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EVALUATION OF RADIOLOGICAL HAZARD INDICES AND EXCESS LIFETIME CANCER RISK ASSOCIATED WITH THE USE OF OGUN RIVER SEDIMENT AS BUILDING MATERIAL

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ABSTRACT
The radioactivity concentrations of ^{40}K, ^{238}U and ^{232}Th in sediment samples of Ogun river at Abeokuta, Ogun State in Nigeria were evaluated by gamma spectrometry using NaI (Tl) detector coupled with a pre-amplifier base to a multiple channel analyzer (MCA), these were then used to determine the radiological indicators; radium equivalent activity, internal and external hazard indices, and absorbed dose rate which translated to the annual effective dose rate. 10 sediment samples were collected from the river. The values of the radiological indicators and excess lifetime cancer risk were found to be within the limits recommended, indicating that the use of the sediment of the river as building material does not have radiological health hazards to the occupants of the buildings.

Keywords: Concentration, Radionuclide, Radiological Indicators, Cancer Risk.

INTRODUCTION
The world is naturally radioactive, thereby exposing humans to naturally occurring quantities of radiation on a daily basis. The exposure of human beings to ionizing radiation from natural sources is a continuing and inescapable feature of life on earth. For most individuals, this exposure exceeds that from all man-made sources combined (UNSCEAR, 2000). Radionuclides are chemical elements with unstable atomic structures called radioactive isotopes. In many parts of the world, building materials containing radioactive materials have been used for generations. As individuals spend more than 80% of their time indoor, the internal and external radiation exposure from building materials creates prolonged exposure situation (ICRP, 1999). Radiation being energy emitted when a radionuclide decays. It can affect living tissue only when the energy is absorbed in that tissue. Radionuclides can be hazardous to living tissue when they are inside an organism where radiation released can be immediately absorbed. They may also be hazardous when they are outside of the organism but close enough for some radiation to be absorbed by the tissue. Radionuclides can be released into the air by human activities. They can also be created in the atmosphere by natural processes such as the interaction of cosmic radiation with nitrogen to produce radioactive Carbon-14. Radionuclides can be removed from the air in several ways. Particles settle out of the atmosphere if air currents cannot keep them suspended. Rain or snow can also remove them. When these particles are removed from the atmosphere; they may land in water, on soil, or on the surfaces of living and non-living things. The particles may return to the atmosphere by re-suspension, which occurs when wind or some other natural or human activity generates clouds of dust containing radionuclides (OEPA, 2005). The main external source of irradiation to the human body is represented by the gamma radiation emitted by naturally occurring radioisotopes, also called terrestrial environmental radiation. These radioisotopes, such as ^{40}K and the radionuclides from the ^{232}Th and ^{238}U series and their decay products, exist at trace levels in all ground formations. Therefore, natural environmental radioactivity and the associated external exposure due to gamma radiation depend primarily on the geological and geographical conditions, and appear at different levels in the soils of each different geological region (UNSCEAR, 1993; 2000). However, it has been observed that the type and concentration vary considerably depending on the soil type. The effects of the radiation emitted by different radionuclides depend on the overlining soil material, its chelating agents and physio-chemical properties (Believermis *et al*, 2009). The exposure of man to gamma radiation from these radionuclides in the aquatic environment is not limited to the internal exposure due to ingestion through the consumption of contaminated aquatic foods. The use of river sediments as a constituent of building materials for flooring, plastering and in molding bricks in the coastal areas of Nigeria has the probability of increasing the external exposure level to man if such sediments have high concentration of radionuclides (Oni *et al*, 2011). Sowole (2014) studied the radioactivity concentrations of ^{40}K, ^{238}U and ^{232}Th in surface soil samples of major markets at Sagamu, in Ogun State, obtained the highest radioactivity concentrations of ^{40}K, and ^{238}U from Falawo market surface soil samples with values of 1274.26 ± 4.26 $Bqkg^{-1}$ and 40.72 ± 3.12 $Bqkg^{-1}$, respectively while that of ^{232}Th was obtained from Sabo market surface soil sample with a value of 115.62 ± 16.39 $Bqkg^{-1}$. The mean external hazard index (H_{ex}) and mean internal hazard index (H_{in}) for all the soil samples from Falawo market were calculated to be 0.616 $Bqkg^{-1}$ and 0.691$Bqkg^{-1}$, respectively, and that of Awolowo market were 0.566 $Bqkg^{-1}$ and 0.634 $Bqkg^{-1}$, respectively. Also for Sabo market the mean values were calculated to be 0.594 $Bqkg^{-1}$ and 0.658 $Bqkg^{-1}$, respectively. All the values obtained were less than 1.0 $Bqkg^{-1}$ as recommended for safety by European Commission (1999) and therefore have no negative radiological health implication to the people within the markets and their environs.

This research work is to evaluate radiological hazard indices and excess lifetime cancer risk associated with the use of Ogun river sediment as building material to occupants.

MATERIALS AND METHODS

Ten (10) samples of Ogun river sediments at Abeokuta in Ogun State of Nigeria were collected at distances of 100 – 400m apart. Sediment samples were collected from the bottom of the river by the help of local divers. The collected sediment samples were kept in clean polythene bags and labelled.

The samples were all oven dried at 80^0C, to remove moisture, which otherwise would result in self-absorption of the radionuclides during radio assay. The sediment samples were sieved after pulverization, packed 120.0g in plastic containers of base diameter 5.0cm so as to sit comfortably on the NaI(Tl) detector of diameter 5.1cm used in this work. The samples were all sealed and kept for 28 days in order to obtain secular equilibrium between radioactive daughter nuclides and their respective progenies.

The method of gamma spectrometry was adopted for the analysis of the samples collected in order to obtain data on ^{40}K, ^{238}U and ^{232}Th. The spectrometer used was a Canberra lead shielded 7.6cm x 7.6cm NaI (Tl) detector coupled to a multichannel analyzer (MCA) through a preamplifier base. The resolution of the detector is about 10% at 0.662MeV of 137Cs. According to Jibiri and Farai (1998) the value is good enough for NaI detector to distinguish the gamma ray energies of most radionuclides in samples. For the analyses of ^{40}K, ^{238}U and ^{232}Th, the photo peak regions of ^{40}K (1.46 MeV), ^{214}Bi (1.76 MeV) and ^{208}Tl (2.615 MeV) were respectively used. The cylindrical plastic containers holding the samples were put to sit on the high geometry 7.6cm x 7.6cm NaI (Tl) detector. High level shielding against the environmental background radiation was achieved by counting in a Canberra 10cm thick lead castle. The counting of each sample was done for 10hrs because of suspected low activities of the radionuclides in the samples. The areas under the photo-peaks of ^{40}K, ^{238}U and ^{232}Th were computed using the Multichannel Analyzer system.

Theoretical Consideration and Calculations

The concentrations of the radionuclides were calculated based on the measured efficiency of the detector and the net count rate under each photopeak over a period of 10 hours using equation 1.0

$$C = \frac{N(E_\gamma)}{\varepsilon(E_\gamma) I_\gamma M t_c} \quad \ldots\ldots\ldots\ldots\ldots\ldots \quad 1.0$$

where:

$N(E_\gamma)$ = Net peak area of the radionuclide of interest, $\varepsilon(E_\gamma)$ = Efficiency of the detector for the γ- energy of interest, I_γ = Intensity per decay for the γ- energy of interest, M = Mass of the sample, t_c = Total counting time in seconds (36000s). The radiological indicators are radium equivalent activity (Ra_{eq}), internal hazard index (H_{in}) and external hazard index (H_{ex}). These were used to estimate the radiological implications of the use of the sediment samples from the river as building material to man. The Ra_{eq} was calculated by the equation described by Beretka and Mathew (1985) and Yang *et al* (2005) as indicated by equation 2.0

$$Ra_{eq} = \frac{10}{130} C_k + \frac{10}{7} C_{Th} + C_{Ra} \quad \ldots\ldots\ldots\ldots \quad 2.0$$

where C_{Ra}, C_{Th} and C_K were the activity concentrations in Bqkg^{-1} of ^{226}Ra, ^{232}Th and ^{40}K, respectively. The external hazard index (H_{ex}) commonly used to evaluate the indoor radiation dose rate due to external exposure to gamma radiation from natural radionuclides in building materials as reported by Hamzah *et al* (2008) was presented in equation 3.0.

$$H_{ex} = \frac{C_k}{4810} + \frac{C_{Th}}{259} + \frac{C_{Ra}}{370} \quad \ldots\ldots\ldots\ldots \quad 3.0$$

The internal hazard index (H_{in}) is a parameter for estimating the negative effect of radioactive materials on lungs and other respiratory organs. The risk internal exposure due to the natural radionuclides ^{40}K, ^{226}Ra and ^{232}Th can be assessed from the value of H_{in} using the equation:

$$H_{in} = \frac{C_k}{4810} + \frac{C_{Th}}{259} + \frac{C_{Ra}}{185} \quad \ldots\ldots\ldots \quad 4.0$$

where C_k, C_{Th} and C_{Ra} are the concentrations of ^{40}K, ^{226}Ra and ^{232}Th, respectively.

In addition, considering the definition of the absorbed dose rate in indoor air D(nGy/h) given by UNSCEAR (1993), European Commission (1999) and Papastefanou *et al* (2005) from the natural radionuclides as:

$$D\ (nGy/hr) = 0.080C_k + 1.1C_{Th} + 0.92C_{Ra} \quad \ldots\ldots\ldots \quad 5.0$$

The absorbed dose rate in air translates to the annual effective dose rate indoor for individuals using the values of the absorbed dose rate in indoor air, D(nGy/hr), the effective dose conversion factor taken to be 0.7 Sv.Gy^{-1} and an indoor occupancy factor as 0.8 with the annual occupancy time approximately 7000h/yr. Hence the annual effective dose rate is estimated as:

$$A_E\ (mSv/yr) = D\ (nGy/hr) \times 7000(hr/yr) \times 0.7(Sv/Gy) \times 10^{-6} \quad \ldots \quad 6.0$$

Excess Lifetime Cancer Risk (ELCR) was calculated using the equation (Taskin *et al*., 2009)

$$ELCR = AEDE \times DL \times RF \quad \ldots\ldots\ldots\ldots\ldots\ldots \quad 7.0$$

Where AEDE was determined using the equations:

$$AEDE\ (Outdoor)\ (\mu Sv/y) = D\ (nGy/h) \times 8760h \times 0.7Sv/Gy \times 0.2 \times 10^{-3} \quad \ldots\ldots\ldots\ldots \quad 8.0$$

$$AEDE\ (Indoor)\ (\mu Sv/y) = D\ (nGy/h) \times 8760h \times 0.7Sv/Gy \times 0.8 \times 10^{-3} \quad \ldots\ldots\ldots\ldots \quad 9.0$$

AEDE is the Annual Equivalent Dose Equivalent, DL is average Duration of Life (estimated to be 70 years), and RF is the Risk Factor (S/v), i.e. fatal cancer risk per Sievert. For stochastic effects, ICRP uses RF as 0.05 for the public (Taskin *et al*., 2009).

RESULTS AND DISCUSSION

The radioactivity concentrations of natural radionuclides obtained from the sediments of Ogun river are shown in table 1. The lowest concentration of ^{40}k was 34.71 ± 3.24 Bqkg^{-1} and the highest was 112.23 ± 9.15 Bqkg^{-1}. For ^{238}U, the lowest concentration was 2.36 ± 1.02 Bqkg^{-1} while the highest was 8.24 ± 3.15 Bqkg^{-1}. The lowest concentration of ^{232}Th was 5.23 ± 0.96 Bqkg^{-1} and the highest was 18.68 ± 4.13 Bqkg^{-1}. Concerning the radium equivalent activity (Ra_{eq}) as shown in table 2, the lowest value was 13.2 Bqkg^{-1}, 34.4 Bqkg^{-1} was the highest and the mean value

was obtained as 24.27 Bqkg^{-1}. These values are lower than the recommended limit value of 370 Bq.kg^{-1} as building material (Matiullah *et al*, 2004).

Table 1: Concentrations of natural radionuclides in Ogun river sediment samples

RIVER	SAMPLE	RADIOACTIVITY CONCENTRATIONS OF OGUN RIVER SEDIMENT SAMPLES (Bqkg^{-1})		
		^{40}K	^{238}U	^{232}Th
OGUN	OSS_1	78.23 ± 5.12	4.17 ± 0.65	7.26 ± 2.23
	OSS_2	52.46 ± 3.86	2.36 ± 1.02	10.85 ± 3.35
	OSS_3	97.46 ± 8.09	5.98 ± 2.64	11.57 ± 3.57
	OSS_4	112.23 ± 9.15	8.24 ± 3.15	9.16 ± 3.12
	OSS_5	48.97 ± 2.86	3.56 ± 0.67	11.69 ± 3.61
	OSS_6	65.24 ± 4.82	2.68 ± 0.57	18.68 ± 4.13
	OSS_7	51.63 ± 4.15	3.86 ± 1.02	13.16 ± 3.87
	OSS_8	72.85 ± 5.34	5.62 ± 1.34	6.78 ± 2.07
	OSS_9	84.62 ± 6.18	4.13 ± 0.79	7.36 ± 2.44
	OSS_{10}	34.71 ± 3.24	3.01 ± 0.42	5.23 ± 0.96

Table 2: Determined values of radiological indicators from the sediment samples

SAMPLE	Ra_{eq}(Bq.kg^{-1})	H_{in}(Bq.kg^{-1})	H_{ex}(Bq.kg^{-1})	D(nGy.h^{-1})	A_E(mSvy^{-1})
OSS_1	20.6	0.067	0.056	18.08	0.022
OSS_2	21.9	0.066	0.059	18.30	0.023
OSS_3	30.0	0.097	0.081	26.03	0.032
OSS_4	30.0	0.103	0.081	26.64	0.033
OSS_5	24.0	0.075	0.065	20.05	0.025
OSS_6	34.4	0.100	0.093	28.23	0.035
OSS_7	26.6	0.082	0.072	22.16	0.027
OSS_8	20.9	0.072	0.057	18.46	0.023
OSS_9	21.2	0.068	0.057	18.67	0.023
OSS_{10}	13.2	0.044	0.036	11.30	0.014

Also, internal hazard indices ranged from 0.044 Bqkg^{-1} to 0.103 Bqkg^{-1} with mean value of 0.077 Bqkg^{-1} and external hazard indices ranged from 0.036 Bqkg^{-1} to 0.093 Bqkg^{-1} with mean value of 0.066 Bqkg^{-1}. All the values are below 1.0 Bqkg^{-1} as recommended limit (European Commission, 1999). The absorbed dose rate ranged from 11.30 nGyh^{-1} to 28.23 nGyh^{-1} with mean value of 20.79 nGyh^{-1}. These are below the worldwide mean of 84.0 nGyh^{-1} (UNSCEAR, 2000), and this translates to the mean annual effective dose rate of 0.026 mSvy^{-1}. Average excess lifetime cancer risk for all the samples was calculated to be 2.23 x 10 $^{-5}$ which is below the world average of 2.9 x 10^{-4} (Taskin *et al*., 2009).

Conclusion

The values of radiological indicators from Ogun river sediment samples had been evaluated using concentration values of natural radionuclides obtained from gamma spectrometry analyses, which were below the recommended values worldwide. Therefore, Ogun river sediment can be used as building material because it poses no radiological health hazard to the occupants.

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