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Assessment of Natural Radioactivity and Estimation of Radiation Dose Parameters around Cement Production Company in Ibese, Ogun State, Nigeria

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Abstract
Natural radionuclides found in soil are capable of disintegrating leading to the release of ionizing radiation that has harmful effects on individual exposed to them. A total of 30 soil samples were collected. Six samples each were collected from area where farming and mining activities were very high; the samples were analyzed using Sodium Iodide detector at the National Institute of Radiation protection and Research. Results showed that the identified radionuclides were ^{238}U, ^{232}Th and ^{40}K with mean activity concentration of 29.68±1.54 Bq/kg, 17.61±1.06 Bq/kg and 175.50±2.73 Bq/kg respectively. The estimated mean absorbed dose rate (D), the outdoor annual effective dose (AED) and the external hazard index (H_{ex}) were found as 31.72nGy/h, 0.04 and 0.16 respectively. The mean radium equivalent was found to be 190Bq/kg while the mean value of excess cancer risk was found to be 0.14. The absorbed dose rate, external hazard index and outdoor annual effective dose are lower than that of the recommended value of 60nGy/h and 1mSv/y. The estimated radium equivalent and excess life cancer risk were found to be within safe limit. Findings from this study shows that the radiation emitted from the radionuclides present in the soil of the study area do not pose any radiological health hazard to the public and the activities carried out on soil such as agriculture.

Keywords: Radioactivity, Radionuclide, Absorbed Dose, Radiological Hazard

INTRODUCTION

The human environment has always been exposed on daily basis to natural radionuclide such as ^{232}Th, ^{226}Ra and ^{40}K. Natural radioactivity exist in various geological formations such as rocks, soils, water and air. In addition to natural sources, soil radioactivity is also affected by anthropogenic activities (Abbady *et al.,* 2005). The radioactivity concentration in soil give information on both natural and man – made sources which is important in radiological monitoring, assessment of radiation dose for public and also their ability to act as excellent biochemical and geochemical traces in the environment. Measurement of natural radioactivity is very important to determine the amount of change of the natural background activity with time as a result of radioactivity release (UNSCEAR, 2002).

Cement is one of the most important building materials as it is used for making concrete, building blocks, conjugate layers between building bricks. It is made from a mixture of natural elements found in natural materials such as limestone, clay, sand and/or shale. Materials used in the building industry that include marl, blast furnace slag, fly ash, Portland clinker, and anhydrite (in the cement industry) may be of radiological effect to human life (Ajayi *et al.,*

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2000). Cement manufacturing may cause environmental contamination at all stages of the production process. The cement as the building material contains various amounts of natural occurring radionuclides of uranium (^{238}U), thorium (^{232}Th) and their decay products and the radioactive isotope of potassium (^{40}K). Although, the origin of these radionuclides is the Earth's crust, they can affect our life via food, air, water and building materials (Prasad *et al.,* 2008). Cement contributes to environmental radioactivity mainly in two ways, i.e. external as well as internal indoor exposure. The external radiation exposure is caused by gamma radiation originating from ^{226}Ra, ^{232}Th and their progenies and ^{40}K. However, the internal radiation exposure, mainly affecting the respiratory tract, is due to short lived daughter product of radon ^{222} Rn after the decay of ^{226}Ra (Ali *et al*., 2012). Natural environmental radioactivity and the associated external exposure due to gamma radiation depend primarily on the geological and geographical conditions and appear at different level in the soil of each different geological region. Natural environmental radioactivity arises mainly from primordial radionuclides, such as ^{40}K, and radionuclide from ^{232}Th and U –series, which occur at a trace levels in all ground formation (Chowdhury *et al.,* 1999).

Data regarding levels of natural radioactivity in soils and the corresponding radiation doses to the population are lacking for most parts of Nigeria. Monitoring of any release of radioactivity material to the environment is necessary for environmental protection (Ahmad *et al*., 2015). However, it has been observed that the type and concentration vary considerably depending on the soil type. The effect of radiation emitted by different radionuclide depends on the over lining soil materials (thickness and type) and its chelating agent physiochemical properties (Tufail *et al.,* 2013). Investigation has shown that natural radioactivity and the associated exposure due to gamma radiation (from radionuclide) depend primarily on geology (soil type). Gamma radiation emitted from primordial radionuclide and their progeny is one of the main external sources of radiation exposure to the humans (Arnedo *et al.,* 2017). Gamma radiation from radionuclides with half- lives comparable to the age of the earth such as ^{40}K and radionuclides from the progeny of ^{238}U and ^{232}Th series are the main contributors of external sources of radiation to human body (UNSCEAR,2000). Of these natural radionuclide potassium is the most abundant and is found in the earth's crust on average of 2.6%, while uranium and thorium are present in levels of parts per million (PPM). The radionuclide contributes to enhanced radiations exposure to human beings and animals (Nada *et al.,* 2009). The two major raw materials in the production of cement, limestone and shale are of geological origin and they are known to contain some natural radioactivity. The radionuclides on limestone and shale bedrocks and their over lining soil materials can become pollutant when present in greater level than their natural concentration. The elevated concentration of the radionuclides either in the exposed bedrocks (i.e., limestone and shales) or cement raw materials and products may be harmful (Sam *et al.,* 1997). Their presence in different food crops grown on the soil around the cement factory may constitute a health hazard. It is envisaged that the by-product resulting from the process of cement production may find ways into the underground water systems, the river and may settle on the soil surface, hence, representing a direct and indirect exposure pathway to man in his environment, through soil to plant man pathway and water to man(Aslam *et al.,* 2012). The gamma radiation from natural radionuclides and cosmic rays constitute the external exposure to human. The greatest threat of radionuclides is the damage to the gene pool (Jibiri *et al.,* 2005). The kind of symptoms experienced by many victims of radiation sickness may not be as significant as childhood leukemia, stillbirths, cancer or birth defects. However, beyond the physiological effects, mental and emotional consequences of trauma of exposure have been documented (Hamid *et al.,* 2002). Radionuclides found in soil are the members of the three naturally occurring radioactive decay series- uranium, thorium and actinium series. Other

radionuclides of terrestrial origin can also find themselves in water. Natural radionuclides have been reported in various concentration in different component of the environment. Limestone and shales deposits, which are bedrocks of concern in this study are used in cement production for building. Investigation have shown that natural radioactivity and associated exposure due to gamma radiation depend on geological conditions(Faheem & Mujahid, 2008). The soil around the cement production company were assessed for their radionuclidles concentration. The values obtained were compared with world public dose rate for the purpose of deducing possible radiological hazard in the area.

MATERIALS AND METHODS

Method

Study Area

The study area is situated in Egbado North, Ogun State, Nigeria. It geographical coordinate are 6° 58′ 0″ North, 3° 2′ 0″ East. The inhabitants are mainly farmers and traders. The mineral resources found in the study area include limestone and shale which

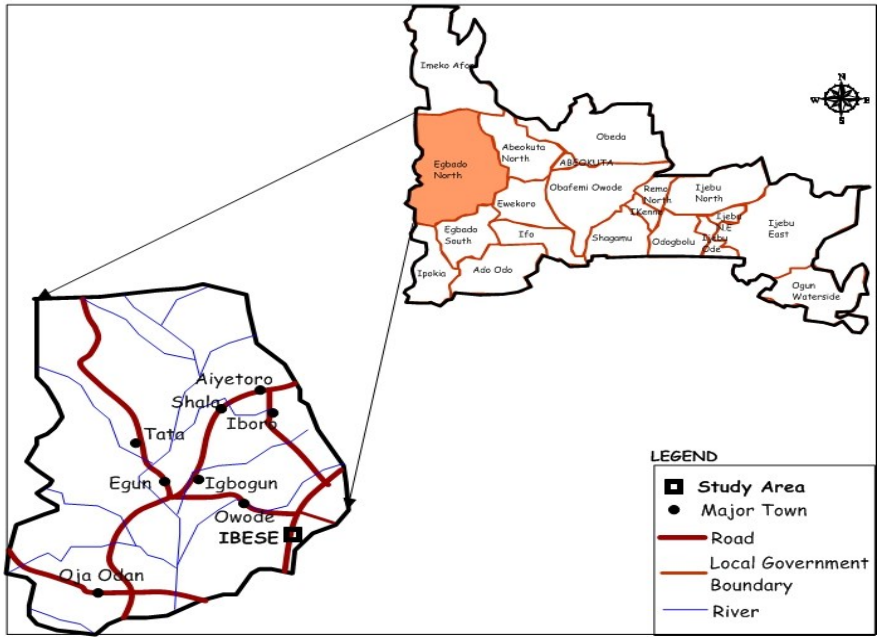

Figure 1: Map of the Study Area

Sample collection and preparation

The study area was divided into four strata and samples were selected randomly from each stratum. A total of 30 soil samples were collected from the site at a depth 30cm using hand trowel. The samples were collected from equidistance location with a distance of 100m from each other. Samples were collected in an area where farming and mining activities is very high around the cement factory. Stones and vegetation was removed from the soil samples. The soil samples were grinded into fine powder and homogenized by filtering them through a 1-mm sieve. The samples were dried at ambient temperature and packed into sample bags. Each sample bag was sealed for about four weeks to ensure secular equilibrium between the parent radionuclides and their gaseous daughter decay product in the uranium and thorium series. Gamma spectrometer measurement for the activity concentration in the soil samples were carried carrying out using Sodium Iodide detector.

Method of Data Analysis

The activity concentration in 30 soil samples were measured by means of gamma spectrometry using NaI(Ti) detector. The gamma ray spectrometry set in the analysis consist of high shielded-up and well calibrated 3cm×3cm NaI(Ti).

Assessment of Radiological Hazards

As more than one radionuclides contribute towards the gamma dose (i.e. ^{238}U, ^{232}Th and ^{40}K), therefore the radiological hazards have been presented in terms of a single quantity called hazard index" and the measured specific activity concentrations have been used to access the radiological hazards in terms of External hazard Index (H_{ex}), internal hazard index (H_{in}) and gamma Index(I_γ)

Internal Hazard Index (H_{in})

In addition to gamma rays, internal dose due to radon and its daughter products is present in the environment, and has the largest contribution towards the average effective dose received by human beings. The combined internal exposure to gamma-rays and radon has been defined by as indoor external hazard criterion (H_{in}). The H_{in} is calculated from the formula given by Ajayi (2005) as:

$$H_{in} = \left({}^{C_U}/_{185} + {}^{C_{Th}}/_{259} + {}^{C_K}/_{4810} \right) \quad (2.1)$$

where C_U, C_{Th}, and C_K are the activity concentrations of ^{238}U, ^{232}Th and ^{40}K in Bq/kg, respectively. For insignificant radiation hazard the indices should be less than unity proposed by Beretka &Mathew, (1985).

External Hazard Index (H_{ex})

Many radionuclides occur naturally in terrestrial soils and rocks and upon decay, these radio nuclides produce an external radiation field to which all human beings are exposed. In terms of dose, the principal primordial radio nuclides are ^{232}Th, ^{238}U and ^{40}K. Thorium and Uranium head series of radio nuclides that produce significant human exposure. The external hazard index (H_{ex}) is calculated from the formula given by Beretka and Mathew (1985) as*:*

$$H_{ex} = \left({}^{C_U}/_{370} + {}^{C_T}/_{259} + {}^{C_K}/_{4810} \right) < 1 \quad (2.2)$$

where C_U, C_T, C_K are the radioactivity concentration in Bq/kg of ^{238}U, ^{232}Th and ^{40}K. The value of this index must be less than unity for the radiation hazard to be negligible and equality to unity corresponds to the upper limit of Ra_{eq} (370Bq/kg) by ICRP.

Gamma Index

The European Commission has proposed an index called the gamma index (I_γ) defined by the following relation (European (Commission), 1999). The gamma index (I_γ) has been introduced to account for the combined impact of^{238}U, ^{232}Th and ^{40}K as radiological hazard associated with soil, vegetation and water.

$$\text{Gamma index } (I_\gamma) = \left({}^{C_U}/_{300} + {}^{C_{Th}}/_{200} + {}^{C_K}/_{3000} \right) \quad (2.3)$$

where C_U, C_{Th}, and C_K are the specific activities of ^{238}U, ^{232}Th and ^{40}K, respectively, in any material.

Absorbed Dose (D)

The external outdoor absorbed gamma dose rates due to terrestrial gamma rays from the nuclides ^{238}U, ^{232}Th and ^{40}K at 1m above the ground level were calculated following the work of Beretka and Mathew (1985) as:

$$(nGyh^{-1}) = 0.462C_U + 0.604C_{Th} + 0.042C_K \quad (2.4)$$

Where, D is the dose rate in nGy/h and C_U, C_{Th} and C_K are the specific activities (Bq/kg) of ^{238}U, ^{232}Th and ^{40}K respectively

Annual Effective Dose (AED)

The annual effective dose received by the population can be calculated using the following equation with a conversion factor of 0.7Sv/Gy to assess the health effects of the absorbed dose and transform absorbed dose in air to the effective dose received by humans, with an outdoor occupancy factor (0.2), which is equivalent to an outdoor occupancy of 20% and 80% for the indoor (0.8). The factor is suitable for determining the term of life in the studied area (UNSCEAR, 2000).The annual effective doses are determined as follows:

$$\text{indoor (mSvs)} = D(nGyh^{-1}) \times \text{conversion factor} \times 8760 \times 0.8 \times 10^{-6}\left(\frac{mSv}{yr}\right) \quad (2.5)$$

$$\text{outdoor (mSv)} = D(nGyh^{-1}) \times \text{conversion factor } x \times 8760 \times 0.2 \times 10^{-6}(mSv/yr) \quad (2.6)$$

where D (nGy/h) is the total absorbed dose, 0.7Sv/Gy is the conversion coefficient from absorbed close to effective dose received by adults; 10^{-6} is the conversion factor from nGy to equal measurements.

Excess Life time Cancer Risk (ELCR)

Outdoor Excess Lifetime Cancer Risk $(ELCR_{out})$ is calculated from outdoor annual effective dose according to Agbalagba and Onoja (2011) by the following equation

$$ELCR_{out} = E_{out} \times LE \times RF \quad (2.7)$$

where LE represents the life expectancy (70 years) and RF represents the fatal risk factor per sievert(Sv^{-1}). ICRP-60uses RF values of 0.05 for the public in case of stochastic effects.

Radium equivalent activity (Ra_{eq})

Radium equivalent (Ra_{eq})is a common index used to compare the specific activities of materials containing ^{238}U,^{232}Th and ^{40}K by a single quantity, which takes into account the radiation hazard associated with them. The radium equivalent activity index as expressed by UNSCEAR(2000) can be given as:

$$Ra_{eq} = C_U + 1.43\ C_{Th} + 0.77\ C_K \quad (2.8)$$

here: C_U, C_{Th}, C_K are the radioactivity concentration in Bq/kg of ^{238}U,^{232}Th, and ^{40}K. The world average of Ra_{eq} in soils is 89Bq/kg. The results obtained are presented in Table 3.2

RESULTS AND DISCUSSION

The results of this study highlights the presence and concentration level of different natural radionuclides in the soil. The mean values for the radionuclidles are shown in Table 3.1

Table 3.1: Present the activity concentration for 30 samples soil around cement production company in Ibese

SAMPLE CODE	CO-ORDINATES		ACTIVITY CONCENTRATION (Bq/kg^{-1})		
	Latitude	Longitude	^{40}K	^{238}U	^{232}Th
IB1	7°00′12″N	3°03′14″E	328.45±4.34	18.16±1.38	19.73±1.24
IB2	7°00′13″N	3°02′54″E	136.51±2.25	31.43±1.54	28.14±1.53
IB3	7°00′00″N	3°02′00″E	248.22±3.35	24.85±1.26	BDL
IB4	7°00′40″N	3°02′30″E	118.43±2.38	27.62±1.60	29.05±1.57
IB5	7°00′45″N	3°03′30″E	157.32±2.24	28.53±1.41	38.50±1.87
IB6	7°00′37″N	3°02′36″E	240.45±3.10	58.24±1.80	27.59±1.49
IB7	6°57′34″N	3°02′11″E	181.14±2.81	34.52±1.35	2.45±0.12
IB8	6°57′22″N	3°02′21″E	242.22±3.12	20.22±1.25	24.82±1.51
IB9	6°57′20″N	3°02′30″E	130.41±2.54	25.36±1.78	11.62±0.82
IB10	6°57′19″N	3°02′52″E	259.21±3.81	29.56±1.64	24.96±1.36
IB11	6°58′14″N	3°03′42″E	165.45±2.28	44.56±2.15	19.79±1.04
IB12	6°58′18″N	3°03′55″E	171.22±2.96	26.15±1.52	7.65±0.44
IB13	6°58′21″N	3°03′28″E	312.52±4.20	55.32±2.31	5.37±0.31
IB14	6°58′39″N	3°03′33″E	269.30±3.45	32.56±1.48	24.33±1.33
IB15	6°58′44″N	3°03′36″E	126.41±2.32	29.82±1.52	10.26±0.58
IB16	6°59′12″N	3°02′45″E	146.75±2.46	24.47±1.21	22.69±1.24
IB17	6°59′17″N	3°02′41″E	164.18±2.75	36.41±1.53	9.03±0.54
IB18	6°59′23″N	3°02′48″E	115.23±2.25	24.22±1.26	12.42 ±0.34
IB19	6°59′43″N	3°02′39″E	192.43±2.46	28.24±1.35	24.53±1.34
IB20	6°59′49″N	3°02′56″E	138.34±2.43	15.83±1.46	BDL
IB21	6°59′56″N	3°02′58 ″E	185.22±2.62	12.47±1.21	38.22±1.12
IB22	7°00′32″N	3°03′24″E	110.41±2.34	60.38±1.86	13.91±1.22
IB23	7°00′39″N	3°03′50″E	228.82±3.37	17.25±1.57	20.33±1.45
IB24	7°00′49″N	3°03′54″E	233.41±3.56	37.43±1.83	25.46±1.37
IB25	7°00′51″N	3°03′46″E	127.34±2.62	48.23±2.19	19.23±1.21
IB26	7°00′54″N	3°03′48″E	92.18±1.90	15.33±1.25	15.84±1.32
IB27	7°00′58″N	3°03′40″E	145.61±2.24	23.52±1.26	16.45±1.28
IB28	6°57′39″N	3°02′37″E	83.12±1.74	19.16±1.28	7.49±0.64
IB29	6°57′53″N	3°02′47″E	90.25±1.78	15.22±1.64	20.13±1.26
IB30	6°57′55″N	3°02′57″E	124.41±2.20	25.22±1.42	8.43±0.12
Mean			**175.50±2.73**	**29.68±1.54**	**17.61±1.06**
		420	**36**	**45**	

The activity concentration of ^{238}U range from 12.47±1.21 to 60.38±1.86Bq/kg with the mean value of 29.68±1.54Bq/kg. The highest value for ^{238}U (60.38±1.86) was found in IB22 and the lowest value (12.47±1.21Bq/kg) was found in IB21. The average radioactive level for ^{238}U (29.68±1.54Bq/kg) is less than the worldwide average value of 36Bq/kg (UNSCEAR, 2000). The ^{232}Th radioactivity concentration varies from BLD to 38.50±1.87Bq/kg with an average of 17.61±1.06Bq/kg. The highest ^{232}Th activity of 38.50±1.87Bq/kg was found in IB5 while the lowest ^{232}Th activity concentration of 2.45±0.12Bq/kg was found in IB3 and IB20.

The average radioactivity level of ^{232}Th (17.61±1.06Bq/kg) is also lower than the worldwide average of 45Bq/kg (UNSCEAR, 2000). The activity concentration of ^{40}K range from 328.45±4.34Bq/kg to 83.12±1.74Bq/kg with an average value of 175.50±2.73Bq/kg. The highest ^{40}K concentration of 328.45±4.34Bq/kg was found in IB1 and the lowest value of 83.12±1.74Bq/kg was found in IB28.The average value of ^{40}K is 175.50±2.73Bq/kg is lower than that of the worldwide average value of 420Bq/kg (UNSCEAR, 2000). The result of the current study reveals that the activity concentration of ^{238}U, is not significantly different from that^{232}Th while that ^{40}K is higher than that of ^{238}U and ^{232}Th in the soil samples. Therefore, the dominant source of gamma radiation measured in samples from all locations must have been from ^{40}K.

Table 3.2: Radiological hazard indices for all samples around cement production company in Ibese.

Sample Code	Absorbed dose rate (nGy/h^{-1})	Annual Effective dose		Ra_{eq} (Bq/kg)	Radiation hazard index			
		Indoor(mSv/y)	outdoor(mSv/y)		$ELCR_{out}$	H_{in}	H_{ex}	I_γ
IB1	34.10	0.17	0.04	299.28	0.14	0.24	0.19	0.27
IB2	37.25	0.18	0.05	176.78	0.18	0.31	0.22	0.29
IB3	21.91	0.11	0.03	215.98	0.11	0.19	0.12	0.17
IB4	35.28	0.17	0.04	160.35	0.14	0.29	0.21	0.28
IB5	43.04	0.21	0.05	204.72	0.18	0.34	0.26	0.34
IB6	53.67	0.26	0.07	282.84	0.25	0.47	0.31	0.41
IB7	25.04	0.12	0.03	177.50	0.11	0.23	0.14	0.19
IB8	34.51	0.17	0.04	242.22	0.14	0.26	0.20	0.27
IB9	24.21	0.12	0.03	142.39	0.11	0.21	0.14	0.19
IB10	39.62	0.19	0.05	264.84	0.18	0.31	0.23	0.31
IB11	39.49	0.19	0.05	200.26	0.18	0.35	0.23	0.30
IB12	23.89	0.12	0.03	168.93	0.11	0.21	0.14	0.18
IB13	41.93	0.21	0.05	303.64	0.18	0.38	0.24	0.32
IB14	41.05	0.20	0.05	274.71	0.18	0.33	0.24	0.32
IB15	25.28	0.12	0.03	141.83	0.11	0.23	0.15	0.19
IB16	31.17	0.15	0.04	169.91	0.14	0.25	0.18	0.24
IB17	29.17	0.14	0.04	175.74	0.14	0.27	0.17	0.22
IB18	23.53	0.12	0.03	130.71	0.11	0.20	0.14	0.18
IB19	35.95	0.18	0.04	211.49	0.14	0.29	0.21	0.28
IB20	13.12	0.06	0.02	122.35	0.07	0.11	0.07	0.10
IB21	36.63	0.18	0.04	209.74	0.14	0.25	0.22	0.29
IB22	40.93	0.20	0.05	165.29	0.18	0.40	0.24	0.31
IB23	29.86	0.15	0.04	222.51	0.14	0.22	0.17	0.24
IB24	42.47	0.21	0.05	253.56	0.18	0.35	0.25	0.33
IB25	39.25	0.19	0.05	173.78	0.18	0.36	0.23	0.30
IB26	20.52	0.10	0.03	108.96	0.11	0.16	0.12	0.16
IB27	26.92	0.13	0.03	159.16	0.11	0.22	0.16	0.21
IB28	16.87	0.08	0.02	93.87	0.07	0.15	0.10	0.13
IB29	22.98	0.11	0.03	113.50	0.11	0.18	0.14	0.18
IB30	21.97	0.11	0.03	133.07	0.11	0.19	0.13	0.17
Average	**31.72**	**0.16**	**0.04**	**190.00**	**0.14**	**0.26**	**0.18**	**0.25**

In other side the values of (AED), (ECLR), (H_{ex}), (H_{in}), (Ra_{eq}) and (Iϒ) respectively, ranged from 0.02mSv/y to 0.07mSv/y for outdoor, 0.06msv/y to 0.26mSv/y for indoor, 0.07 to 0.25, 0.07 to 0.31, 0.11 to 0.47, 93.87 Bq/kg to 303.64 Bq/kg, 0.10 to 0.41 The mean values of (D), (AED), (ECLR), (H_{ex}), (H_{in}), (Ra_{eq}) and (Iϒ) are lower than international limits of 60nGyh^{-1}, 1mSvy^{-1}, 1,1, 370Bq/kg and 1 for (D), (AED), (ECLR), (H_{ex}), (H_{in}), (Ra_{eq}) and (Iϒ) respectively.

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CONCLUSION

The assessment of natural radioactivity and estimation of radiation dose around cement production company in Ibese has been carried out . The mean annual effective dose are lower than the maximum permitted limit of 1mSv/y, hence may not have adverse effect on the populace. The activity concentration of ^{238}U,^{232}Th and ^{40}K was observed to be lower than world average value. The results obtained have shown that radiological hazards are well below the world average value. The radiation emitted from the radionuclides present in the soil of the study area do not pose any radiological health hazard to the public of the area and the activity carried out on soil such as agriculture. However, there may be increase in the values obtained with longer period of operation.

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