

# Gamma Spectrometric Analysis of Soil, Sediment and Water Samples of Granitic-Type Solid Mineral Mining Activities

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**Abstract:** The activity concentrations of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K in soil, sediment and water from tantalite mine in Iluku-Ofiki, Oke-Ogun, Oyo State, Southwestern Nigeria were determined using Thallium doped Sodium Iodide (NaI (TI)) detector-based gamma spectrometry. This was with a view to determine the health implication of the miners and the populace. The average activity concentrations of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K were found to be  $8.42 \pm 2.50$ ,  $10.28 \pm 3.24$  and  $337.08 \pm 63.65$  Bq kg<sup>-1</sup> in soil;  $6.37 \pm 2.00$ ,  $14.49 \pm 5.57$  and  $320.40 \pm 41.10$  Bq kg<sup>-1</sup> in sediment; and  $4.26 \pm 1.52$ ,  $9.13 \pm 3.80$  and  $83.79 \pm 21.39$  Bq L<sup>-1</sup> in water, respectively. The average activity concentrations of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K in the soil and sediment samples were lower than the UNSCEAR activity limits of 32, 45 and 420 Bq kg<sup>-1</sup> respectively. The estimated average annual effective dose due to these radionuclides in soil was  $44.65 \pm 11.34$  μSv y<sup>-1</sup> and in sediment  $46.3 \pm 11.07$  μSv y<sup>-1</sup>. These respective average annual effective doses from the said radionuclides in soil and sediment were each lower than the UNSCEAR dose limit of 70 μSv y<sup>-1</sup>.

The radiological variables were treated with statistical analysis so as to determine the similarities and correlations among various samples. Two component representations of the data acquired were generated by the principal component analysis in which 94.60 % of the total variance was explained.

**Keywords:** Tantalite, Sediment, Radionuclides, Radiation hazard, Loading plot

## 1 Introduction

The naturally occurring radioactive materials pose exposure risks to man. The radionuclides can be categorized into cosmogenic and primordial. The primordial radionuclides are originated in the earth's crust and these natural radionuclides of interest mainly include <sup>238</sup>U, <sup>232</sup>Th, and <sup>40</sup>K. About 85% of the radiation dose received by man comes from the natural radionuclides [1]. The radionuclide in soil and sediment is unevenly distributed; hence natural radioactivity in the earth's environment depends primarily on the geographical and geological conditions, resulting in the various levels of radionuclides in the matrices [2]. The formation, chemical and biochemical interactions of soil and sediment influence the distribution patterns of the primordial radionuclides such as uranium, thorium and their decay products [3].

When mineral resources are mined from natural radioactive material-hosted site, the wastes generated are rich in technologically enhanced naturally occurring radioactive materials [4]. In the course of mining the resources, the

activities which involve drilling, leaching, handling, storage, transportation of mineral ores as well as the use of likely contaminated tools result in environmental pollution. These lead to the increased level of natural radionuclides, resulting in the potential radiation exposure of the miners and the public [5]. For instance, Columbite-tantalite, which is the major source of tantalum, occurs mainly as accessory minerals disseminated in granitic rocks or in pegmatite associated with granites [6, 7].

The assessments of concentrations of thorium, uranium and the potassium in soil samples from tantalite mining sites have been carried out [6, 7]. The mining and processing of tantalite generate wastes which are commonly left within and around the mining and processing sites. The heaps of tailings are distributed by wind and erosion.

For this reason, it is very crucial to assess the level of naturally occurring radionuclides as well as their daughter nuclides along with the <sup>40</sup>K in the tailings generated from the tantalite processing sites. This is so crucial especially in an area such as Iluku in Oyo State, Nigeria where the

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miners and settlers in the region are exposed through inhalation of radioactive dust fallout from the mine dumps, ingestion of food crop grown on the land, usage of mine water for bathing and through the discharge of surface water run-off into water bodies.

In this study, we determined the level of radionuclides in the soil, sediment and water from the mining site, using gamma spectrometric technique centered on the NaI(Tl) detector. The data obtained was used to assess the possible radiological hazards by estimating the doses and the derived doses.

## 2 Experimental Sections

### 2.1 Geology of the study area

The study area is situated in Iluku, Atisbo Local Government, Oke-ogun, Oyo state South-western Nigeria. Iluku lies between latitudes N8° 20' and N8° 30' and longitudes E03° 10' and E03° 15'. The area is within the Pre-Cambrian Basement Complex of South-western Nigeria, which contains magnetite, gneiss and schist. Ogun River is known to be the major river in Oke-Ogun. The undulating lowland terrain and small hills which occurs as isolated mountains are the common relief in Oyo State. In the study area, the prominent rock unit is the meta-sediments. While the granite, granitogneiss and porphyritic granite occur as nominal rocks. The mineral deposits in the region include tantalite, amphibolite which serves as the major hosts for the pegmatite intrusions and granites [8, 9].

### 2.2 Sample Collection and Preparation

Soil, sediment and water samples were collected from Iluku, tantalite mining site in Ofiki, Oke-Ogun, Oyo State. Fifteen soil samples were collected at various points, 20 metres apart and at a depth of about 10 to 15 cm. Each sample was collected into a sample bag to avoid cross contamination. Two sediment samples were collected from two processing ponds located within the site, each sample from each pond. Two water samples were also collected from the two processing ponds, each sample from each processing ponds. The sediment samples were collected into sample bags and water samples into sample bottle.

The wet soil and sediment samples were air dried at the laboratory to constant weight. The dried samples were crushed with Rocklab Ring Mill and sieved through 2-mm mesh at the Centre for Energy Research and Development (CERD), ObafemiAwolowo University, Ile-Ife, Osun State, Nigeria. The water samples were acidified with 11 M HCl at the rate of 10 ml per litre in order to prevent adsorption of the radionuclides to the walls of the containers. The soil and sediment samples were, each, weighed to 200 g while the water samples were 200 ml. The soil, sediment and water samples were each sealed in a 250 ml air-tight PVC container [10] and stored for a period of one month to

achieve secular equilibrium between  $^{226}\text{Ra}$  and their short-lived progenies [11].

### 2.3 Radioactive measurements

The activity concentrations of radionuclides in the samples were determined using a 7.62 cm by 7.62 cm NaI (Tl) detector. The detector was housed in a cylindrical lead shield of approximately 100 mm thickness with a complete electronic instrumentation coupled to a PC-based multichannel analyzer for data acquisition and gamma spectra analysis. The energy calibration was carried out using standard sources of known gamma-ray energies and activities prepared by the Isotope Products Laboratories, Burbank, California, USA. An empty container having the same geometry as the sample container was counted. This count was subtracted from the gross count so as to determine the background count.

The activity concentrations of  $^{238}\text{U}$  and  $^{232}\text{Th}$  were determined using the 1120.3 keV line of  $^{214}\text{Bi}$ , and the 911.1 keV line of  $^{228}\text{Ac}$ , respectively. The activity concentration of  $^{40}\text{K}$  was determined directly by measuring the gamma-ray transitions at 1460.8 keV. The detection limits of the NaI(Tl) detector system were calculated to be 6.77, 11.40, and 12.85 Bq kg<sup>-1</sup> for  $^{40}\text{K}$ ,  $^{232}\text{Th}$ , and  $^{238}\text{U}$ , respectively. For appreciable counts to be obtained under the photo peaks, the prepared samples were placed in the detector and counted for 25,200 s.

### 2.4 Statistical analysis

To identify the relationship that exists among the various radiological parameters, principal component analysis, Pearson's correlation analysis and loading plot have been used to treat these parameters.

## 3 Results and Discussion

### 3.1 Activity concentrations of $^{238}\text{U}$ , $^{232}\text{Th}$ and $^{40}\text{K}$

Table 1 shows the average activity concentrations of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in the soil, sediment and water samples. In soil samples, the activity concentrations ranged from 1.76 ± 0.36 to 12.28 ± 4.32 Bq kg<sup>-1</sup>, 6.50 ± 3.11 to 16.15 ± 4.21 Bq kg<sup>-1</sup> and from 203.70 ± 51.20 to 659.95 ± 112.21 Bq kg<sup>-1</sup>, with the mean values of 8.42 ± 2.50 Bq kg<sup>-1</sup>, 10.28 ± 3.24 Bq kg<sup>-1</sup> and 337.08 ± 63.65 Bq kg<sup>-1</sup>, respectively. In sediment samples, the activity concentrations ranged from 6.15 ± 1.97 to 6.58 ± 2.02 Bq kg<sup>-1</sup>, 13.32 ± 5.11 to 15.66 ± 6.02 Bq kg<sup>-1</sup> and 313.30 ± 42.30 to 327.50 ± 39.80 Bq kg<sup>-1</sup>, with mean values of 6.37 ± 2.00 Bq kg<sup>-1</sup>, 14.49 ± 5.57 Bq kg<sup>-1</sup> and 320.40 ± 41.10 Bq kg<sup>-1</sup>, respectively. In the water samples, the activities ranged from 1.76 ± 0.73 to 6.76 ± 2.31 Bq L<sup>-1</sup>, 7.47 ± 3.36 to 10.79 ± 4.24 Bq L<sup>-1</sup> and 78.04 ± 20.89 to 89.54 ± 21.88 Bq L<sup>-1</sup>, with the mean values of 4.22 ± 1.52 Bq L<sup>-1</sup>, 9.13 ± 3.80 Bq L<sup>-1</sup> and 83.79 ± 21.39 Bq L<sup>-1</sup> respectively. These ranges of values for the respective

**Table 1-** Activity concentrations (Bq Kg-1). ND=Not detected

Sample code	<sup>238</sup> U	<sup>232</sup> Th	<sup>40</sup> K
SL 1	10.3±3.08	11.48±3.41	404.14±98.33
SL 2	9.84±2.17	7.36±2.32	498.87±99.81
SL 3	9.74±3.11	7.39±1.81	203.70±51.20
SL 4	1.76±0.36	6.50±3.11	275.74±43.60
SL 5	9.22±2.15	10.45±4.90	368.34±57.21
SL 6	10.06±2.31	16.15±4.21	353.03±49.15
SL 7	2.66±1.10	ND	245.68±51.67
SL 8	5.15±2.03	13.87±4.25	314.42±48.09
SL 9	11.47±3.02	12.72±4.13	252.74±67.66
SL 10	7.90±2.12	13.19±2.75	231.94±48.23
SL 11	11.77±4.31	8.17±2.66	229.00±39.31
SL 12	12.28±4.32	13.14±3.32	334.16±58.99
SL 13	7.02±2.10	14.37±5.01	659.95±112.21
SL 14	8.40±2.31	10.53±4.05	317.54±53.19
SL 15	8.71±3.02	8.89±2.60	366.90±76.06
Average	8.42±2.50	10.28±3.24	337.08±63.65
SD 1	6.15±1.97	13.32±5.11	313.30±42.30
SD 2	6.58±2.02	15.66±6.02	327.50±39.80
Average	6.37±2.00	14.49±5.57	320.40±41.10
PW 1	6.76±2.31	10.79±4.24	78.04±20.89
PW 2	1.76±0.73	7.47±3.36	89.54±21.88
Average	4.26±1.52	9.13±3.80	83.79±21.39

**Table 2-** Annual absorbed dose (DR), Annual effective dose (Ad).

Sample code	Annual absorbed dose	Annual effective dose
	D <sub>R</sub> (nGyh <sup>-1</sup> )	A <sub>d</sub> (μSvy <sup>-1</sup> )
SL 1	28.67± 7.61	52.77± 14.01
SL 2	29.94± 6.60	55.12± 12.14
SL 3	17.52± 4.68	32.25± 8.62
SL 4	16.32± 3.88	30.04± 7.14
SL 5	26.04± 6.36	47.94± 11.70
SL 6	29.23± 5.67	53.81± 10.45
SL 7	11.55± 2.68	21.26± 4.93
SL 8	23.96± 5.52	47.11± 10.17
SL 9	23.6± 6.73	43.44± 12.39
SL 10	21.36± 4.67	39.32± 8.59
SL 11	19.99± 5.25	36.8± 9.66
SL 12	27.64± 6.48	50.89± 11.93
SL 13	39.64± 8.71	72.97± 16.03
SL 14	23.58± 5.75	43.4± 10.58
SL 15	24.8± 6.16	45.66± 11.34
Average	24.26± 5.79	44.65± 10.65
SD 1	24.05± 5.77	44.26± 10.63
SD 2	26.25± 6.24	48.33± 11.49
Average	25.15± 6.01	46.3± 11.07
PW 1	12.92± 4.51	23.78± 8.29
PW 2	9.09± 3.29	16.73± 6.05
Average	11± 3.90	20.25± 7.17

radionuclides of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K in soil and sediment samples are lower than the activity limits of 32, 45 and 420 Bq kg<sup>-1</sup>[12].

<sup>238</sup>U gives the least contribution of 2.37 % to the total activity in the area. <sup>232</sup>Th gives the second highest contribution to the total activity in the study area and constitutes about 3.24 % of the total activity in the mine. <sup>238</sup>U has been reported to have higher solubility and mobility than <sup>232</sup>Th even if they occur at the same concentration in the source [13, 14]. This is confirmed in the activity concentrations of <sup>232</sup>Th and <sup>238</sup>U estimated from both the soil and sediment samples as the values of <sup>232</sup>Th are higher than the values of <sup>238</sup>U.

The higher abundance of <sup>232</sup>Th than <sup>238</sup>U in the water samples further established the fact that <sup>238</sup>U has higher mobility than <sup>232</sup>Th, hence, <sup>232</sup>Th is readily more retained in the water than <sup>238</sup>U.

In the same vein, the use of generated wastes may not pose immediate risk to the miners and populace, but long-term exposure to radiation from these radionuclides may adversely affect human health.

### 3.2 Absorbed dose rate (D<sub>R</sub>)

The absorbed dose rate in air at 1 metre above the ground has a direct relationship between activity concentrations of natural radionuclides <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K. The calculated absorbed gamma dose rate in air at each location was estimated thus [12]:

$$D \text{ (nGy h}^{-1}\text{)} = 0.462 A_U + 0.604 A_{Th} + 0.042 A_K \quad (1)$$

where A<sub>U</sub>, A<sub>Th</sub> and A<sub>K</sub> are the activity concentrations of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K in Bq kg<sup>-1</sup> respectively. As shown in Table 2, the absorbed dose rate due to <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K in the soil samples ranged from 11.55 ± 2.68 to 39.60 ± 8.11 nGyh<sup>-1</sup>, with an average value of 24.26 ± 5.79 nGy h<sup>-1</sup> and in sediment samples it ranged from 24.05 ± 5.77 nGyh<sup>-1</sup> to 26.25 ± 6.24 nGyh<sup>-1</sup>, with an average value of 25.15 ± 6.01 nGyh<sup>-1</sup>. In the water samples, it ranged from 12.92 ± 4.51 to 9.09 ± 3.29 nGy h<sup>-1</sup>, with the average value of 11.00 ± 3.90 nGy h<sup>-1</sup>. The mean dose rate in soil was found to be lower than the dose limit of 59nGy h<sup>-1</sup> [12].

### 3.3 Annual effective dose equivalent (A<sub>d</sub>)

In order to estimate the annual effective dose, the conversion coefficient from absorbed dose and the outdoor occupancy factor were considered. The mean numerical values of these parameters vary with the age of the population and the climate at the study location. In some studies, the value of 0.7 Sv Gy<sup>-1</sup> for the conversion coefficient and 0.456 for the outdoor occupancy factor were used. In Iluku, Nigeria, the miners spend about 7 hours in the mining site. This implies that about 30% of time is spent outdoors. Thus, the outdoor effective dose rate due to the activities of the sample matrices was estimated using

[4]:

$$\text{AnnualEffectiveDose}(\mu\text{Svy}^{-1}) = \text{Dose}(\text{nGy h}^{-1}) \times 24(\text{h}) \times 365.25(\text{d}) \times 0.3(\text{OCPF}) \times 0.7\text{Sv Gy}^{-1}(\text{CONVC}) \times 10^{-3} \quad (2)$$

where OCPF and CONVC are the occupancy factor and the conversion coefficient respectively.

In the soil and sediment samples, average annual outdoor effective dose (Table 2) ranged from  $21.26 \pm 4.93$  to  $72.79 \pm 16.03 \mu\text{Sv y}^{-1}$  and  $44.26 \pm 10.63$  to  $48.33 \pm 11.49 \text{nGy h}^{-1} \mu\text{Sv y}^{-1}$ , with average annual effective doses of  $44.65 \pm 10.65 \mu\text{Sv y}^{-1}$  and  $46.30 \pm 11.07 \mu\text{Sv y}^{-1}$  respectively. In water samples, it ranged from  $16.73 \pm 6.05$  to  $23.78 \pm 8.29$  with an average value of  $20.25 \pm 7.17 \mu\text{Sv y}^{-1}$ . The respective mean annual effective doses in soil and sediment samples were each lower than the dose limit of  $70 \mu\text{Sv y}^{-1}$  [12].

### 3.4 Radium equivalent ( $R_{eq}$ )

As a result of the distribution of natural radionuclides As a result of the distribution of natural radionuclides which is not uniform in the samples, the actual activity level of naturally occurring radionuclides in the samples can be assessed by the radium equivalent activity ( $R_{eq}$ ). This gives a single index which describes the gamma output from different mixtures of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in the samples. The radium equivalent activity was calculated thus [15]:

$$R_{eq} (\text{Bq Kg}^{-1}) = A_U + 1.43 A_{Th} + 0.077 A_K \quad (3)$$

where  $A_U$ ,  $A_{Th}$  and  $A_K$  are the activity concentrations of  $^{226}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in  $\text{Bq kg}^{-1}$ , respectively. Table 3 shows that the mean value of radium equivalent ranged from  $21.58 \pm 5.08$  to  $78.39 \pm 17.90 \text{Bq kg}^{-1}$ , with the average value of  $49.08 \pm 12.02 \text{Bq kg}^{-1}$  for soil,  $49.32 \pm 12.50$  to  $54.19 \pm 13.70 \text{Bq kg}^{-1}$ , with the average value of  $51.76 \pm 13.70 \text{Bq kg}^{-1}$  for sediment and  $28.20 \pm 9.98$  to  $19.34 \pm 7.22 \text{Bq L}^{-1}$ , with an average value of  $23.77 \pm 8.60 \text{Bq L}^{-1}$  for water. The respective values for soil and sediment samples were each lower than the dose limit of  $370 \text{Bq kg}^{-1}$ . This implies that the radionuclides in the samples do not pose immediate radiological hazard to the mine workers and the public, but the accumulation of radiation dose over a long term may cause stochastic effects.

### 3.5 Hazard indices ( $H_{ex}$ and $H_{in}$ )

To limit the external radiation exposure to natural radionuclides present in the samples to the allowed dose equivalent limit of  $1 \text{mSv y}^{-1}$ , the external hazard index,  $H_{ex}$ , was introduced using [15]:

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (4)$$

As shown in Table 3, the external hazard mean value calculated for soil samples in this study was found to be

$0.13 \pm 0.03$  which is below the dose limit of unity. The quantity of internal exposure to radon and its short-lived decay products is given by internal hazard index ( $H_{in}$ ). The value of  $H_{in}$  must be less than unity to have insignificant hazardous effect of radon and its short-lived decay products

to the respiratory organs [16]. The estimated internal hazard index,  $H_{in}$ , is given by [15]:

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (5)$$

From Table 3, the internal hazard mean value calculated for soil samples was found to be  $0.16 \pm 0.04$  and is below the dose limit of unity.

### 3.6 Representative level index (RLI)

Representative level index implies the gamma activity level that is associated with various concentrations of some radionuclides and can be determined using [17]:

$$RLI = \frac{1}{150} C_u + \frac{1}{100} C_{Th} + \frac{1}{1500} C_K \quad (6)$$

where  $C_u$ ,  $C_{Th}$  and  $C_K$  are the average concentrations of  $^{238}\text{U}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  in  $\text{Bq Kg}^{-1}$ , respectively. The value of RLI as provided in Table 3 varies from 0.18 to 0.63 with average value of 0.38. The average value is less than the maximum limit of 1 [17]  $1.76 \pm 13.70 \text{Bq kg}^{-1}$  for sediment and  $28.20 \pm 9.98$  to  $19.34 \pm 7.22 \text{Bq L}^{-1}$ , with an average value of  $23.77 \pm 8.60$

### 3.7 Annual gonadal equivalent dose $AG_d$ ( $\mu\text{Svy}^{-1}$ )

The gonads, the active bone marrow and the bone surface cells are chosen as the interested organs [18]. The annual gonadal equivalent dose ( $AG_d$  in  $\mu\text{Svy}^{-1}$ ) for the miners and the general public in the study area as a result of the specific activities of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  was calculated as [19]:

$$AG_d (\mu\text{Svy}^{-1}) = 3.09A_{Ra} + 4.18A_{Th} + 0.314A_K \quad (7)$$

where  $A_{Ra}$ ,  $A_{Th}$  and  $A_K$  are the activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$ , respectively. The average value of annual gonadal effective dose ( $AG_d$ ) estimated was  $174.83 \mu\text{Sv y}^{-1}$  in soil and  $180.86 \mu\text{Sv y}^{-1}$  in sediment (Table 3). Each of these values is lower than the dose limit of  $300 \mu\text{Sv y}^{-1}$  [18]. This shows that the dose received yearly by the gonads, the active bone marrow and the bone surface cells of the mine workers and the public poses insignificant radiological hazard to them.

### 3.8 Comparison of activity concentrations of the present study with other environments in the world

Table 4 shows the comparative data found in the literature. Lower activity concentration was determined by [20] and [21], in Nigeria for  $^{238}\text{U}$  compared to this study but lower

**Table 3-**Radium equivalent, External hazard index, Internal hazard index, Representative level index, Annual gonadal equivalent.

Sample code	Radium equivalent Ra <sub>eq</sub> (Bq kg <sup>-1</sup> )	External hazard index H <sub>ex</sub>	Internal hazard index H <sub>in</sub>	Representative level index RLI	Annual gonadal equivalent AG <sub>d</sub> (μSvy <sup>-1</sup> )
SL 1	57.84±15.50	0.16±0.04	0.18±0.05	0.45	206.71
SL 2	58.78±13.20	0.16±0.04	0.19±0.04	0.47	217.82
SL 3	35.99±9.64	0.10±0.03	0.12±0.04	0.27	124.95
SL 4	32.29±8.17	0.09±0.02	0.09±0.02	0.26	119.19
SL 5	52.53±13.60	0.14±0.04	0.17±0.04	0.41	187.83
SL 6	60.34±12.10	0.16±0.03	0.19±0.04	0.46	209.44
SL 7	21.58±5.08	0.06±0.02	0.07±0.02	0.18	85.36
SL 8	49.19±11.80	0.13±0.03	0.15±0.04	0.38	172.62
SL 9	49.12±14.10	0.13±0.04	0.16±0.05	0.37	167.97
SL 10	44.62±9.77	0.12±0.03	0.14±0.03	0.34	152.37
SL 11	41.09±11.10	0.11±0.03	0.14±0.04	0.31	142.43
SL 12	56.80±13.60	0.15±0.04	0.19±0.05	0.44	197.8
SL 13	78.39±17.90	0.21±0.05	0.23±0.05	0.63	288.98
SL 14	47.91±12.20	0.13±0.03	0.15±0.04	0.37	169.68
SL 15	49.67±12.60	0.13±0.03	0.16±0.04	0.39	179.28
Average	49.08±12.02	0.13±0.03	0.16±0.04	0.38	174.83

**Table 4-**Comparison of activity concentrations of soils in Bq Kg-1 of the present study with other environments in the world.

References	Country	<sup>238</sup> U	<sup>232</sup> Th	<sup>40</sup> K
Present study	Nigeria	8.42	10.28	337.08
[25]	Egypt	16.3	12.94	200.21
[20]	Nigeria	7	10	153
[24]	Egypt	17.26	92.85	
[15]	Nigeria	51.5	48.1	114.7
[5]	Nigeria	12.1	60.1	426.5
[26]	Egypt	117.6	65	126
[27]	Iraq	83.34	19.15	284.86
[24]	Pakistan	26-31	50-55	500-610
[25]	India	22.8	39.9	253.16
[28]	Spain	46	49	650
[30]	Tripoli	10.5	9.5	270
[29]	USA	40	35	370
[12]	World Average	32	45	420

than the activity concentration obtained by [22], [8] and [23], in Nigeria; [24], [25] and [26] in Egypt; [27] in Pakistan; and [28] in India; The activity of <sup>238</sup>U measured in this study is lower than the world average [12]. The average activity concentration of <sup>232</sup>Th obtained in this study is lower than that obtained by [15], [5] and [21] in Nigeria, [29] in Ghana; [24] in Egypt; [30] in Iraq; [31] in Spain; and [32] in USA. The mean activity concentration <sup>232</sup>Th in this study is higher than the activity determined by [30], however, the average activity of <sup>232</sup>Th in this study is lower than the world average [12]. The activity of <sup>40</sup>K in this study is higher than the activity obtained by [20] in Nigeria; [25] and [26] in Egypt; [28] in India; and [33] in Tripoli. The activity of <sup>40</sup>K in this study is lower than the world average [12].

## 4 Statistical analysis

### 4.1 Principal component analysis

Principal components analysis is a process of identifying a smaller number of uncorrelated variables, called “principal

components”, from a large set of data. The principal components analysis is aimed at explaining the maximum amount of variance with the fewest number of components. It is useful in reducing the number of variables and avoids multicollinearity.

**Table 5-**Principal components of the variables

Variables	Principal Component 1	Principal Component 2
<sup>238</sup> U	0.16178	0.75958
<sup>232</sup> Th	0.25614	0.3676
<sup>40</sup> K	0.28784	-0.49482
D <sub>R</sub>	0.34392	-0.07029
A <sub>d</sub>	0.34392	-0.07036
Ra <sub>eq</sub>	0.34478	-0.0014
H <sub>ex</sub>	0.34422	-0.02174
H <sub>in</sub>	0.34022	0.1277
RLI	0.3437	-0.07927
AG <sub>d</sub>	0.34289	-0.10056

The principal component analysis produces two component representation of the acquired data in which 95.14 % of the total variance was explained. The principal components of the variables are shown in Table 5. The principal component one is strongly correlated with seven of the original variables namely, radium equivalent, external hazard index, absorbed dose, annual effective dose, internal hazard index, representative level index, annual gonadal equivalent dose and potassium. This component can be defined as a measure of the qualities of the above radiological variables and the lack of quality of potassium. It can be seen that absorbed dose and annual effective dose have the same correlation. It also suggests that the principal component one correlates most strongly with radium equivalent, showing that the component is primarily a measure of radium equivalent. The second principal component increases with increasing uranium and thorium and decreases with potassium.

## 4.2 Pearson correlation analysis

**Table 6**-Pearson correlation matrix

Variables	$^{238}\text{U}$	$^{232}\text{Th}$	$^{40}\text{K}$	$D_R$	$A_d$	$R_{a_{eq}}$	$H_{ex}$	$H_{in}$	RLI	$AG_d$
$^{238}\text{U}$	1									
$^{232}\text{Th}$	0.45261	1								
$^{40}\text{K}$	0.04255	0.32003	1							
$D_R$	0.41405	0.70403	0.87328	1						
$A_d$	0.41396	0.70403	0.87331	1	1					
$R_{a_{eq}}$	0.45769	0.75649	0.83018	0.99652	0.99651	1				
$H_{ex}$	0.44683	0.73726	0.8429	0.99662	0.9966	0.99783	1			
$H_{in}$	0.59505	0.74698	0.76556	0.97443	0.97444	0.98379	0.97772	1		
RLI	0.40016	0.70867	0.87479	0.99983	0.99983	0.99632	0.99632	0.97147	1	
$AG_d$	0.38977	0.68566	0.88863	0.99946	0.99947	0.99337	0.99426	0.96771	0.99948	1

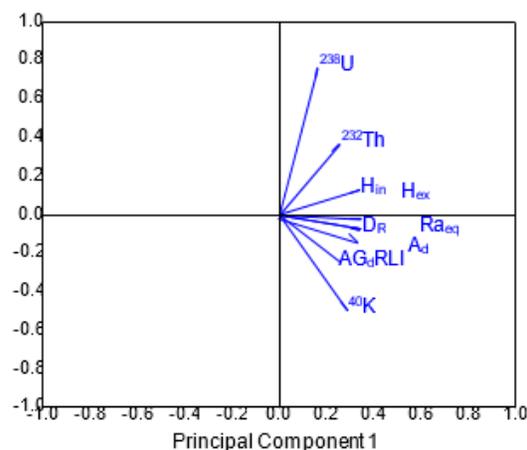
Correlation is an approach for studying by close examination the relationship between two quantitative, continuous variables. Pearson correlation coefficient is a measure of degree of association between two variables. The radiological variables were subjected to Pearson correlation analysis to quantify the relations between pair of variables. Table 5 shows the Correlation Coefficient Matrix of all the radiological variables for the samples from the mining site. Correlation strength is defined for the absolute value of Pearson correlation coefficient as 0.00-0.19 (very weak), 0.20-0.39 (weak), 0.40-0.50 (moderate), 0.60-0.79 (strong), and 0.80-1.0 (very strong) [34]. It is observed from the Table 6 that a high positive correlation exists among all the radiological variables as all values are higher than 0.3, except the one that exists between  $^{238}\text{U}$  and  $^{40}\text{K}$ , where it shows a very weak correlation, indicating that  $^{238}\text{U}$  and  $^{40}\text{K}$  decay series do not occur together. This suggests that as one variable increases the other variable has a tendency to also increase. The Pearson correlation coefficient value of 1 implies that changes in one variable are strongly correlated with changes in the second variables.  $^{232}\text{Th}$  and  $^{40}\text{K}$  have very strong correlations with all other radiological variables. The strong positive correlation coefficient is found between  $^{232}\text{Th}$  and other radiological variables and a strong positive correlation exists between  $^{238}\text{U}$  and other radiological variables. Hence, these relationships indicate that  $^{232}\text{Th}$  and  $^{238}\text{U}$  radionuclides contribute to the emission of gamma radiation in all the locations Senthilkumar and Narayanaswamy, [28].

## 4.3 Loading plot

The Loading plot describes the relationships that exist between the original variables and subspace dimension. From Fig. 1, we can see that the loading of  $^{238}\text{U}$  is close to principal component 2. Variables such as Internal hazard index ( $H_{in}$ ), radium equivalent activity

( $R_{a_{eq}}$ ), external hazard index ( $H_{ex}$ ), annual equivalent dose ( $A_d$ ), representative level index, annual gonadal equivalent dose ( $AG_d$ ) and absorbed gamma dose rate ( $D_R$ ) formed a large cluster to the right side of the plot indicating they are highly correlated.

Principal Component 2



**Figure 1**

## 5 Conclusions

The activity concentrations of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in the soil samples hosting mineral ores in Ofiki tantalite mine have been obtained using gamma-ray spectrometry. The activity concentrations of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  are lower than the worldwide averages. It was noted that the geological structure and the minerals such as Monazite, Zircon, ilmenite and Xenotime, which contained small amount of uranium and potassium radionuclides, are the factors responsible for the low activity concentrations in the mine. Though the levels of activities of the radionuclides in the soil, sediment and water samples and the associated doses

are lower than the worldwide averages, there is still a possibility of accumulation of radiation dose over a long term which might result into severe radiological hazard.

It is recommended that the site should be under regular monitoring and control in case of any increased level of radionuclides.

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