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Measurement of Natural Radioactivity levels in the Topsoil of Abandoned Tantalite Mine in Southwestern, Nigeria

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Abstract

Activity concentration of naturally occurring radionuclides in the topsoil of an abandoned tantalite mine at Awo in Osun State was measured using gamma ray spectrometry method. The results indicate that activity concentration of ^{238}U, ^{232}Th and ^{40}K vary from 36.91 ± 3.44 to 517.21 ± 30.24 Bq kg^{-1}, 92.12 ± 21.06 to 261.61 ± 52.88 Bq kg^{-1}, and 1001.06 ± 6.88 to 1902.25 ± 11.07 Bq kg^{-1} respectively. The mean activity concentration of radionuclide: ^{238}U, ^{232}Th and ^{40}K are 282.86 ±17.44,164.29 ±34.73 and 1391.99 ±8.78 respectively. The mean radium equivalent activity concentration, Ra_{eq} obtained in this study is 618.33 Bq kg^{-1}. This is higher than the recommended reference value of 370 Bq kg^{-1} for safe use. Moreover, the mean external (H_{ex}) and internal (H_{in}) health hazard indicesare greater than unity, indicating that intervention is necessary in the present study location. The result of the average total external gamma dose rate (288.24 nGy h^{-1}) is higher than Indian average value (90 nGy h^{-1}) and the world average (86 nGy h^{-1}) . The results of this study show that the soil of the study area contains a relatively higher activity concentration above the recommended reference value and could be considered unsafe for building construction.

Keywords: Natural radioactivity, activity concentration, radium equivalent activity, health hazard index, tantalite mine.

1.0 Introduction

Radiation in the environment originates from man-made and natural sources. The man-made sources include conventional X-rays, Computed Tomography (CT), and the use of radionuclide in imaging and treatment. Medical X-rays are by far the largest man-made source of public exposure to ionizing radiation [1]. Radioactive materials (e.g. uranium, thorium and potassium) occur naturally everywhere in the environment that man lives in. By far the largest proportion of human exposure to radiation comes from natural source, from external sources of radiation including cosmic and terrestrial radiation, and from inhalation or ingestion of radioactive materials [2].

Human beings and plants are exposed to natural background radiation daily from the ground [soil, rock], building materials, air, water, food, the universe and even elements in the their own bodies. In certain rare instances, nuclear accidents could be another source of radiation to man [3]. Moreover, natural environmental radioactivity and the associated external exposure due to gamma radiation depends primarily on geological and geographical conditions, and appears at different levels in the soils of each region in the world[4,5,6]. The distribution of natural radionuclides is not uniform, and the external radiation exposure generally varies by a factor of three or more around the world [3].

1.1 Geological Setting

Natural radiation is the largest contributor to the collective dose of the world population. Natural background radiation represents a substantial fraction of the total radiation exposure of most individuals [7,8].

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In Nigeria, illegal mining is a common place. The study location presented in this study is located within the latitudes $7^o45^1\ N\ and\ 7^o48^1\ N$, longitude $4^o20^1\ E\ and 4^o24^1\ E$ of the Southwestern (Osun State), Nigeria. The study location has witnessed illegal mining of tantalite in the past and this could have led to bringing the underlying sub-soils, minerals and radionuclides to the surface, thus exposing the occupant to natural radiation.

The study area (Awo) is a military zone which falls within Osun State, Southwestern Nigeria basement complex. Four main lithological groups are usually distinguished within the Nigerian basement complex, namely: a polycyclic magnetite-gneiss, quartzite complex schists belt (newer sediments) [9], younger metasediment [10], non-magnetized to slightly magnetized paraschists and metaigneous rocks [11]. In specific terms, the area is underlain by basement rock types such as migmatite metasedimentary and metaigneous rocks and granite rocks [12]. The topsoil ranges from sandy clay to dark loamy soil and gravel aggregates. The study in [12] earlier carried out on ground magnetic study and estimation of tantalite deposit of this study location, indicates a low grade deposit of tantalite of density 5.7 kg m^{-3} as against 8.6^+ kg $m^{-3}$for a pure tantalite. The estimated tonnage deposit is between 4.86×10^3 and 1.226×10^4 tonnes. In the study area, the tantalite is associated with other minerals of economic value; such as barite, quartz, mica and gravel.

The aim of this study was to determine the activity concentration of natural radionuclides (^{238}U, ^{232}Th and ^{40}K) in the abandoned tantalite mine at Awo (Southwestern, Nigeria) and its health hazard implications.

2.0 Materials and Methods

2.1 Sampling and Preparation of Samples.

Fifteen samples of topsoil were collected at a depth of 5 cm from an abandoned tantalite mine at Awo, Osun State. Pebbles, roots and vegetation in the samples collected were removed, packed in black polythene and labeled for easy identification. The samples were then moved to the laboratory for analysis. The soil samples were pulverized into a fine powder using the pulverizing machine at the Centre for Energy Research and Development (CERD). The samples were placed inside trays and dried under room temperature. The dried samples were sieved with 2mm mesh. Each dried sample of 250 g was packed in a Marinelli beaker and thus present equal amount of samples on top of and around the detector head. The Marinelli beaker with the sample inside was sealed with a paper tape. The samples were kept for a period of 28 days before gamma spectrometric analysis to allow establishment of secular equilibrium between^{238}U, ^{232}Th and ^{40}K and their decay products, so that the activity of the parent is the same as that of the daughter [13].

2.2 Measurement of Activity Concentration by Gamma Ray Spectrometry Method

The activity concentration of natural radionuclides (^{238}U, ^{232}Th and ^{40}K) in the soil samples were measured using gamma spectrometer at CERD, Obafemi Awolowo University, Ile Ife. Each prepared sample was placed in a shielded gamma ray spectrometer unit during a counting period of 25,200 s (7 hr). The measuring device is a 7.62 x7.62 calibrated NaI (Tl) detector- a 2048 channel analyser and a lead shield with inside diameter of about 20 cm and wall thickness of 5-10 cm. The spectral analysis was performed with the aid of computer software, obtained from Canberra(Canberra S100 gamma ray acquisition). The activity concentration of the soil sample were obtained from the intensity of each line in the spectrum, taking into account the mass, the geometry of the samples, the counting time and the efficiency of the detector using comparative method shown in equation (1)

$$S_x = \frac{A_x\ S_s M_s}{M_x\ A_s} \qquad (1)$$

Where S_x is the concentration of the sample, A_x is the peak area of the sample, $M_x$the mass of the sample, M_s mass of the standard used, A_s is the peak area of the standard and S_s is the concentration of the standard source. When this method is used, the sample container is of the same geometry as the standard and the counting time times are the same.

3.0 Results and Discussion

The specific average activity concentrations of U, Th and K in Bq kg^{-1} in fifteen different sample locations are shown in Table1. The mean activity concentration and the corresponding range of ^{238}U, ^{232}Th and ^{40}K are 282.86 ±17.14 (36.91 - 517.21), 164.29 ± 34.73 (92.12-261.61) and 1391.99 ±8.78 (1001.06-1902.25)Bq kg^{-1}respectively. The variation in the activity concentrations of naturally occurring radionuclides depend on geological conditions such as formation of rocks and transport process [14]. It is evident from Table 1that ^{40}K has relatively higher concentration. This trend is in agreement with the studies in[15,16]. Natural potassium has three isotopes ^{39}K, ^{40}K and ^{41}K, however, ^{40}K is abundance in nature (0.012% of all potassium). During decay, ^{40}K produces two decay isotopes ^{40}Ca and ^{40}Ar, with the emission of of beta and gamma radiation.

The mean values of the specific concentration of^{232}Th is higher than those measured in a worldwide scale- 50 Bq kg^{-1}[17]. This implies that, it is greater than normal value. The result in Table 1 in addition indicates that the mean value of ^{40}K is higher than the normal average value of 500Bq kg^{-1}[18]. The use of potassium containing fertilizer is one of the causes of its high activity in soil [19].

Table 1: The average specific activity of U, Th and K in Bq kg^{-1} in fifteen sample locations

Sample	^{238}U (Bq kg^{-1})	^{232}Th (Bq kg^{-1})	^{40}K(Bq kg^{-1})
S1	324.33±19.43	172.64±36.20	1731.61±10.42
S2	517.21±30.24	261.61±52.88	1178.78±7.65
S3	350.41±20.95	210.11±43.05	1202.38±7.83
S4	36.91±3.44	113.57±25.29	1760.71±10.33
S5	441.77±26.02	208.88±42.95	1339.21±8.45
S6	272.29±16.52	251.70±51.33	1007.35±7.04
S7	212.38±13.19	156.78±33.33	1436.72±9.13
S8	243.89±14.96	147.25±31.55	1725.32±10.30
S9	262.78±16.05	92.12±21.06	1555.46±9.59
S10	211.49±13.15	93.29±21.45	1221.25±8.04
S11	320.30±19.22	144.36±31.10	1293.60±8.42
S12	253.28±15.46	168.67±35.54	1301.46±8.55
S13	323.42±19.38	152.17±32.79	1001.06±6.88
S14	185.61±11.72	164.74±34.86	1902.25±11.07
S15	286.76±17.35	126.49±27.62	1222.82±8.02
Mean (SD)	282.86 ± 17.14	164.29 ± 34.73	1391.99 ± 8.74

The radionuclides (^{238}U, ^{232}Th and ^{40}K) are not uniformly distributed in the soil. This is due to disequilibrium between ^{226}Ra and its decay product. Therefore, for uniformity in exposure estimate, the radionuclide concentrations have been defined in terms of radium equivalent activity (Ra_{eq}) in Bq kg^{-1}. It is a single index or number to describe the gamma output from different mixture of radionuclides in materials[20,21, and 22]. This allows comparison of the specific activity of materials containing different amounts of ^{238}U, ^{232}Th and ^{40}K. The mathematical equation which defines radium equivalent activity is given by equation (2)[23]. This is based on the earlier study in[24] who proposed that activity concentration of 259 Bq kg^{-1} of ^{232}Th, 4810 Bq kg^{-1} of ^{40}K are equivalent to 370 of ^{226}Ra and in this quantities, each of the radionuclides gives an effective dose of 1.5 mSv per year.

$$Ra_{eq} = \left(\frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810}\right) X\ 370 \qquad (2)$$

Where A_{Ra}, A_{Th} and A_K are the activity concentration (Bq kg^{-1}) of ^{226}Ra [which can be that of ^{238}U] [21], ^{232}Th and ^{40}K respectively. The results of Ra_{eq} for each sample are presented in Table 2. The results for the fifteen samples ranged between 334.89 and 897.42 Bq kg^{-1} with sample S4 and S2 having the lowest and the highest values respectively. Aside the impact of the mining activity which took place in the present study location, the relative high activity concentration of ^{40}K in the soil as seen in Table 1 could be attributed to the use of fertilizer for growing crops [25] chiefly grains in the study location. The mean value of the radium equivalent activity for all the samples is618.33Bqkg^{-1}. The mean value calculated in this study is higher than the recommended reference value of 370 Bqkg^{-1} by the Organization of for Economic Cooperation and Development [26] for save use by a factor of 1.67. The results in Table 2 also indicate that radium equivalent activities of all the samples are higher than the reference value except for sample 4 (S4) which is a little less than it.

The soils (laterite and gravel) of this study area are used for construction of building or filling during building construction. In addition, active farming (planting of root crops and grains are carried out). The acres of land within and around the study area are used by herdsmen as grazing field for their cattle. The soils could contribute to the external gamma dose rates in the houses built with soil sample taken from the study area.

Table 2: Radium equivalent activity Bq kg^{-1}

Sample	Ra_{eq} (Bq kg^{-1})
S1	704.54
S2	897.42
S3	743.45
S4	334.89
S5	843.59
S6	709.79
S7	547.20
S8	587.31
S9	514.28
S10	438.93
S11	626.34
S12	594.69
S13	618.11
S14	567.66
S15	561.79
Mean	618.33 ± 145.36

Based on these fact, the external hazard index (H_{ex})due to the emitted gamma rays of the samples are calculated according to the expressions in equation (3)[15, 27]. Similarly, the results of the internal hazard index (H_{in}) - indicator of internal exposure to ^{222}Rn and it radioactive progeny[28,25] are also presented in Table 3.

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (3)$$

Also presented in Table 3 are the results of the absorbed dose rate in air (D), in nGy h^{-1} due to the natural specific activity concentration of ^{238}U, ^{232}Th and ^{40}K at a height of 1 m above the ground was calculated using equation (4)[17]:

$$D = 0.427A_U + 0.662A_{Th} + 0.0432A_K \quad (4)$$

WhereA_U is the activity concentration of uranium in Bqkg^{-1}.

The results of external health hazard index, H_{ex} and internal health hazard index, H_{in} in Table 3 show that they are greater than unity in the entire soil samples except in sample S4.The average value of external health hazard index for all soil sample was found to be above 1 (≈1.7) and the corresponding value of internal health hazard is also greater than 1(≈2.5). According to the recommended value, the calculated meanH_{ex} indicates that intervention is necessary for the location under study.

Table 3: External and internal health hazard and absorbed dose ratein air (nGy h^{-1})

Sample	H_{ex}	H_{in}	Absorbed dose rate (D) nGy h^{-1}
S1	1.90	2.78	326.65
S2	2.65	4.05	446.64
S3	2.01	2.96	339.2
S4	0.90	1.00	159.11
S5	2.28	3.47	386.55
S6	1.92	2.65	320.10
S7	1.48	2.05	252.94
S8	1.59	2.25	273.81
S9	1.39	2.09	242.17
S10	1.19	1.76	205.19
S11	1.69	2.56	289.44
S12	1.61	2.29	273.42
S13	1.67	2.54	283.39
S14	1.53	2.04	264.76
S15	1.52	2.29	260.16
Mean	1.69	2.45	288.24

The results above indicate that the soil sample from the study site could be hazardous to the health of the grazing animals (and consumers of beef), farmers and the unsuspecting occupant of the building or structures constructed with the laterite taken from the abandoned mine.

The results of the total absorbed external gamma dose rate were found to vary (Table 3) from 159.11 to 446.64 nGy h^{-1}, with an average value of 288.24 nGy h^{-1}. Comparison of the mean absorbed dose rate in this study with similar results from India and the world average show that the result of this study (288.24 nGy h^{-1}) is higher than the Indian (90 nGy h^{-1}) result and world average [86 nGy h^{-1}] [3] by at least a factor of 3.This level of dose rate further strengthens the need for intervention in the study location.

To establish a linear relation and the correlation between the uranium concentration in the sample and the radium equivalent activity concentration, the variation of radium equivalent activity concentration in the samples are plotted against uranium activity concentration in Figure 1. Apparently, there is a positive correlation (R^2 =0.8545) between Ra_{eq} and A_U and the linear relation between them for the study location is shownin equation (5):

$$Ra_{eq} = 1.2108A_u + 276.85 \quad (5)$$

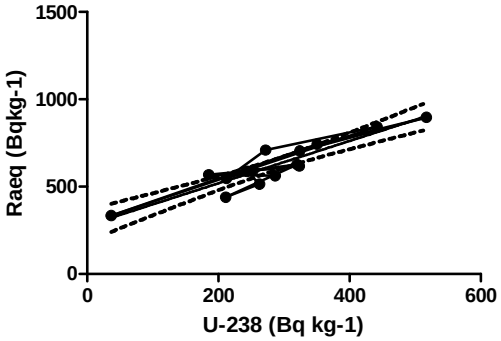

Figure 1: The relation between the uranium(^{238}U)concentration and radium equivalent activity concentration (Ra_{eq}) the line showing 95% confidence band of the best fit.

Figure 2 shows the plotof radium equivalent activity concentration against activity concentration of thorium-232. There seems to be a positive correlation between them (R^2 = 0.7382), and the linear relation between the two is given by equation (6)

$$Ra_{eq} = 2.4483A_{Th} + 217.1 \quad (6)$$

Using multiple regression analysis, a relation connecting radium equivalent activity concentration (Ra_{eq}) and activity concentration of uranium-238, thorium-232 and potassium-40 was established as shown in equation (7):

$$Ra_{eq} = 53.212 + 0.9068A_U + 1.315A_{Th} + 0.06727A_k \quad (7)$$

The correlation coefficient (R^2=0.9863) is also positive.

The indoor and outdoor annual effective dose (mSv) likely to be delivered to the occupant of the building constructed using soil sample obtained from the site or the general public farming on the study area were calculated from absorbed dose rate. Occupancy factors of 0.8 and 0.2 were used for indoor and outdoor assessments respectively. The results show that the annual outdoor effective dose range from 0.19 to 0.55 mSv with an average of 0.35, while the annual indoor effective dose range between 0.78 and 2.19 mSv, with an average of 1.41.

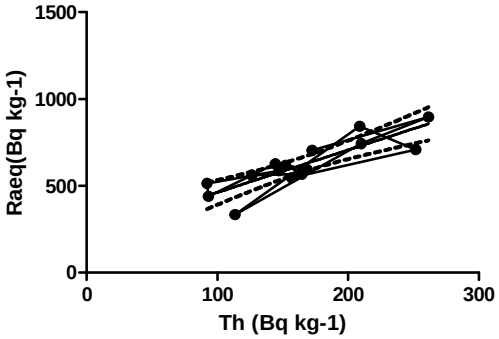

Figure 2: The relation between the thorium (232) concentrations and radium equivalent activity concentration(Ra_{eq}) the line showing 95% confidence band of the best fit.

The results of annual effective dose in this study show that the mean value of the outdoor calculated in this study is higher thanthe global average outdoor terrestrial radiation value of 0.07 mSv y^{-1}[3] for individual member of the public.

4.0 Conclusion

The results of this study showed that terrestrial radionuclides are not evenly distributed in the soil of the study location, and by all standard of measurement of health hazards, there are clear indications that the activity concentration of the soil of the study location is relatively higher than the recommended dose level. The internal and external health hazard indices are greater than unity indicating that intervention is required for the location under study. However, a positive correlation of each has been established between radium equivalent activity concentration and uranium-238 and thorium-232 activity concentrations respectively.

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6.0 References

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