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RADIONUCLIDE CONCENTRATIONS AND IMPACT ASSESSMENT OF THE JOS TIN MINING SOIL RESIDUES

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This study was performed to measure the radionuclides in the tin mining soil mounds from the Jos Plateau, Nigeria and to evaluate the impact of radiation on the environment where the soils are used as building materials. Gamma spectrometry was employed via a NaI(Tl) detector to determine the activities of the radionuclides ^{40}K, ^{238}U and ^{232}Th in ten (10) samples from points within a distance of 20 km along the mining trail. The results of measurements of natural radionuclide (^{238}U, ^{232}Th and ^{40}K) in soil samples show that the concentrations of ^{238}U, ^{232}Th and ^{40}K ranged between 1.51 – 4.98, BDL - 8.64 and 10.3 – 35.2 Bq/kg with mean concentrations of 3.20±1.16, 1.31±2.75 and 25.60±8.89 Bq/kg, respectively. The external hazards ranged between 0.008 – 0.044 with mean value of 0.019±0.009 while the internal hazards ranged between 0.014 – 0.048 with mean value of 0.028±0.009. These hazard values are less than 1. The annual gonadal dose equivalent (AGDE) ranged between 10.28 – 55.87 μSvy^{-1} with mean value of 24.22±12.16 μSvy^{-1}. The radium equivalent activities ranged from 3.07 – 16.23 Bq/kg with a mean value of 7.04±3.53 Bq/kg. The external absorbed dose rate ranged from 5.35– 18.76 nGyh^{-1} with a mean value of 12.95±4.26 nGyh^{-1}. The internal absorbed dose rate ranged from 10.34 – 35.53 nGyh^{-1} with an average value of 24.87±8.13 nGyh^{-1}. The external absorbed dose equivalent rate ranged from 0.007- 0.023 μSvy^{-1} with a mean value of 0.016±0.005 μSvy^{-1}. The internal absorbed dose equivalent rate ranged from 0.051 – 0.174 μSvy^{-1} with an average value of 0.122±0.040 μSvy^{-1}. All the calculated radiological indices fall within the recommended safe limits and world averages. The soil mounds, therefore, do not constitute environmental radiation risks and the soils could be used in construction.

Keywords: Radiological risk indices, Soil mounds, Tin, Mining, Wasteland, Environment, Annual Gonadal Dose Equivalent (AGDE), Gamma spectrometry, Overburden, Gangue

1. Introduction

The mining of ores has been carried out from ages past for the production of materials utilized by humans. However, this process affects the environment negatively resulting in disfigurement of landscape with pollutants being released to the atmosphere. This has been the case with the mining of tin on the Jos Plateau, Nigeria. The Jos Plateau lies in the north central part of Nigeria about 1100 m above sea level, and has an area of about 18120 km^2 [1, 2]. Its geology is made up of the basement complex with younger granites and older basalts [3, 4]. The younger granites dominate, forming intrusive hills and the area is mineralized with tin and columbite [5]. The major tin ore in this area is cassiterite which has been mined and smelted following the discovery of rich alluvial deposits [6-10].

The climate is temperate on the plateau, but, hot and humid on the lowlands. It is warm during the rainy season (April- October) and cold during the harmattan period (December-February).

The topography has two main features, viz, the natural young granite intrusions and soil mounds (overburden and gangue) which are residues from tin mining activities. The granite hills are being blasted and crushed as aggregates, chips and dust which are being employed for various construction purposes. However, the soil mounds remain largely un-utilized, constituting environmental eyesores. The dug out pits and their ponds are sources of danger to humans and cattle, while about 320 km^2 of farmland use is hindered by the area taken up by the mounds [11, 12]. The lithology of the Jos plateau is deeply associated with gneiss granite, and is known to contain the radioactive elements – uranium, potassium and thorium [13]. Tin ore and the discarded tailings obtained during smelting have been found to be radioactive [9, 14-17]. Due to the haphazard and indiscriminate manner in which tin mining residues have been handled, it is believed that water bodies and farm soils have been contaminated, and are radioactive [18, 19]. Buildings constructed with mill tailings have also been reported to be radioactive [20].

Land reclamation has been proposed, in which heavy equipment would be used to move the soil into the pits. As the land area involved is extensive, this is an expensive procedure. Moreover, most of the earlier prospectors were foreigners who have left the country and the reclamation is therefore left to third parties to

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carry out. It is no wonder that not much success has been achieved in the reclamation efforts. Land is becoming scarce and has resulted in tenements springing up around the soil mounds. The populace around the soil mounds could be exposed to the vagaries of radiation in the atmosphere and indoors via emissions from building materials.

Radioactivity in the environment and in buildings is of major concern. This is because high levels of radioactivity are detrimental to the well being of animals and humans. The Jos province is known to be associated with higher radioactivity levels than in other parts of Nigeria [21- 23]. The presence of natural radionuclides in building materials results in external and internal radiation hazards. It is, therefore, imperative to monitor materials for environmental safety. The present study aims to determine activities of ^{238}U, ^{232}Th and ^{40}K in soil mounds and evaluate their radiological impacts on the environment and the populace.

2. Materials and Methods

Samples were collected by traversing the mining trail from Jos north around Lamingo Dam, via Du and Bukuru, across the Jos – Barkin Ladi Road, to Vom. Samples were collected at ten (10) locations from the beginning of the trail to the Police Staff College within a distance of about 20 km. The sampling area is shown in Figure 1. The coordinates of the sampling points were logged with Garmin Nüvi and are as listed in Table 1. Concentration of sample points was greatest around Du, because of the number/density and diversity of the soil mounds. About 50 kg of disturbed soil free from organic matter was collected from each point, at a depth of 1m from the top. The samples were bagged, labelled and transported to the laboratory for analyses where they were pulverized and dried. A mass of 200 g of each sample was packed in labelled cylindrical plastic containers. The containers were sealed and left for 28 days to allow for attainment of secular equilibrium between Ra, Th and their short lived progenies.

The gamma radiation spectrometer consisted of a planar NaI(Tl) detector with diameter of 4.0 cm (Detector ID: ANS P-Type 1) coupled to a computer resident Quantum™ multichannel analyzer (MCA) produced by Princeton Gamma Tech., USA. The source to detector distance was 1.0 cm. The photopeak regions of ^{40}K (1460 keV); ^{214}Bi (1764 keV) and ^{208}Tl (2615 keV) respectively were used for the analyses of the activities of ^{40}K, ^{238}U and ^{232}Th in the samples. Gamma spectra were obtained for a time period of 36000 s (10 hours) for each sample.

3. Radiological Indices

3.1. Radionuclide Activities

The corresponding γ-ray peaks in the energy spectrum were used to compute the activity concentrations in the sample according to the equation [24, 25]

$$C = \frac{C_n}{\varepsilon I_\gamma m_s} \tag{1}$$

Where C is the activity of the radionuclide in the sample ($Bq\,kg^{-1}$), C_n is the count rate under the photo peak, ε is the detector efficiency at the specific γ-ray energy, I_γ is the absolute transition probability of specific γ-ray and m_s is the mass of the sample (kg).

3.2. External and Internal Hazard Indices

The following indices were calculated:

1. The external hazard index is a measure of the radiation exposure due to radioactivity in the samples. The value of this index must be less than 1 so as to limit the radiation dose to the admissible dose equivalent of 1.5 mSv/y. The external radiation hazard, H_{ex}, of each material is determined from the equation [26]:

$$H_{ex} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \tag{2}$$

Where C_{Ra}, C_{Th} and C_K are the activities of ^{236}Ra, ^{232}Th and ^{40}K.

2. Radon and its radioactive products which may accumulate in buildings are hazardous to the respiratory system [27]. This risk is measured by the internal hazard index, H_{in}, which is determined according to the following expression [26- 28]:-

$$H_{in} = \frac{C_{Ra}}{185} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \tag{3}$$

3. The radium equivalent activity, Ra_{eq}, is a common index used to compare the radiological effect or activity of materials that contain ^{226}Ra, ^{232}Th and ^{40}K [26, 29-31]. It is calculated by:

$$Ra_{eq} = C_{Ra} + 1.43\,C_{Th} + 0.07\,C_K \tag{4}$$

4. Some organs in the human body are more susceptible to ionizing radiations and are considered to be of interest. Cells in the gonads and bone marrow are prone to damage and modifications by radioactive materials. The annual gonadal equivalent dose (AGED) for a resident in a house is calculated using the equation [32, 33]:

$$AGED\ (\mu Svy^{-1}) = 3.09C_{Ra} + 4.18C_{Th} + 0.314C_K \tag{5}$$

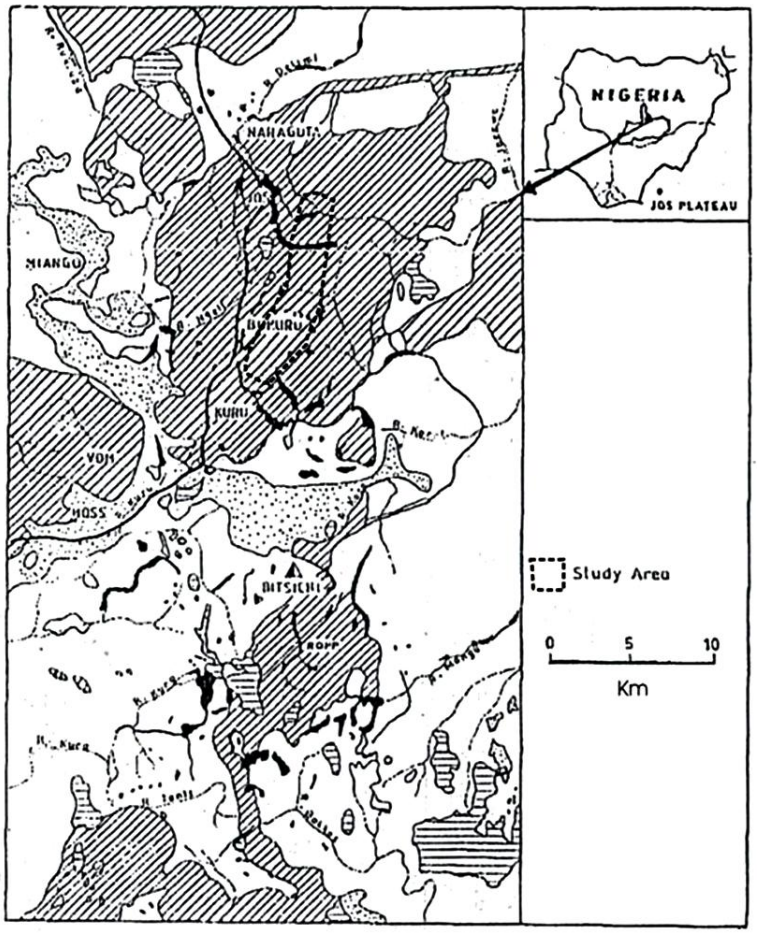

Figure 1. Area of study.

Table 1. Sampling point coordinates

Sample Number	Latitude (North)	Longitude (East)	Description or land use
1	9° 51′ 02.0″	8° 54′ 51.6″	Massive mound with pond. Not farmed.
2	9° 50′ 50.1″	8° 55′ 43.0″	Massive mound with pond. Not farmed.
3	9° 51′ 10.2″	8° 55′ 25.7″	Massive mound with pond. Not farmed.
4	9° 50′ 51.2″	8° 55′ 44.7″	Massive mound with pond. Not farmed.
5	9° 44′ 46.9″	8° 51′ 24.4″	Low mound with pond. Not farmed
6	9° 44′ 50.1″	8° 51′ 24.4″	Low mound with pond. Farmed.
7	9° 44′ 52.0″	8° 51′ 26.1″	Low mound with pond. Farmed.
8	9° 44′ 52.3″	8° 51′ 26.9″	Low mound with pond. Farmed.
9	9° 52′ 31.3″	8° 55′ 28.6″	Massive mound without pond. Not farmed.
10	9° 52′ 28.8″	8° 55′ 30.2″	Low mound without pond. Farmed.

Table 2. Radionuclide activity concentrations

Sample Number	Activity ^{238}U (Bq/kg)	Activity ^{232}Th (Bq/kg)	Activity ^{40}K (Bq/kg)
1	4.78	BDL	32.87
2	2.30	1.57	28.12
3	3.62	BDL	35.21
4	3.17	BDL	20.98
5	2.34	2.88	11.34
6	4.12	BDL	27.50
7	2.88	BDL	24.95
8	4.98	BDL	33.96
9	2.28	BDL	10.30
10	1.51	8.64	30.79
Range	1.51-4.98	1.57-8.64	10.30-35.21

BDL = below detectable limit of the system which was 0.26 Bq/kg for ^{232}Th

The above equation compares the radioactivity in a material with that in the model of a house with infinitely thick walls built with soil having activities of ^{236}Ra, ^{232}Th and ^{40}K equal to those of the world averages of 25, 25 and 370 Bqkg^{-1}, respectively [33].

3.3. Absorbed Dose Rates

1. The risk associated with exposure to gamma radiation in the environment in outdoor air at 1.0 m above ground level for radioactive soil is calculated using the external absorbed gamma dose rate D_{ext} (nGyh^{-1}) via the equation [23, 34]:

$$D_{ext} = \sum_{R} A_R \,..DC_{ext.R} \qquad (6)$$

Where D_{ext} is the external absorbed dose rate, $DC_{ext.R}$ is the coefficient of dose rate per unit activity of radionuclide (nGyh^{-1}/Bqkg^{-1}) and A_R is the activity of the radionuclide R in the sample (Bqkg^{-1}). The United Nations Scientific Committee on Effects of Atomic Radiation (UNSCEAR) [34] has recommended DC_{extR} coefficients of ^{238}U as 0.462 nGyh^{-1}/Bqkg^{-1}, ^{232}Th as 0.604 nGyh^{-1}/kgBq^{-1}, ^{40}K as 0.0413 nGyh^{-1}/Bqkg^{-1} and ^{137}Cs as 0.03 nGyh^{-1}/Bqkg^{-1}. The activity of ^{137}Cs was considered as zero because it was not detected in any of the soil samples.

2. The absorbed dose rate in indoor air, D_{in} (nGyh^{-1}), inside a standard room of dimensions 4 m×5 m×2.8 m, with the assumption that the wall thickness is 20 cm [35] and the density of the aggregates is 2.35 g/cm^3 is given by the expression [36-38]

$$D_{in}\,(nGyh^{-1}) = 0.92C_{Ra} + 1.1C_{Th} + 0.08C_K \qquad (7)$$

3. The absorbed dose rates, D_{ext} and D_{in} would result in the annual effective dose rates, E_{ext} and E_{in}, for outdoor and indoor respectively. These parameters are evaluated by assuming an outdoor occupancy factor of 0.2 and an indoor occupancy factor of 0.8. We have that [39]:

$$E_{ext}(nSvy^{-1}) = D_{ext}(nGyy^{-1}) \times 8760(h) \times 0.2 \times 0.7(Sv/Gy) \qquad (8)$$

$$E_{in}(nSvy^{-1}) = D_{in}(nGyy^{-1}) \times 8760(h) \times 0.8 \times 0.7(Sv/Gy) \qquad (9)$$

Where h is hours.

4. Results and Discussion

The results of measurements of natural radionuclide activities (^{238}U, ^{232}Th and ^{40}K) in the soil samples are shown in Table 2. From this table it can be seen that the activities of ^{238}U, ^{232}Th and ^{40}K ranged between 1.51 – 4.98, BDL - 8.64 and 10.3 – 35.2 Bq/kg with mean concentrations of 3.20 ± 1.16, 1.31 ± 2.75 and 25.60 ± 8.89 Bq/kg, respectively. These values lie within the very low ranges of the world averages as given in UNSCEAR [34]. High activities were observed by Oresegun and Babalola [9] on the Jos Plateau, with 112 kBq/kg for ^{228}Th, 21 kBq/kg for ^{226}Ra and 5.1 kBq/kg for ^{40}K. The disparity with the present result could be attributed to the fact that the tailings used in the former work were obtained mostly from mills, whereas, overburden and gangue from mining fields were used in this study. Mill tailings are derived from concentrated ores and are expected to be more radioactive. However, mean activities of 7.2 kBq/kg for ^{238}U, 16.8 kBq/kg for ^{232}Th and ranges from 8.7 kBq/kg - 51 kBq/kg for ^{238}U, 16.8 kBq/kg - 98 kBq/kg for ^{232}Th were recorded in [17] and [40], respectively, for tests conducted on mining

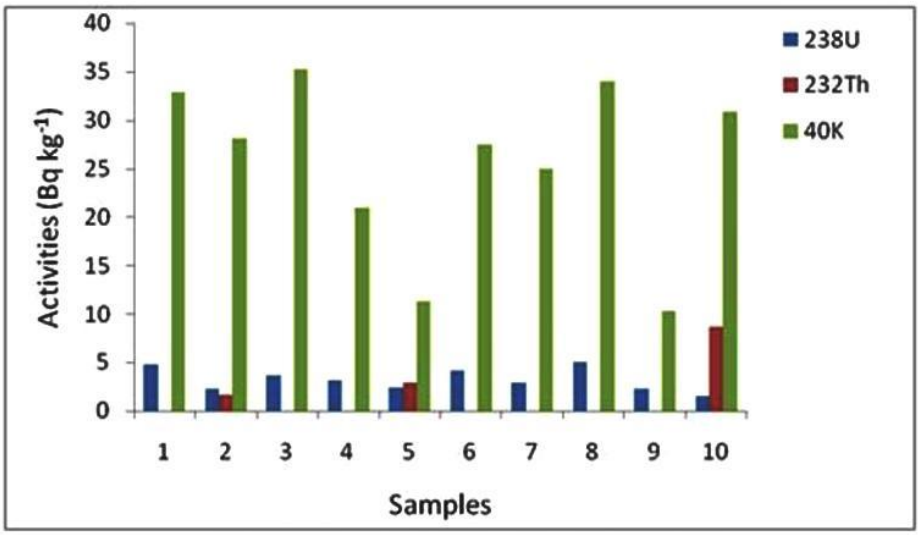

Figure 2. Activities of samples.

Table 3. Calculated radiological indices

€	External Hazard H_{ex}	Internal Hazard H_{in}	Annual Gonadal Equivalent Dose AGED (μSvy^{-1})	Radium equivalent activity Ra_{eq} (Bq/kg)	External absorbed gamma dose rate D_{ext} (nGyh^{-1})	External annual effective dose rate E_{ext}(nSvy^{-1})	Internal absorbed gamma dose rate D_{in} (nGyh^{-1})	Internal annual effective dose rate E_{in}(nSvy^{-1})
1	0.020	0.033	25.15	7.33	15.93	0.020	30.72	0.151
2	0.018	0.024	23.48	6.71	13.74	0.017	26.34	0.129
3	0.017	0.027	22.26	6.34	16.36	0.020	31.50	0.155
4	0.013	0.022	16.39	4.79	10.21	0.013	19.70	0.097
5	0.020	0.026	24.65	7.33	7.55	0.009	14.40	0.071
6	0.017	0.028	21.35	6.23	13.37	0.016	25.79	0.127
7	0.013	0.021	16.73	4.80	11.73	0.014	22.61	0.111
8	0.021	0.034	26.04	7.59	16.46	0.020	31.75	0.156
9	0.008	0.014	10.28	3.07	5.35	0.007	10.34	0.051
10	0.044	0.048	55.87	16.23	18.75	0.023	35.52	0.174

field soils at Bisichi. As shown in Figure 1, Bisichi is far from the present study area and is also on a different geological formation. This may account for the differences in activities. It is to be noted that no activity was detected for ^{40}K [17, 42], whereas, this was the highest contributor to activities in the present study. Therefore the geological origins of the soils may be a contributing factor.

There is a wide variation in ^{40}K activities in the present study samples. As shown in Table 1, low mounds with associated ponds are likely to be used for farming. Since all the samples have the same geological origin, use of potassium based fertilizer could marginally account for the higher activities at farmed sites as depicted in Figure 2. Some of the samples could also be overburden while others could be gangue. Activities from the former are expected to be lower than in the latter case as overburden has not contacted the ore.

The values of the calculated radiological indices are shown in Table 3. From this table it can be seen that the external hazards ranged between 0.008 – 0.044 with mean value of 0.019$\pm$0.009 while the internal hazards ranged between 0.014 – 0.048 with mean value of

0.028±0.009. These hazard values are less than 1. Internal hazard index has higher value than external hazard index because of the higher occupancy factor in the former case.

The AGDE ranged between 10.28 – 55.87 μSvy^{-1} with mean value of 24.22±12.16 μSvy^{-1}. These values are well below the world average for soil of 300 μSvy^{-1}. The radium equivalent activities ranged from 3.07 – 16.23 Bq/kg with a mean value of 7.04±3.53 Bq/kg. These values are considered safe since they are well below the limit of 370 Bq/kg recommended by Beretka and Mathew [26].

The external absorbed dose rate ranged from 5.35 – 18.76 $nGyh^{-1}$ with a mean of 12.95±4.26 $nGyh^{-1}$. This is lower than the world average of 56 $nGyh^{-1}$ [34]. The internal absorbed dose rate ranged from 10.34 – 35.53 $nGyh^{-1}$ with an average of 24.87±8.13 $nGyh^{-1}$. The external effective dose rate ranged from 0.007 – 0.023 $mSvy^{-1}$ with a mean of 0.016± 0.005 $mSvy^{-1}$. The internal effective dose rate ranged from 0.051 – 0.174 $mSvy^{-1}$ with an average of 0.122± 0.040 $mSvy^{-1}$. The world wide average effective dose rate is approximately 0.46 $mSvy^{-1}$ [34]. Therefore, the calculated effective doses are smaller than the world average. The derived indices were low because the activities used to calculate were also low. High dose equivalents were obtained in [16] because mill tailings were used which is not the case here.

5. Conclusion

The activities of ^{238}U, ^{232}Th and ^{40}K in the tin mining soil residues of the Jos Plateau in Nigeria have been measured in this wok. The values obtained were found to be low as compared to ealier studies. Differences could be attributed to samples being collected from regions of different geological origins. In other instances, mill tailings from concentrated ores (with associated high activities) were studied. This should be called mill slag to distinguish from mining site tailings.

Results from the current study show that the overburden soils are safe and can be used as construction materials without radiological threat to the populace, since the overburden has not made contact with the radioactive ore. However care should be taken during the utilization of the soil for construction to distinguish between overburden and gangue. The soil from the latter will show higher radioactivity since it has been in contact with radioactive elements in tin before extraction.

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