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Article

Gamma radioactivity levels and their corresponding external exposure of soil samples from tantalite mining areas in Oke-Ogun, South-Western Nigeria

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Abstract **The radioactivity concentrations of ^{226}Ra, ^{232}Th and ^{40}K were measured using gamma-ray spectroscopy with NaI (Tl) detectors in four tantalite mining sites in the Oke-Ogun area, South-Western Nigeria. The measured values of the activities of ^{40}K, ^{226}Ra and ^{232}Th in the soil samples were found to lie in the ranges 123.7 ± 3.8 – 1372.3 ± 8.6, 16.8 ± 1.6 – 71.1 ± 2.53 and 3.0 ± 0.7 – 31.9 ± 1.0 Bq kg^{-1}, respectively. These samples were also found to have radium equivalent activity in the range 74.2–121.0 Bq kg^{-1}. Values of 0.2–0.3 for the external and 0.3–0.4 for the internal hazard indices were estimated for the samples and the annual effective dose varied from 70.3 ± 13.5 to 100.8 ± 42.8 μSv with a mean of 87.5 ± 18.6 μSv y^{-1}. The annual effective dose is higher than the world average.**

Keywords: Oke-Ogun / radionuclide / tantalite / natural radioactivity / effective dose

1. Introduction

Human beings are exposed to background radiation that stems from both natural and man-made sources. Natural background radiation, which is equivalent to 2.4 mSv per person, makes up approximately 80% of the total radiation to which a person is exposed during one year (IAEA, 1996). Most of the radioactivity in the terrestrial environment, whether it is natural or man-made, is bound to the components of the soil. Transportation of this radioactivity from soil to vegetation is possible via dust deposition or root uptake and then to humans through inhalation, breathing and soil ingestion. Therefore, all pathways of exposure that originate from soil are potentially important for the purpose of radiation risk assessment. The two prominent sources of external radiation are cosmic rays and terrestrial gamma rays. Terrestrial gamma rays are essentially due to radionuclides belonging to the ^{238}U and ^{232}Th series and singly occurring ^{40}K that are present in the earth's crust.

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In the last two decades, many studies have been carried out on the natural background and artificial radiation levels in Nigeria (Oresegun *et al*., 1990; Obed *et al*., 2005; Jibiri *et al*., 1998; Jibiri *et al*., 2011; Farai *et al*., 2006; Ademola *et al*., 2006).

The Oke-Ogun area is naturally endowed with a variety of minerals, especially tantalite, iron ore, marble and aquamarine, which contribute enormously to the economic progress of the region. These stones are of granite type and may contain a relatively high concentration of natural radioactivity. So, with a lot of mining activities taking place in this area, there is the possibility of high background radiation in this area. Also, the representative gamma irradiation may be indirectly transferred to dwellings, thereby creating a radioactive environment due to indoor radon. Therefore, there is a need to carry out a survey of background radiation in these mining sites to yield data that may be used to assess the health effects on the population.

The present study was designed to enhance the existing information on radioactivity in mining sites. The objectives of the present study are therefore: (i) determination of activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the representative soil samples; (ii) determination of the internal (H_{in}) and external (H_{ex}) radiological hazard indices, radium equivalent (Ra_{eq}), absorbed dose rate (D), and the representative gamma index ($I_{\gamma r}$) of the representative soil samples, and (iii) estimation of the effective dose.

2. Geology of the study area

The study area, Oke-Ogun, is located in the North-Western part of Oyo State, Nigeria, and is bounded by River Ogun (Fig. 1). The geographical location of this area is (38° 35'- 48° 13' E, 88° 05'- 98° 08' N). The area is between 277 and 456 m above sea level within the Pre-Cambrian Basement Complex of South-Western Nigeria and is at the fringes of the border between Nigeria and the Republic of Benin. Geologically, the study area is underlain by the Pre-Cambrian Basement Complex, composed of magnatite, gneiss and schist, which extends from Iseyin to Kisi in the far north. The major rock unit in the study area is the undifferentiated meta-sediments in addition to granite, granitegneiss and porphyritic granite. However, syenites are common and well exposed within and around Saki and Okeho in the study area, while there is occurrence of several pegmatite veins, as intrusion into the undifferentiated crystalline basement rocks, most of which are in gemstones, especially tourmaline (Tijani *et al*., 2003). Apart from the small hills and inselbergs, the bedrocks are generally covered by the weathered regolith usually composed of clay and sandy soils, which are lateritized in places, depending on the underlying bedrock types. The mineral deposits in the region include tantalite in Atisbo, Iwajowa, Iseyin and Itesiwaju districts, amphibolites in Atisbo, Kajola, Saki East and Saki West, Kaolin in Iseyin, Saki East and Saki West, gemstones in Saki

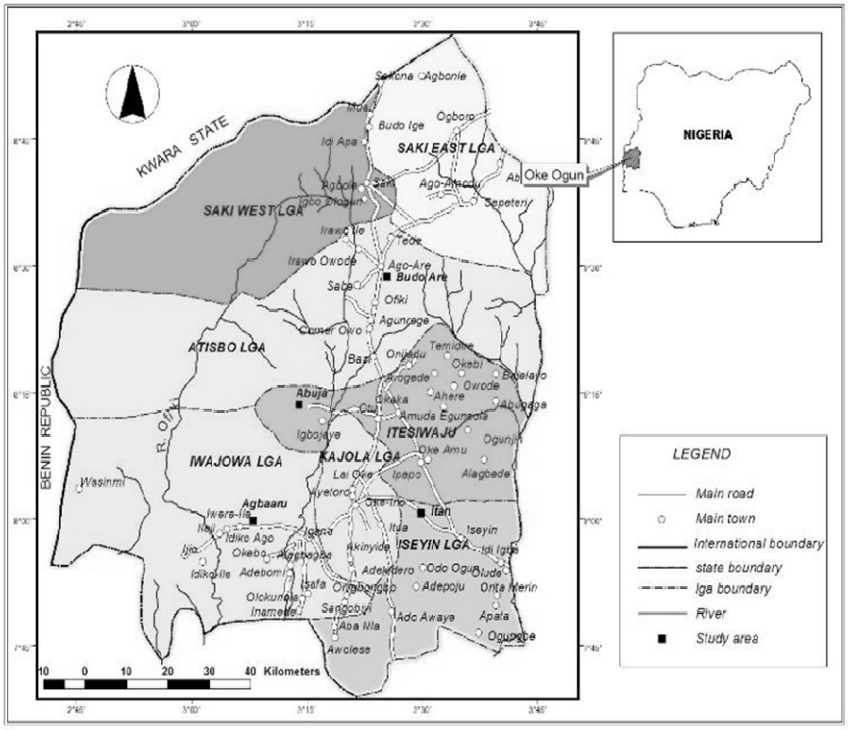

Figure 1 – Map of the Oke-Ogun area in Nigeria and localization of investigation sampling points.

West, Saki East, Atisbo, Itesiwaju and Kajola districts, and granites, sand and gravel in all the districts. These stones are of granite type and may contain a relatively high concentration of natural radioactivity.

3. Materials and methods

3.1. Sample collection and processing

Soil samples were collected from four tantalite mining sites in the Oke-Ogun area (Fig. 1). Ten samples were collected from each mine about 500 cm apart to allow an even collection of the soil samples. The samples were collected at a depth of about 20-30 cm at the various locations. The samples were processed following the standard procedures (EML Procedure Manual, 1983). Soil samples were well mixed after removing extraneous materials such as roots, pieces of stones and

gravel. Samples were weighed and then dried in an oven at 105 °C overnight and re-weighed to find the water content. The samples were crunched and were made to pass through a 0.2-mm sieve. Sieved samples were weighed and a mass of 200 g of each sample was placed in a plastic container (6.5 cm in diameter and 7.0 cm in height). The plastic containers were hermetically sealed with adhesive tape (AERB, 2003) for 30 d, which is enough for secular equilibrium to take place (Olomo *et al*., 1994).

3.2. Activity determination

Analysis of radionuclide concentrations was performed by gamma-ray spectrometry with sodium iodide detectors. The counting assembly was a scintillation detector and a Canberra multi-channel analyzer. The detector was a 7.6-7.6 cm^2 NaI (Tl) manufactured by Bicron. A cylindrical lead shield of approximately 5 cm thickness with a fixed bottom and a movable cover shielded the detector from background radiation. The spectrometer was tested for its linearity and then calibrated for energy using gamma sources supplied by the International Atomic Energy Agency, Vienna. The detection efficiency calibration of the system was carried out using a reference standard gamma source prepared by Rocketdyne Laboratories, Canoga Park, CA, USA, which is traceable to a mixed standard gamma source (No. 48722-356) by Atlantic Inc., Atlanta, GA, USA. The detector assembly has a resolution of ~8% at 0.662 MeV of ^{137}Cs. The reference sources have activity concentration of 479.15, 566.47 and 11.60 Bq for ^{40}K, ^{226}Ra and ^{232}Th, respectively.

The background count was determined by counting an empty container of the same dimensions as the one containing the samples and subtracting from the gross count. The counting time was set at 36 000 s (10 h) to obtain the gamma spectrum with good statistics.

From the net area, the activity concentrations in the samples were obtained using equation (1):

$$C\,(\text{Bq kg}^{-1}) = kC_n \tag{1}$$

where $$k = 1/\varepsilon P_\gamma M_s,$$

C is the activity concentration of the radionuclide in the sample given in Bq kg^{-1}, C_n is the count rate under the corresponding peak, ε is the detector efficiency at the specific γ-ray energy, P_γ is the absolute transition probability of the specific γ-ray, and M_s is the mass of the sample (kg).

TABLE I
Activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K (Bqkg^{-1}) in mining sites of the Oke-Ogun area.

Sites	^{226}Ra	^{232}Th	^{40}K
Itan, Iseyin	41.3 ± 12.3	19.1 ± 2.7	153.0 ± 16.5
Budo-Are Ofiki	42.1 ± 12.7	16.4 ± 4.6	541.7 ± 372.5
Agbaaru, Iwere-Ile	40.3 ± 28.6	15.7 ± 8.9	506.6 ± 361.1
Abuja, Komu	35.4 ± 7.5	19.1 ± 3.0	335.6 ± 121
Mean	39.8 ± 3.0	17.7 ± 1.8	384.2 ± 178.5

The detection limit of a measuring system describes its operating capability without the influence of the sample. The detection limit (DL) given in Bq kg^{-1}, which is required to estimate the minimum detectable activity in a sample, was obtained using equation (2):

$$\mathrm{LLD} = 4.65 \sqrt{C_b / t_b} \times f \qquad (2)$$

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}).

The detection limits obtained were 17.3 Bq kg^{-1}, 4.2 Bq kg^{-1} and 5.1 Bq kg^{-1} for ^{40}K, ^{226}Ra and ^{232}Th, respectively. Values below these numbers were taken as being below the detection limit.

The concentration of ^{226}Ra was determined by a 1.764 MeV gamma ray from ^{214}Bi. The gamma-ray energy of 2.614 MeV from ^{208}Tl was used to determine the activity concentration of ^{232}Th and a gamma ray of 1.460 MeV from ^{40}K was used to determine the concentration of ^{40}K in the samples.

4. Results and discussions

The results of activity measurements are presented in Table I. ^{226}Ra concentration varied in the range 16.8 ± 1.6 – 71.1 ± 2.5 Bq kg^{-1} with arithmetic mean ± standard deviation (AM ± SD) of 39.8 ± 3.0 Bq kg^{-1}, ^{232}Th in the range of below the detection limit 3.0 ± 0.7 – 31.9 ± 1.0 Bq kg^{-1} with a mean of 17.7 ± 2.5 Bq kg^{-1} and ^{40}K in the range of 123.7 ± 3.8 – 1372.3 ± 81.6 Bq kg^{-1} with a mean of 384.2 ± 178.5 Bq kg^{-1}. The activity concentration of ^{40}K was lowest in Itan, Iseyin mining site (153 ± 16.5 Bq kg^{-1}) and highest in Budo-Are, Ofiki (541.7 ± 372.5 Bq kg^{-1}). The high concentration of ^{40}K in Ofiki may be due to extensive farming activities involving the use of fertilizers. The mean activity concentration of ^{226}Ra

TABLE II

The absorbed dose rate of soil samples in mining sites of the Oke-Ogun area (nGy h^{-1}).

Sites	Absorbed Dose Rate in Air (nGy h^{-1})
Itan, Iseyin	38.2 ± 5.5 (34.0 ± 0.9 – 51.8 ± 0.9)
Budo-Are Ofiki	54.8 ± 17.4 (31.0 ± 0.8 – 87.7 ± 1.0)
Agbaaru, Iwere-Ile	52.7 ± 19.8 (23.5 ± 0.8 – 77.0 ±1.0)
Abuja, Komu	44.5 ± 11.1 (29.8 ± 0.8 – 74.5 ± 1.0)
Mean	47.6 ± 7.7

was highest in Budo-Are, Ofiki (42.1 ± 12.7 Bq kg^{-1}) and lowest in Abuja, Komu (35.4 ± 7.5 Bq kg^{-1}). The mean activity concentration of ^{232}Th was highest in Abuja, Komu (19.1 ± 3.0 Bq kg^{-1}) and lowest in Agbaaru, Iwere-Ile (15.7 ± 8.9 Bq kg^{-1}).

4.1. Absorbed dose rates

From the results of ^{226} Ra, ^{232}Th and ^{40}K activities in Table I, the gamma dose rates in air were calculated using the dose coefficients (nGy h^{-1} per Bq kg^{-1}) 0.462, 0.604 and 0.0417 given in UNSCEAR (2000) for ^{226}Ra sub-series, ^{232}Th series and ^{40}K, respectively. The absorbed dose is given by equation (3):

$$D = 0.446A_{\mathrm{Ra}} + 0.662A_{\mathrm{Th}} + 0.048A_{\mathrm{K}} \tag{3}$$

where D is the dose rate (nGy h^{-1}) at 1 m above the ground due to ^{226}Ra, ^{232}Th and ^{40}K in the soil sample. A_{Ra}, A_{Th} and A_{K} are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq kg^{-1}, respectively.

The absorbed dose values thus calculated are presented in Table II. The total absorbed dose delivered by these radionuclides ranged from 23.5 ± 0.8 – 87.7 ± 1.0 nGy h^{-1} with a mean of 47.6 ± 7.7 nGy h^{-1}, which is lower than the world average value of 51 nGy h^{-1} (UNSCEAR, 2000). The annual effective dose to the population due to the soil activity was estimated using the dose coefficient (0.7 Sv Gy^{-1}) and occupancy factor (0.2) for outdoors given in UNSCEAR (2000). The average effective dose (Tab. III) varies in the range 70.3 ± 13.5 in Itan, Iseyin to 100.8 ± 42.8 μSv in Budo-Are, Ofiki, with a mean annual effective dose of 87.5 ± 18.6 μSv y^{-1}, which is about 25% higher than the world average of 70 μSv y^{-1} (UNSCEAR, 2000).

TABLE III

Effective dose estimated in mining sites of the Oke-Ogun area.

Sites	Mean Effective dose (μSv y^{-1})
Itan, Iseyin	70.3 ± 13.5
Budo-Are Ofiki	100.8 ± 42.8
Agbaaru, Iwere-Ile	96.7 ± 48.1
Abuja, Komu	81.9 ± 37.1
Mean	87.5 ± 18.6

TABLE IV

Radium Equivalent (Ra_{eq}), the external hazard index, (H_{ex}), internal hazard index (H_{in}) and gamma index ($I_{\gamma r}$) in the tantalite mining sites.

Sites	Radium Equivalent (Bq kg^{-1})	External Hazard Index (H_{ex})	Internal Hazard Index (H_{in})	Gamma Index ($I_{\gamma r}$)
Itan, Iseyin	79.9 ± 12.3	0.2	0.4	0.6
Budo-Are Ofiki	113.2 ± 29.1	0.3	0.4	0.8
Agbaaru, Iwere-Ile	102.8 ± 36.7	0.3	0.4	0.8
Abuja, Komu	88.5 ± 23.3	0.2	0.3	0.7
Mean	96.1 ± 14.8	0.3	0.4	0.7

4.2. Radium equivalent, internal hazard, external hazard and gamma index

The radium equivalent activity, internal hazard index and external hazard index are presented in Table IV. The radium equivalent activity is calculated from equation (4) (UNSCEAR, 1982) and it ranged from 79.9 ± 12.3 to 113.2 ± 29.1 Bq kg^{-1} (mean = 96.1 ± 14.8 Bq kg^{-1}):

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_{K} \quad (4)$$

where A_{Ra}, A_{Th} and A_{K} are the activity concentrations in Bq kg^{-1} of ^{226}Ra, ^{232}Th and ^{40}K, respectively.

The current regulation is that Ra_{eq} should be inferior to 370 Bq kg^{-1} for any material that will be used in building of dwellings (UNSCEAR, 1982). It is assumed that 370 Bq kg^{-1} of ^{226}Ra, 259 Bq kg^{-1} of ^{232}Th and 4810 Bq kg^{-1} of ^{40}K produce the same gamma-ray dose rate (Straden, 1979). The mean radium equivalents obtained are presented in Table IV.

To limit the external gamma-radiation dose from building materials, the external hazard index (H_{ex}) was calculated from equation (5) (Beretka *et al*., 1985):

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1. \quad (5)$$

The calculated mean external hazard index in the present study is 0.3, which is much lower than unity, as desirable.

To assess the radiation hazard due to internal exposure from radon and its short-lived decay products to the respiratory organs, the internal exposure to radon and its decay products is quantified by the internal hazard index (H_{in}), which is given in equation (6) (Beretka *et al*., 1985):

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1. \quad (6)$$

The internal hazard index (H_{in}) for soil calculated in this study ranged from 0.3 to 0.4 with a mean of 0.4, which is lower than unity, as desirable.

According to the European Commission, the gamma activity concentration index ($I_{\gamma r}$) is derived to identify whether a dose standard is met (EC, 1999) and is estimated from equation (7):

$$I_{\gamma r} = \frac{A_{Ra}}{150} + \frac{A_{Th}}{100} + \frac{A_K}{1500}. \quad (7)$$

The index $I_{\gamma r}$ is correlated with the annual dose due to the excess external gamma radiation caused by external material. Values of the index $I_{\gamma r} = 1$ correspond to 0.3 mSv y^{-1}. Thus, the activity concentration index $I_{\gamma r}$ should be used only as a screening tool for identifying materials which might be of concern to be used as the covering material (Al-Saleh *et al*., 2007). The index $I_{\gamma r}$ estimated is 0.7, which is less than unity.

Table V compares the results of this study with those obtained in other parts of the country. The mean activity concentrations of ^{40}K, ^{232}Th and ^{226}Ra in this study were 384.6 ± 178.5, 17.7 ± 1.8 and 39.8 ± 3.0 Bq kg^{-1}, respectively. These values were lower than the world average of 400 and 30 Bq kg^{-1} for ^{40}K and ^{232}Th, respectively, but slightly higher for ^{226}Ra (35 Bq kg^{-1}). These values were also higher than those obtained in Ibadan and Lagos in the South-West, South-Eastern cities and Southern cities but lower than the results obtained in Abeokuta and Akure in the South-Western Zone, North-East, North-West and North Central cities (Obed *et al*., 2005; Ademola *et al*., 2010). The calculated mean absorbed dose rate in air was 47.6 ± 7.7 nGy h^{-1}. This is lower than the zonal mean of 51 ± 48 nGy h^{-1} obtained for the South-Western zone (Obed *et al*., 2005). The mean

TABLE V
Comparison of the calculated results with those obtained from other parts of the country.

References	Area	^{226}Ra (Bq kg^{-1})	^{232}Th (Bq kq^{-1})	^{40}K (Bq kg^{-1})	Absorbed dose rate (nGy h^{-1})	Annual Effective dose (μSv)
Present Study	Oke-Ogun	39.8 ± 3.0	17.7 ± 1.8	384.2 ± 178.5	47.6 ± 7.7	87.5 ± 18.6
Obed *et al*., 2005	Onitsha	31 ± 9	13 ± 4	93 ± 36	26 ± 4	31.9
Obed *et al*., 2005	Abeokuta	43 ± 15	84 ± 57	329 ± 175	88 ± 44	107.9
Ademola *et al*., 2010	Ondo	13.3 ± 8.0	40.0 ± 10.2	240.2 ± 133.5	41.5 ± 8,5	50.7 ± 10.9
Obed *et al*., 2005	Warri	33 ± 8	7 ± 3	87 ± 48	20 ± 7	25.8
Obed *et al*., 2005	Bauchi	35 ± 11	24 ± 11	961 ± 287	71 ± 18	87.1
UNSCEAR, 2000	World Average	35	30	400	54	70

annual effective dose in the study area was estimated as 87.5 ± 18.6 μSv y^{-1}. This value for the effective dose is about 25% higher than the world average of 70 μSv (UNSCEAR, 2000) and is also higher than the average of 62.9 μSv obtained for South-Western cities in Nigeria (Farai *et al*., 2006).

5. Conclusions

Although the study did not cover the entire country, this is the first report on radionuclide data in soil of the Oke-Ogun area, Nigeria. The environmental monitoring of natural background radiation in Oke-Ogun using sodium iodide detectors revealed the distribution of the natural radiation level in all the soil samples measured. From the results obtained, the distribution of radionuclide was not uniform. Also, no artificial radionuclide was detected in any of the measured soil samples.

The measured values of the activities of ^{40}K, ^{226}Ra and ^{232}Th in the soil samples were found to lie in the ranges 123.7 ± 3.8 – 1372.3 ± 8.6, 16.8 ± 1.6 – 71.1 ± 2.53 and 3.0 ± 0.7 – 31.9 ± 1.0 Bq kg^{-1}, respectively. These samples were also found to have radium equivalent activity in the range 74.2–121.0 Bq kg^{-1}. Samples were found to have values of 0.2–0.3 for the external and 0.3–0.4 for the internal hazard indices. The annual effective dose rate in air varied from 70.3 ± 13.5 to 100.8 ± 42.8 μSv with a mean of 87.5 ± 18.6 μSv y^{-1}. The estimated representative (I_r) hazard index is 0.7. The annual effective dose is higher than the world average (UNSCEAR, 2000).

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