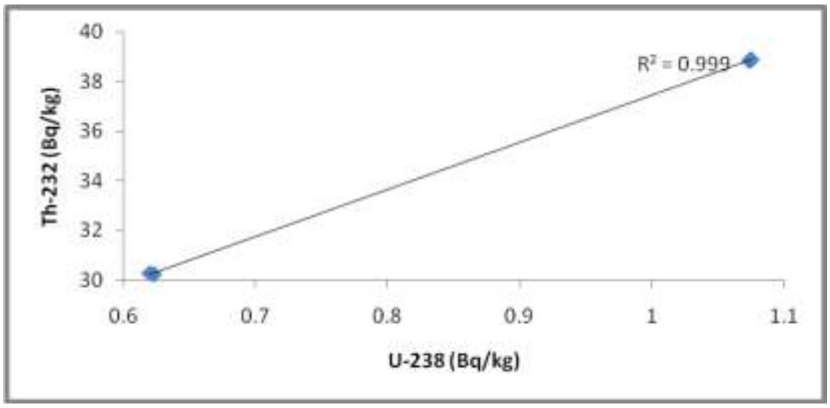

Figure 4c Uranium-thorium relationship at Zone 3

Table 4 The estimated radiological hazards in the present study

Stations	**Zone 1 (Ewekoro Cement Factory)**						
	$D_{External}$ ($nGyh^{-1}$)	$AED_{External}$ ($mSvy^{-1}$)	ELCR $\times 10^{-3}$	H_{Out}	Ra_{Eq} ($Bqkg^{-1}$)	I_{γ}	AG (μSvy^{-1})
1	46.982	0.058	0.202	0.283	296.211	0.767	331.314
2	42.977	0.053	0.185	0.258	276.955	0.702	303.328
3	42.153	0.052	0.181	0.248	329.035	0.687	299.533
4	42.977	0.053	0.184	0.258	276.955	0.702	303.328
Mean	43.772	0.054	0.188	0.262	294.789	0.715	309.376
Station	**Zone 2 (Cement Neighbouring settlement)**						
	$D_{External}$ ($nGyh^{-1}$)	$AED_{External}$ ($mSvy^{-1}$)	ELCR $\times 10^{-3}$	H_{Out}	Ra_{Eq} ($Bqkg^{-1}$)	I_{γ}	AG (μSvy^{-1})
1	48.726	0.060	0.209	0.296	278.804	0.797	342.627
2	48.384	0.059	0.208	0.291	313.062	0.799	341.482
3	43.239	0.053	0.186	0.259	290.240	0.706	305.591
4	42.977	0.053	0.184	0.257	291.463	0.702	303.835
Mean	45.832	0.056	0.197	0.276	293.392	0.749	323.384
Station	**Zone 3 – Control (CU)**						
	$D_{External}$ ($nGyh^{-1}$)	$AED_{External}$ ($mSvy^{-1}$)	ELCR $\times 10^{-3}$	H_{Out}	Ra_{Eq} ($Bqkg^{-1}$)	I_{γ}	AG (μSvy^{-1})
1	31.222	0.038	0.134	0.181	275.806	0.508	223.041
2	28.765	0.035	0.141	0.169	231.225	0.469	204.684
3	36.035	0.044	0.176	0.213	277.779	0.587	255.971
4	36.095	0.044	0.177	0.213	279.213	0.588	256.427
Mean	33.030	0.041	0.162	0.194	266.006	0.538	235.031
Global mean	**59**	**0.07**	**0.29**	**1**	**370**	**1**	**300**

4. CONCLUSION

The radioactivity and risk assessment of quarrying in Ewekoro Cement Factory and its environs has been carried out. The radionuclides in this present study are in the order ^{238}U < ^{232}Th < ^{40}K respectively. By comparing the radionulcides with the global average, it was found out that ^{40}K is higher by a factor of 2.05, 2.20 and 1.38 for Zone 1, Zone 2 and Zone 3 respectively. Very strong positive correlations between ^{238}U and ^{232}Th were observed from the three zones, which are 0.806; 0.906; and 0.999 respectively. The negative strong correlations of -0.769 and -0.578 experienced between ^{40}K and ^{232}Th at the cement factory and its environs are linked with quarrying activities in the environment. The estimated radiological hazards showed that all the parameters fall below the global standard, except the annual gonadal doses from the cement factory and its environs, which are higher that the global standard by the factors of 1.03 and 1.07 respectively. It is affirmed from this study that there might be long term effect on over exposure to γ-radiation workers and dwellers as a result of contributions from ^{40}K and AG respectively.

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