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Investigation of gamma radiation levels in soil samples collected from some locations in Ogun State, Nigeria Usikalu M.R.\*, Akinyemi M.L. and Achuka J.A Department of Physics, Covenant University, P.M.B.1023, Ota, Ogun State, Nigeria

## Abstract

This study measured the terrestrial gamma radiation and associated dose rates from primordial radionuclides  $^{238}$ U,  $^{40}$ K and  $^{232}$ Th in sixty soil samples collected from north, west, east and south of Ewekoro cement factory premises, Owowo village situated adjacent the factory and Covenant University, Ogun State using gamma ray spectrometry method. In order to estimate the radiological hazard index of the natural radioactivity, the gamma absorbed dose rate, annual effective dose and hazard indices were estimated. The measured activity concentrations of radionuclides in Ewekoro cement factory soils [ $^{238}$ U {1.60±1.60 Bqkg<sup>-1</sup> (east) - 2.56±0.08 Bqkg<sup>-1</sup> (north)},  $^{232}$ Th {44.78±1.83 Bqkg<sup>-1</sup> (east) - 56.62±1.96 Bqkg<sup>-1</sup> (north)},  $^{40}$ K {261.54±12.67 Bqkg<sup>-1</sup> (south) - 342.08±14.17 Bqkg<sup>-1</sup> (east)}] and Owowo village [ $^{238}$ U {1.78±0.09 Bqkg<sup>-1</sup> (east) - 2.62±0.08 Bqkg<sup>-1</sup> (north)},  $^{232}$ Th {50.07±1.93 Bqkg<sup>-1</sup> (west) - 61.69±1.89 Bqkg<sup>-1</sup> (north)},  $^{40}$ K {244.11±13.38 Bqkg<sup>-1</sup> (north)}] soils are higher than that of Covenant University soils [ $^{238}$ U {0.62±0.07 Bqkg<sup>-1</sup> (south) - 1.07±0.06 Bqkg<sup>-1</sup> (north)},  $^{232}$ Th {30.23±1.87 Bqkg<sup>-1</sup> (south) - 38.87±1.78 Bqkg<sup>-1</sup> (east)},  $^{40}$ K { 243.35±12.57 Bqkg<sup>-1</sup> (south) - 301.15±13.55 Bqkg<sup>-1</sup> (north)}]. The radiation hazard indices estimated were less than unity, the mean absorbed dose and annual effective dose rates are 40.88 nGyr<sup>-1</sup> and 0.05 mSv respectively. The study found that the activity concentrations and radiological hazard index from samples from Ewekoro and the neighbouhood are consistently higher than those from Covenant University however, the values are less than the recommended safe levels.

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# 1. Introduction

Natural radioactivity arises mainly from the primordial radionuclides, such as <sup>40</sup>K, and the radionuclides from <sup>238</sup>U and <sup>232</sup>Th series and their decay products, which are present at trace levels in all ground formations [1]. Human population is exposed to radiation from these radionuclides directly, as a result of external exposure, or through incorporation of these radionuclides into the body through inhalation or ingestion [2]. Estimation

of the radiation dose distribution is important in assessing the health risk to a population and serve as the reference in documenting changes to environmental radioactivity in soil due to anthropogenic activities. Since these radionuclides are not uniformly distributed, the knowledge of their distribution in soil and rock play an important role in radiation protection and measurement [3]. Cement is produced majorly from limestone and some small quantities of other materials such as clay, shale ash and ion oxide. They also contain some elements like gypsum, which contains silicate and aluminates that have ionization tendency [4]. Limestone which is the main constituent in cement is largely abundant in the earth crust (the residence of primordial radionuclides). Therefore, the materials taken from the Earth's crust and used in the construction of dwellings are one of the sources of (indoor) external exposure [5]. A crucial process in cement production is the quarry process. Research has shown that this process increases the activity concentration of radionuclides in the production environment [6, 7]. However, there are dearths of information on the radioactivity measurement of the effect of Cement manufacturing company in Ewekoro to the human population. Therefore, this work measures the specific activity of <sup>40</sup>K, <sup>234</sup>Th, and <sup>238</sup>U and estimates the radiological hazard associated with them in soil samples obtained in Ewekoro cement plant, Owowo a neigbouring community and Covenant University Ota all in Ogun State.

### 1.2 Materials and Methods

### Sample Collection and Preparation

Soil Samples were collected from Ewekoro cement plant cited in Itori local government located at 6°56'N and 3°13'E Nigeria Owowo village situated adjacent to the cement factory and Covenant university located in Ota 6°41'N and 3°41'E the local government headquarter of Ado-odo Ota Ogun State, Southwestern Nigeria. The locations are within the Eastern Dahomey Basin of Nigeria. A total of 60 soil samples were collected from north, south, west and east of each study area. The study area was divided into three zones: Ewekoro cement factory, neighborhood of the factory site and Covenant University Ota. An area of  $3 \times 3 m^2$  was marked at every sampling location, and the top 0.5-cm surface layer along with vegetation was removed prior to taking the sample. About 2 kg of soil sample comprising five cores was collected from selected area using coring device. The depth of each sample was 5 cm, having a diameter of 10 cm. Such five samples were thoroughly mixed to prepare one representative sample of the sampling site. The samples were dried at the ambient temperature of about 30°C for many days until a constant weight was attained indicating the lowest water content. The dried samples were pulverized and 240 g of each was then filled into an empty cylindrical plastic container of uniform size (9 cm height and 7 cm diameter) and sealed for about 30 days to allow <sup>238</sup>U and <sup>232</sup>Th and their progenies to reach secular equilibrium before radiometric analysis was carried out.

# 1.3 Gamma Spectrometry Analysis

The samples were counted for 36,000 seconds to achieve minimum counting error in a low-level gamma counting spectrometer comprising a 7.6 cm x 7.6 cm NaI (Tl) detector (Model No 802-series) by Canberra Inc., which is coupled to a Canberra Series 10 plus Multichannel Analyzer (MCA) (Model No 1104) through a preamplifier base. The detector has a resolution of about 8% at 0.662 MeV of <sup>137</sup>Cs which is capable of distinguishing the gamma ray energies used for the measurements. The photopeak at 1.460 MeV was used for the measurement of <sup>40</sup>K while those at 1.760 MeV peak from <sup>214</sup>Bi and 2.614 MeV from <sup>208</sup>Tl were used for the measurement of <sup>226</sup>Ra and <sup>232</sup>Th, respectively. The detector is coupled to a set of amplifier, Analog-to-Digital Converter (ADC) that bring out an output through a Canberra S100 Multi-Channel Analyzer (MCA) using a spectrum analysis program SAMPO 90. The detector was quoted at 25% efficiency and calibration was achieved using an IAEA-375 Reference soil supply by the International Atomic Energy Agency (IAEA) [8]. Each sample was counted for. For quality assurance the gamma ray spectroscopy analysis adopted in this work was used by several researchers [9, 10, 11, and 12]. The mean specific activity was computed using equation (1)

$$A_c = \frac{A_{net}}{M_s . t_c . P_{\gamma} . \xi}$$

1

where  $A_c$  is the activity concentration of radionuclide (Bq kg<sup>-1</sup>), Anet is the net area under the peak of radionuclide,  $M_s$  is the mass of the sample (kg), tc is the counting time (s), P $\gamma$  is the emission probability and  $\xi$  is the efficiency of the detector.

#### 1.4 **Results and Discussion**

### Activity Concentration

The specific activity of the natural radionuclides ( $^{238}$ U,  $^{232}$ Th, and  $^{40}$ K) in the samples was presented in Table 1.  $^{232}$ Th activity in the soil samples is distinctly higher than that of  $^{238}$ U and it ranges between 30.23 Bqkg<sup>-1</sup> 1. In activity in the soil samples is distinctly higher than that of  $^{-1}$  U and it ranges between 30.23 Bqkg (NU2) and 61.69 Bqkg<sup>-1</sup> (NN1) with a mean activity of 46.91±1.87 Bqkg<sup>-1</sup>.  $^{238}$ U concentration in the soil samples ranges between 0.62 Bqkg<sup>-1</sup> (NU2) and 2.61 Bqkg<sup>-1</sup> (NN1) with a mean activity of 1.67±0.35 Bqkg<sup>-1</sup> and was found to be lesser than that of both  $^{232}$ Th and  $^{40}$ K. The activity of  $^{40}$ K in all the samples was found higher when compared to that of  $^{232}$ Th and  $^{238}$ U in all sampling locations studied, it ranges between 243.35 Bqkg<sup>-1</sup> (NU3) and 342.08 Bqkg<sup>-1</sup> (NE3) with a mean activity of 280.52±14.04 Bqkg<sup>-1</sup>. In order to compare the activity concentrations of three natural radionuclides in soil samples, correlations between them was have a first order to be the sample of the activity of the samples activity of  $^{238}$ U is a mean activity of  $^{238}$ U in  $^{238}$ U in plotted Figure 2(I-III) presents correlations between the activity concentration of  $^{238}$ U and  $^{232}$ Th,  $^{238}$ U and  $^{40}$ K, and <sup>232</sup>Th and <sup>40</sup>K respectively, with a trend line drawn among the data points using regression technique. In all the three cases, the regression was found to be linear and positive. The correlation coefficient between  $^{238}$ U and  $^{232}$ Th was high (0.96) whereas correlation between  $^{238}$ U and  $^{40}$ K and  $^{232}$ Th and  $^{40}$ K was very low. It is not surprising, since  $^{238}$ U and  $^{232}$ Th come from natural decay series  $^{238}$ U and  $^{232}$ Th whereas  $^{40}$ K, although a naturally occurring radionuclide, is not part of any such decay series. However, a positive correlation may still be attributed to the property of the soil in retaining these radionuclides under varying weather conditions. It was also observed from Table 1 that the mean value of  $^{40}$ K was the highest and that of  $^{238}$ U was the lowest in all the study area. The spatial distribution of the radionuclides across the three locations under study revealed that the concentration of <sup>40</sup> K and <sup>234</sup>Th was highest at the neighboring settlement while Covenant University has the lowest concentration of <sup>238</sup>U and <sup>234</sup>Th. The result also revealed that highest concentrations of the three radionuclides were consecutively obtained in the samples from Ewekoro cement and neighbourhoods by factor 23% of <sup>238</sup>U, 49% of <sup>232</sup>Th and 71% of <sup>40</sup>K compared to Covenant University that is about 12 km to the cement factory which suggest that the cement factory increased radiation level of the population within the vicinity. The results obtained in this study are comparable to worldwide average concentration of these radionuclides in soils reported [13].

#### 1.5 Gamma Absorbed Dose Rates and Estimation of Hazard Indices

The absorbed dose of the naturally occurring radionuclide from the samples was calculated using equation 2 (UNSCEAR, 2000).

 $D (nGyh^{-1}) = 0.604C_{th} + 0.462C_{u} + 0.042C_{k}$ (2)

Annual effective dose  $(Sv y^{-1}) = D (nGyh^{-1}) x (8760 h y^{-1}) x 0.2 x 0.7 (SvGy^{-1})$ (3)

Annual effective dose (Sv y ) = D (nGyn ) x (6700 h y ) x 0.2 x 0.7(SvGy ) (37) where,  $C_{th}$ ,  $C_u$ ,  $C_k$  are the activity concentrations (Bq kg<sup>-1</sup>) of <sup>232</sup>Th, <sup>238</sup>U and <sup>40</sup>K, respectively in the soil sample and 0.604, 0.462 and 0.042 (nGy h<sup>-1</sup> per Bq kg<sup>-1</sup>) are the activity concentration-to-dose conversion factors. The outdoor gamma absorbed doses in air ranging between 28.77 nGy.h<sup>-1</sup> and 48.38 nGy.h<sup>-1</sup> with a mean of 40.88 nGy. $h^{-1}$  for the study area, which is less than the world average value of 60 nGyh<sup>-1</sup>. The differences are considered to be due to the geological settings and the influence of the cement manufacturing in the area, which vary from one place to another and from one locality to another in the same zone. The mean

dose rate is important for determining radiation detriment to the population as a whole, but some members of the population may receive higher doses due to high concentration of radionuclides. A common feature in any environmental radiation measurements is the considerable variation in soil radioactivity with location depending on soil physiochemical parameters. The largest contribution from natural radionuclides in the study area soil samples to the absorbed doses in air is due to <sup>232</sup>Th. In order to estimate the annual effective dose, the conversion coefficient from absorbed dose in air to effective dose, the indoor occupancy factor of 20% base on the average, spent of their time outdoors, using the dose rate data obtained from the concentration values of natural radionuclides in soil and adopting the conversion factor of 0.7 Sv/Gy was substituted into equation 3 [13]. The calculated values of annual effective dose range from 0.035 to 0.06 mSv, with a mean value of 0.05 mSv, which is lower than the world average of 0.48 mSv [13].

Radiation hazards due to naturally occurring radionuclides  $^{238}$ U,  $^{232}$ Th, and  $^{40}$ K may be indoor or outdoor depending upon the location of a receptor indoor (inside a dwelling) or outdoor (outside a dwelling). The radiation hazards are defined in terms of indoor or outdoor radiation hazard index and are denoted by H<sub>in</sub> and H<sub>out</sub>, respectively. They were calculated using equation (4) and (5) respectively [14].

$$H_{in} = C_U / 370 + C_{Th} / 185 + C_K / 4810 \le 1$$

$$H_{ex} = C_U/370 + C_{Th}/259 + C_K/4810 \le 1$$

(4) (5)

The indoor hazard index was calculated to determine the radiation hazard to respiratory organs due to <sup>222</sup>Rn and its other short-lived decay products. The values of  $H_{out}$  and  $H_{in}$  calculated from the measured activity concentrations of <sup>238</sup>U, <sup>232</sup>Th, and <sup>40</sup>K. The results indicated that indoor radiation hazard index was greater than outdoor radiation hazard index at every sampling site. The indoor radiation hazard index ( $H_{in}$ ) range from 0.17 to 0.30 with a mean value of 0.25, whereas the outdoor radiation hazard index ( $H_{out}$ ) range between 0.17 to 0.30 with a mean value of 0.24. The mean values of the indoor and the outdoor radiation hazard index free from the radiation hazards. The value of  $H_{out}$  must be lower than unity to keep the radiation hazard insignificant. These values are far below the criterion limit ( $H_{ex}$  less than or equal to one) as per the European Commission on Radiation Protection reports, the terrestrial soils from this city has no high exposure for either inhabitants and can be used as a construction material without posing any significant radiological threat to the population.

### 1.6 Conclusion

Gamma ray spectrometry has been used to investigate the radioactivity concentrations of  $^{238}$ U,  $^{40}$ K and  $^{232}$ Th in soil samples collected from Ogun State. The activity profile of radionuclides showed the existence of low level activity in the studied locations. The mean activity concentrations of  $^{238}$ U,  $^{40}$ K and  $^{232}$ Th, is 1.67±0.35 Bqkg<sup>-1</sup>, 280.52±1.87 Bqkg<sup>-1</sup> and 46.91±1.87 Bqkg<sup>-1</sup> respectively, were obtained from all the soil samples studied. These values fall within the lowest range of those measured at worldwide scale reported by other authors. The mean gamma absorbed dose rates and the annual effective dose rate calculated is 40.88 nGy.h<sup>-1</sup> and 0.05 mSv respectively, these which are lower than the worldwide average value. The radiation hazard indices obtained are far less than the critical value of unity. Though, the result showed that the radiation levels in the cement factory and adjacent village are higher than those obtained in Covenant University location far from the factory all the obtained were within the recommended safe limit. The present study is the first to establish a baseline data of naturally occurring and anthropogenic radionuclides for the study area most especially for the cement factory. Based on our results, we conclude that the soil of the study area do not pose any radiological health hazard to the people of the area.



Fig.2 (I) Correlation between  $^{238}$ U, and  $^{232}$ Th concentrations; (II) correlation between  $^{238}$ U and  $^{40}$ K concentrations; (III) correlation between  $^{232}$ Th and  $^{40}$ K concentrations

Code	Location	Number of Samples	<sup>238</sup> U (Bq kg <sup>-1</sup> )	<sup>40</sup> K (Bq kg <sup>-1</sup> )	<sup>232</sup> Th (Bq kg <sup>-1</sup> )
NE1	Ewekoro North	5	2.56±0.08	276.21±13.25	56.62±1.96
NE2	Ewekoro South	5	1.83±0.08	261.54±12.67	51.70±1.79
NE3	Ewekoro East	5	$1.60 \pm 1.60$	342.08±14.17	44.78±1.83
NE4	Ewekoro West	5	1.83±1.83	261.54±13.38	51.57±1.89
NN1	Owowo North	5	2.62±0.08	244.11±13.38	61.69±1.89
NN2	Owowo South	5	2.61±0.10	296.40±14.90	57.50±2.02
NN3	Owowo East	5	1.78±0.09	280.42±14.62	50.73±1.85
NN4	Owowo West	5	1.81±0.08	283.17±15.43	50.07±1.93
NU1	Cov. Univ. North	5	0.62±0.07	301.1513.55	30.28±1.81
NU2	Cov. Univ. South	5	0.62±0.06	243.35±12.57	30.23±1.87
NU3	Cov. Univ. East	5	1.07±0.06	287.17±15.43	38.87±1.78
NU4	Cov. Univ. West	5	1.07±0.07	289.10±15.19	38.84±1.80
Range			0.62-2.61	243.35-342.08	30.23-61.69
Mean			1.67±0.35	280.52±14.04	46.91±1.87

Table 1: Activity Concentrations in the soil samples

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