

# Natural Radionuclides and Hazards of Sediment Samples Collected from Osun River in Southwestern Nigeria.

O.A. Oyebanjo<sup>1\*</sup>; E.O. Joshua<sup>2</sup>; and N.N. Jibiri<sup>2</sup>

<sup>1</sup>Department of Physics and Telecommunication, Tai Solarin University of Education, Ijebu-Ode, Nigeria.

<sup>2</sup>Department of Physics, University of Ibadan, Ibadan, Nigeria.

E-mail: [oyebanjookemi@yahoo.com](mailto:oyebanjookemi@yahoo.com)\*

## ABSTRACT

The concentrations of the natural radionuclides in sediments of the Osun River were determined using gamma ray spectrometer NaI(Tl) detector coupled to a Multichannel Analyzer for spectral analysis. A total of 106 sediment samples were collected from 25 locations spanning the whole length of the river course from Oke-Imesi Ridge, about 5km North of Efon Alaiye to Lekki Lagoon.

Sampling locations were spread out over the five states that the river traversed. The number of locations per state was partly due to accessibility. Hazard indices were used to determine the suitability of the sediment samples for building in the context of radiological hazard. Results obtained showed that the mean concentrations of <sup>40</sup>K, <sup>238</sup>U, and <sup>232</sup>Th in the sediments varied between 175.6 ± 6.1 Bq/kg in lower course to 188.5 ± 7.2 Bq/kg in upper course, 28.4 ± 2.0 Bq/kg in upper course to 13.1 ± 1.4 Bq/kg in lower course and 11.4 ± 0.3 Bq/kg in upper course to 16.3 ± 0.4 Bq/kg in middle course. The mean radium equivalent varied between 42.9 ± 9.0 and 65.7 ± 13.0 Bq/kg while external hazard index ranged from 0.1 ± 0.1 to 0.2 ± 0.1. The radium equivalent and external hazard index values were below the recommended international limits of 370 Bq/kg and 1.0, respectively.

(Keywords: radioactivity, river sediments, hazard indices, gamma ray spectrometer)

## INTRODUCTION

Sediments have been widely used as environmental indicator and their ability to trace contamination sources and monitor contaminants is widely recognized (Sansone, et al., 2008). Radioactivity in the environment consists of the three well known radioactive series of uranium,

thorium, and actinium. The uranium series originates with <sup>238</sup>U is less than 1%, hence the contribution of <sup>235</sup>U to the environmental dose is very small (Walley El-Dine, et al., 2001). Several singly occurring radionuclides are also present in the environment. The most important of these is <sup>40</sup>K because it is a gamma ray emitter in addition to beta decays and therefore contributes significantly to gamma radiation exposure (Xinwei, et al., 2006).

Sediments originate from soils and rocks. Radionuclides are present in rocks in varying amounts, and they are easily mobilized into the environment. Radioactivity in sediments results from the parent rock which they are derived. The distribution of naturally occurring radionuclides depend on the distribution of rocks from which they originate and the processes which result to their removal from the soil and migrate them (Joshua, et al., 2009). Therefore, the naturally environmental radioactivity mainly depends on geological and geophysical conditions (Florou and Kritidis, 1992). The concentration of natural radionuclides in the rock varies considerably depending on the rock formation and lithologic character (Anjos, et al., 2005; Tzortis, et al., 2003).

Little is known about the concentrations of radionuclides in the sediments of Osun River basin which is located in an area whose boundaries lie approximately between latitudes 8° 20' N and 6° 30' N, and longitudes 5° 10' E and 3° 25' E. The greater part of the basin is located in Osun and Ogun States with less than 7 percent of the total area located in Oyo, Ekiti and Lagos States. The drainage system of Osun River rises in Oke-Mesi ridge, about 5 km North of Efon Alaiye on the border between Oyo and Ondo States of Nigeria, and flows North through the Itawure gap to latitude 7° 53' before winding its way Westwards through Osogbo and Ede and

Southwards to enter Lagos lagoon about 8 km east of Epe (Tahal 1979).

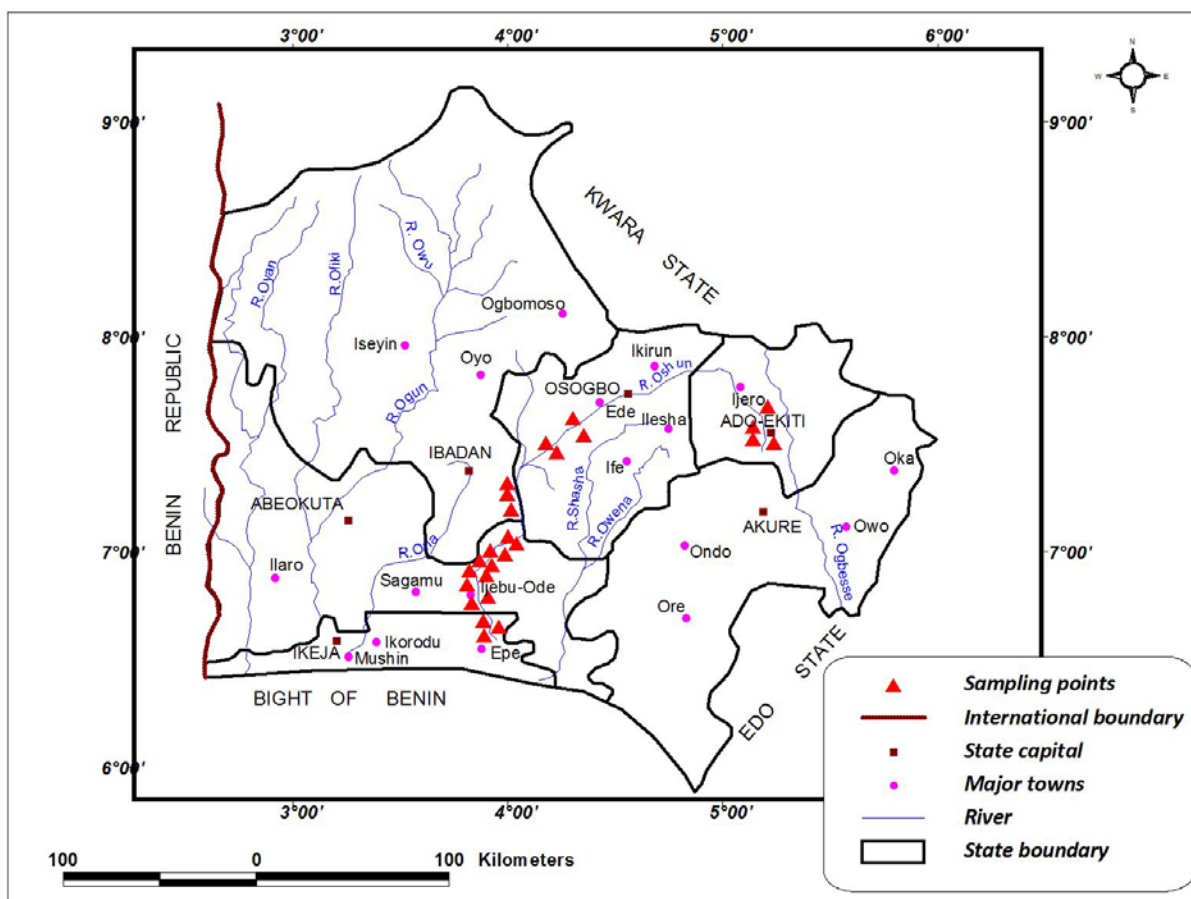
The objective of this study is to determine the activity concentrations of natural radionuclides ( $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$ ) in sediment samples collected from Osun River in south western part of Nigeria and estimate the radiological hazard associated with the sediments; which serve as components of building materials.

### SAMPLE PREPARATION AND MEASUREMENT

A total of 106 sediment samples were collected from 25 locations spanning the whole length of the river course as shown in Figure 1. The number of locations per state is partly due to accessibility, the distance traversed by the River in each state and largely the level of human activities along the River basin. The sediment

samples were pulverized, sieved through mesh at the department of Physics, university of Ibadan. The pulverized samples were dried, weighed and sealed in cylindrical plastics containers. The sealed samples were stored for four weeks before counting. This was to allow for the reaching of secular equilibrium between  $^{226}\text{Ra}$  and its short lived decay products.

A gamma-ray scintillation spectrometry system was used for the measurements of the natural radionuclides content of the rock samples. The spectrometry is a Canberra 7.6cm x 7.6 cm NaI (TI) detector (Model No. 802-series). The spectroscopic system is connected to Canberra series 10 plus Multichannel Analyses (MCA) (Model No. 1104) through a preamplifier base, which enables data acquisition, storage and display of the acquired spectra. The resolution of the detector is about 8% at 0.662 MeV  $^{137}\text{Cs}$  which is capable of distinguishing the gamma-ray energies used for the measurements.



**Figure 1:** Map Showing the Sampling Points in Five States along Osun River Basin in Southwestern Nigeria. (Source: Modified from Ajayi, 2000)

The efficiency calibration of the system was done using reference soil sample prepared from Rocketdyne Laboratories, California, USA. The reference soil sample is traceable to a mixed standard gamma source (No. 48722-356) by Analytix Inc., Atlanta, Georgia, USA. The net count under each photo peak was related to the specific activity of the radionuclide using the calibration factors obtained for  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  by the following relation (Tzortzis, et al., 2003):

$$A_{Ei} = \frac{N_{Ei}}{M_E \times t \times Y \times M_s} \quad (1)$$

Where  $A_{Ei}$  is the specific activity in Bq Kg<sup>-1</sup> of a nuclide I and for a peak at energy E. The net peak count of a peak energy E is  $N_{Ei}$ .

mean = standard deviation.

$M_E$  is the detection efficiency at energy E and t is the counting live time. The gamma-ray yield per disintegration of the specific nuclide for a transition at energy E is Y while  $M_s$  is the mass in kg of the measured sample.

Each sample was put on the shielded NaI (TI) detector and counted for an accumulating period of 36, 000 seconds (10 hours). Correction was made for background radiation level. The determination of  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  were based on the measurements of photo-peaks from  $^{214}\text{Bi}$  (1.76 MeV) and  $^{208}\text{Tl}$  (2.62 MeV), respectively.

The primary decay of  $^{40}\text{K}$  (1.46 MeV) was measured directly (Chiozzi, et al., 2000). The lower limits of detection (LLD of  $^{226}\text{Ra}$  ( $^{214}\text{Bi}$ ),  $^{232}\text{Th}$  ( $^{208}\text{Tl}$ ) and  $^{40}\text{K}$  were determined (IAEA,1989) from the background radiation. The LLD obtained are 4.0,4.8 and 17.0, Bq kg<sup>-1</sup>, respectively, for  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$ .

## RESULTS AND DISCUSSION

**Table 1:** Mean Activity Concentrations of  $^{40}\text{K}$ ,  $^{238}\text{U}$ , and  $^{232}\text{Th}$  with Standard Deviations in Osun River Sediments.

States	No of Samples	$^{40}\text{K}$ (Bq/Kg)		$^{238}\text{U}$ (Bq/Kg)		$^{232}\text{Th}$ (Bq/Kg)	
		Range	Mean ± SD	Range	Mean ± SD	Range	Mean ± SD
Ekiti	16	609.78 - 83.35	214.63 ± 164.45	56.5 - 8.79	32.70 ± 14.00	48.85 - 6.01	17.7 ± 10.40
Osun	18	423.67 - 8.33	162.38 ± 182.00	53.6 - 0.86	24.00 ± 23.90	35.7-1.25	14.20 ± ± ± 35.70
Oyo	12	340.31 - 64.76	188.43 ± 94.50	44.13 - 5.68	23.90 ± 8.95	19.32 - 2.69	11.30 ± 8.03
Ogun	46	610.90 - 25.32	253.00 ± 158.00	46.72 - 4.65	22.10 ± 10.20	32.52 - 2.23	15.00 ± 6.70
Lagos	14	549.50 - 99.22	278.00 ± 136.00	43.27 - 1.63	21.98 ± 13.00	33.48 - 3.10	16.47 ± 8.68

**Table 2:** Mean Activity Concentrations of  $^{40}\text{K}$ ,  $^{238}\text{U}$ ,  $^{232}\text{Th}$  with Standard Deviations in the Upper, Middle and Lower Courses in Sediments of Osun River.

River Course	No of Samples	$^{40}\text{K}$ (Bq/Kg)		$^{238}\text{U}$ (Bq/Kg)		$^{232}\text{Th}$ (Bq/Kg)	
		Range	Mean ± SD	Range	Mean ± SD	Range	Mean ± SD
Upper	34	8.33 - 609.78	188.50 ± 173.20	0.86 - 56.5	28.35 ± 18.40	1.25 - 48.85	15.95 ± 7.20
Middle	17	340.31 - 64.76	184.11 ± 92.00	5.68 - 44.13	25.07 ± 9.00	2.69 - 35.7	16.33 ± 8.40
Lower	55	610.90 - 25.32	175.60 ± 94.50	1.63 - 46.72	13.06 ± 7.05	2.23 - 33.48	11.40 ± 6.01

From Table 1, the mean concentration values of  $^{40}\text{K}$  with standard deviations showed a significant contributor to the radioactive elements present in the river sediment throughout the five states along the river basin. However, appreciable mean concentration values of  $^{238}\text{U}$  and  $^{232}\text{Th}$  (32.7±14.0 Bq/Kg, 17.7±10.4 Bq/Kg) were recorded in Ekiti State while low mean concentration values of  $^{238}\text{U}$  and  $^{232}\text{Th}$  were found to be  $21.98 \pm 13.00 \text{ Bqkg}^{-1}$  and  $11.30 \pm 8.03 \text{ Bqkg}^{-1}$  in Lagos and Oyo states, respectively. In all the states, the mean concentration of  $^{238}\text{Th}$  has the lowest value and  $^{238}\text{U}$  mean concentration value decreases as we move down from the source to the mouth of the river.

The mean Activity Concentrations of  $^{40}\text{K}$ ,  $^{238}\text{U}$ , and  $^{232}\text{Th}$  with standard deviations in the upper, middle and lower courses in sediments of Osun River are presented in Table 2. The mean activity concentration of  $^{40}\text{K}$  decreases downward from the upper course of the river to the lower courses (ranges from  $188.5 \pm 173.3 \text{ Bq/Kg}$  to  $175.60 \pm 94.5 \text{ Bq/Kg}$ ). The distribution of mean concentration of  $^{238}\text{U}$  also follow the same pattern

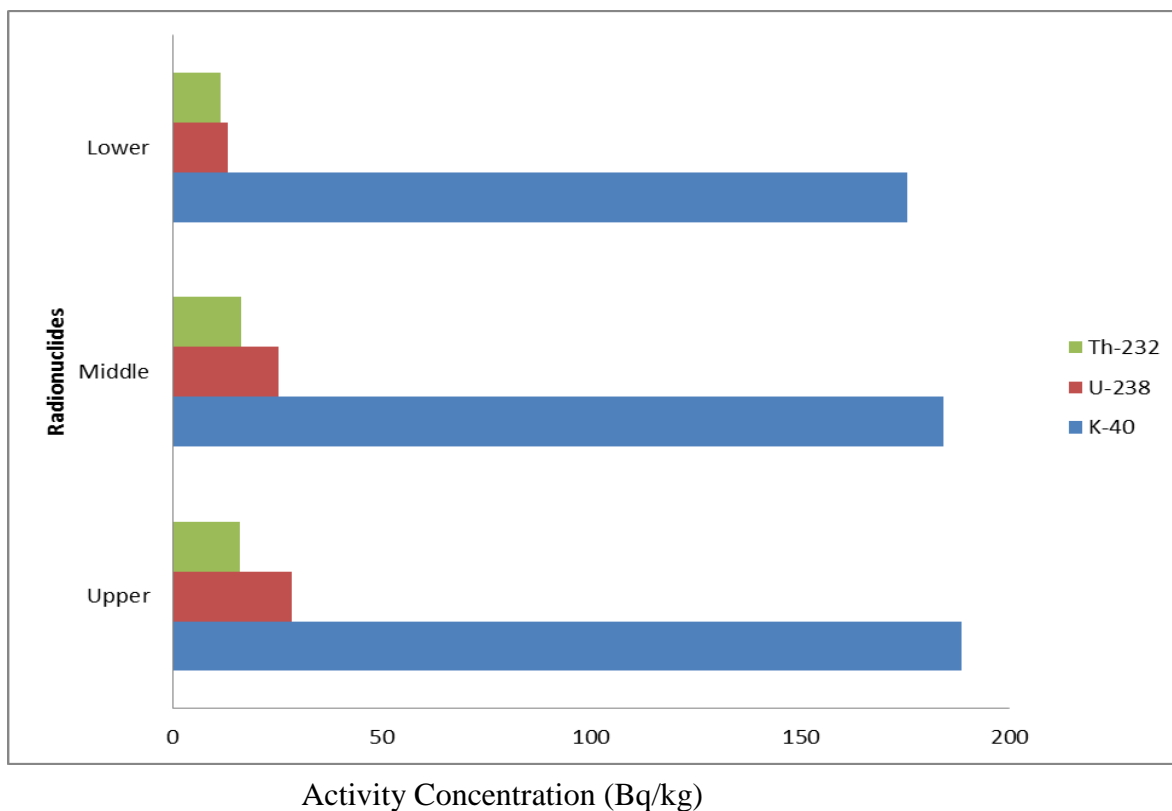
as it ranged from  $28.35 \pm 18.4 \text{ Bq/Kg}$  in upper course to  $13.06 \pm 7.05 \text{ Bq/Kg}$  in lower course.

The highest mean concentration value for  $^{232}\text{Th}$  was found in middle course followed by the upper course and then the lowest value of  $11.40 \pm 6.01 \text{ Bq/Kg}$  was recorded in the lower course of the river. The mean activity concentration were below the recommended international limits of  $370 \text{ Bq/Kg}$  (UNSCEAR 2000) and as such the sediments may be considered suitable for use along the river basin.

Radium equivalent activity is a widely used hazard index (Bereka and Mathew, 1985). The index is based on the estimation that  $370 \text{ Bq/kg}$  of  $^{226}\text{Ra}$ ,  $4180 \text{ Bq/kg}$  of  $^{40}\text{K}$  and  $259 \text{ Bq/kg}$  of  $^{232}\text{Th}$  produce the same gamma ray dose rate and can be expressed as:

$$\text{Ra}_{\text{eq}} = C_{\text{Ra}} + 1.43C_{\text{Th}} + 0.077C_{\text{K}} \quad (2)$$

Where  $C_{\text{Ra}}$ ,  $C_{\text{Th}}$ , and  $C_{\text{K}}$  are the activity concentrations of  $^{226}\text{Ra}$ ,  $^{40}\text{K}$ , and  $^{232}\text{Th}$ , respectively, in Bq/kg.



**Figure 2:** Mean Activity Concentration of Radionuclides of Sediments Samples in the Upper, Middle and Lower Courses of Osun River.

The  $Ra_{eq}$  calculated using the mean activity concentrations obtained in the lower, middle and upper courses are 42.88, 62.54, and 65.68 Bq/kg, respectively. These values are lower than the recommended maximum value of 370 Bq/kg (NEA-OECD, 1979; Bereka and Mathew, 1985).

The external hazard index  $H_{ex}$  defined as:

$$H_{ex} = \frac{\frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810}}{\quad} \quad (3)$$

This is used to evaluate the suitability of a material for building. The value of  $H_{ex}$  must be less than unity in order for the radiation exposure owing to radioactivity in construction materials to be limited to 1.5 mSv/y (Bereka and Mathew, 1985; Xinwei, et al., 2006).

The value of  $H_{ex}$  obtained for the upper course is 0.12 which is lower than the value recorded in the middle course 0.16. The  $H_{ex}$  value for 0.11 which is closer to the value obtained at the upper level of the river. These values are lower than the recommended limit of one.

## CONCLUSION

This study presents the activity concentrations of natural radionuclides, radium equivalent activity and external indices of sediment samples of Osun River. The activity concentration of  $^{40}K$  was highest in all the samples. The radium equivalent activity and the external hazard index calculated for the samples were below the recommended limits. The results obtained from this study show that the sediment samples do not pose any significant radiation hazard when used as building components.

## RECOMMENDATION

More systematic studies of Osun River basin geological rock units are required in order to express the exposure levels due to terrestrial gamma radiation. However, the present data provide useful information to investigate future occurrence of radioactive elements in the region.

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