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Determination of Specific Activity of ^{238}U, ^{232}Th and ^{40}K and Radiological Hazard Assessment of Tuomo River Sediments in Burutu, Delta State, Nigeria

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ABSTRACT: The activity concentrations of ^{238}U, ^{232}Th and ^{40}K in sediments samples from Tuomo river in Burutu, Delta State, Nigeria have been determined using gamma spectrometry NaI(TI) detector. The concentrations of ^{238}U, ^{232}Th and ^{40}K in the sediments range from 18.69±1.88–77.99±4.35 $Bqkg^{-1}$, 59.67±11.85–149.03±23.97 $Bqkg^{-1}$ and 580.19±3.89–873.04±4.86 $Bqkg^{-1}$ respectively. The mean values were found to be above their respective world average values. Radiological risk assessments were carried out to ascertain the possible radiological effects associated with the use of the sediments. The results for the radiological hazard parameters revealed that the sediments are in tandem with recommended safe limits. Therefore, the use of Tuomo river sediment for purpose of building and other human needs is generally of negligible radiological concern for human health.

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Human beings are constantly exposed to ionizing radiations which are naturally present in different environmental media. Natural environmental radioactivity arises mainly from primordial radionuclides, such as ^{40}K, and the radionuclides from the ^{232}Th and ^{238}U series, and they exists in various geological formations like soils, rocks, plant, sand, water, air and building materials in amounts that vary significantly, depending on the geological and geographical features of the region (Gholami *et al.*, 2011; SureshGandhi *et al.*, 2014). Gamma radiation emitted from naturally occurring radionuclides is the main external source of radiation to human populace.

Assessment of activity concentrations of natural radionuclides in the environmental matrices has great importance because natural radiation is the largest contributor to the external dose to the world population (UNSCEAR, 2000). Estimating the radiation doses and radiological hazards associated with detected natural radionuclides from different environmental samples is paramount in examining the health exposure to a populace. Studies of natural radioactivity are thus necessary not only because of their radiological impact, but also act as excellent biochemical and geochemical tracers in the environment (Singh *et al.*, 2003). Such studies are also useful for both assessment of external public dose rates and also serve as a guide in assessing the performance of epidemiological studies of diseases traceable to radiation, as well as keeping reference data records to ascertain possible changes in the environmental radioactivity due to nuclear, industrial, and other human activities (Tzortzis *et al.*, 2004; Ugbede, 2019).

The present study is one of such aimed at determining and quantifying the natural radionuclides present in sediments of Tuomo River in Burutu local government area of Delta State, Nigeria. The study also seeks to assess the radiological risks associated with the use of the sediments, possibly as construction materials. It is hoped that the result of the study will form part of the radiological data already existing in the country and also provide a useful baseline data for further studies into the radiation levels of the immediate environments.

MATERIALS AND METHODS

Samples collection and preparation: Five sediment samples of about 1.0 kg were collected at different locations from Tuomo river in Burutu local government area of Delta State, Nigeria into a polyethene bag and labeled accordingly to avoid mix up. The sampling points were carefully chosen to represent areas where human populations are involved in various activities (Onjefu *et al.*, 2017). The samples were transported to the laboratory and air dried at

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room temperature for five days. Thereafter, samples were homogenized and pulverized by grinding and sieving through a 2 mm mesh sieve. The homogenized sediments were then oven-dried at 105 ºC until they attained constant weight for 12 hours. Two hundred grams (200 g) of each sample was measured into cylindrical plastic containers of uniform size and sealed for 30 days before gamma spectrometric analysis, in order to allow for secular equilibrium between radon and its short lived decay products.

Gamma-ray spectrometry and activity measurements: Gamma spectrometric analysis using 7.6 cm × 7.6 cm NaI(Tl) detector (Model 802 series) by Canberra Inc. was employed in determining the activity concentration of natural radionuclides in the samples. The detector is adequately shielded with 15 mm lead shield which reduced the background by a factor of about 95%. The output of the detector was connected to a Canberra series 10 plus Multichannel Analyzer (MCA) with computer-based MAESTRO (ORTEC) program used for data acquisition and analysis of the gamma spectra. Energy and efficiency calibrations of the gamma-counting systems were done for various energies of interest in the selected sample geometry using standard IAEA sources. The calibration procedures described in IAEA (1989) were followed. The background count was determined by counting an empty container having same dimensions as the one containing the samples and subtracted from the gross count. The counting time of each sample and background was set at 36000 s to obtain the gamma spectrum with good statistics. 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. After correcting for background and Compton contribution, the activity concentrations of ^{238}U, ^{232}Th, and ^{40}K were determined using the gamma ray photo peaks corresponding to energy of 1120.3 keV (^{214}Bi), 911.21 keV (^{228}Ac) and 1460.82 keV (^{40}K) respectively.

Estimation of radiological hazards parameters: The radiological hazard parameters were calculated as follows:

Radium equivalent activity, Ra_{eq}: The radium equivalent is the weighted sum of activities of ^{238}U, ^{232}Th and ^{40}K. Ra_{eq} was calculated using equation 1 as defined by Beretka and Mathew (1985).

$$Ra_{eq} = A_u + 1.43A_{Th} + 0.077A_k \qquad (1)$$

Where A_u, A_{Th} and A_k are the activity concentrations of ^{238}U, ^{232}Th and ^{40}K ($Bqkg^{-1}$) respectively.

Hazard index: To account for the external and internal exposure of the radiation emanating from ^{238}U, ^{232}Th and ^{40}K in the sediment samples, the external hazard index (H_{ex}) and internal hazard index (H_{in}) were evaluated using equations 2 and 3 respectively (Sivakumar *et al*, 2014; Darwish *et al*, 2015).

$$H_{ex} = \frac{A_u}{370} + \frac{A_{Th}}{259} + \frac{A_k}{4810} \leq 1 \qquad (2)$$

$$H_{in} = \frac{A_u}{185} + \frac{A_{Th}}{259} + \frac{A_k}{4810} \leq 1 \qquad (3)$$

Where A_u, A_{Th} and A_k are the activity concentrations of ^{238}U, ^{232}Th and ^{40}K in sediments samples respectively. The prime objective of H_{ex} and H_{in} is to limit the radiation dose to dose equivalent limit of 1 $mSvy^{-1}$ (ICRP, 1990). Thus, their values must be less than unity for the radiation hazards to be negligible.

Absorbed dose rate (D_R):The radiation absorbed dose rate in air at 1.0 m above the ground level was evaluated in units of $nGyh^{-1}$ by applying conversion factors of 0.462, 0.604 and 0.0417 for ^{238}U, ^{232}Th and ^{40}K respectively (Beretka and Mathew, 1985; UNSCEAR, 2000) as follows:

$$D_R = 0.462A_u + 0.604A_{Th} + 0.0417A_k \qquad (4)$$

Where A_u, A_{Th} and A_K are the activity concentration of ^{238}U, ^{232}Th and ^{40}K respectively.

Annual effective dose (AED): The outdoor annual effective dose in $mSvy^{-1}$ was calculated using equation 5 as follows;

$$AEDE = D_R \times 8760 \times 0.2 \times 0.7 SvGy^{-1} \times 10^{-6} \qquad (5)$$

Where D_R is the absorbed dose rate in air in $nGyh^{-1}$, 8760 is the total hours in a year; 0.7 $SvGy^{-1}$ is the dose conversion factor from absorbed dose in air to the effective dose and 0.2 is the occupancy factor for outdoor exposure as recommended by UNSCEAR (2000).

Excess Lifetime Cancer Risk (ELCR): The excess lifetime cancer risk is used in radiation protection assessment to predict the probability of cancer development by an individual over a lifetime due to low radiation exposure level (Darwish *et al*, 2015; Ugbede and Benson, 2018). Based upon calculated values of annual effective dose, the ELCR was estimated using equation 6 (Darwish *et al.*, 2015; Ugbede and Benson, 2018).

$$ELCR = AEDE \times DL \times RF \quad (6)$$

Where AEDE is the annual effective dose equivalent, DL is average lifetime duration assumed to be 70 years (Isinkaye and Emelue, 2015; Ugbede and Benson, 2018) and RF represents fatal cancer risk factor per Sievert taken to be 0.05 Sv^{-1} as contained in ICRP-103 (2007) and ICRP-106 (2008) recommendations.

Annual gonadal dose equivalent (AGDE): It is a measure of the genetic significance of the yearly dose equivalent received by the population's reproductive organs (gonads) (Morsy *et al.*, 2012; Ramasamy *et al*., 2014). In this study, AGDE due to the specific activities of ^{238}U,^{232}Th and ^{40}K was calculated using the following formula (Ravisankar *et al*., 2014).

$$AGDE = 3.09A_u + 4.18A_{Th} + 0.314A_k \quad (7)$$

Activity utilization index (AUI): In order to facilitate the calculation of dose rates in air from different combinations of the three radionuclides in sediments, an activity utilization index was constructed and it is given by the following equation (Sivakumar *et al*, 2014; Ravisankar *et al*., 2014; Isinkaye and Emelue, 2015)

$$AUI = \frac{A_u}{50\,Bqkg^{-1}} f_U + \frac{A_{Th}}{50\,Bqkg^{-1}} f_{Th} + \frac{A_k}{500\,Bqkg^{-1}} f_K \quad (8)$$

Where A_u, A_{Th} and A_K are the activity concentration of ^{238}U, ^{232}Th and ^{40}K respectively and f_U $(= 0.462)$, f_{Th} $(= 0.604)$and f_K $(= 0.041)$are the fractional contributions to the total dose rate in air due to gamma radiation from the actual concentrations of the radionuclides.

RESULTS AND DISCUSION

Specific activity concentrations of ^{238}U, ^{232}Th and ^{40}K in sediment samples: The activity concentrations of ^{238}U, ^{232}Th and ^{40}K in the sediment samples are presented in Table 1. The results showed that these natural radionuclides were present in all the samples at varying amount. The activity concentrations of ^{238}U, ^{232}Th and ^{40}K vary from 18.69±1.88 to 77.99±4.35 $Bqkg^{-1}$, 59.67±11.85 to 149.03±23.97 $Bqkg^{-1}$ and 580.19±3.89 to 873.04±4.86 $Bqkg^{-1}$ with mean values of 43.89±22.68 $Bqkg^{-1}$, 99.21±37.73 $Bqkg^{-1}$ and 793.99±121.82 $Bqkg^{-1}$ respectively. Potassium–40 has the highest activity in the sediment samples followed by ^{232}Th and ^{238}U (that is ^{40}K >^{232}Th >^{238}U). Similar trend has also been observed in river sediments of Northern Pakistan (Qureshi *et al*, 2014). The activity concentrations varied with sample location, because the river bottom exhibit large variations in chemical and mineralogical properties and rare-earth elements (Ramasamy *et al*, 2011).The wide variation observed in the activity concentration values can also be attributed to the physical, chemical and geochemical properties of the marine environment (Khatir *et al.*, 1998; El Mamoney and Khater, 2004; SureshGandhi *et al.*, 2014). The activity of ^{232}Th was found to be higher than that of ^{238}U in all the sediment samples due to the low geochemical mobility and insoluble nature of thorium in water (Suresh et al, 2011; Isinkaye and Emelue 2015). The results also revealed that the mean activity concentrations of ^{238}U, ^{232}Th and ^{40}K are higher when compared with the world average values of 35, 30 and 400 $Bqkg^{-1}$ respectively (UNSCEAR, 2000). The total (^{238}U + ^{232}Th + ^{40}K) activity concentration ranges from 699.59 to 1066.92 $Bqkg^{-1}$ with an average of 937.09 $Bqkg^{-1}$. This value is higher than that of river sediments of Northern Pakistan (Qureshi *et al*, 2014). The percentage contribution of ^{238}U, ^{232}Th and ^{40}K to the total activity of the sediments are 4.68%, 10.59% and 84.73% respectively with ^{40}K displaying the highest contribution. This is expected because ^{40}K is a naturally occurring radionuclide which abounds in the earth crust (Ibikunle *et al*, 2016). The variations of activity concentrations of the detected radionuclides with sediments samples are shown in figure. 1.

Table 1: Activity concentration of ^{238}U, ^{232}Th and ^{40}K in Tuomo River sediments

Sample code	Activity concentration $Bqkg^{-1}$		
	^{238}U	^{232}Th	^{40}K
S1	77.99±4.35	121.40±20.13	867.53±4.98
S2	31.47±2.39	149.03±23.97	816.89±4.71
S3	18.69±1.88	100.71±17.43	580.19±3.89
S4	38.61±2.78	65.24±12.88	873.04±4.86
S5	52.68±3.34	59.67±11.85	832.31±4.83
Range	18.69±1.88 - 77.99±4.35	59.67±11.85 - 149.03±23.97	580.19±3.89 - 873.04±4.86
Mean	43.89±22.68	99.21±37.73	793.99±121.82

The mean concentration of the radionuclides reported here are higher than values reported in sediments from Tema Habour, Ghana (Botwe *et al*, 2017), Ras Tanura, Saudi Arabia (El-Taher *et al*, 2018) and Ogun river, Nigeria (Jibiri and Okeyode, 2011). The mean value of 793.99 $Bqkg^{-1}$ for ^{40}K in this study is lower than 1023 $Bqkg^{-1}$ value found in sediments of Oguta Lake, Nigeria (Isinkaye and Emelue 2015). Mean concentration of ^{232}Th found in sediments of North east coast of Tamilnadu, India (SureshGandhi *et al*, 2014) is higher than the value found in this present study. Figures 2, 3 and 4 show the frequency histogram graph of ^{238}U, ^{234Th} and ^{40}K respectively.

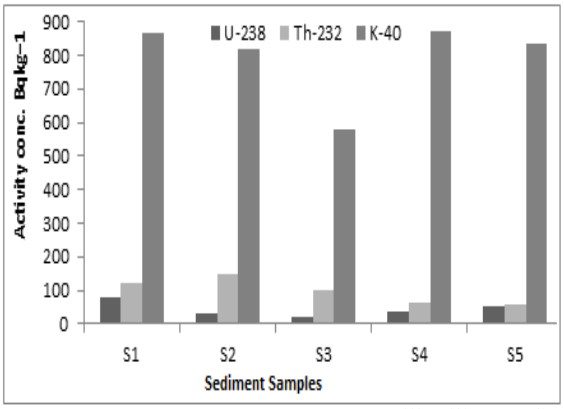

Fig 1: Comparison of activity concentration of ^{238}U, ^{232}Th and ^{40}K of Tuomo sediment samples

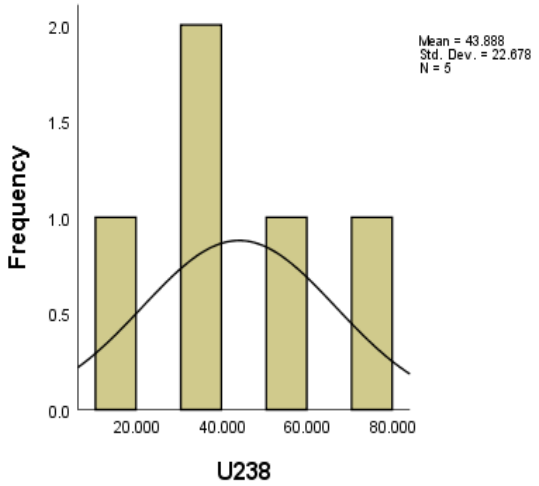

Fig. 2: Frequency histogram of ^{238}U in Tuomo sediment

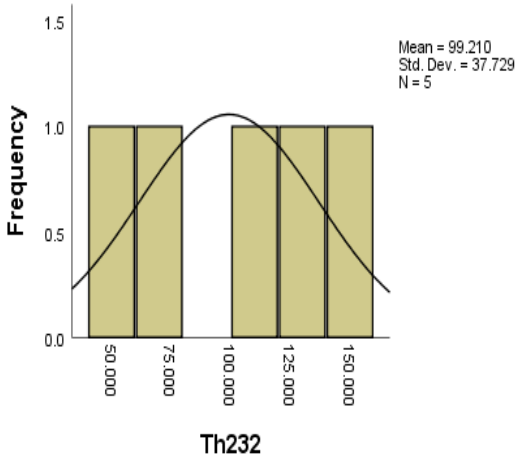

Fig. 3: Frequency histogram of ^{232}Th in Tuomo sediment

Radiological hazards assessment: Radiological hazard assessment using well established radiation hazard parameters was carried out to ascertain the health effects that might arise with the use of Tuomo sediments possibly as building materials by human populace. The results are shown in Table 2.

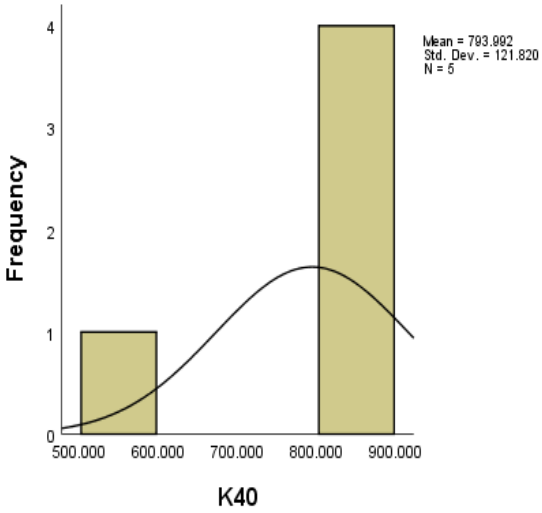

Fig. 4: Frequency histogram of ^{40}K in Tuomo sediment

In nature the concentration of radionuclides are not uniform so it is necessary to express the radiological effects by a single term which takes into account the hazards associated with all the primordial radionuclides. The ^{238}U, ^{232}Th and 40K emit different gamma doses even if present in the same amount in any material. The Radium equivalent activity, Ra_{eq} is a common radiological index used to compare the specific activities of materials containing ^{238}U, ^{232}Th and ^{40}K by a single quantity. It is the weighted sum of activities of ^{238}U, ^{232}Th and ^{40}K in the sediment samples and it was evaluated based on the assumption that 370 $Bqkg^{-1}$ of ^{238}U (or ^{226}Ra), 259 $Bqkg^{-1}$ of ^{232}Th, and 4810 $Bqkg^{-1}$ of ^{40}K produce the same gamma radiation dose rate (SureshGandhi *et al*, 2014; Qureshi *et al*, 2014). As shown in Table 2, the values of Ra_{eq} range from 199.13 (sample S4) to 318.39 $Bqkg^{-1}$ (sample S1) with mean value of 206.48 $Bqkg^{-1}$. The maximum value of Ra_{eq} must be less than 370 $Bqkg^{-1}$which corresponds to the dose limit of 1.0 mSv for the general population (UNSCEAR, 2000).It is seen that for all the sediment samples analyzed including the mean value, the Ra_{eq} values are well below the permissible limit of 370 $Bqkg^{-1}$. Ra_{eq} values in this study are comparable to values of 52.59–377.53$Bqkg^{-1}$ with an average of 205.67±70.84 $Bqkg^{-1}$ obtained in Oguta lake sediment in Owerri, Nigeria (Isinkaye and Emelue, 2015), and 75.54–393.29 $Bqkg^{-1}$ obtained in rivers sediments of Northern Pakistan (Qureshi *et* al, 2014). But however, higher than mean value of 84.57 $Bqkg^{-1}$ for east coastal sediments of Tamilnadu, India (Ravisankar *et al*., 2014). The variations of Ra_{eq} with respect to sample location are shown in Figure 5.

Table 2: Estimated radiological hazards parameters in sediment samples of Tuomo River

Sample code	Ra_{eq} $Bqkg^{-1}$	H_{ex}	H_{in}	D_R $nGyh^{-1}$	AEDE $mSvy^{-1}$	ELCR $\times10^{-3}$	AUI	AGDE $mSvy^{-1}$
S1	318.39	0.860	1.071	145.53	0.178	0.623	2.258	1.021
S2	307.48	0.830	0.915	138.62	0.170	0.595	2.158	0.977
S3	207.38	0.560	0.610	93.66	0.115	0.403	1.437	0.661
S4	199.13	0.538	0.642	93.65	0.115	0.403	1.216	0.666
S5	202.10	0.546	0.688	95.09	0.117	0.410	1.276	0.674
Mean value	206.48	0.667	0.785	113.31	0.139	0.487	1.669	0.799

To account for the external and internal exposure of the radiation emanating from ^{238}U, ^{232}Th and ^{40}K in the sediment samples, the external hazard index (H_{ex}) and internal hazard index (H_{in}) were evaluated. The prime objective of H_{ex} and H_{in} is to limit the radiation dose to dose equivalent limit of 1 $mSvy^{-1}$(ICRP, 1990). The external exposure is caused by direct gamma radiation whereas internal exposure is caused by the inhalation of radon (^{222}Rn), thoron (^{220}Rn) and their short-lived decay products present in air as a result of the decay of ^{238}U and ^{232}Th. The gaseous short-lived decay product of ^{226}Ra called Radon (^{222}Rn), poses a threat to the respiratory organs. The values of the hazard indices H_{ex} and H_{in} must be less than unity for the radiation hazard to be negligible. The results for H_{ex} in Tuomo sediments range between 0.538 and 0.860 with average of 0.667 and that for H_{in} are between 0.610 and 1.071 with average of 0.785. Only one sample (S1) was found to have H_{in} value greater than the permissible limit. The average value of H_{ex} and H_{in} in the present study is lower when compared respectively to values of 1.87 and 2.9 in Malaysia (Almayahi *et al.*, 2012) and 0.70 and 1.18 in Henties Bay, Namibia (Onjefu *et al.*, 2017), but higher than values of 0.56 and 0.67 in Oguta lake, Nigeria (Isinkaye and Emelue, 2015) and 0.228 and 0.233 in east coastal of Tamilnadu, India (Ravisankar *et al*., 2014).Generally, the mean values are less than the permissible limits of 1, indicating that the radiation hazard in the sediment is insignificant and can thus be used as building materials without restriction.

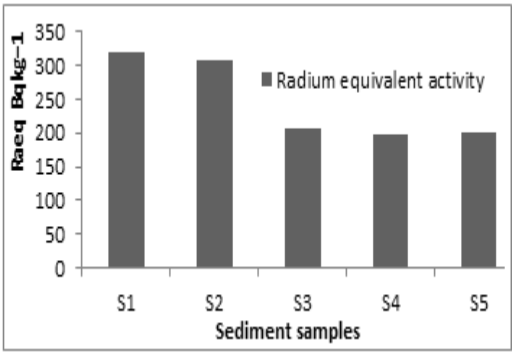

Fig 5: Radium equivalent activity of Tuomo sediment samples

In order to further assess the radiological risk of external exposure to radiation arising from naturally radionuclides ^{238}U, ^{232}Th and ^{40}K, the absorbed dose rate (D_R) was determined. The absorbed dose rate (in $nGyh^{-1}$) in air at 1 m above the ground surface was evaluated by applying conversion factor of 0.462, 0.604 and 0.0417 for ^{238}U, ^{232}Th and ^{40}K respectively (Beretka and Mathew, 1985; UNSCEAR, 2000; Onjefu *et al.*, 2017). The outdoor annual effective dose equivalent (AEDE) associated with the D_R was also estimated using a dose conversion factor of 0.7 $SvGy^{-1}$ and an occupancy factor of 0.2 as recommended by UNSCEAR (2000). The D_R which depends on the concentrations of various radionuclides in the sediment samples range from 93.65 to 145.53 $nGyh^{-1}$ with average value of 113.31 $nGyh^{-1}$. These values are above the allowed maximum value of 59 $nGyh^{-1}$ UNSCEAR (2000). The high activity concentration found in ^{40}K contributed significantly to the elevated D_R in the sediments of Tuomo river. The AEDE values were found to range from 0.115 to 0.178 $mSvy^{-1}$ with an average value of 0.139 $mSvy^{-1}$ which is within permissible limit of 1.0 $mSvy^{-1}$ (UNSCEAR, 2000; ICRP, 2007). This is an indication that the ionizing radiation doses that may be derived from external radiation from short-lived uranium and thorium progenies are within the internationally recommended annual effective dose threshold level thus making the radiation effects insignificant over a period of one year.

Direct interaction of gamma radiation with living cells could result in cell death or genetic mutation of the cell. United Nations Scientific Committee on Effects of Atomic Radiation (UNSCEAR, 2000) considered the gonads, bone marrow and bone surface to be the organs of importance since they are active. The annual gonadal dose equivalent (AGDE) is used to measure the genetic significance of the yearly dose equivalent received by the population's reproductive organs (gonads) (Morsy *et al.*, 2012). The calculated AGDE values range from 0.661 to 1.021 $mSvy^{-1}$ with an average of 0.799 $mSvy^{-1}$. The present average value is less than the recommended value of 1.00 $mSvy^{-1}$ (UNSCEAR, 2000) indicating the negligible effects of radiations emanating from the detected radionuclides

in Tuomo sediments. Sample S1 however have AGDE value slightly higher than the limit. Our average AGDE value slightly differs from that of 701μSvy^{-1} found in Oguta lake sediments (Isinkaye and Emelue, 2015). The excess lifetime cancer risk which predicts the probability of cancer development by an individual over a lifetime as a result of continuous low radiation exposure was also calculated based on lifetime duration of 70 years (Isinkaye and Emelue, 2015; Ugbede and Benson, 2018). The result ranges from 0.403×10^{-3} to 0.623×10^{-3} with an average of 0.487×10^{-3}. The average value is 67.93% higher than the world average value of 0.290×10^{-3}. This value of ELCR is a direct consequence of the external dose levels of radiation derived from the detected radionuclides present in the sediments. Another parameter called activity utilization index (AUI), is usually computed in radiological health assessment studies in order to estimate the dose rates in air from different combinations of the various radionuclides present in sediments (Isinkaye and Emelue, 2015). The calculated values for AUI vary from 1.216 (sample S4) to 2.258(sample S1) with an average of 1.669. Apart from samples S1 and S2, the values including the mean value show that AUI are less than 2, which corresponds to an annual effective dose of <0.3 mSvy^{-1} (Sivakumar *et al.*, 2014).This result further indicates that the Tuomo river sediments are safe for use as building materials and other human needs.

Conclusion: Gamma spectrometric study of activity concentrations of ^{238}U, ^{232}Th and ^{40}K in sediments of Otuomo River had been studied. The results had shown that these radionuclides are present in all the samples at varying amount. The average activity of ^{238}U, ^{232}Th and ^{40}K in all the samples are above their respective world average value. The results for the radiological hazards assessment are in tandem with recommended safe limits. Therefore, the use of the river sediment for all purposes of building and other human needs is generally of negligible radiological concern for human health.

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