



ASSESSMENT OF RADIONUCLIDES CONCENTRATION AND RADIOLOGICAL IMPACT OF ABANDONED TIN MINES SOILS IN SOME COMMUNITIES OF BARKIN LADI, PLATEAU STATE, NIGERIA

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ABSTRACT

The concentration of natural radionuclides in the soil around the world has become an environmental concern for international and national authorities because of the harmful effect of radiation exposure on human health. The samples of soil used for this study were collected from Foron and Heipang districts of Barkin Ladi Local Government Area of Plateau State. The samples were analyzed using Canberra Model 727/727R Lead Shield Gamma-ray Spectrometer with NaI(Tl) detector. The result shows that ^{40}K ranges from 301.91 ± 0.24 Bq/kg to 124.97 ± 0.31 Bq/kg from the mining spot to 400 m away from the mining spots. ^{226}Ra ranges from 105.43 ± 1.76 Bq/Kg to 43.10 ± 0.62 Bq/Kg from mining spots to 400 m away from the mining spots, and ^{232}Th also ranges from 92.79 ± 0.26 Bq/kg to 26.97 ± 0.39 Bq/kg respectively. The absorbed dose rate in the air (Dr) has a mean value of 82.88 nGh $^{-1}$. The annual effective dose rate (AEDR) mean value was $0.1\text{mSv}\text{y}^{-1}$, and the radium equivalent (Ra eq) mean value was 180.23 Bq/kg, H_{ex} and H_{in} have mean values of 0.49 and 0.69 respectively. The calculated excess lifetime cancer risk (ELCR) ranges from 0.0003 to 0.00039 with a mean value of 0.00035 in the soil samples. Therefore, this result suggested that the area under study could be termed radiologically unsafe for people living around or close to the area.

KEYWORDS: Radionuclides; Abandoned tin mining spot; Dose rate; Soil; Cancer risk

1.0 INTRODUCTION

The production of tin in the Jos plateau during the colonial era started at 1.5 metric tons in 1914 and then it began to increase until 1943 when Nigeria became the 6th producer (Onwuka *et al.*, 2013). Tin tailings were usually processed to extract valuable minerals such as columbite, monazite, zircon, and xenotime, which have been proven to contain a high concentration of naturally occurring radioactive materials (NORMs) such as ^{40}K , ^{226}Ra and ^{232}Th and any of their decay products, such as radium and radon (Faanu *et al.*, 2011 and Ibeanu *et al.*, 2013). The exposure of the human population to ionizing radiation is a major health hazard. The largest source of this ionizing radiation is the natural radioactivity present in the human environment. (UNSCEAR, 2000).

Mining generally has a very high impact on man and the environment. Mining could cause an increase in employment and economic profit for the community and the operators, but represent a threat to natural reserves due to landscape changes and pollution (Zaini *et al.*, 2008). This often results in destroyed ecosystems and polluted environments, which represents a hazard to the local population and the operators. The by-product of heavy minerals from tin mining is rich in uranium and thorium. External exposures occur when workers or persons are close to stockpiles of radioactive minerals, soils, and tailings which they might not have knowledge of. Abandoned dumping sites (tailings) are usually subject to the leaching of uranium and thorium when they come into contact with an aqueous medium (Ibeanu, 2003). Ibeanu (2003) acknowledge that, tin ore contains radioactive mineral rich in monazite and

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zircon. During the tin ore processing operations, uranium and thorium concentrate are responsible for the observed high concentrations levels measured in the tailing dump and contaminated soil samples. Funtua and Elebga (2005), stated that several years of mining and processing of cassiterite (tin ore) and columbite (niobium ore) in the Jos Plateau have generated large quantities of tailing that are rich in these radioactive minerals and are mostly dumped haphazardly in the environment.

Furthermore, there is a growing concern over the radiological impact of these tailings since it has been established that these ores are associated with accessory minerals like zircon, monazite, xenotime, and thorite, which have high concentrations of thorium and uranium. Atipo *et al.* (2020) studied the high terrestrial radiation level in active tin mines in the Rayfield -Du area of Jos South Nigeria and their findings showed a high level of radiation above regulatory limits. Also study conducted by Olise *et al.* (2014) around tin mining dumpsite in the Rayfield area of Jos reported high values in the concentration of ²²⁶Ra, ²³²Th, and ⁴⁰K and associated radionuclides in the soil of the dumpsites. Similarly, elevated levels have been reported by many other previous kinds of research conducted in Jos and its environment. (Shibdawa *et al.*, 2019; Jwanbot *et al.*, 2012; Usikalu *et al.*, 2011; Arogunjo *et al.*, 2009; Ademola *et al.*, 2008). Most of these reports attributed the high levels of radiation primarily to the influence of tin and its mining activities in the area, while the influence of the geological formation of the area was also identified

as one of the factors. This study was therefore aimed at Assessing the radionuclides content in soils and the radiological hazard index of abandoned tin mines in some communities of Barkin Ladi, Plateau State, Nigeria

2.0 MATERIALS AND METHODS

2.1 MATERIALS/EQUIPMENT

The materials that were used for the research work are the Canberra Model 727/727R Lead Shielded Gamma-ray Spectrometer with NaI(Tl) detector, Oven Gallenham England, Beakers, Mortar, and pestle.

2.2 AREA OF STUDY

Barkin Ladi, a Local Government Area in Plateau State is situated in the central part of Northern Nigeria, on a rugged terrain of lowlands. A total of fifteen samples were collected. Three samples each from five villages in Foron and Hiepang districts of Barkin Ladi Local Government area of Plateau State. The samples were collected from, Foron Junction on Lat 9.69164N and Long. 8.9119E at an altitude of 1259m above sea level, Nkan on Lat.96686S and long.8.96256E at an altitude of 1197m, Newo on Lat. 9.66882S and Long. 8.96265E on an altitude of 1187m, Kwat on Lat. 9.66847N and Long. 8.92711E on an altitude of 1188m Ban Heipang on Longitude 9.67465, Latitude 8.88261, Altitude 1252 m. These areas are part of the principal centre of tin and columbite mineralization, which forms the focal area of younger granites.

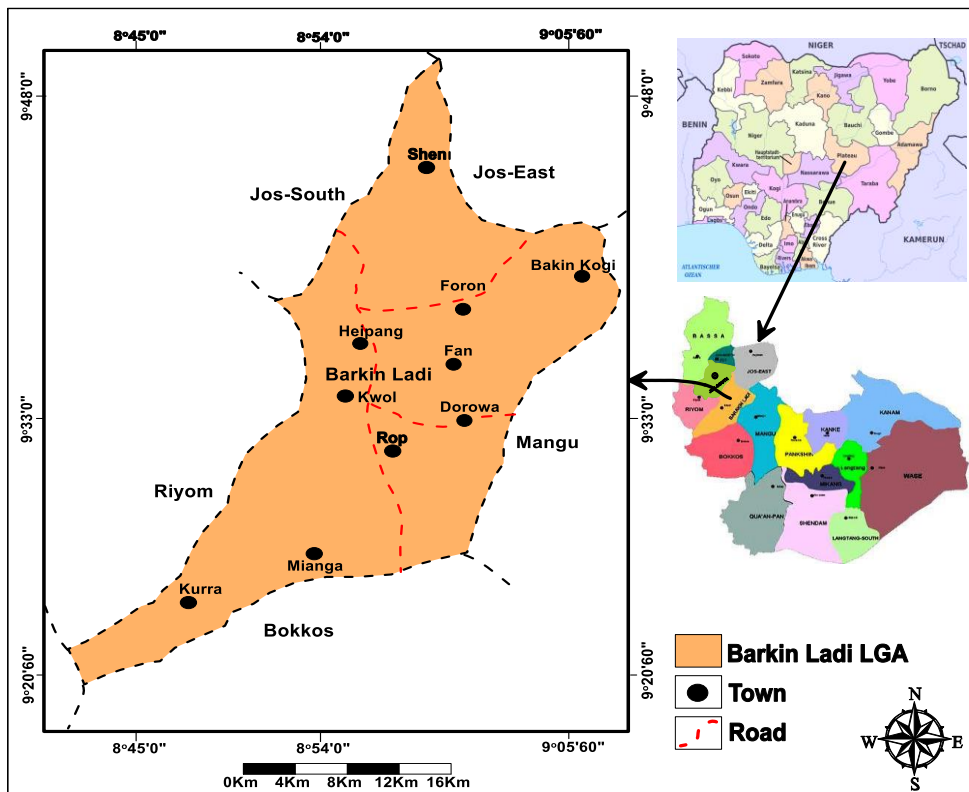


Figure 1: Map of Barkin Ladi

2.3 SAMPLE COLLECTION AND ANALYSIS

At each sampling point, about 0.50 kg of the soil sample was collected from between 0- 15 cm from the surface of the soil, using a clean stainless-steel spoon at a distance of 1m away from each other, and within an area of one square meter from each sampling site. In each of the villages, three samples were collected at a distance of 200 m away from each other. The soil samples were collected into a very clean polythene bag and well-labelled to avoid mixed up of samples.

The samples after weighing were transferred to radon-impermeable cylindrical plastic containers of uniform size (70 mm height by 60 mm diameter) and were sealed for about 30 days. This was done to allow for Radon and its short-lived progenies to reach secular radioactive equilibrium before gamma spectroscopy. The reference soil was also transferred to a container of the same material and dimensions as were used for the soil samples. A lead-shielded 76 x 76 mm NaI(Tl) detector crystal (Model No. 727 series, Canberra Inc.) that is coupled to a Canberra Series 10 plus Multichannel Analyzer (MCA) (Model No.1104) through a preamplifier was used for the radioactivity measurements. It has a resolution (FWHM) of about 8% at the energy of 662.0 KeV (¹³⁷Cs) which is considered adequate to distinguish the gamma-ray energies of interest in the present study. The choice of gamma-ray peaks of the radionuclides to be used for measurements was made because the NaI(Tl) detector used in this study had a modest energy resolution. This was to ensure that the photons emitted by the radionuclides would only be sufficiently discriminated if their emission probability and their energy were high enough, and the surrounding background continuum low enough. Therefore, the activity concentration of ²¹⁴Bi (determined from its 1760 KeV γ - ray peak) was chosen to provide an estimate of ²²⁶Ra (²³⁸U) in the samples, while that of the daughter radionuclide ²⁰⁸Tl (determined from its 2615 KeV γ -ray peak) was chosen as an indicator of ²²⁸Th (²³²Th). Potassium-40 was determined by measuring the 1460 KeV γ -rays emitted during its decay.

The soil samples were placed symmetrically on top of the detector and measured for 29000 seconds. The net area under the corresponding peaks in the energy spectrum was computed by subtracting counts due to the Compton scattering of higher peaks and other background sources from the total area of the peaks.

2.3.1 Radiological Hazard Assessment

2.3.1.1 Assessment of radium equivalent (Ra_{eq}). Gamma radiation hazard caused by specific radionuclides of ²²⁶Ra, ²³²Th, and ⁴⁰K was calculated using different indices (UNSCEAR, 2000). Ra_{eq} is a weighted sum of activities of the three radionuclides based on the supposition that 370Bq/kg ²²⁶Ra, 259Bqkg⁻¹ ²³²Th, and 481Bqkg⁻¹ ⁴⁰K produce the same gamma-ray dose rate.

$$Ra_{eq} \left(\frac{Bq}{kg} \right) = CRa + 1.43CTh + 0.077CK \text{ -----1}$$

CRa, CTh, and CK are the activity concentrations of ²²⁶Ra, ²³²Th, and ⁴⁰K (in Bqkg⁻¹) respectively. To keep the annual radiation dose below 1.5 mGy⁻¹, the maximum value must be less than 370 Bqkg⁻¹

2.3.1.2 Absorbed dose rate in the air (D_R).

According to the guidelines provided by UNSCEAR (2000), the absorbed gamma dose rate D_R (nGyh⁻¹) in the air was estimated at 1 m above the ground surface to ensure uniform distribution of radionuclides. This parameter can be used to assess any radiological hazards and radiation exposure from radionuclides in the soil; the absorbed dose rate was calculated using the following formula (Veiga *et al.*, 2006 in Ghazwa *et al.*, 2016)

$$D_R (nG/h) = 0.427CRa + 0.623CTh + 0.043CK \text{ -----2}$$

Where D_R is the dose rate in nGyh⁻¹ and CRa, CTh and CK are the activity concentrations (Bqkg⁻¹) of radium ²²⁶Ra, thorium ²³²Th, and ⁴⁰K respectively.

2.3.1.3 Annual effective dose rate (AEDR):

Annual effective dose rate was calculated to assess the health effect of the absorbed dose by using a conversion coefficient (0.7SvGy⁻¹) to transform absorbed dose in air to the effective dose received by humans, with an outdoor occupancy factor of (0.2), which is equivalent to an outdoor occupancy of 20% and 80% for the indoors (Veiga *et al.*, 2006 in Ghazwa *et al.*, 2016). This factor is suitable for determining the pattern of life in the study area. The annual effective dose rate (AEDR, in mSvy⁻¹) received by the population can be expressed as reported by UNSCEAR (2000).

$$AEDR = D_R \times 8760 \times 0.2 \times 10^{-6} \text{ -----3}$$

Where AEDR is the annual effective dose rate (mSv/y), D_R is the gamma dose rate (nGy/h), the coefficient 0.7 Sv/Gy is the conversion coefficient from the absorbed dose in the air to the effective dose received by the adult, 0.2 for outdoor occupancy factor, 8760 hour per year is equal to 365 days x 24 h per year.

2.3.1.4 External hazard index H_{ex}:

The external hazard index for the samples under investigation was calculated using the equation defined by (El-TaHER, 2010).

$$H_{ex} = \frac{CRa}{370} + \frac{CTh}{259} + \frac{CK}{4810} \leq 1 \text{ -----4}$$

CRa, CTh, and CK are the activity concentrations of ²²⁶Ra, ²³²Th, and ⁴⁰K in (Bq/Kg) respectively. The maximum value of H_{ex} equal to unity corresponds to the upper limit of Ra_{eq} (370Bq/kg)

2.3.1.5 Internal Hazard Index H_{in}:

The internal hazard index (H_{in}) was introduced to describe the hazard of radon and its short-lived products in the soil and recommended to be less than unity (Haribala, 2017; Korkulu and Ozkan, 2013)

$$H_{in} = \frac{CRa}{185} + \frac{CTh}{259} + \frac{CK}{4810} \text{ -----5}$$

CRa, CTh, and CK are the activity concentrations of ²²⁶Ra, ²³²Th, and ⁴⁰K in (Bq/Kg) respectively.

2.3.1.6. Excess life expectancy cancer risk (ELCR) was calculated using the equation described by (Agbalagba, 2017)

$ELCR = AEDR \times L \times RF \times 10^{-6}$ -----6
Where AEDR is annual effective dose rate, L is life expectancy taken as 70 years and RF is risk factor taken as 0.05/Sv. The global average value of 0.00027 is given by (ICRP, 1991) as the safety limit.

3.0 RESULT AND DISCUSSION

Table 1: Activity concentration of ^{40}K at mining spots, 200 m and 400 m away from mining spots in Bq/kg

Sample ID	Mining spot	200 m away	400 m away
KW	210.26 ± 0.24	276.01 ± 0.08	124.97 ± 0.31
FJ	315.23 ± 0.26	204.14 ± 0.32	142.87 ± 0.46
B	231.87 ± 0.34	249.41 ± 0.25	210.27 ± 0.10
NK	301.91 ± 0.24	236.32 ± 0.34	139.92 ± 0.24
N	208.12 ± 0.24	157.12 ± 0.48	167.33 ± 0.15

n=3

Table 2: Activity concentration of ^{226}Ra in mining spots, 200 m and 400 m away from mining spots in Bq/kg

Sample ID	Mining spot	200 m away	400 m away
KW	98.71 ± 0.64	97.32 ± 0.40	51.99 ± 0.30
FJ	98.21 ± 0.42	85.88 ± 0.24	67.86 ± 0.12
B	85.97 ± 0.43	63.43 ± 0.52	46.20 ± 0.80
NK	83.42 ± 0.30	58.02 ± 0.36	43.10 ± 0.62
N	105.43 ± 1.76	99.91 ± 1.11	57.02 ± 0.22

n=3

Table 3: Shows the Activity concentration of ^{232}Th in mining spots, 200 m and 400 m away from mining spots in Bq/kg

Sample ID	Mining spot	200 m away	400 m away
KW	92.79 ± 0.26	39.08 ± 2.40	26.97 ± 0.39
FJ	89.23 ± 0.33	72.48 ± 0.22	64.23 ± 0.10
B	69.08 ± 0.51	62.22 ± 0.55	41.88 ± 0.08
NK	76.20 ± 0.37	58.57 ± 0.44	35.74 ± 0.51
N	86.66 ± 1.73	67.39 ± 0.94	61.20 ± 0.23

n=3

The activity concentration of ^{40}K , ^{226}Ra , and ^{232}Th expressed in Bq/kg for the samples obtained from mining areas in Foron and Heipang district of Barkin Ladi Local Government Area of Plateau State are presented in Table 1-3 above. The samples were taken directly from the mining spots, 200 m and 400m away from the mining spots respectively. This was done to assess the distribution patterns of the radionuclides from the mining spots to other parts of the environment. The result of the analyses shows that ^{40}K has the highest concentrations compared to the other radionuclides in the study area. The values of ^{40}K range from 301.91 ± 0.24 Bq/kg to 124.97 ± 0.31 Bq/kg from the mining spot to 400 m away from the mining spots. ^{226}Ra ranges from 105.43 ± 1.76 Bq/kg to 43.10 ± 0.62 Bq/kg in the mining spots to 400 m away from mining spots, and ^{232}Th also ranges from 92.79 ± 0.26 Bq/kg to 26.97 ± 0.39 Bq/kg respectively from the mining spots to 400 m

away from the mining spots. The concentration of radionuclides tends to decrease as the distance is moved further away from the mining spots. The concentration of ^{40}K , ^{226}Ra , and ^{232}Th obtained from this study was found to be lower compared to those reported by (Masok, *et al.*, 2015; Jibiri and Okeyode, 2011; Mangset and Sheyin, 2009 and Ajayi, 2008). Results reported by Jwanbot *et al.* (2013) in some soils and food crops in Barkin Ladi are similar to those of this study, except for samples taken from mining spots where the value is higher. The decrease in the concentrations of the radionuclides over some years now may be a result of the leaching and uptake of the radionuclides by plants from the soil. The result of the ^{40}K in this study were all lower than the recommended limits of 420 Bq/kg (UNSCEAR, 2000). Though ^{226}Ra and ^{232}Th were found to be higher than the recommended limits of 35 Bq/kg and 45 Bq/kg respectively (UNSCEAR,

2000). ^{226}Ra is a decay product of ^{238}U , when ^{226}Ra undergoes alpha decay, ^{222}Rn is produced. ^{226}Ra is primarily deposited in the bone replacing Ca^{2+} , whereas ^{222}Rn is deposited in the lungs upon inhalation. The accumulation of these radionuclides

over a long period could increase one's risk of cancer (Masok *et al.*, 2015). Factors such as erosion (water and wind), animal movement, transportation of the minerals from the mining spot, and improper tailings disposal could be responsible for the distribution of these radionuclides into the environment.

Table 4: Result of the radiological hazard assessment of the study area

Sample Location	$D_R(\text{nGh}^{-1})$	AEDR (mSvy^{-1})	Ra eq Bq/Kg	H_{ex}	H_{in}	ELCR
KW	77.00	0.09	172.72	0.46	0.69	0.00030
FJ	92.27	0.11	208.67	0.56	0.78	0.00039
B	84.67	0.10	165.12	0.45	0.62	0.00035
NK	73.36	0.09	155.52	0.43	0.60	0.00032
N	87.09	0.11	199.10	0.54	0.76	0.00039

Table 4, gives the summary of the result of the radiological hazard assessment in the study area. The absorbed dose rate in the air (D_R) was calculated and the values ranged from 77.00nGh^{-1} to 92.27nGh^{-1} with a mean value of 82.88nGh^{-1} . This value is higher than the permissible limit of 60nGh^{-1} (UNSCEAR, 2000). The annual effective dose rate (AEDR) also has the highest value of 0.11mSvy^{-1} , with the lowest value of 0.09mSvy^{-1} and a mean value of 0.1mSvy^{-1} . All the AEDR values in this study are lower than the global average indoor terrestrial radiation value of 0.5mSvy^{-1} . The radium equivalent (Ra eq) values in these analysis ranges from 155.52Bq/kg to 208.67Bq/kg , with a mean value of 180.23Bq/kg , these values are lower than the maximum permissible limit of 370Bq/kg which for safety purpose is equivalent to the 1mSvy^{-1} (UNSCEAR, 2000). The external and internal hazard index (H_{ex} and H_{in}) were calculated from the activity concentration of the soil samples. The result for the H_{ex} ranges from 0.43 to 0.56 with a mean value of 0.49 , whereas the H_{in} ranges from 0.60 to 0.78 with a mean value of 0.69 , which suggests that there is insignificant radiation on the people living around the area since the values obtained are less than unity. The calculated excess lifetime cancer risk (ELCR) as presented in Table 4, shows that the values range from 0.0003 to 0.00039 with a mean value of 0.00035 in the soil samples. The ELCR values in the study area are generally high compared to the global average value of 0.00027 given by (ICRP, 1991). Generally, the result was lower than those reported by (Masok *et al.*, 2015), but higher than those of (Jwanbo *et al.*, 2013)

CONCLUSION

The radionuclides, ^{226}Ra , ^{232}Th , and ^{40}K was analyzed for their concentration and the radiological risk assessment was also calculated from some mining spot of the Foron and Heipang district of Barkin Ladi local government Area. The result of the study shows that the concentrations of ^{226}Ra and ^{232}Th in almost all the sampling spots were above the world average, whereas the concentrations of ^{40}K in

all the sampling locations were below the acceptable limits. Annual effective dose rate, radium equivalent, H_{ex} , and H_{in} were all below permissible limits, whereas, the absorbed dose in the air (D_R) and excess lifetime cancer risk (ELCR), were above acceptable limits. Therefore, the area under study could be termed radiologically unsafe for people living around or close to the area.

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