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Determination of Radiological Hazards Associated with the use of Oba-Ile River Sediments as Building Material

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Abstract

This study was conducted to determine the radioactivity level in the sediment samples collected from Oba-Ile River, Olorunda Local Government Area of Osun State, Nigeria and evaluate the radiological hazards associated with the use of the river sediments as building material. Twenty four (24) sediment samples were obtained along three different points within the River for a period of eight months. The activity concentration of ^{40}K in the sediment sample ranged from 273.36 ± 5.41 $Bqkg^{-1}$ to 1662.41± 28.19 $Bqkg^{-1}$with an average concentration 892.22 ± 14.66 $Bqkg^{-1}$. The values of ^{232}Th in the sediment samples ranged between 27.22 ± 5.12 $Bqkg^{-1}$ to 186.14 ± 6.22 $Bqkg^{-1}$ with an average concentration 87.95 ± 7.91 $Bqkg^{-1}$. The values of ^{238}U ranged from 12.61 ± 2.01 $Bqkg^{-1}$ to 241.12 ± 5.91 $Bqkg^{-1}$ with an average 62.83 ± 3.18 $Bqkg^{-1}$. The radiological hazards due to natural radionuclides content, such as absorbed dose rate (D_R), annual effective dose rate (AEDE),excess lifetime cancer risk (ELCR), internal radiation hazard (H_{in}) and external radiation hazard (H_{ex}) in the sediment samples were calculated. The calculated radiological parameters were compared with recommended safety limits and internationally approved values.

Keywords: Radionuclides; Activity concentration; Water; Radiological Hazards; Sediment; Fish

1. Introduction

Radionuclides can be transported from water phase to the sediment phases through physical processes such as sedimentation, chemical means as well as biological processes. Sediments usually act as sinks and sources of radionuclides in the aquatic environment. Though, when the primary sources have stopped or been significantly reduced, remobilization of radionuclides from earlier contaminated sediments may upsurge in importance as a diffuse secondary source (Hunt and Kershaw, 1990; Oughton *et al*., 1997). A secondary source also involves mobilization from contaminated land-based areas. The exposure of man to gamma radiation from radionuclides in the aquatic environment is not limited to the internal exposure due to consumption of contaminated aquatic foods alone. The use of river sediments as an essential building material for flooring, plastering and moulding bricks in Nigeria (Fig. I) has a great probability of increasing the external exposure level to man especially, if such sediments have high level of radionuclide concentration (Oni *et al*., 2011). In building, it has been reported that the highest radionuclides concentrations are observed in mineral based materials such as stone, sand, bricks, cement and sediments (Turtianen *et al*., 2008). Nevertheless, these radionuclides are known to be widely spread in the environment and their concentrations (Iqba *et al*., 2000) largely depend on the geological setting of a specific environment. Hence, the concentration of these radionuclides varies from one location to the other (Oni *et al*., 2011). Radionuclides can be transported from the water to the sediment phases through physical processes such as sedimentation, chemical means as well as biological processes. Estimating the concentrations of radionuclides in the environment is key in examining the health exposure of the populace and provides a locus for recording variations in environmental radioactivity as a

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result of anthropogenic works (Obed *et al*., 2005). Radiation can arise not only from natural radionuclides but it can also be from man-made sources. The samples of different environmental media such as sediment, soil, water, vegetation, and so on are measured to determine background levels of radiation, or to assess the levels of contamination as a consequence of manmade radioactivity. This study serves as a reference material in future studies for subsequent assessments of the possible future environmental contamination or threats due to activities surrounding Oba-Ile River.

Figure 1 (a) and (b) Pile of Sharp Sand packed for building purpose

2. Materials and method

2.1. Description and Suitability of the Study Area

Oba-Ile River traverses two states namely; Oyo and Osun States in Nigeria. It lies between Latitude7^{0}54'30"N and Longitude 4^0 33'30"E. It is the main tributary of the Osun River and the people who live along its length practice farming and fishing. The area constitutes a part of the basement complex of Southwestern Nigeria and characteristically underlained by crystalline igneous and metamorphic rocks. These rocks constitute the major outcrops and inselbergs that explained the topographic highlands of the studied area. Though the rocks are basically of granite-gneiss complex, they are mainly monolithologic (Adeboye and Alatise, 2008). The study area is below Koppen's Af humid tropical rainforest weather. The mean annual rainfall is about 1400 mm with the rainy and dry season covering eight months with its beginning and end marked by heavy rains and storms. The rainy season is typically considered to have two maxima rainfall with peaks in July and September/October. Temperatures are generally very high and almost uniform (Iloeje, 1978).

2.2. Sample Collection

Sediment samples were collected from the aquatic ecosystems for a period of 8 months (September, 2013, November, January, February - June, 2014). Three (3) sampling points were identified along the river. The first sampling point was the downstream, second sampling point was the middle and the third point was at the upstream of the river. Twenty four (24) samples were collected from a depth of 15 cm using a hand trowel during the period of study. The sampling points were recognized using a Global Positioning System coordinates (GPS). About 2 kg of sample was obtained from each point. The study area, apart from the fact that it is close to residential homes, it also entices activities such as fishing and agricultural activities. The samples were conveyed down to Centre for Energy Research and Development, Obafemi Awolowo University, Ile-Ife for Gamma Spectrometry analysis.

2.3. Preparation of Samples for Gamma Ray Spectroscopy Study

The sediment samples were dried in an oven at a temperature of 60^{o}C for 72 h. The dried samples were pulverized and weighed. The samples were packed into plastic containers which were then hermetically sealed with the aid of PVC tape to prevent the escape of airborne ^{222}Rn and ^{220}Rn from the samples. As part of measures to obtain a reliable result, the plastic containers used have the same geometry with that of the background and the prepared standard sample, which had the same base as the detector surface area. Each empty container with its cover was weighed by the use of an electronic weighing balance. Then each of the pulverized samples was placed in each container, covered, labeled and

re-weighed. They were stored for a period of 28days prior to measurement to attain secular equilibrium and to avoid the escape of ^{226}Ra. Each sample was placed on the NaI detector enclosed in a thick lead shield and counted for 25200s. Gamma ray spectrometry was used to determine and measure the concentration of radionuclides in the samples. The activity concentration was determined using a 7.62 cm x 7.62 cm NaI (TI) detector surrounded with adequate lead shielding that reduces the background by a factor of approximately 95%. A counting time of 25,200s was used. The activities of various radionuclides were determined in Bq/kg from the count spectra obtained from each of the samples 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) for ^{238}U, ^{232}Th and ^{40}K, respectively. For a counting time of 25,200s, the detection limits of the NaI (TI) detector system were calculated as 6.77, 11.40, and 12.85 Bq/kg for ^{40}K, ^{232}Th, and ^{238}U, respectively.

Table 1 Activity Concentration of Oba-Ile Sediment samples in Bq/kg

Sample	**^{40}K**	**^{238}U**	**^{232}Th**
September S1	398.88±10.02	241.12±5.91	27.22±5.12
September S2	339.35±8.55	189.05±6.04	29.80±4.84
September S3	305.02±8.44	91.06±3.11	34.42±5.82
November S1	364.54±9.94	82.29±5.40	35.82±6.91
November S2	312.46±8.07	78.81±5.5	35.33±6.92
November S3	273.36±5.41	69.22±2.41	32.11±4.28
January S1	1662.41±28.19	68.28±1.18	186.14±6.22
January S2	1390.31±46.12	65.52±4.01	133.97±7.91
January S3	933.43±19.31	61.55±5.1	119.91±8.34
February S1	1591.63±28.62	61.94±3.72	163.21±11.22
February S2	1278.10±22.42	59.04±5.31	124.29±9.55
February S3	908.27±18.87	52.56±4.72	101.91±8.54
March S1	1517.71±26.22	51.04±2.66	141.91±10.23
March S2	723.43±19.65	38.25±0.93	81.90±6.58
March S3	1115.29±16.09	45.14±1.04	112.71±9.87
April S1	1470.27±12.25	48.57±3.92	126.96±11.42
April S2	1001.21±7.11	39.38±4.12	97.40±10.78
April S3	601.64±9.07	25.18±3.71	65.20±7.21
May S1	1329.501±10.02	36.28±1.62	113.97±3.51
May S2	980.31±9.32	29.32±0.61	85.49±12.72
May S3	509.06±4.31	17.93±1.13	58.16±9.41
June S1	1262.41±11.53	25.00±2.22	108.71±10.28
June S2	721.63±6.91	18.66±0.04	62.71±7.18
June S3	423.05±5.23	12.61±2.01	31.38±5.06
Mean Value	892.22±14.65	62.83±3.18	87.95±7.91

*S1=first sampling point, *S2= second sampling point, *S3= third sampling point

Table 2 Calculation of Ra_{eq}, Dose Rate, Annual Effective Dose Equivalent (AEDE), Effective Life Cancer Risk (ELCR), External Hazard Index (H_{ex}) and Internal Hazard Index (H_{in})

Sample	$Ra_{(eq)}$Bq/kg	D_R (Abs.gamma dose rate) nGy/h	AEDE $mSvy^{-1}$	ELCR Male10^{-3}	ELCR Female 10^{-3}	H_{ex}	H_{in}
September S1	179.09	61.58	0.08	0.20	0.21	0.67	1.16
September S2	167.44	48.44	0.06	0.16	0.16	0.64	1.10
September S3	160.81	43.69	0.05	0.14	0.15	0.63	1.07
November S1	171.62	56.47	0.07	0.19	0.19	0.68	1.14
November S2	160.13	38.74	0.05	0.13	0.13	0.63	1.07
November S3	151.53	29.89	0.04	0.10	0.10	0.59	1.0
January S1	275.55	118.23	0.15	0.39	0.40	1.81	2.55
January S2	251.83	100.84	0.12	0.33	0.34	1.49	2.17
January S3	209.63	75.93	0.09	0.25	0.26	1.22	1.79
February S1	281.76	126.21	0.15	0.41	0.43	1.72	2.48
February S2	248.71	105.37	0.13	0.35	0.36	1.42	2.09
February S3	213.76	82.14	0.10	0.27	0.28	1.16	1.74
March S1	279.16	129.80	0.16	0.44	0.44	1.62	2.37
March S2	243.28	105.87	0.13	0.34	0.36	1.12	1.78
March S3	204.21	78.39	0.10	0.26	0.26	1.22	1.77
April S1	272.97	124.09	0.15	0.41	0.42	1.53	2.27
April S2	227.73	91.97	0.11	0.30	0.31	1.20	1.82
April S3	188.77	66.20	0.08	0.22	0.23	0.89	1.40
May S1	245.91	101.75	0.12	0.34	0.34	1.38	2.05
May S2	212.06	73.12	0.09	0.24	0.25	1.11	1.68
May S3	168.39	45.26	0.06	0.15	0.15	0.79	1.24
June S1	232.53	86.57	0.11	0.28	0.29	1.31	1.94
June S2	180.49	43.40	0.05	0.14	0.15	0.88	1.37
June S3	161.91	30.54	0.04	0.10	0.10	0.65	1.08
Mean Value	212.05	77.69	0.10	0.26	0.26	1.11	1.67

*S1=first sampling point, *S2= second sampling point, *S3= third sampling point

2.4. Determination of Radiological Hazard Index

2.4.1. Radium equivalent activity concentration index(Ra_{eq})

Radium equivalent (Raeq) index in Bq/kg is a widely used radiological hazard index. It is a convinient index to compare the specific activities of samples containning different concentrations of ^{238}U,^{232}Th and ^{40}K. This radium equivalent

concept allows a single index or number to describe the gamma output from different mixtures of uranium, thorium and potassium in the sediments of Oba-Ile river (Beretka and Matthew,1995; Ravinsankar *et al.,* 2014). It was calculated using the formula;

$Ra_{eq} = A_U + 1.43A_{Th} + 0.077A_K$ ..... [1]

where A_U, A_{Th} and A_K are the specific activity concentrations of ^{238}U, ^{232}Th and ^{40}K in Bq/kg respectively.

2.5. The Absorbed Gamma Dose Rate

The input of natural radionuclides to the absorbed dose rate in air (D_R) at average height of one meter above the surface of ground depends on the natural specific activity concentration of ^{238}U, ^{232}Th and ^{40}K. If a radionuclide activity is known then, its exposure dose rate in air at 1m above the ground can be estimated using the formular given byKurnaz *et al.,* 2007 and Ravinsankar *et al*., 2014:

$D_R\ (nGyh^{-1}) = 0.43A_U + 0.666A_{Th} + 0.042A_K$ ..... [2]

2.5.1. The Annual Effective Dose Equivalent

The annual effective dose equivalent (AEDE) to the population, the UNSCEAR, United Nations Scientific Committee on the Effects of Atomic Radiation (2000) reports a value of 0.7 $SvGy^{-1}$ for the conversion coefficient from absorbed dose in air to effective dose received by adults, the indoor to outdoor ratio (1:4), the outdoor occupancy factor 0.2 and the indoor occupancy factor 0.8 (Kaleel and Mohanad, 2012). Therefore, the annual effective doses outdoors and indoors equivalent are calculated by using the relations:

$D_{outdoor}$ (mSv/yr) = [D_r (mGy/hr) x 24hr x 365.25d x 0.2 x 0.7Sv/Gy] x 10^{-6} ..... [3]

D_{indoor} (mSv/yr) = [D_r (mGy/hr) x24hr x 365.25d x 1.4 x 0.8 x 0.7Sv/Gy] x 10^{-6} ..... [4]

The corresponding worldwide values of D_{out}, D_{ind} and D_{tot} are 0.08, 0.42 and 0.50$mSvy^{-1}$ respectively.

The Excess Lifetime Cancer Risk

The Excess Lifetime cancer risk (ELCR) was calculated using the following equation:

$$ELCR = AEDE \times DL \times RF \quad \text{..... [5]}$$

where, AEDE is the Annual Equivalent Dose Equivalent, DL is the average duration of life (estimated to 54 years), and RF is the Risk Factor (Sv^{-1}), i.e. fatal cancer risk per Sievert. For stochastic effects, ICRP uses RF as 0.05 for public (Taskin et al., 2009).

2.6. External Hazard Index

The external hazard index (H_{ex}) is used to estimate the indoor radiation dose rate due to external exposure to gamma radiation from natural radionuclides in building materials. The equation for determining as reported by Hamzah *et al*., (2008) was presented in equation 6.

$H_{ex} = C_k/4810 + C_{Th}/259 + C_{Ra}/370 \leq 1$ ..... (6)

Where C_{Ra}, C_{Th} and C_K are the activity concentrations in $Bqkg^{-1}$ of ^{226}Ra, ^{232}Th and ^{40}K respectively.

2.7. Internal Hazard Index

The internal hazard index (H_{in}) is a parameter for assessing the negative effect of radioactive materials on lungs and other respiratory organ. The risk was due to the natural radionuclides ^{40}K, ^{226}Ra and ^{232}Th was determined using the equation:

$H_{ex} = C_k/4810 + C_{Th}/259 + C_{Ra}/370 \leq 1$ ..... (7)

3. Result and discussion

3.1. Activity Concentrations of Oba-Ile Samples

The activity values (Table 1) of ^{40}K ranged from 273.36 ± 5.41 Bqkg^{-1} to 1662.41± 28.19 Bqkg^{-1}with an average concentration 892.22 ± 14.65Bqkg^{-1}. The values of ^{232}Th in the sediment samples ranged between 27.22 ± 5.12 Bqkg^{-1} to 186.14 ± 6.22 Bqkg^{-1} with an average concentration 87.95 ± 7.91 Bqkg^{-1}. The values of ^{238}U ranged from 12.61 ± 2.01 Bqkg^{-1} to 241.12 ± 5.91 Bqkg^{-1} with an average 62.83 ± 3.18 Bqkg^{-1}. World average concentrations are 50 Bqkg^{-1}, 50 Bqkg^{-1} and 500 Bqkg^{-1} for ^{228}U, ^{232}Th and ^{40}K respectively (UNSCEAR, 2000).In all the sampling points, mean activity concentration of the natural radionuclide is of the order $^{238}U < ^{232}Th < ^{40}K$. All mean concentrations of ^{40}K, ^{238}U and ^{232}Th in Oba-Ile River were higher than the global background world average concentrations (UNSCEAR, 1994).Potassium-40 (^{40}K) has the highest mean activity concentration, followed by ^{232}Th and ^{238}U in all the samples. Potassium-40 is found in most terrestrial materials with an abundance of 0.012% while ^{232}Th is 1.5 times higher than Uranium in the earth's crust (Kabir *et al*., 2009). Therefore, the activity level of ^{232}Th is expected to be higher than those of ^{238}U (Silver *et al*., 2016).The recorded high values of the radionuclides in the samples may be due to the presence of radioactive-rich granite. Any type of rock could contain naturally occurring radioactive elements like radium, uranium and thorium. Some pieces of granite contain more of these elements than others, depending on the composition of the molten rock from which they formed. Geologists provided an explanation of this behavior in the course of partial melting and fractional crystallization of magma, which enables U and Th to be concentrated in the liquid phase and become incorporated into the silica-rich products. For that reason, igneous rocks of granitic composition are strongly enriched in U and Th (on an average 5 µg/ml of U and 15 µg/ml of Th) (Uosif,*et al*., 2015), compared to the Earth's crust natural concentrations of ^{238}U and ^{232}Th 2.9 and 9.6 µg/ml respectively, which are equivalent to activity concentrations of 36 and 39 Bq/kg, respectively(Agalga *et al*., 2013).

3.2. Radium Equivalent Activity

The radium equivalent activity (Ra_{eq}) in Table 2, ranged from 151.53to 281.76 Bqkg^{-1} with an average value 212.05; it was clearly shown that the obtained values were lower than the recommended maximum value of 370 Bqkg^{-1}(UNSCEAR, 1993).This revealed the investigated samples were within the recommended safety limit.

3.2.1. Absorbed Gamma Dose Rate ($D_{R)}$

The absorbed dose rate (Table 2) ranged from 29.89 to 126.21 nGy.h^{-1} with a mean value 77.69 nGy.h^{-1}. The value was below the worldwide mean of 84 nGy·h–1 for soil matrix as reported by UNSCEAR, 2000. The absorbed dose rate itself does not give an indication of possible biological effects until it is converted to the effective dose equivalent, which is measured in Sieverts (Sv) (Nwankwo and Olubo, 2016).

3.2.2. Annual Effective Dose Equivalent (AEDE)

The annual effective dose equivalent of Oba-Ile sediment (Table 2) varied from 0.04 µSv to 0.16 µSv (average = 0.10 µSv). The calculated values is found to be lower than the recommended value of 1 µSv implying that the radiation hazard is insignificant for the population in Oba-Ile community as at the time when this study was carried out (Nwankwo and Olubo, 2016).

3.2.3. Excess Lifetime Cancer Risk (ELCR)

The potential carcinogenic effects are characterized by estimating the probability of cancer incidence in a population of individuals for a specific lifetime from projected intakes (and exposures). *The calculated values of the excess lifetime cancer risk (ELCR) for males and females (Table 2) ranged between* 0.10 x10^{-3} to 0.43 x10^{-3} having a mean value 0.26 x10^{-3} for males and females ranged between 0.10x10^{-3} to 0.37 x10^{-3} with a mean value 0.26x10^{-3}. When compared with United Nations Scientific Committee on the Effect of Atomic Radiation (UNSCEAR, 2000) who recommended a safe limit of 0.29 x10^{-3}, the calculated value were observed to be lower as at the time this research was undertaken.

External (H_{ex}) and Internal (H_{in}) Hazard Indices

The mean values for the external (H_{ex}) and internal (H_{in}) hazard indices are 1.11 and 1.67. The values obtained are higher than unity. Therefore, sediment obtained from Oba-Ile river should be handled with thoughtfulness to avoid excessive exposure of the inhabitants of the town to radiation since the use of these materials may pose immediate as well as long term negative health implications.

4. Conclusion

The mean activity concentrations of ^{40}K, ^{238}U and ^{232}Th in Oba-Ile River were higher than the global background world average concentrations (50 $Bqkg^{-1}$, 50 $Bqkg^{-1}$ and 500 $Bqkg^{-1}$ for ^{228}U, ^{232}Th and ^{40}K respectively UNSCEAR, 1994).The recorded high values of the radionuclides in the samples may be due to the presence of radioactive-rich granite. The health hazard indices from Oba-Ile study areas are *however, below the permissible limit*. The hazard indices indicated that continuous exposure could lead to some level of health risk. This study specifically confirmed the possible use of the sediments from Oba-Ile river as sharp sand or gutter sand for building purposes at different stages owing to the recorded insignificant levels of the calculated hazard indices. It also provides important information in future studies for subsequent evaluations of the possible future environmental contamination due to activities surrounding this river.

Compliance with ethical standards

Disclosure of conflict of interest

The authors declare no conflict of interest on this work.

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