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Correlation of gamma emitting radionuclides and radiological health hazards indices around Lancaster dam

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ABSTRACT

Background: All human beings and living organisms are exposed to natural radiations on daily basis, which is mainly due to the activity concentration of primordial radionuclides ^{232}Th, ^{238}U and their products of decay together with the natural radionuclide ^{40}K present in the earth's crust. ***Materials and Methods***: A total of 59 soil samples were collected around the banks and surroundings of the Lancaster dam using an auger at a depth of about 0.75 meters from the top surface. The samples were analysed using low background co-axial n-type High Purity Germanium (HPGe) detector of high-resolution and the resulting spectrum were analysed using Canberra Genie software. ***Results:*** This study revealed that the activity concentration values are in the order of ^{232}Th <^{40}K<^{238}U in all sampling sites. A statistical analysis based on Pearson correlation showed a positive correlation between the radiological parameters and the two primordial radionuclides ^{232}Th, ^{238}U and the natural radionuclide ^{40}K. ***Conclusion:*** This implied that the two primordial radionuclides ^{232}Th, ^{238}U and their products of decay including the natural radionuclide ^{40}K contribute to the emission of gamma radiation in all the locations of the study area. The calculated ^{238}U/^{232}Th concentration ratio in soils of present study was almost six times higher compared with different countries of the world.

Keywords: *Annual gonadal dose equivalent; Gamma ray spectrometer; Radiological hazards; Excess lifetime risk; Lancaster dam.*

INTRODUCTION

All living organisms are exposed to natural radiations on daily basis, which is mainly due to the activity concentration of primordial radionuclides ^{232}Th, ^{238}U and their products of decay together with the natural radionuclide ^{40}K present in the earth's crust [1]. Radioactive material is found everywhere in the universe, and also on earth. More than 60 radionuclides are found in nature, and they can be placed in two general categories [2-4]:

Primordial radionuclides have existed since the creation of the Earth. Their half-lives are comparable to the age of the universe (15 billion years) and they have survived since their generation in stellar nucleosynthesis.

Cosmogenic radionuclides are continually formed and replenished by cosmic ray interactions with material in the atmosphere and in the biosphere.

Radionuclides are found in air, water and soil, and also in the human body [5]. Every day, we ingest/inhale radionuclides in the air we breathe, in the food we eat and the water we drink. Radioactivity is common in the rocks and soil that makes up our planet, in the water and oceans, and even in our building materials and homes [6-12].

The environment and health are interrelated,

hence the health risks related to natural radioactivity are of great concern and require assessment in order to estimate the risks. Naturally occurring radioactive materials generally contain terrestrial origin radionuclides (primordial radionuclides), left over since the creation of the earth [13].

Owing to the general shortage of naturally available water in and around Johannesburg, competition for water from the many small streams was severe among early miners whose economic survival depended on it for running steam engines and other mining processes [14, 15]. In order to store water of the non-perennial streams for the dry period artificial reservoirs and dams were created and streams diverted to bring water close to the mines. Inversely, mine infrastructures, such as tailing dams, also tended to be placed in the vicinity of water sources [17]. Consequently many mine waste deposits are now located near, or even in water courses, dams, wetlands and canals. In some instances return water dams for recycling slurry water from tailings dams were placed directly into water courses such as the Tudor and Lancaster dams. As a consequence, the distance between potential pollution sources and receiving water courses was often considerably shortened thereby promoting water pollution [15].

Lancaster Dam in Gauteng province is surrounded by tailings dams holding millions of tonnes of dangerous metals and is the source of the Wonderfonteinspruit [18, 19]. Most of the stream around this locations are filled with acid mine water and its wetlands had been classified as the radiological hotspot by the Nuclear Regulator of South Africa. Lancaster dam historically was indeed the source of the most pristine water [20]. Radioactivity monitoring around the bank of the Lancaster dam to know the extent of radiological hazards impact is based on analyses of specific nuclides in the surrounding environments. The knowledge of the concentrations and distributions of natural radionuclides is of interest since it provides useful information in the monitoring the surrounding environments due to the present of the dam. The activity concentration of the progenies from the natural decay series in the surrounding environments as a result of the dam may not necessarily be in equilibrium with their parents.

There is a lack of information about the radioactivity levels around the Lancaster dam in the province of Gauteng in South Africa. Therefore, recently, considerable attention has been given, to allow the creation of scientific hazard data base of the radiological baseline levels around the dam using γ-ray spectrometry. The baseline data can be used to assess any changes in the radioactivity background level due to various activities involving radioactive materials or any fallout in the near future due to surrounding mine tailing dams.

This study aims to identify the potential effects of primordial radionuclides ^{232}Th, ^{238}U and natural ^{40}K to members of public due to the contaminated dam in Lancaster, South Africa. This work further correlates the gamma emitting radionuclides and the radiological health hazards parameters and compared the results obtained from this study to others available from different countries of the world.

MATERIALS AND METHODS

The study area

Lancaster Dam in Gauteng province of South Africa is located at Latitude: -26.13333 and Longitude: -27.78333. The dam is found in Krugersdorp and is surrounded by tailing dams. Its source is of the Wonderfonteinspruit and the streams around are filled with acid mine water and its wetlands had been classified as the radiological hotspot by the Nuclear Regulator. Lancaster dam historically was primarily the source of most of the pristine water. The Lancaster dam is filled with water of a pH of about 2.6. figure 1 shows the sampling locations around the dam and the immediate surroundings.

Sample preparation and measurements

Two kilograms each of 59 soil samples were collected around the banks and surroundings of the Lancaster dam using an auger at a depth of

about 0.75 meters from the ground as shown in figure 1. The samples were transported to Centre for Applied Radiation Science and Technology, the environmental laboratory in North-West University, Mafikeng province in South Africa. In the laboratory, the soil samples were sieved using a 2 mm mesh to remove larger objects and then ground using mortar and pestle to fine powder in order to have the same matrix as the reference sample. The samples were dried in an oven at a temperature of 378 K for two hours until constant weights were obtained. The samples were then placed in desiccators to avoid moisture absorption. After that, the homogenized sample was packed in a standard 1500 ml air-tight labelled Marinelli beakers and sealed using silicon and plastic tapes. The samples were left for a minimum of 28 days to allow radioactive equilibrium among the radon-222 (^{222}Rn), radon-220 (^{220}Rn), and their short lived progenies.

Before measurement, the low background co-axial n-type High Purity Germanium (HPGe) detector of high-resolution gamma spectrometry system was calibrated using a primary standard obtained from the International Atomic Energy Agency. The detector has a resolution of 2.0 keV at 1332 keV and relative efficiency of 20 %. The output of the detector was analyzed using a 4 K analogue to digital converter (ADC) system connected to a desk-top computer. The resulting spectrum were analysed using Canberra Genie software "Genie-2000".

The detector was shielded using 4'' lead on all sides to reduce the background level of the system [30]. The system was calibrated and each of the soil sample was placed in gamma spectrometer and was counted for 43200 seconds using a gamma spectroscopy device connected to a coaxial HPGe detector, Canberra [15].

The activities of the samples were determined using the total net counts under the selected photo-peaks, the measured photo-peak efficiency, gamma intensity and weight of the samples [15]. After correcting for background and Compton contribution, the activity concentrations of ^{238}U, ^{232}Th, and ^{40}K were determined. The ^{238}U and ^{232}Th were calculated assuming secular equilibrium was established with their decay products ^{238}U series: ^{226}Ra (186.0 keV), ^{214}Pb (351.9 keV) and ^{214}Bi (609.2 keV); ^{232}Th series: ^{228}Ac (911 keV), ^{208}Tl (583.1 keV).

Radium equivalent (Raeq)

The universally accepted index for analysing the radiation exposure created by the primordial radionuclides is the radium equivalent activity index measured in Bq kg^{-1}. This index allows one to describe the gamma output from different mixtures of ^{238}U, ^{232}Th and ^{40}K in soil samples from the study area (figure 1) and it is calculated using equation (1) below [15]:

$$Ra_{eq} = C_{U238} + 1.43C_{Th232} + 0.077C_{k40} \quad (1)$$

where C_{U238}, C_{Th232} and C_{K40} are the specific activity concentrations of ^{238}U, ^{232}Th, and ^{40}K in Bq kg^{-1}, respectively.

Effective dose rate

Various radiological hazards delivered to the surrounding living biota can be assessed based upon the activity concentration of primordial radioactive elements. The effective dose rate (EDR) in soils was evaluated based on the Dose-rate conversion factors and the concentrations of the radionuclides [28]. Hence, the effective dose rate (EDR) to a human standing on the soil was calculated using equation (2) below:

$$EDR = f_c D \quad (2)$$

where $f_c = 0.72\,Sv.Gy^{-1}$ and is the conversion factor from absorbed dose rate D in air, to effective dose rate EDR for an adult [1]. The absorbed dose rate D is given by equation (3) below [29]:

$$D = DCF_{U238}C_{U238} + DCF_{Th232}C_{Th232} + DCF_{K40}C_{K40} \quad (3)$$

where the dose conversion factors DCF for ^{238}U, ^{232}Th and ^{40}K have the values of 0.462, 0.604 and 0.0417 nGyh^{-1}/Bq kg^{-1} respectively. C_{U238} = Concentration ^{238}U in soil (Bq kg^{-1}), C_{Th232} = Concentration ^{232}Th in soil (Bq/kg) and C_{K40} = Concentration ^{40}K in soil (Bq/kg).

From equations (2) and (3), it follows that the concentration of ^{238}U in soil depends on the effective dose rate (EDR) and the concentrations of the other two main radionuclides in soil as shown in equation (4):

$$C_{U238} = \frac{\left(\frac{EDR}{f_c} - DCF_{Th232}C_{Th232} - DCF_{K40}C_{K40}\right)}{DCF_{U238}} \quad (4)$$

Annual effective dose equivalent (AEDE)

The conversion coefficient from absorbed dose in air to effective dose and the indoor/ outdoor occupancy factors were taken into account to estimate the annual effective doses. The UNSCEAR 2000 report [1] revealed that, the conversion coefficient from absorbed dose in air to effective dose received by adults is 0.7 Sv Gy^{-1} and the fraction of time spent indoors and outdoors is 0.8 and 0.2 respectively. Hence, the annual effective dose equivalent (AEDE) in indoor and outdoor air is determined using equation (5):

$$AEDE\,(mS\,y^{-1}) = D \times DCF \times F_{IO} \times T \quad (5)$$

The annual effective dose external is given by equation (6) [30]:

$$AEDE_{EX}\left(mSv\,y^{-1}\right) = AEDE_{outdoor} + AEDE_{indoor} \quad (6)$$

Where F_{10} = the indoor and outdoor occupancy factors (0.8 and 0.2), DCF =dose conversion factor (0.7 Sv Gy^{-1}) and T= time (8760 h y^{-1}).

To assess the gamma ray radiation hazards due to the ^{238}U, ^{232}Th, and ^{40}K in the soil samples are achieved by calculating the following two hazard indices using equations (7) and (8) below [31]:

$$H_{ex} = C_{U238}\,1/370Bq\,kg^{-1} + C_{Th232}\,1/259Bq\,kg^{-1} + C_{K40}\,1/4810Bq\,kg^{-1} \quad (7)$$

$$H_{in} = C_{U238}\,1/185Bq\,kg^{-1} + C_{Th232}\,1/259Bq\,kg^{-1} + C_{K40}\,1/4810Bq\,kg^{-1} \quad (8)$$

where C_{U238}; C_{Th232} and C_{K40} are the activity concentrations of ^{238}U, ^{232}Th, and ^{40}K in Bq kg^{-1}.

The level of gamma radioactivity associated with different concentrations of some specific radioactive elements are evaluated by using the representative level index (RLI) as shown in equation (9) below [33],

$$RLI = 1/150\,C_{U238} + 1/100\,C_{Th232} + 1/1500\,C_{K40} \quad (9)$$

where C_{U238}; C_{Th232} and C_{K40} are the activity concentrations of ^{238}U, ^{232}Th, and ^{40}K in Bq kg^{-1}, respectively.

The activity utilization index (AUI) was calculated from equation (10) based on the dose rates in air from different combinations of ^{238}U, ^{232}Th, and ^{40}K (Bq kg^{-1}) in soil samples and applying the suitable conversion factors as [34];

$$AUI = C_{U238}f_{U238}\,1/50 + C_{Th232}f_{Th232}\,1/50 + C_{K40}f_{K40}\,1/500 \quad (10)$$

where C_{U238}; C_{Th232} and C_{K40} are the activity concentrations of ^{238}U, ^{232}Th, and ^{40}K in Bq kg^{-1} in soil samples, respectively, and f_{U238}(0.462), f_{Th232} (0.604) and f_{K40}(0.042) are the respective fractional contributions from the actual activities of ^{238}U, ^{232}Th, and ^{40}K to the total dose rate in air [6].

According to UNSCEAR [13], the activity in the bone marrow and the bone surface cells are considered as the organs of interest. Therefore, the annual gonadal dose equivalent (AGDE) due to the activities of ^{238}U, ^{232}Th, and ^{40}K is calculated as shown in equation (11) below [35],

$$AGDE = 3.09C_{U238} + 4.18C_{Th232} + 0.314C_{K40} \quad (11)$$

The excess lifetime cancer risk ($ELCR$) was calculated by using equation (12) below [15],

$$ELCR = AEDE_{outdoor} \times E_{LD} \times C_{RF} \quad (12)$$

where E_{LD} = Expected lifetime duration (70 yrs.) and C_{RF} = Fatal cancer risk factor. For stochastic effects, [29] uses a value of 0.05 for the general public [36].

Multivariate statistical analysis

The statistical software package "Statistical Program for Social Science (SPSS)" was used to identify the variation of the various parameters obtained from natural radionuclides. The multivariate statistical analyses (Pearson's correlation analysis) were performed. This was

done to understand the mutual relationships which exist among all the measured radiological parameters.

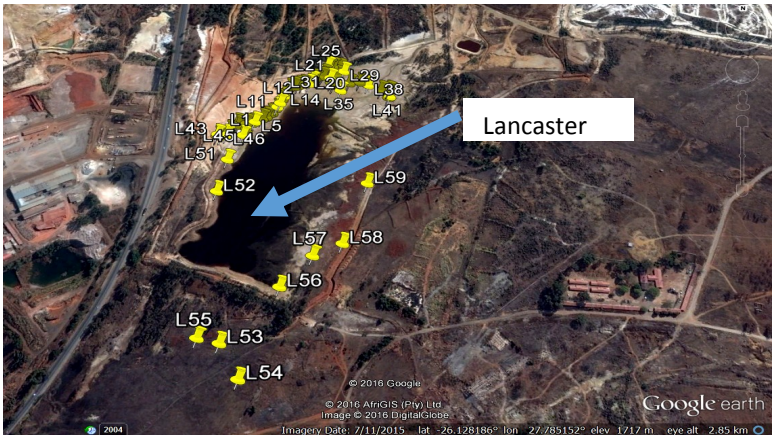

Figure 1. Soil sampling distribution around the bank and immediate surroundings of Lancaster dam.

RESULTS

The results of the activity concentrations of ^{238}U, ^{232}Th and ^{40}K are presented in figure 2 *(all locations with significant low activity concentrations values were omitted from the plots)*.

The results of the universally accepted index for analysing the radiation exposure created by the primordial radionuclides evaluated using equation (1) for some samples collected at locations closer to the dam as well as further away from the dam is show in figure 3.

Making use of the activity concentration of ^{232}Th (C_{Th232}) = 216.12 Bq kg^{-1} and activity concentration of ^{40}K (C_{K40}) = 502.00 Bq kg^{-1}, the evaluation of the dependence of the ^{238}U concentration on the measured effective dose rate (EDR) calculated using equation (4) are shown in figure 4.

The results of the mean values of external, indoor and outdoor AEDE values, the calculated hazard indices for H_{ex} and H_{in} at maximum location L1, the calculated average value of RLI and the average values of AUI and AGDE are shown in figure 5.

The Pearson correlation coefficient matrix between radionuclides and radiological parameters are presented in the form of graph as shown in figure 6.

The comparison of $^{238}U/^{232}Th$ concentration ratio in soils of present study with other countries are presented in table 1.

A comparison of radiological parameters of present study with other countries were shown in table 2.

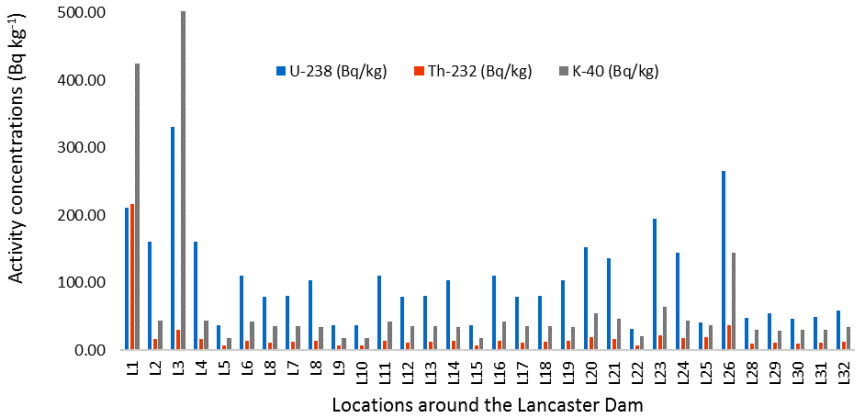

Figure 2. Activity concentrations of ^{238}U, ^{232}Th and ^{40}K.

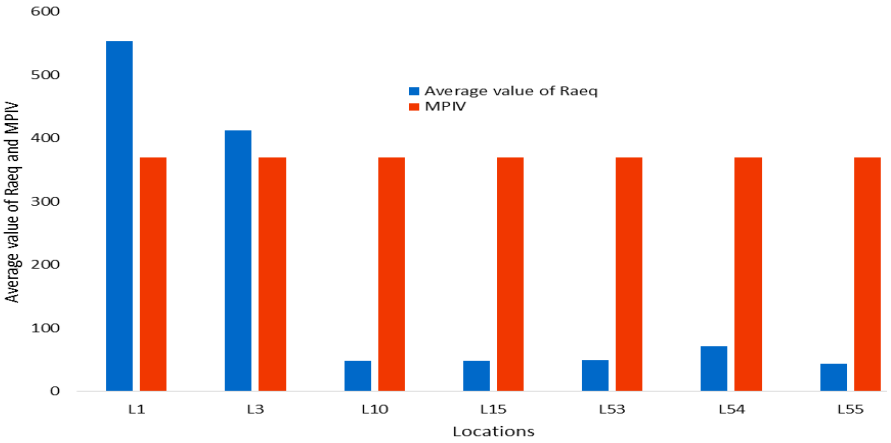

Figure 3. Evaluated mean values of Raeq and MPIV and some selected locations.

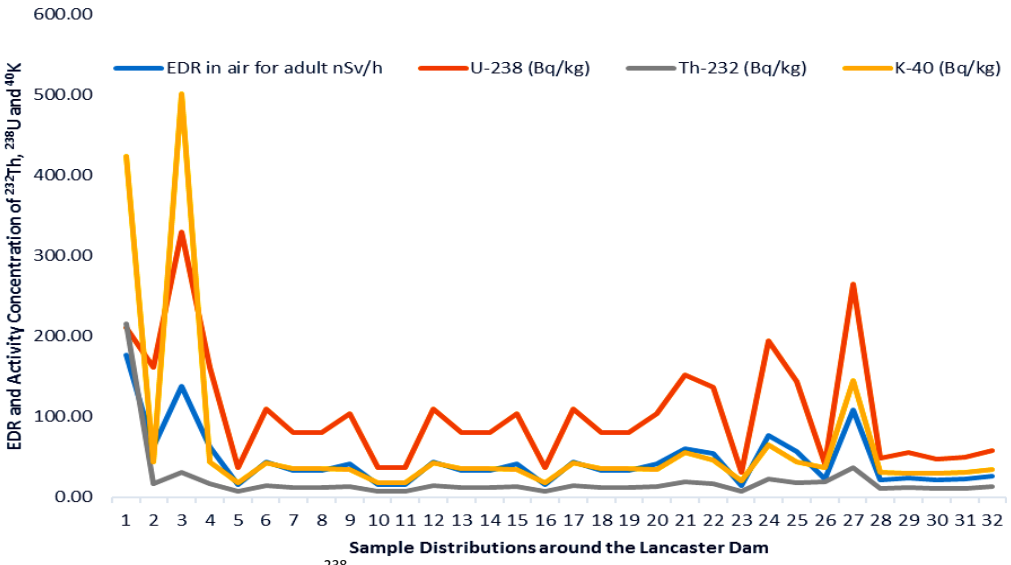

Figure 4. Dependence of ^{238}U concentration on the measured effective dose rate (EDR).

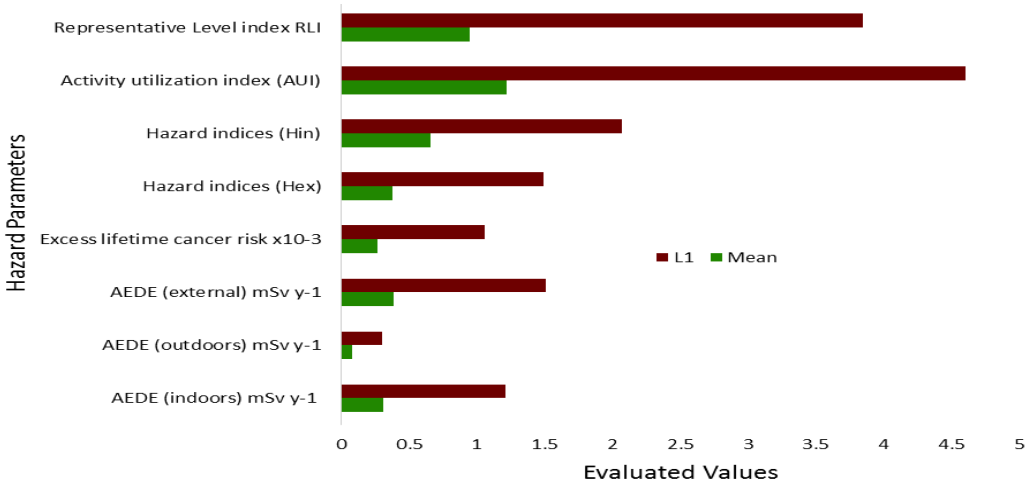

Figure 5. Evaluated Hazard index values.

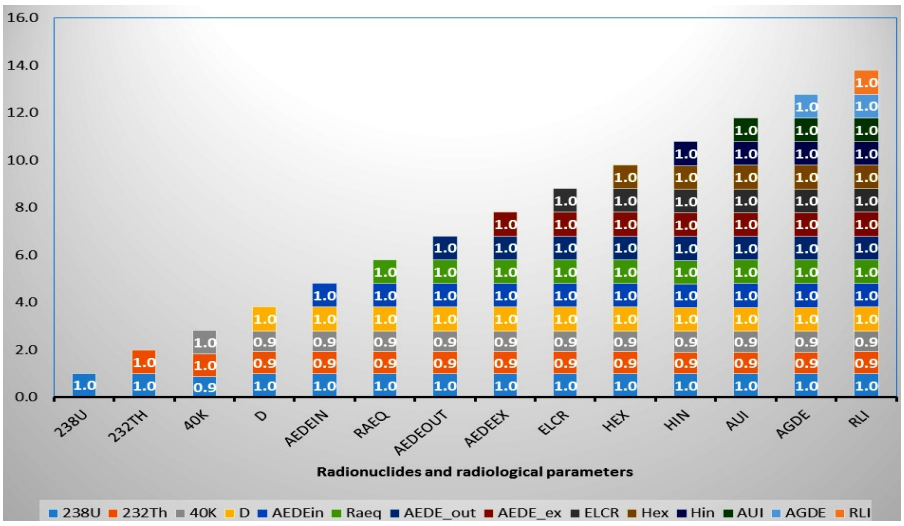

Figure 6. Pearson correlation coefficient matrix between radionuclides and radiological parameters.

Table 1. Comparison of $^{238}U/^{232}Th$ concentration ratio in soils of present study with other countries.

Country	$^{238}U/^{232}Th$	Reference
Istanbul, Turkey	0.57	(23)
Kalpakkam, India	0.09	(12)
Western Ghats	0.49	(9)
India	0.69	(24)
Algeria	1.09	(25)
Brazil	0.32	(26)
Egypt	1.11	(27)
Pakistan	0.88	(28)
Tudor Shaft (South Africa)	5.71	(14)
Worldwide	1.17	(1)
This study	6.14	Present study

Table 2. Comparison of radiological parameters of present study with other countries

Sampling site	Ra_{eq} (Bq kg^{-1})	RLI	D (nGy h^{-1})	AGDE (mSv y^{-1})	AEDE (outdoor) (mSv y^{-1})	AEDE (indoor) (mSv y^{-1})	Reference
Soil around gold mine tailings Tudor Shaft, South Africa			163.17				(15)
Sethiyathope,Tamilnadu, India		0.72	45.189	316.72	0.06	0.22	(40)
Olode mining site, Nigeria	45.07		45.7		0.032		(37)
Soils around cement factory, Nigeria			40.88		0.05		(35)
Soil from petroleum Industry, Tunisia	38.6		18.5		0.022		(5)
Soil around aluminium industry, Nigeria	134		60	420	0.084		(42)
Soil around gold mining, Nigeria	132.14		66.3	439.73	0.081		(16)
Soil around phosphate fertilizer, Mumbai, India	211						(29)
Soil, phosphate fertilizer, Egypt	126.2		67.3		0.32	1.28	(27)
Quarry products, Nigeria	310		260		0.07		(11)
Soil from oil field, Niegeria	98.5		54.6		0.25		(38)
Petroleum waste, Saudi Arabia	116.46		59.38				(39)
Uranium mining area, South India			148			0.97	(8)
Soil of industrial dumpsites, Nigeria	61.02		29.79		0.037		(16)
Worldwide		1	55	300	1		(1)
Present study	139.2	0.95	63.55	430.22	0.08	0.31	This study

DISCUSSION

As seen in figure 2, the average values of ^{238}U, ^{232}Th and ^{40}K are 99.72 ± 13.81 Bq kg^{-1}, 14.79 ± 8.70 Bq kg^{-1} and 43.07 ± 9.67 Bq kg^{-1} respectively. The world average concentration of ^{238}U, ^{232}Th and ^{40}K are 35, 30 and 400 Bq kg^{-1} respectively [13]. When compared the activity concentration of the obtained radionuclides with the world average value we found out that ^{238}U is higher by a factor of 2.85 whereas ^{232}Th and ^{40}K are lower by a factor of 2.03 and 9.20 respectively. The high value of the activity concentration of ^{238}U could be attributed to the washed-off tailings which have completely filled the former mine water reservoirs (return water dams) such as the Tudor and Lancaster dams.

As revealed in figure 2, the mean activity concentration of ^{238}U, ^{232}Th and ^{40}K were high along the locations closer to the dam and locations that were filled with water during raining season (wetland) before drying up. The wetlands act as pollution sinks with high concentration of the radionuclides and other metals. As revealed by [14, 17], uranium migrates together with other dissolved heavy metals in seepage from tailings into groundwater and further into adjacent streams. The deposits (tailings) liquefy during raining seasons, combine with liquid effluents and enter the natural surface and ground water systems. These liquids/effluents are cocktails of the direct discharges of both process and fissure water from mines, run-offs and infiltration from mine waste materials and contaminated areas, and other industrial and domestic wastewaters from the cities and towns that have developed around the Lancaster dam.

The average $^{238}U/^{232}Th$ concentration ratio in surface soil samples were compared to other countries in the world as presented in table 1. The mean ratio of this study is higher compared to Istanbul, Turkey [23], Kalpakkam, India [12], Western Ghats [9], India [24], Algeria [25], Brazil [26], Egypt [27], Pakistan [28], and Worldwide [1] and slightly above Tudor Shaft in South Africa [15].

As shown in table 2, the calculated average value of radium equivalent activity (Ra_{eq}) is 139.2 Bq kg^{-1}. This study as indicated in figure 3 shows that almost all the values from the sampling sites were low when compared to the maximum permissible index value (MPIV) of 370 Bq kg^{-1} except L1 and L3 with values of 553.16 and 412.81 Bq kg^{-1} respectively because these two locations are closer to the dam. Figure 3 revealed that as we moved away from the dam to locations L10, L15, L53, L54 and L55 we obtained low values of 48.13, 48.13, 49.12, 70.88 and 43.48 Bq kg^{-1} respectively.

As seen in table 2, the evaluated average dose rate is 63.55 nGy h^{-1} and is higher when compared to the maximum permissible level [1] of 55 nGy h^{-1}. As revealed in figure 5, some locations closer to the dam have values greater than the maximum permissible level of 55 nGy h^{-1}. As shown in figure 2, it has been realized that the activity concentration of ^{238}U is higher compared to ^{232}Th and ^{40}K and therefore ^{238}U dominate and hence the effective dose rate (EDR) will depend more on the ^{238}U as shown in figure 5.

As shown in table 2, the mean results of the indoor and outdoor AEDE values were calculated to be 0.31 and 0.08 mSv y^{-1} respectively. The worldwide average value of the annual effective dose is 0.48 mSv y^{-1} and this value is 10 % higher for children and 30% higher for infants. As revealed in figure 5, at location L1 the AEDE value recorded was 1.51 mSv y^{-1} for external and 1.21 mSv y^{-1} for indoors. These values were higher compared to worldwide average value shown in table 2 by a factor of 3.14 and 2.51 respectively.

To control the internal exposure to radon, we used the internal hazard index (H_{in}) and its short -lived products which are also dangerous to the respiratory organs [32]. As shown in figure 5, the calculated hazard indices for H_{ex} and H_{in} at maximum was found at L1 to be 1.49 and 2.07 respectively. However the average value of 0.38 for H_{ex} and 0.66 for H_{in} were obtained. The recommended limit by UNSCEAR [1] report for the hazard indices must be less than unity. It is clear from our results that the average hazard indices calculated were below the recommended value except for location L1.

As seen in figure 5, the calculated average

value of RLI for all the soil samples is 0.95 and according to [33], the maximum limit for RLI should be 1. Also from figure 5, the average values of AUI is 1.22 and for AGDE is 430.22 mSv y^{-1}. This clearly indicates that the average AGDE value of this study is above the world average value of 300 mSv y^{-1}.

Risk of cancer increases as the dose of radiation increases [33]. Exposure to one Sievert of radiation spread out over time is evaluated to increase the lifetime risk of fatal cancer in an average adult by around 4% and a 0.8% chance of hereditary defect in future offspring.

As shown in figure 5, the average value of ELCR is 2.7×10^{-4} and is lower when compared to the world average value of 3.0×10^{-4} obtained using recommended value of 0.25 mSv yr^{-1}. However at L1 the ELCR value of 1.06×10^{-3} was obtained which is 5.33 times higher compared to the world average value of 3×10^{-4}.

As shown in table 2 the comparative data found in literature for the average values of Ra_{eq}, D, AGDE, RLI were compared to this study. It was observed that all the parameters in our study were higher compared to that of Olode mining site Nigeria [37], Soil from petroleum Industry, Tunisia [5], Soils around cement factory Nigeria [34], Soil from oil field Nigeria [38] Petroleum waste, Saudi Arabia [39], Sethiyathope, Tamilnadu India [40] and Uranium mining area South India [8]. Also, our study showed lower values of D and AEDE as compared with the soils around gold mine tailings Tudor Shaft [15], Quarry products Nigeria [11], soils around gold mining Nigeria [16] and world average values [1].

The Pearson correlation coefficient matrix between radionuclides and radiological parameters show a strong positive correlation coefficient of 0.9 to 1.0 as shown in figure 6. Hence, these relationships show that ^{238}U, ^{232}Th radionuclides and ^{40}K contribute to the emission of gamma radiation in all the locations in the study area. Contrary to the study of [42], these radiological parameters have a relatively strong correlation with ^{40}K in this study.

CONCLUSION

The activity concentration of ^{238}U was higher by a factor of 2.85 whereas ^{232}Th and ^{40}K were lower by a factor of 2.03 and 9.20 respectively when compared to the world average value. The Pearson correlation analysis matric shows a positive correlation with a value of 0.9 to 1.0 indicating that the three nuclides contribute to the emission of gamma radiation in all the locations. The mean value of ELCR is 2.7×10^{-4} and is below the world mean value except location L1 which is 5.33 times higher compared to the world average value of 3×10^{-4}. The studied $^{238}U/^{232}Th$ ratio was almost six times higher compared with different countries of the world.

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***Conflicts of interest*: Declared none.**

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