Nigerian Research Journal of Chemical Sciences, Vol. 6, 2019

Radiometric and Spatial distribution of natural radionuclides concentrations and excessive lifetime cancer risks in sediments from selected rivers in Ilobi and Erinja communities, Southwest, Nigeria

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ABSTRACT

The concentration and spatial distribution of naturally occurring radionuclides ^{238}U, ^{232}Th and ^{40}K in the river sediments in Ilobi and Erinja communities were assayed using well calibrated (HPGe detector) γ–spectrometry in order to assess the potential radiological health hazards and lifetime cancer risks associated with the use of the river sediments. The radio-analytically calculated ranges of activity concentrations of ^{238}U, ^{232}Th and ^{40}K were $0.0 \leftrightarrow 166.8 \pm 19.5 Bqkg^{-1}$, $0.0 \leftrightarrow 17.9 \pm 6.7 Bqkg^{-1}$ and $181 \pm 3.3 \leftrightarrow 814 \pm 29.4 Bqkg^{-1}$ respectively. The mean activity concentrations of ^{238}U and ^{40}K exceeded the typical admissible world average values. To assess the complete radiological hazards of the river sediments, ten radiological and health hazard indices were calculated. The results indicated that the average value of each radiological hazard parameter were below the world average value reported in UNSCEAR except annual gonadal dose equivalent and internal hazard index whose mean value exceeded their recommended limits. It follows therefore that there is a fingering potential radiological hazard directly associated with the sediments from those locations. Sediments from a specific location in both communities have the ELCR values higher than the world average value, which is suggestive of its likelihood of inducing cancers over a period of exposure to humans. The statistical treatment result shows that the distribution of the radionuclides (^{238}U & ^{232}Th) was asymmetrical and peaked with an observed flat distribution in ^{40}K level in the sediments. Extensive research needs

to be carried out on Okooko River in Ilobi community as its sediment shows consistent higher concentration in both radionuclides and radiological hazard indices.

Keywords: cancer risks, radiological health hazards, radiological indices, radionuclides

INTRODUCTION

Natural radionuclides, of recent, are a subject of discussion. It has been found to be widely spread in the earth environment and in various geological formations such as soils, rocks, water, sediment, air and in building materials [1]. Radionuclides are unstable nuclides and their decay series emits harmful radiations such as alpha, beta particles and gamma rays [2]. Exposure to gamma radiation leads to the damage of living cells and lung tissues [3]. While artificial radionuclides emanate from nuclear weapon tests, nuclear accidents, medical and industrial applications, etc., about 87 % of the radiation doses received by humans are from natural radiation sources, which come from the naturally occurring radioactive isotopes of ^{238}U and ^{232}Th and their progeny as well as ^{40}K [4,5]. A few building materials that contain more radionuclides may contribute to excessive outdoor exposure to the open atmosphere where most humans spend about 20 % of everyday life, and within the building, it causes indoor exposure due to the gamma rays (radiating from the building material) and alpha radiation (from the decay of radon) where we spend about 80 % of everyday life [6]. In addition to being a major source of radiation exposure to aquatic biota, sediments, act as a medium of migration for the transfer of radionuclides in the aquatic environment. The sediments deposited at the bottom of rivers and lakes, most frequently, consist of sand and gravel of different grain sizes, which are very valuable for building constructions [1]. Aside the naturally present radionuclides, external orchestrated concentrations referred to technologically enhanced natural radiation (TENAR) by [7], from soil weathering, agricultural lands, disposal of radioactive waste materials [8], mining (especially uranium and thorium), phosphate fertilizers manufacture, agricultural applications, coal combustion, cement production, street construction and other human activities have produced and redistributed increasing amounts of radioactive matter leading to a considerable contribution to the radio-ecological pollution [9] in the riverine bodies by the run-off of rainwater and streams. It is known that long-term exposures to radioactivity and inhalation of

radionuclides have serious health effects such as chronic lung cancer and leukemia especially from within and without buildings constructed using such sediment loaded with NORMs (Naturally occurring radionuclide materials). In addition, the long-term exposure to uranium and radium through inhalation has several health effects as chronic lung diseases, acute leucopoenia, anemia and necrosis of the mouth while Thorium exposure can cause lung, pancreas, hepatic, bone, kidney cancers and leukaemia [10]. Radioactive isotopes concentration in soil is an indicator of radioactive accumulation in the environment, which impacts humans, plants and animals. Naturally occurring radioactive materials generally contain terrestrial origin radionuclides (primordial radionuclides), left over since the creation of the earth [11]. These rivers are of enormous importance to the local population resident around the basin as a source of utility water, fish, sand for building construction, tourism and also as an outlet for sewages. Imperatively therefore, gamma dose rate and radionuclides activity concentrations should be monitored as the knowledge of the level of natural radioactivity and other radiological indices in the building materials would help assess the possible radiological hazards and excessive life cancer risk probability to human health and to develop standards and guidelines for the use and management of these materials.

Hence, the objective of this study is to evaluate the natural radionuclides concentration and excessive lifetime cancer risks probability of the river sediments in Ilobi and Erinja communities Southwest, Nigeria.

MATERIALS AND METHODS

Description of the study area

Ilobi and Erinja are towns in Yewa South Local Government Area of Ogun state, Nigeria, within the latitude 6º44’00”N and 6º45’00”North of the Equator and longitude 3º3′00″E and 3º2′0″ East of the Greenwich Meridian (Fig. 1). Ilobi and Erinja are sleepy neighboring towns between Ilaro and Owode-Yewa. The inhabitants are mainly farmers, traders and sand dredgers. There is abundance of mineral resources such as limestone, shale, e.t.c. which are yet to be exploited. The sediments of its rivers are excavated only for building constructions. The small hydraulic structure and barrages were constructed for drinking and agriculture purposes, respectively on the study area. On both sides of the bank of these rivers, so many residential buildings and some

industries are situated. None of the industries have proper and controlled outlet. The discharged wastes and toxic metals from the industries and residences are directly let out into the river. Also along the riverbank, a lot of agricultural lands are available, where excess chemical fertilizers and pesticides are washed into the river [12]. These are parts of the main factors for enrichment of pollutants in the study area. The graph is shown below.

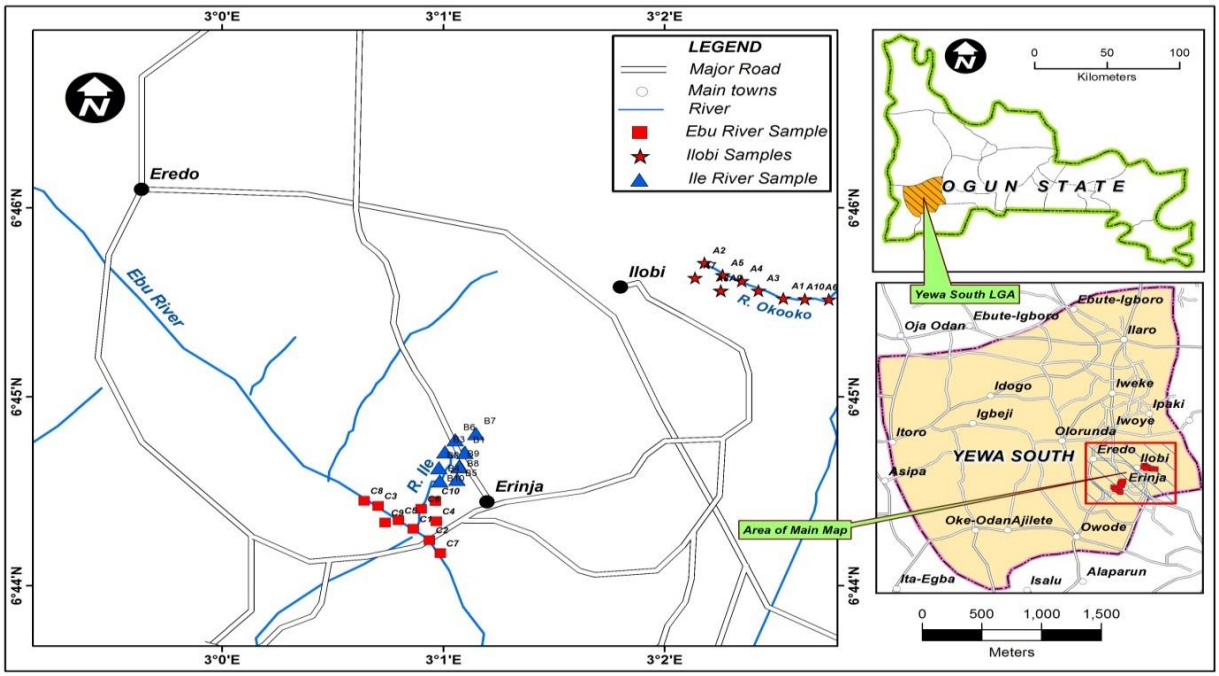

Fig 1: Map of the study area showing sample locations

Sample coding

The sediment samples were coded as follows in order to prevent identification error. The sample codes consist of one alphabet and one number. The alphabet connotes the study area of each river and the number stands for each point where the sediment samples were taken. A_1 to A_{10} represents Okooko River sediment in Ilobi Community, B_1 to B_{10} for Ile River and C_1 to C_{10} for Ebu River sediment both in Erinja Communities. A stands for Ilobi community river sediments, while B and C were from Erinja community river sediments.

Sample collection and preparations for radiochemical analysis

A total of thirty sediment samples were obtained from three different rivers in the study area using a flat stainless steel auger. Collected samples were introduced into a stainless sieve to drain

the water content, which were then transferred into a polythene bag and were labeled accordingly. The first ten sediment samples labeled A were collected from Okooko river in Ilobi, while the other twenty samples were collected from the two rivers (Ile and Ebu) in Erinja communities. Each river was segmented into four sections. In each section, two sediment samples were collected and the other two at randomly to make ten samples at each river. All sediment samples were collected at a depth of about 10 cm (using a meter-rule) and then taken to the laboratory in labeled polythene bags where they were air-dried until a fairly constant dry weight was obtained. The samples were pulverized by further grinding and filtered through 2 mm mesh sieve. About 500 g of pulverized sediment samples were subsequently measured using an analytical weighing balance with a precision of ±0.01g and packed into a polythene bag. These samples were safely conveyed to National Institute of Radiation Protection and Research, University of Ibadan, South-West Nigeria. At the laboratory, the plastic were hermetically sealed with adhesive tape and kept for 30 days to ensure that the parent and daughter nuclide in the samples were at secular equilibrium between radium and its gaseous decay progenies. At the end of the 4 weeks in-growth period, the samples were subjected to gamma-ray spectroscopy counting.

Radioactivity measurements

System used for measurements

Activity concentrations ^{238}U, ^{232}Th and ^{40}K were measured by high resolution, low background γ-ray spectrometer with sodium iodide detector (NaI) detector with a relative efficiency of 20.2%. The spectrometer was tested for its linearity and calibrated for energy and efficiency using the well calibrated standard gamma source obtained from an International Atomic Energy Agency (IAEA), laboratories, Vienna, Austria [13]. Efficiency is the measure of percentage of radiation at a given detector detect from the overall yield that is emitted from the source into a solid angle of usually 4π in the photo peaks [14]. Accuracy of efficiency calibration of detector is necessary to obtain the high precision measurements with radioactive samples. The resolution of the detector is 7.5% at 0.662 MeV of 137Cs. This resolution is capable of distinguishing the gamma ray energies of interest in the study. All the samples were counted for 36,000 s in order

to obtain good statistics for ^{238}U, ^{232}Th and their daughter products and ^{40}K. Also measurements were repeated at intervals for quality assurance purposes as well as to ascertain the stability of the measuring system. The background radiation due to the naturally occurring radionuclides in the environment around the detector was measured using an empty plastic container; the empty plastic container was measured in the same manner as the soil samples for the same counting time of 36,000s (10h). The background spectrum was subtracted from the measure spectra to obtain the net radionuclides activities. The background, reference and the sediment samples were measured under the same conditions [15]. The γ-ray photo-peaks corresponding to ^{214}Pb at (242.0 keV, 295.2 keV& 351.9 keV), ^{214}Bi (609.3 keV, 768.4 keV, 806.19 keV, 1120.3 keV, 1377.669 keV and 1401.516 keV respectively) and ^{234}Pa (1001.025 keV) were considered for identifying ^{238}U. The γ-ray photo-peaks of ^{228}Ac at (209.3 keV, 338.3 keV, 409.5 keV and 911.1 keV), ^{208}Ti at (277.4 keV, 583.2 keV and 860.6 keV), ^{212}Bi at (727.33 keV and 785.37 keV), ^{212}Pb (238.6 keV and 300.1 keV and ^{224}Ra (240.986 keV) were used to identify ^{232}Th in the samples. The radioisotope ^{40}K was estimated from emits gamma ray with energy of 1460.8 keV. Hence the determination of ^{40}K was considered as direct [16]. The activity concentration (A_c) of ^{238}U, ^{232}Th and ^{40}K in Bq kg^{-1} was obtained using the relation in Eq. (1) [17]:

$$\text{Sample activity } (A_C)\ (\text{Bq kg}^{-1}) = \frac{C_i}{\varepsilon(E_\gamma)P\gamma(E)tm} \quad (1)$$

where C_i is the net peak area after subtraction of background of the gamma-ray line at energy E, ε(Eγ) is the detector efficiency of such gamma-ray line at photopeak energy (E), $P_\gamma(E)$ is the emission probability of the gamma-ray photons of energy (E) under consideration, t is the time of measurement in seconds and m is the mass of the sample in kg.

Each radionuclide's below detectable limit (BDL) was determined from the background radiation spectrum for the same counting time for the dumpsites soil samples. The detection limits (DL) required estimating the minimum detectable activities in a sample were obtained using Eq. (2) [18]

$$LLD\ (\text{Bq kg}^{-1}) = 4.65\frac{\sqrt{C_B}}{t_b}f \quad (2)$$

where C_b is the net background count in the corresponding peak t_b is the background counting time(s) and f is the factor that converts cps(counts per second) to activity concentration (Bq kg^{-1}).

Radiation hazard parameters

In order to estimate the radiation hazards incurred by the population due to the activity levels of the measured sediment enhanced naturally occurring radionuclides (NORM) in the selected rivers in Ilobi and Erinja towns, some radiation hazard indices were calculated and the formulas used for the calculations are given in Table 1. These are used to reassess the statistical information about excessive lifetime cancer risk (ELCR), γ-ray absorbed dose rate (D_R), outdoor and indoor annual effective dose equivalent ($AEDE_{outdoor}$ and $AEDE_{indoor}$), Annual gonadal dose equivalent (AGDE), radium equivalent (Ra_{eq}), activity utilization index (AUI), exposure rate (ER), internal and external hazard indices (H_{int} and H_{ext}) and external (γ-radioactivity) level index (Iγr) for the present study. Even though the total activity concentration of radionuclides is calculated, it does not provide the exact indication about the total radiation hazards due to uneven distribution of the waste enhanced naturally occurring radionuclide (^{40}K, ^{238}U and ^{232}Th) in the sediment.

Statistics of all the calculated radiological parameters and their recommended levels by [11] are given in Tables 2 and 3.

RESULTS AND DISCUSSION

The results obtained from NIRPR shows different activity concentrations of the radionuclides as shown below.

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

Table 2 shows the activity concentration of naturally occurring radioactive elements. The activity concentration of ^{238}U, ^{232}Th and ^{40}K in the river sediments ranged from 0.0 ↔ 166.8 ± 19.5 Bq kg^{-1}, 0.0 ↔ 17.9 ± 6.7 Bq kg^{-1} and 181 ± 3.3 ↔ 814 ± 29.4 Bq kg^{-1}respectively. In comparison with the recommended limit, activity concentrations of radionuclides ^{238}U and ^{40}K are higher than the worldwide average value of 35 and 400 Bq kg^{-1} respectively (UNSCEAR) and that ^{238}U is about 3 to 5 times higher than the world average value in two sampling points (A5 and A4). ^{40}K level is twice the limit in sample B6 (Fig. 2), whereas only ^{232}Th average values were below the recommended average value [11]. The presence of ^{232}Th in the analyzed samples is low in all locations, which indicates low level of contamination. According to the results obtained in this

study, activity concentration of the radionuclides increased in order of $^{232}Th<^{238}U<^{40}K$ in all the sampling sites (Fig. 2). The wide variation in the activity concentrations as shown in the large values of standard deviation is a measure of the spatial distribution of the radionuclides within the rivers. It also indicates the influence of physical and geo-chemical processes on the accumulation of radionuclides in the sediment within the river basin [1]. The high activity concentration of ^{238}U in the all the samples in the present study may be due to the solubility and mobility of U (VI)O_2^{+2}[11] and ^{40}K due to the high content of monazite [19]. The presence of mineral monazite which contains radioactive elements results in higher natural radioactivity [20]. Since the natural radionuclides in the sample under investigation is randomly distributed (not uniform), further radiological index has been introduced to evaluate the radiation hazard associated with these radionuclides.

Table 1: Summary of the radiological parameter of all the River sediment samples

S/N	Radiological Parameters	Units	Used Formula
1	Absorbed dose rate (D_R)	nGy hr^{-1}	$D_R= (0.462A_U + 0.604A_{Th} + 0.0417A_K)$
2	Radium equivalent (Ra_{eq})	Bq kg^{-1}	$Ra_{eq}= (A_U + 1.43A_{Th} + 0.077A_K)$
3	External Hazard index (H_{ext})	-	$H_{ext} = A_U/370 + A_{Th}/259 + A_K/4810 \leq 1$
4	Internal Hazard index (H_{int})	-	$H_{int} = A_U/185 + A_{Th}/259 + A_K/4810 \leq 1$
5	Annual effective dose equivalent ($AEDE_{outdoor}$)	µSv yr^{-1}	$AEDE_{outdoor} = D_R$ x 8766h x 0.7Sv/Gyx0.2x 10^{-3}
6	Annual effective dose equivalent ($AEDE_{indoor}$)	µSv yr^{-1}	$AEDE_{indoor} = D_R$x8766hx0.7Sv/Gyx0.8x 10^{-3}
7	Annual gonadal dose equivalent (AGDE)	µSv yr^{-1}	AGDE = $3.09A_U + 4.18A_{Th} + 0.314A_K$
8	Gamma level index ($I_\gamma r$)	-	$I_{\gamma r} = A_U/300 + A_{Th}/200 + A_K/3000 \leq 1$
9	Activity utilization index (AUI)	-	$AUI = A_U/50 + A_{Th}/50f_{Th} + A_K/500f_K \leq 2$
10	Exposure rate (ER)	µR hr-1	$ER = 1.90A_U + 2.82A_{Th} + 0.179A_K$
11	Excess lifetime cancer risk ($ELCR_{outdoor}$)	-	$ELCR = AEDE_{outdoor}$ x DL x RF

[a][25]

where A_U, A_{Th} and A_K are the activity concentrations of ^{238}U, ^{232}Th and ^{40}K (in Bq kg^{-1}) present in river sediments respectively. f_U (0.462), f_{Th} (0.604) and f_K (0.0417) are the fractional contributions to the total dose rate due to γ-radiation from the actual radionuclide of ^{238}U, ^{232}Th and ^{40}K, respectively. DL and RF is duration of life (70 years) and risk factor (Sv^{-1}), fatal cancer risk per sievert. For stochastic effects, ICRP 60 uses values of 0.05 for the public.

Evaluation of radiological parameters

Radium equivalent (Raeq)

In terms of the radiological health safety assessment, the maximum permissible limit of $Ra_{eq} \leq 370$ Bq kg^{-1} had been set for materials to be used as component of building construction [11] and it was calculated using equation in Table 1. The calculated values of Raeq ranged from 33.7 ± 0.8 ↔ 215.3 ± 22.1Bq kg^{-1} with a mean value of 99.9 ± 9.1Bq kg^{-1}, which is lower than the safety limit set for this index. Thus, from the radiological protection point of view, the sediments from these rivers are safe for use as building materials.

Table 2: Activity concentration of radionuclides (A_c) in the river sediments

Sample ID	Activity Concentrations (Bqkg^{-1})			D_R (nGy hr^{-1})	Raeq (Bq kg^{-1})
	^{238}U	^{232}Th	^{40}K		
A1	42.34 ± 5.44	13.2 ± 2.45	342.43 ± 16.54	41.8 ± 4.7	87.6 ± 10.2
A2	67.23 ± 7.88	11.31 ± 1.76	453.23 ± 17.76	56.8 ± 5.4	118.3 ± 11.8
A3	90.23 ± 12.56	8.26 ± 3.13	278.23 ± 14.56	58.3 ± 8.3	123.5 ± 18.2
A4	166.77 ± 19.47	9.74 ± 0.58	449.22 ± 23.18	101.7 ± 10.3	215.3 ± 22.1
A5	107.06 ± 13.06	7.27 ± 0.44	287.99 ± 9.89	65.9 ± 6.7	139.6 ± 14.5
A6	BDL	15.55 ± 0.96	181.09 ± 9.48	16.9 ± 1.0	36.2 ± 2.9
A7	54.56 ± 7.67	8.89 ± 4.5	554.23 ± 17.89	53.7 ± 7.0	109.9 ± 15.5
A8	76.89 ± 9.66	BDL	654.23 ± 21.34	62.8 ± 5.4	127.3 ± 11.3
A9	BDL	14.56 ± 1.32	772.67 ± 29.42	41.0 ± 2.0	8.3 ± 4.2
A10	39.56 ± 5.22	BDL	532.54 ± 21.11	40.5 ± 3.3	80.6 ± 6.8
B1	40.34 ± 5.05	12.54 ± 2.11	633.34 ± 23.21	52.6 ± 4.6	107.0 ± 9.9
B2	60.54 ± 6.22	9.56 ± 0.99	542.34 ± 22.12	56.4 ± 4.4	116.0 ± 9.3
B3	BDL	BDL	722.54 ± 19.78	30.1 ± 0.8	55.6 ± 1.5
B4	BDL	12.09 ± 2.13	213.54 ± 7.76	16.2 ± 1.6	33.7 ± 3.6
B5	82.88 ± 8.67	11.55 ± 2.11	311.76 ± 12.77	58.3 ± 5.8	123.4 ± 12.7
B6	62.76 ± 7.22	BDL	813.87 ± 24.22	62.9 ± 4.3	125.4 ± 9.1
B7	55.55 ± 4.67	9.12 ± 6.66	677.21 ± 17.77	59.4 ± 6.9	120.7 ± 15.6
B8	23.45 ± 3.52	7.24 ± 1.01	444.32 ± 11.54	33.7 ± 2.7	68.0 ± 5.9
B9	32.43 ± 3.11	BDL	321.22 ± 8.15	28.4 ± 1.8	57.2 ± 3.7
B10	45.33 ± 4.12	11.21 ± 2.17	206.43 ± 6.78	36.3 ± 3.5	7.3 ± 7.7
C1	64.21 ± 3.21	BDL	489.73 ± 9.12	50.1 ± 1.9	101.9 ± 3.9
C2	34.22 ± 2.35	6.75 ± 1.01	221.32 ± 3.34	29.1 ± 1.8	60.9 ± 4.1
C3	45.41 ± 3.22	8.98 ± 2.10	344.23 ± 4.14	40.8 ± 2.9	84.8 ± 6.5
C4	46.77 ± 4.51	7.23 ± 1.78	544.23 ± 8.16	48.7 ± 3.5	99.0 ± 7.7
C5	67.54 ± 8.22	10.98 ± 2.10	433. 54 ± 6.88	55.9 ± 5.4	116.6 ± 11.8
C6	61.88 ± 7.77	9.16 ± 1.55	632.65 ± 6.55	60.5 ± 4.8	123.7 ± 10.5
C7	64.52 ± 6.75	9.18 ± 1.75	599.32 ± 9.32	60.3 ± 4.6	123.8 ± 10.0

C8	55.21 ± 8.17	17.89 ± 3.45	732.63 ± 10.76	66.9 ± 6.3	137.2 ± 13.9
C9	BDL	BDL	662.76 ± 9.87	27.6 ± 0.4	51.0 ± 0.8
C10	61.55 ± 6.10	BDL	455. 12 ± 12.2	47.4 ± 3.3	96.6 ± 7.0
Min.	0.0 ± 0.0	0.0 ± 0.0	181 ± 3.3	16.2 ± 0.4	33.7 ± 0.8
Max.	166.8 ± 19.5	17.9 ± 6.7	814 ± 29.4	101.7 ± 10.3	215.3 ± 22.1
Range	166.8 ± 19.5	17.9 ± 6.7	633 ± 26.1	85.5 ± 9.9	181.6 ± 21.3
Mean	52 ± 5.8	7.6 ± 1.5	488 ± 13.8	*48.7 ± 4.2*	*99.9 ± 9.1*
Kurtosis	5.07	2.53	1.87	*1.86*	*1.92*
Skewness	1.96	1.19	0.46	*0.99*	*1.07*
World Average	35[a]	30[a]	400[a]	84[a]	370[a]

[a]UNSCEAR, 2000; BDL: Below Detection limit

Absorbed dose rate (D_R)

The energy imparted per unit weight of the irradiate material was also calculated using the equation in Table 1 and the results are shown in Table 2. The absorbed dose rate (D_R) values varied from 16.2 ± 0.4 ↔ 101.7 ± 10.3nGy hr^{-1} with a mean value of *48.7 ± 4.2* nGy hr^{-1}. From the present study, the estimated mean value of D_R is below the recommended limits of 84 nGy hr^{-1}[11] but higher by a factor of 2.0 times in only one of the sampling point (A4) (Fig. 3). The contribution of natural radionuclides to the absorbed dose rates depends on the concentrations of various radionuclides in the sediment [21].

Annual effective dose equivalent (AEDE), Rate of Exposure (ER) and Annual gonadal dose equivalent (AGDE).

The calculated AEDE values ranged from 19.9 ± 0.5 ↔ 124.8 ± 12.7 µSv yr^{-1} with a mean value of 59.8 ± 5.1 µSv yr^{-1}. This is below the world average value of 70 µSv yr^{-1}[11]. Highest value of AEDE was obtained in sample A4 (124.8 ± 12.7 µSv yr^{-1}) (Fig. 3) which is about 2 times higher than the recommended level. Only these locations (Okooko River in Ilobi community) have the AEDE value above the world average value. This was due to a high activity concentration radionuclide distribution in that location [18]. The calculated value for the rate of exposure (ER) of individuals and scavengers to these radionuclides in the selected river sediments was below the maximum limit of (600 µR hr^{-1}) in all samples, with only one sample point i.e. A4 having an exposure rate value of 424.74 ± 42.78 µR hr^{-1} which is moderately high but below the recommended limits of 600 µR hr^{-1} in all samples. This implies that human exposure from the

ionizing radiation from the used of sediments as a building materials is insignificant in about 97 % of the sampling points.

The annual genetically significant dose equivalent (AGSDE) or annual gonadal dose equivalent (AGDE) is a measure of the genetic significance of the yearly dose received by the population's reproductive organs [22]. Organs with rapidly dividing cells such as gonads, the active bone marrow and bone surface cells are considered as organs of interest (since they are active) by the United Nations Scientific Committee on the Effects of Atomic Radiation [23]. This means that not all living cells are equally sensitive to radiation. Those cells which are actively reproducing are more sensitive than those which are less or not reproducing [24]. A direct interaction of radiation with reproductive cells could result in the death or genetic mutation of the cell, whereas a direct interaction with the DNA of a dormant cell could have less effect [24]. Hence the AGSDE or AGDE due to specific activities of ^{238}U, ^{232}Th and ^{40}K was calculated and ranged from 117.6 ± 3.1 ↔ 697.1 ± 69.9 µSv yr^{-1}, with an average value of 343.8 ± 28.7 µSv yr^{-1} (Table 3), which is higher than the UNSCEAR value of 300 µSv yr^{-1}[11], reported as world value.

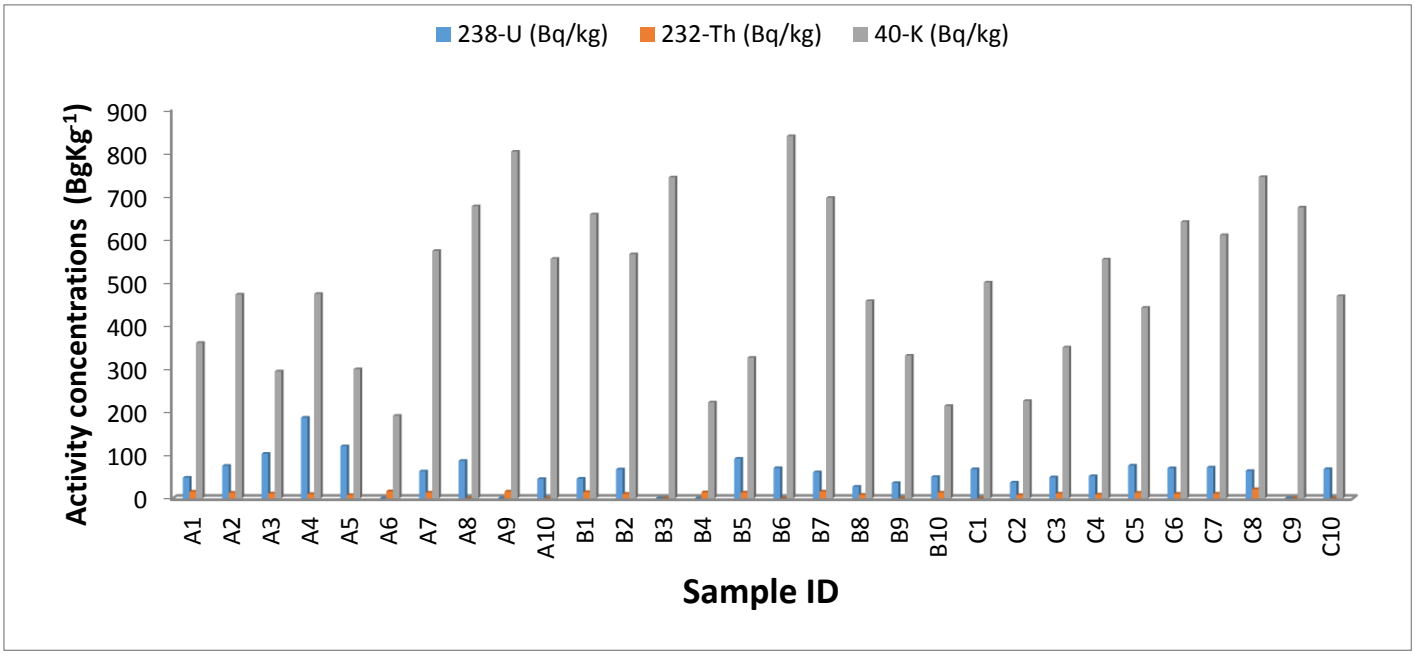

Fig.2: Activity concentration of ^{238}U, ^{232}Th, and ^{40}K

Internal, external hazard & activity utilization index (H_{int}, H_{ext} & AUI)

In order to assess the suitability of any material to be used for building construction, and also to determine the dose delivered externally to individuals in a house constructed with such material, a dose criterion called external hazard index (Hext.) [1] was calculated using the equation in Table 1. In addition to external hazard index, Uranium and its daughter products pose radiological risks to the respiratory organs. To assess these risks, an internal hazard index (Hint) was estimated using the equation in Table 1. The index value must be less than unity in order to keep the radiation hazard insignificant. The calculated external hazard values are between 0.1 ± 0.0 ↔ 0.6 ± 0.1 with an average value of 0.3 ± 0.0 (Table 3), which is lower than the recommended value of 1(unity). As such, there is no radiological risk whatsoever from external exposure of the inhabitant or people working/living in buildings constructed with these sediments to these deleterious natural radionuclides.

The calculated values of internal hazard index (Hint) ranged from 0.1 ± 0.0 ↔ 1.0 ± 0.1 (Fig. 4) with an average value of 0.4 ± 0.04. The value of internal hazard exceeded the recommended permissible value in one of the sampling points i.e. A4. Therefore the sediment from this location poses potential internal radiological risk while the value for other sediments further confirms suitability for use as building materials.

In radiological health assessment studies, activity utilization index (AUI) is usually computed in order to estimate the dose rates in air from different combinations of the various radionuclides present in sediments [1]. The calculated values for activity utilization index (AUI), which can be used to determine the possibility of using the sediments for building construction ranged from 0.06 ± 0.00 (C9) ↔ 3.49 ± 0.40 (A4) with an average of 1.17 ± 0.14 (Table 4). This calculated average value is lower than the world average of 2 [11]. Beyond the recommended upper limit is noted in four different samples (A3, A4, A5 and B5), with the highest value of 3.49 ± 0.40 observed at (A4). These higher AUI values can be attributed to the higher concentration of radionuclides in these sites. Here also, average relative contribution of the γ-index is due to higher 40K followed by the contributions due to ^{238}U and ^{232}Th.

Sample ID	H_{ext}	H_{int}	ER ($\mu R\ hr^{-1}$)	AGDE ($\mu Sv\ yr^{-1}$)	$I_{\gamma r}$	AUI	AEDEout ($\mu Sv\ yr^{-1}$)	AEDEin ($\mu Sv\ yr^{-1}$)	ELCR x 10^{-3} outdoor
A1	0.24±0.03	0.35±0.04	178.96±20.2	193.53±32.2	0.32±0.04	1.03 ± 0.14	51.3±5.7	205.3 ± 23.0	0.18 ± 0.02
A2	0.32 ± 0.03	0.50 ± 0.05	240.76±23.1	397.33±37.3	0.43±0.04	1.52 ± 0.18	69.7±6.7	278.8 ± 26.7	0.24 ± 0.02
A3	0.33 ± 0.05	0.58 ± 0.08	244.53 ± 35.30	400.70 ± 56.47	0.43 ± 0.06	1.93 ± 0.29	71.5±10.2	286.1 ± 40.7	0.25 ± 0.04
A4	0.58 ± 0.06	1.03 ± 0.11	424.74 ± 42.78	697.09 ± 69.87	0.75 ± 0.08	3.49 ± 0.40	124.8±12.7	499.1 ± 50.6	0.44 ± 0.04
A5	0.38 ± 0.04	0.67 ± 0.07	275.47 ± 27.83	451.63 ± 45.30	0.49 ± 0.05	2.25 ± 0.27	80.8 ± 8.2	323.3 ± 32.9	0.28 ± 0.03
A6	0.10 ± 0.01	0.10 ± 0.01	76.27 ± 4.40	121.86 ± 6.99	0.14 ± 0.01	0.20 ± 0.01	20.8 ± 1.2	83.2 ± 4.8	0.07 ± 0.0
A7	0.30 ± 0.04	0.44 ± 0.06	227.94 ± 30.47	379.78 ± 48.13	0.41 ± 0.05	1.24 ± 0.21	65.9 ± 8.6	263.6 ± 34.4	0.23 ± 0.03
A8	0.34 ± 0.03	0.55 ± 0.06	263.20 ± 22.17	443.02 ± 36.55	0.07 ± 0.04	1.59 ± 0.19	77.1 ± 6.6	308.3 ± 26.3	0.27 ± 0.02
A9	0.22 ± 0.01	0.22 ± 0.01	179.37 ± 8.99	303.48 ± 14.76	0.33 ± 0.02	0.24 ± 0.02	50.3 ± 2.5	201.3 ± 9.9	0.18 ±0.01
A10	0.22 ± 0.02	0.32 ± 0.03	170.19 ± 13.70	289.46 ± 22.76	0.31 ± 0.02	0.84 ± 0.11	49.7 ± 4.0	198.7 ± 16.2	0.17 ± 0.01
B1	0.29 ± 0.03	0.40 ± 0.04	225.38 ± 19.70	375.94 ± 31.71	0.41 ± 0.04	1.01 ± 0.13	64.6 ± 5.6	258.3 ± 22.5	0.23 ± 0.02
B2	0.31 ± 0.03	0.48 ± 0.04	239.06 ± 18.57	397.32 ± 30.3	0.43 ± 0.03	1.37 ± 0.14	69.2 ± 5.4	276.7 ± 21.6	0.24 ± 0.02
B3	0.15 ± 0.00	0.15 ± 0.00	129.33 ± 3.54	226.88 ± 6.21	0.24 ± 0.01	0.06 ± 0.00	37.0 ± 1.0	147.9 ± 4.0	0.13 ± 0.0
B4	0.09 ± 0.01	0.09 ± 0.01	72.32 ± 7.40	117.59 ± 11.34	0.13 ± 0.01	0.16 ± 0.03	19.9 ± 2.0	79.6 ± 7.9	0.07 ± 0.01
B5	0.33 ± 0.03	0.56 ± 0.06	245.85 ± 24.71	402.27 ± 39.62	0.44 ± 0.04	1.82 ± 0.20	71.5 ± 7.1	286.0 ± 28.5	0.25 ± 0.02
B6	0.34 ± 0.02	0.51 ± 0.04	264.93 ± 18.05	449.48 ± 29.91	0.48 ± 0.03	1.32 ± 0.18	77.2 ± 5.3	308.9 ± 21.3	0.27 ± 0.02
B7	0.33 ± 0.04	0.48 ± 0.05	252.48 ± 30.84	422.42 ± 47.85	0.46 ± 0.05	1.28 ± 0.18	72.9 ± 8.5	291.7 ± 34.0	0.26 ± 0.03
B8	0.18 ± 0.02	0.25 ± 0.03	144.51 ± 11.60	242.24 ± 18.72	0.26 ± 0.02	0.59 ± 0.08	41.4 ± 3.3	165.6 ± 13.3	0.14 ± 0.01
B9	0.15 ± 0.01	0.24 ± 0.02	119.12 ± 7.37	201.07 ± 12.17	0.22 ± 0.01	0.68 ± 0.06	34.8 ± 2.2	139.3 ± 8.7	0.12 ± 0.01
B10	0.21 ± 0.02	0.33 ± 0.03	154.69 ± 15.16	251.75 ± 23.93	0.28 ± 0.03	1.06 ± 0.11	44.6 ± 4.3	178.3 ± 17.2	0.16 ± 0.02
C1	0.28 ±0.01	0.45 ± 0.02	209.66 ± 7.73	352.18 ± 12.78	0.38 ± 0.01	1.33 ± 0.06	61.5 ± 2.3	245.9 ± 9.1	0.22 ± 0.01
C2	0.16 ± 0.01	0.26 ± 0.02	123.67 ± 7.91	203.45 ± 12.53	0.22 ± 0.01	0.78 ± 0.06	35.7 ± 2.3	142.9 ± 9.0	0.13 ± 0.01
C3	0.23 ± 0.02	0.35 ± 0.03	173.22 ± 12.78	285.94 ± 20.03	0.31 ± 0.02	1.05 ± 0.09	50.0 ± 3.6	200.1 ± 14.4	0.18 ± 0.01

C4	0.27 ± 0.02	0.39 ± 0.03	206.67 ± 15.05	345.63 ± 23.94	0.37 ± 0.03	1.07 ± 0.11	59.7 ± 4.3	238.9 ± 17.2	0.21 ± 0.02
C5	0.32 ± 0.03	0.50 ± 0.05	236.89 ± 22.77	390.73 ± 36.34	0.42 ± 0.04	1.52 ± 0.19	68.6 ± 6.6	274.5 ± 26.3	0.24 ± 0.02
C6	0.33 ± 0.03	0.50 ± 0.05	256.65 ± 20.31	428.18 ± 32.55	0.46 ± 0.04	1.40 ± 0.17	74.3 ± 5.9	297.0 ± 23.6	0.26 ± 0.02
C7	0.33 ± 0.03	0.51 ± 0.05	255.75 ± 19.43	425.93 ± 31.10	0.46 ± 0.03	1.45 ± 0.16	74.1 ± 5.6	296.2 ± 22.4	0.26 ± 0.02
C8	0.37 ± 0.04	0.52 ± 0.06	286.19 ± 27.18	475.42 ± 43.04	0.52 ± 0.05	1.38 ± 0.21	82.1 ± 7.7	328.2 ± 31.0	0.29 ± 0.03
C9	0.14 ± 0.00	0.14 ± 0.00	118.63 ± 1.77	208.11 ± 3.10	0.22 ± 0.00	0.06 ± 0.00	33.9 ± 0.5	135.7 ± 2.0	0.12 ± 0.0
C10	0.26 ±0.02	0.43 ± 0.04	198.41 ± 13.77	333.10 ± 22.68	0.36 ± 0.02	1.27 ± 0.12	58.2 ± 4.1	232.8 ± 16.3	0.20 ± 0.01
Min.	0.14±0.0	0.14 ± 0.0	72.3 ± 1.8	117.6 ± 3.1	0.1 ± 0.0	0.06 ± 0.00	19.9±0.5	79.6 ± 2.0	0.07 ± 0.002
Max.	0.58±0.06	1.03 ± 0.11	424.7 ± 42.8	697.1 ± 69.9	0.8 ± 0.1	3.49 ± 0.40	124.8±12.7	499.1 ± 50.6	0.44 ± 0.04
Range	0.5±0.1	0.9 ± 0.1	352.4 ± 41	579.5 ± 66.8	0.6 ± 0.1	3.44 ± 0.40	104.9±12.2	419.5 ± 48.6	0.37 ± 0.04
Mean	*0.3 0.0*	*0.4 ± 0.04*	206.5 ± 17.8	343.8 ± 28.7	0.4 ± 0.03	1.17 ± 0.14	59.8 ± 5.1	239.1 ± 20.5	0.21 ± 0.02
Kurtosis	*1.92*	*3.64*	1.12	1.61	1.45	4.10	1.86	1.71	1.86
Skewness	*1.07*	*1.52*	1.01	0.90	0.89	1.67	0.99	0.72	0.99
World Average	≤1[a]	≤1[a]	600[a]	300[a]	0.5[a]	≤2[a]	70[a]		0.29

[a][25]

In the present case all the radionuclides but one (Potassium, ^{40}K) have positive Kurtosis values (Table 5) which indicates a peaked distribution. This multi-modal feature of the radio elements demonstrates the complexity of minerals in sediments sample.

Excessive lifetime cancer risk (ELCR)

Excessive lifetime cancer risk is the probability of an individual developing a cancerous cell as a result of exposure to cancer-inducing substance over a passage of time. Radioactivity in building materials are known to produce carcinogenic effects due to accumulation in indoor air such that the probability or extra risk of developing lung cancer due to exposure to gaseous radionuclides indoors incurred over the lifetime of an individual is estimated using the excess lifetime cancer risk (ELCR). The potential carcinogenic and mutagenic effects are characterized by evaluating the probability of cancer incidence in a population of individuals for a specific lifetime from projected intakes cum exposure with chemical-specific dose response data (i.e. slope factors) [18]. Excessive lifetime cancer risk (ELCR) was calculated using the equation presented in Table 1 and the calculated values ranged from $0.07 \pm 0.002 \leftrightarrow 0.44 \pm 0.04$ with an average value of 0.21 ± 0.02 (Table 3). The present average ELCR value is less than the world average value of (0.29) (UNSCEAR, 2000) (Table 3) (Fig. 4). Three (3) of the sampling points (A4, A5 & C8) (10 %) have the ELCR values higher than the world average value. These results show that the lifetime cancer risk due to exposure through the use of these sediments as building material (or any anthropogenic purposes) for a maximum duration of 70 years is high in these three locations. Hence, the use of these sediments for building, construction, agriculture or for soil remediation studies and other purpose should be discouraged.

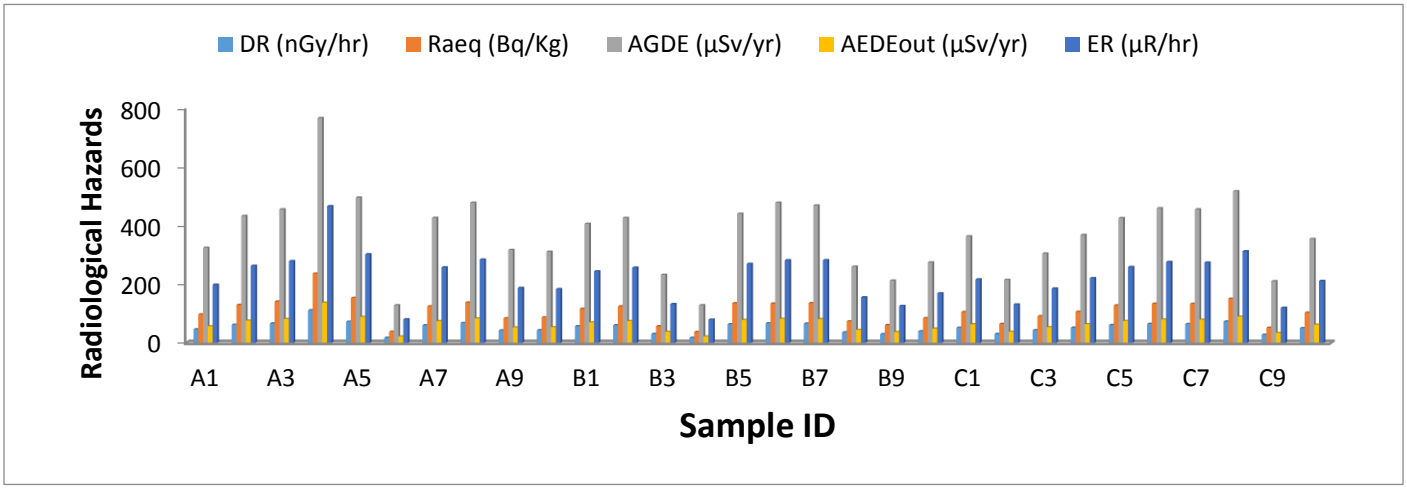

Fig. 3: Radiological parameters

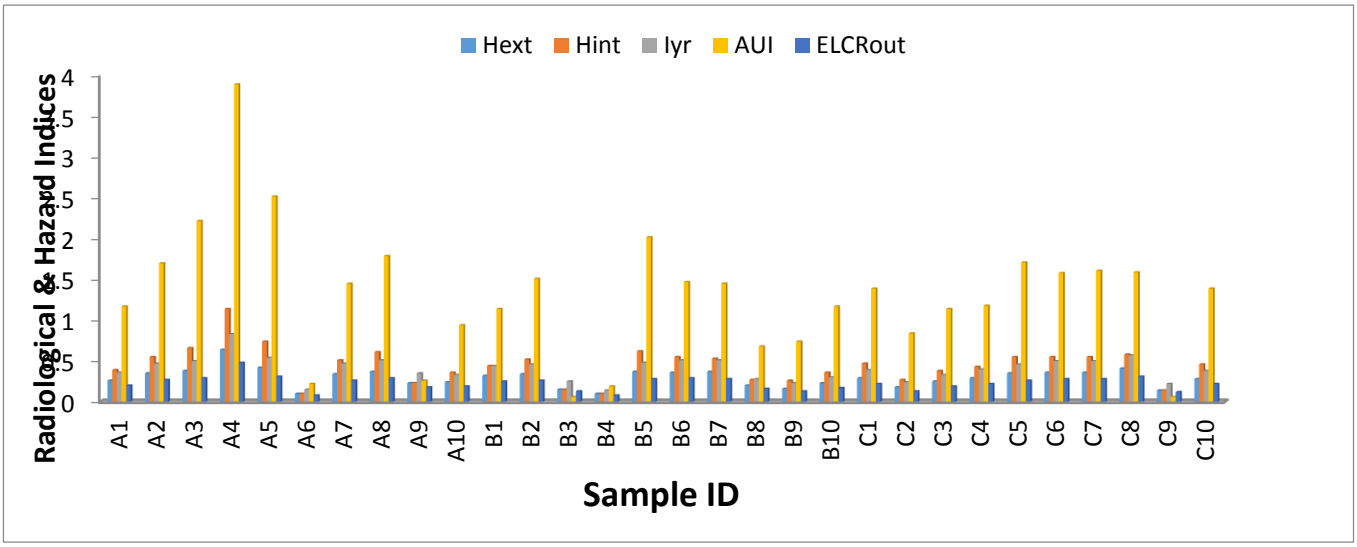

Fig. 4: Radiological parameters and hazard index

Spatial distributions

The investigation of ^{238}U:^{232}Th activity concentrations in the river sediments revealed that ^{238}U activity concentrations were 18 times higher than the ^{232}Th activity concentration in the measured sediment from A4 (Okooko River sediment in Ilobi community) with other samples showing appreciable high concentrations like A3 and A5. The ratio $^{238}U/^{232}Th$ ratios (Table 4) were higher than the world's average value of 1 in almost all the sediment samples. The activity concentration of ^{40}K shows that on average ^{40}K are about 16 - 55 times higher in Ilobi community and 29 – 61 times (Erinja community) than ^{238}U and ^{232}Th activity concentrations in the collected sediment samples. These ratios ^{232}Th:^{238}U, ^{238}U:^{232}Th, ^{232}Th:^{40}K and ^{238}U: ^{40}K gave indication that the samples from this region have higher uranium and potassium than thorium concentrations to be economical feasible for the ^{238}U and ^{40}K extraction. On the average, activity concentration of ^{40}K and ^{238}U is high in all the samples which may be due to the use of potassium rich fertilizer like phosphate in the agricultural lands around the location which consequently could have been washed into the surrounding rivers.

The $^{232}Th/^{238}U$ ratio was also calculated (Table 4). From these results, the ratios were lower than the stipulated range of 0.7–0.4 mentioned by [25]. All the soil samples of the study area have

$^{232}Th/^{238}U$ ratio above this range. This present study confirmed the presence of xenolith zircon and monazite in the sediments

Table 4: Activity ratios

Sample ID	$^{238}U/^{232}Th$	$^{238}U/^{40}K$
A1	3.05	0.13
A2	5.75	0.16
A3	9.02	0.35
A4	18.05	0.39
A5	15.58	0.40
A6	0.00	0.00
A7	4.65	0.11
A8	-	0.13
A9	0.00	0.00
A10	-	0.08
B1	3.10	0.07
B2	6.33	0.12
B3	-	0.00
B4	0.00	0.00
B5	6.70	0.28
B6	-	0.08
B7	3.82	0.09
B8	3.27	0.06
B9	-	0.11
B10	3.70	0.23
C1	-	0.14
C2	4.71	0.16
C3	4.39	0.14
C4	5.69	0.09
C5	5.79	0.17
C6	6.50	0.11
C7	6.52	0.12
C8	2.97	0.09
C9	-	-
C10	-	-

CONCLUSION

The activity concentrations of ^{238}U, ^{232}Th and ^{40}K in sediments collected from Ilobi and Erinja communities, Southwest Nigeria, had been radio-metrically assessed. The obtained activity concentrations of these radionuclides were used to evaluate radiological hazard indices in order to determine the effects of the natural radionuclides in the sediments. The results indicated that the average value of each radiological hazard parameter were below the world average value reported in UNSCEAR except Annual gonadal dose equivalent (AGDE) and internal hazard index whose mean value exceeded their recommended limits. It follows therefore that there is a fingering potential radiological health hazard directly associated with the sediments from these locations. Sediments from specific locations in both communities have the ELCR values higher than the world average value, which is suggestive of its likelihood of inducing cancers over a period of exposure to humans. The statistical treatment result shows that the distribution of the radionuclides (^{238}U and ^{232}Th) was asymmetrical and peaked with an observed flat distribution in ^{40}K level in the sediments. Extensive research needs to be carried out on Okooko River in Ilobi community as its sediment shows higher concentrations in both radionuclides and radiological hazard indices.

Conflict of interest

None

Acknowledgements

Authors are grateful to Mrs. Owoade, L.R. and Mr. Olubunmi, Emmanuel T. of the National Institute of Radiation and Protection, University of Ibadan, Ibadan for the support given during the radionuclides analysis.

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