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Radioactivity Measurements of the Jos Tin Mine Tailing in Northern Nigeria

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ABSTRACT

The activity concentrations of ^{226}Ra and ^{232}Th in Jos Tin mine tailings and its environment were measured using Hyper-pure Germanium detector (HPGe). Preset counting time of 30,000s was used for each sample to avoid statistical errors. Results showed the activities varied from one site to another. The activities of ^{226}Ra and ^{232}Th are much higher than the recommended average limit given by UNSCEAR in the mining sites and even at 500 m away from the mining site. At 1 Km away from the mining site the activity measured is within the limit for normal background. The ratio of activity of Thorium to Radium is greater by factor of 5 in all the samples in the mining site and 500 m away which should be about unity. The mean absorbed doses (1828.66, 252.08 and 171.07 nGyh^{-1}) for Site1, Site2 and 500m from mining site respectively in air are equally high compared with (51 nGyh^{-1}). Also, the estimated effective dose equivalents are much higher than the recommended limit except for 1 Km from the mining site. There is need to improve waste management practices in this industries in order not to expose those living and working in this environment to health hazards associated with these radionuclides.

Keywords: Radioactivity, Absorbed dose, Effective dose, Tin mine tailings, HPGe

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INTRODUCTION

The knowledge of distribution of radionuclides and radioactivity levels in the environment is important for assessing the radiation exposure to the population. There are two main contributors to natural radiation exposures: high-energy cosmic ray particles incident on the earth's atmosphere and radioactive nuclides that originated in the earth's crust and are present everywhere in the environment, including human body [1]. Jos area is located in the Northern part of Nigeria and its lithological formations are basement complex, new basalt and

Biotite Granite. Biotite Granite is associated with Tin and Columbine and the primary purpose of this industry is to mine Tin, mill and by-product. Tin is one of the mineral resources that Nigeria is endowed with. It has contributed immensely to the national wealth and provides Nigeria with some socio-economic benefits. However, extensive mining operations can degrade the environment affecting the human population in the surrounding vicinity. Also, the large scale mining activities have the potential to enhance the background radiation levels prevailing in the region [2]. Different types of environmental damage and hazards inevitably accompany the harnessing of this important mineral [1; 3]. Most specially the Tin tailings that is, the by-product from the mining and processing of Tin such as monazite, thorite, zircon etc., which are dumped in the mining site due to lack of a market. The tailings contain some heavy metals that are highly radioactive. These tailings may result in radiation exposure of the population living in the neigbourhood through (i) leached activity which may be directly ingested through drinking water or may indirectly enter the food chain by uptake through vegetation, fish, milk and meat, (ii) an enhanced external radiation background in the area, and (iii) higher radon levels due to ground emanation [4]. The low level doses of ionizing radiation emanating from the tailings may pose health hazard to the immediate human population. The most common radiation induced health effects are incidence of cancers and genetic effects. Lung cancer induction is the most common effect in inhalation radiation exposure. Exposure to these radionuclides for a period of time may results in life shortening [5; 6]. The aim of this present work is to determine the activity concentrations of ^{226}Ra and ^{232}Th in the tailing sites and mining neighbourhood. Activities of ^{226}Ra and ^{232}Th were considered because both head series of radionuclides that produce significant human exposure [1]. The data obtained were used to assess the potential radiological hazards associated with these soils by estimating the absorbed dose rates in air (D_R) and the annual outdoor effective dose equivalent rates (H_R).

MATERIALS AND METHOD

Sample Collection

Five samples from five positions from the two tailings sites were cored out using digger and spade. Also five samples from five positions were taken at 500 m and 1 Km away from the mining site. One kilogram samples were taken from each site and stored in a polyethylene bag. The samples were processed following the standard procedures [7]. These samples were oven dried at 60^{o}C to attain a constant weight. The samples were grounded and sieved with a fine mesh, packed in 0.02kg mass and properly sealed up in a marinelli beaker. These were left for 30days before measurement so that the ^{226}Ra and ^{232}Th would attain secular equilibrium with their respective daughter and grand-daughters. Proper sealing was ensured by providing double seal to the lid of the container to avoid ^{222}Rn escaping.

Activity Determination

The activity concentration of ^{232}Th and ^{226}Ra were determined by hyper-pure germanium (HPGe) detector (Canberra co-axial type), with a relative efficiency of 50% and an energy resolution of 2.4 keV(FWHM) at 1.33 MeV of ^{60}Co. The detector was properly shielded to suppress external background radiation coming from building materials. The electronics of the system was set up to cover a photon energy range of about 2.0 MeV with 4K channels. The detector was calibrated using the IAEA certified soil reference standard materials, RG U-

238, RG Th-232, with densities similar to the pulverized samples to be measured. The ^{226}Ra activity concentration was calculated using the 609.3 keV peak from the ^{214}Bi decay and the 351.9 keV peak from the ^{214}Pb peak. In the case of ^{232}Th was measured using the peaks at 911.2 keV from the ^{228}Ac and 583.2 keV from ^{208}Tl. The counting time was 30000s; the time is large so as to minimize statistical errors. The activity of each radionuclide in the samples was determined using the relation given in equation 1 [8].

$$A_s = \frac{C_{net}}{\varepsilon_\gamma Y_\gamma m_s} \qquad (1)$$

ε_γ is the detector efficiency at γ-energy of interest
C_{net} is the count per second of the sample
m_s is the mass of the sample
Y_γ is the intensity of gamma ray at the particular energy being counted

RESULTS AND DISCUSSION

The results of the radioactivity measured from the tailing sites are displayed in Table 1 with their counting errors. The activity of ^{226}Ra measured range from 231.98 ± 5.7 $Bqkg^{-1}$ to 784.250 ± 3.9 $Bqkg^{-1}$ with mean value of 512.24 $Bqkg^{-1}$, while that of ^{232}Th range from 1737.38 ± 20.81 to 3616.0 ± 38.08 $Bqkg^{-1}$ with mean value of 2635.78 $Bqkg^{-1}$ in the tailing Site1. In the tailing Site2 activity of ^{226}Ra range from 43.11 ± 2.74 to 63.48 ± 3.77 $Bqkg^{-1}$ with mean value of 51.36 $Bqkg^{-1}$, while that of ^{232}Th is between 298.68 ± 3.09 and 415.71 ± 4.07 $Bqkg^{-1}$ with mean value of 378.08 $Bqkg^{-1}$. The mean activity of the two sites was not calculated together because of large variation in the measured concentrations from the two sites. It was observed from the measurements that the activity of Site1 is 10 and 7 times greater than that of Site2 for ^{226}Ra and ^{232}Th respectively. The variation in the activity measured from the two sites is probably due to difference in the radioactive separation disposal point, for instance, Site1 is disposal point for the monazite separation tailings. [9; 10; 11] have reported high concentration of radionuclides associated with the presence of radioactive minerals most especially monazite in the rocks. These values are far above the world average for soil, 33 and 45 $Bqkg^{-1}$ for ^{226}Ra and ^{232}Th respectively [1]. The activity of ^{226}Ra measured at 500 m away from the mining site range from 31.83 ± 5.35 to 42.65 ± 3.10 $Bqkg^{-1}$ with a mean value of 38.30 $Bqkg^{-1}$, while for ^{232}Th varied between 194.41 ± 5.40 and 312.43 ± 6.31 $Bqkg^{-1}$ with a mean value of 261.32 $Bqkg^{-1}$. At 1 Km away from the mining site the measured activity of ^{226}Ra range from 2.03 ± 0.18 to 7.68 ± 0.45 $Bqkg^{-1}$ with a mean value of 5.04 $Bqkg^{-1}$, while that of ^{232}Th range from 2.08 ± 0.19 to 29.09 ± 1.44 $Bqkg^{-1}$ with a mean value of 16.28 $Bqkg^{-1}$. The activity measured at 500 m away was higher compared to 1 Km away from the site, this may be due to the closeness to the mining site and through leached activity via the water course. Although, the activity measured at 500 m away from the site was much lower than that obtained in the mining site, the activity measured still exceed the recommended limit for normal background [1]. It was observed from the measurements that the activity measured at 1 Km away from the site was within the recommended average limit for normal background [1]. It is obvious from the measurements that the activity decreases with distance from the mining site. These results show that the high background radiation level at the mining site is mainly due to the mining activities and poor waste management. Figure 1 shows the pictorial presentation of the concentrations at various positions with Site1 values scaled by 5 for clear comparison, from the Figure, ^{232}Th exhibited higher activity concentrations in the entire samples at various positions. This can be attributed to the biotite granitic origin of Jos area geology and that biotite granites may contain high concentration of thorium than radium. A comparison of the mean activity concentrations values obtained in this work with values from other regions of the world are displayed in Table 3. The mean activity of both radionuclides for Site1 is greater than the values obtained other countries. In Site2 the activity of ^{226}Ra obtained in this study are higher than those obtained in other countries except for Turkey and China, while the activity obtained for ^{232}Th in other countries are lower than the value obtained in this site. At 500 m away from the mining site the activity of ^{226}Ra obtained in this study is less than other countries except for India and Pakistan, while that of ^{232}Th is greater than the value obtained in other countries. 1 Km away from the mining site the activity of both radionuclides obtained in this study are less than the values obtained in the other countries.

Tailings Radiation Hazard Assessment

In order to assess the radiation hazard of the measured natural ^{226}Ra and ^{232}Th activity concentrations, absorbed dose rate D_R (nGyh^{-1}) in air and annual effective dose equivalent H_R (mSv year^{-1}) were calculated. According to the results of ^{226}Ra and ^{232}Th activities in the sample (Table 1) the gamma dose rates in air were estimated using the dose coefficients factor 0.462, 0.604 (nGyh^{-1}) given by [1] for ^{226}Ra and ^{232}Th respectively. The absorbed dose is estimated by:

$$D = [(0.462 \times A_{Ra}) + (0.604 \times A_{Th})]nGyh^{-1} \qquad (2)$$

where, A_{Ra} and A_{Th} are the concentrations of ^{226}Ra and ^{232}Th respectively, in Bq kg^{-1}. The calculated absorbed dose values for Site1 and 2 are displayed in Columns 5–7 of Table 1. The total absorbed dose delivered by these radionuclides for Site1 ranged between 1302.88-2546.38 nGyh^{-1} with a mean value of 1828.66 nGyh^{-1} which is about 36 times higher than the world average value of 51 nGyh^{-1}. For Site2 the total absorbed dose range 200.32-275.66 nGyh^{-1} with a mean value of 252.08 nGyh^{-1} this is still higher than the world average value [1]. The calculated absorbed dose values for 500m and 1 Km away from mining site are displayed in Columns 5–7 of Table 2. The mean total absorbed dose obtained at 500 m away (171.07 nGyh^{-1}) from the site is also greater than the average world value but at 1 Km away the mean total absorbed dose obtained (12.16 nGyh^{-1}) is within the recommended limit for normal background. The annual effective dose equivalent to the population due to the radioactivity was estimated using the dose coefficient (0.7 SvGy^{-1}) and occupancy factor (0.2) for outdoors

[1]. This translates the absorbed dose rate (nGy h^{-1}) in air to effective dose (mSvyr^{-1}) which can be determined as:

$$H_R = D_R N_h \times 0.7 SvGy^{-1} \times 0.2 \qquad (3)$$

where D_R is the absorbed dose rates in air (nGyh^{-1}) and N_h (8,760 h) is the number of hours in one year.

Table 1: Radioactivity of ^{238}U and ^{232}Th in Tailing Samples

	Sample Name	Activity ($Bqkg^{-1}$)		Absorbed Doses ($nGyh^{-1}$)		Total Absorbed Doses ($nGyh^{-1}$)	Effective Dose ($mSvyr^{-1}$)
		^{226}Ra	^{232}Th	^{226}Ra	^{232}Th		
Site 1	APT1_Dru 1	784.25 ± 3.9	3616± 38.08	362.32	2184.06	2546.38	3.18
	APT1_Dru 2	645.15 ± 4.3	2718.93 ± 7.11	298.06	1642.23	1940.29	2.42
	APT1_Dru 3	351.09 ± 3.1	2431.41 ± 23.52	162.20	1468.57	1630.77	2.03
	APT1_Dru 4	231.98 ± 5.7	2675.19 ± 31.48	107.17	1615.81	1722.98	2.15
	APT1_Dru 5	548.71 ± 6.8	1737.38 ± 20.81	253.50	1049.38	1302.88	1.63
	Range	231.98 - 784.25	1737.38 - 3616	107.17 – 362.32	1049.38 – 2184.06	1302.88 – 2546.38	1.63 – 3.18
	Mean	512.24	2635.78	236.65	1592.01	1828.66	2.28
Site 2	THS 1	46.28 ± 1.69	400.23 ± 5.00	21.38	241.74	263.12	0.33
	THS 2	63.48 ± 3.77	397.44 ± 8.05	29.33	240.05	269.38	0.34
	THS 3	53.18 ± 2.09	415.71 ± 4.07	24.57	251.09	275.66	0.34
	THS 4	50.73 ± 5.13	378.25 ± 3.21	23.44	228.46	251.90	0.31
	THS 5	43.11 ± 2.74	298.68 ± 3.09	19.92	180.40	200.32	0.25
	Range	43.11 – 63.48	298.68 – 415.71	19.92 – 29.33	180.40 – 251.09	200.32 – 275.66	0.25 – 0.34
	Mean	51.36	378.02	23.73	228.35	252.08	0.31

± *Value indicates the error associated with the measurement.*

Table 2: Radioactivity Measured from the Mining Neighbourhood

Distance	Sample Name	Activity ($Bqkg^{-1}$)		Absorbed Doses ($nGyh^{-1}$)		Total Absorbed Doses ($nGyh^{-1}$)	Effective Dose ($mSvyr^{-1}$)
		^{226}Ra	^{232}Th	^{226}Ra	^{232}Th		
	APT1_A1	40.13 ± 0.39	312.43 ± 6.31	18.54	166.48	185.02	0.23
	APT1_A2	39.19 ± 1.82	298.88 ± 8.97	18.11	180.52	198.63	0.25
	APT1_A3	37.68 ± 1.17	225.26 ± 5.73	17.41	136.05	153.46	0.19
500 m	APT1_A4	42.65 ± 3.10	275.63 ± 5.03	19.70	166.48	186.10	0.23
Away	APT1_A5	31.83 ± 5.35	194.41 ± 5.40	14.71	117.42	132.13	0.17
	Range	31.83 – 42.65	194.41 – 312.43	14.71 – 19.70	117.42 – 180.52	132.13 – 198.63	0.17 – 0.25
	Mean	38.30	261.32	17.69	153.39	171.07	0.21
	APMC_1	5.95 ± 0.36	29.09 ± 1.44	2.75	17.57	20.32	0.03
	APMC_2	7.68 ± 0.45	24.50 ± 1.92	3.55	14.80	18.33	0.02
	APMC_3	5.80 ± 0.45	16.83 ± 0.56	2.68	10.17	12.85	0.016
1 Km	APMC_4	3.75 ± 0.36	8.89 ± 0.92	1.73	5.37	7.10	0.009
Away	APMC_5	2.03 ± 0.18	2.08 ± 0.19	0.94	1.26	2.20	0.003
	Range	2.03 – 7.68	2.08 – 29.09	0.94 – 3.55	1.26 – 17.57	2.20 – 20.32	0.003 – 0.03
	Mean	5.04	16.28	2.33	9.83	12.16	0.016

± *Value indicates the error associated with the mea surement.*

The effective doses calculated for Site1 and 2 are presented in Column 8 of Table 1. This ranged between 1.63–3.18 mSv with a mean value of 2.28 mSv for Site1 while the mean for Site2 is 0.31 mSv. The mean value of effective doses obtained for these sites are much higher than the recommended limit (0.07 mSv) for the normal background. The effective doses calculated for 500 m and 1 Km away from the site are presented in Column 8 of Table 2. It was found that the mean effective doses obtained at 500m away (0.21 mSv) from the site is higher than the recommended limit, but at 1 Km away (0.016 mSv) from the site the mean effective doses is much lower than the recommended limit for normal background [1].

Table 3: Comparison of the mean concentration of ^{226}Ra and ^{232}Th in present study with other countries

Country	^{226}Ra	^{232}Th	Reference
India	31	63	[12]
Ireland	60	26	[13]
Turkey	115	192	[14]
China	112	71.5	[15]
Spain	46	49	[16]
Pakistan	32.9	53.6	[4; 17]
Site 1	512.24	2635.78	
Site 2	51.36	378.02	
500 m Away	38.30	261.32	Present Study
1 Km Away	5.04	16.28	

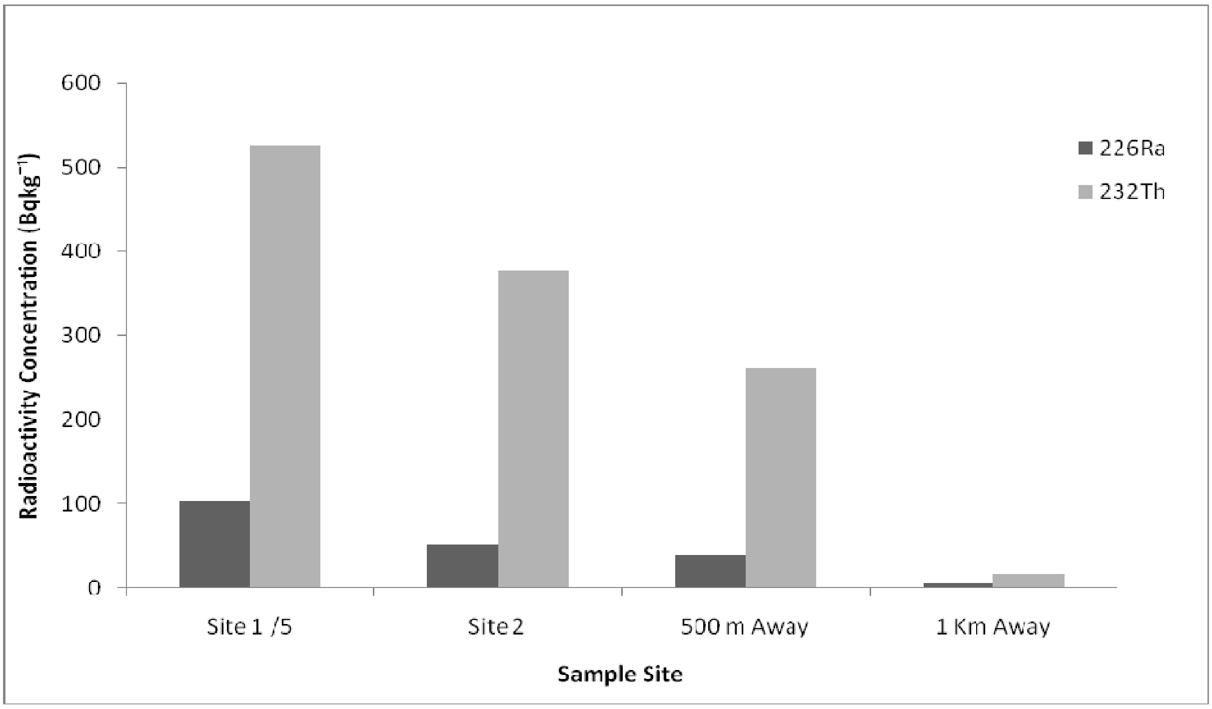

Fig. 1: Radioactivity Concentrations of ^{226}Ra and ^{232}Th in the samples

CONCLUSION

Hyper pure Germanium detector has been used to determine the activity of ^{226}Ra and ^{232}Th in the Tin tailing sites and its surroundings. The activity concentrations were much higher than the world average value for soil in the mining site and at 500 m away, but at 1 Km away the activity is within the recommended limit. This may suggest that the dumping of tailings around the site has increased the level of background radiation on the surroundings and the mining area. Potential radiological effects on miners and those living less than 1 Km to the site due to external irradiation are significant. The high concentrations of natural radionuclides can act as a source for extended radon exhalation. Dwellings in the vicinity less than 1 Km from mining site are candidates for higher radon burden. There is need to improve waste management practices in this industries in order not to expose those living and working in this environment to health hazards associated with these radionuclides.

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