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Research Paper

Radiological safety assessment of some mine sites at Gusau and environs, Nigeria

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Abstract. Investigations were carried out to determine radiological hazard indices of NORM associated with selected abandoned mine sites at Gusau and its environs in Zamfara State, Nigeria. The γ-ray spectrometry setup at the Centre for Energy Research and Training, Ahmadu Bello University Zaria, Nigeria, which consists of 3" × 3" NaI(Tl) detector and associated electronic modules was used to determine the quantity and quality of radioactivity in the soil samples randomly collected from 10 abandoned mine sites in area of study. The results obtained show that the activity concentrations for ^{40}K, ^{226}Ra and ^{232}Th ranged from 227.10 ± 7.54 to 590.44 ± 10.57 Bq.kg^{-1}, 4.68 ± 3.52 to 18.98 ± 0.84 Bq.kg^{-1} and 40.58 ± 1.85 to 94.92 ± 2.76 Bq.kg^{-1}, respectively. The values of the radiological hazard indices further show that radium equivalent activity ranged from 94.41 ± 5.64 to 191.34 ± 7.11 Bq.kg^{-1} with mean value of 130.93 ± 4.94 Bq.kg^{-1}. The mean values of gamma index level, external and internal hazard indices are 0.48, 0.35 and 0.39, respectively. The mean absorbed dose rate of the samples was found to be 59.7 nGy.h^{-1} while the mean annual effective dose was estimated to be 0.073 mSv.y^{-1}. From the hazard indices information, it can be concluded that the radiation level in the study areas is within the safety limit.

Keywords: Radionuclides, soil, mining, NaI (Tl) detector, hazard indices.

INTRODUCTION

Our environment is naturally radioactive and has been so, since the creation of the world. Naturally occurring radioactive materials (NORMs) are wide spread in the earth's environment and exist in various geological formations such as rocks, soils, vegetation, water and air. It has been known that, naturally occurring radioactive materials became the focus of regulatory interest with the publication of the International Atomic Energy Agency (Augustine and Aku, 2013). Investigating the levels of radionuclide distribution in the environment provides essential radiological information. Soil from abandoned mine sites may contain naturally occurring radionuclide's in significant amounts and the resulting external radiation exposure pathway to the population have been the subject for study in the field of radiation protection and measurement (Faweya and Babalola, 2010).

It is disturbing to note that some of the abandoned mined sites are being turn into economic sites by some of the villagers, who engage in activities such as moulding of mud bricks, washing bicycles in the waterlogged ones, and using some as relaxation joints by herdsmen with their cattle, thereby spending ample time there almost every day. It is therefore, important to monitor the terrestrial background radiation mainly due to natural radionuclides in soil of those abandoned mine sites to ascertain the level of associated hazard indices. The interest of this work is to evaluate the radiological hazard indices using the activity concentrations of the prominent gamma emitting-NORMs (^{40}K, ^{226}Ra and ^{232}Th) associated with mine sites. It is hoped that this research work will

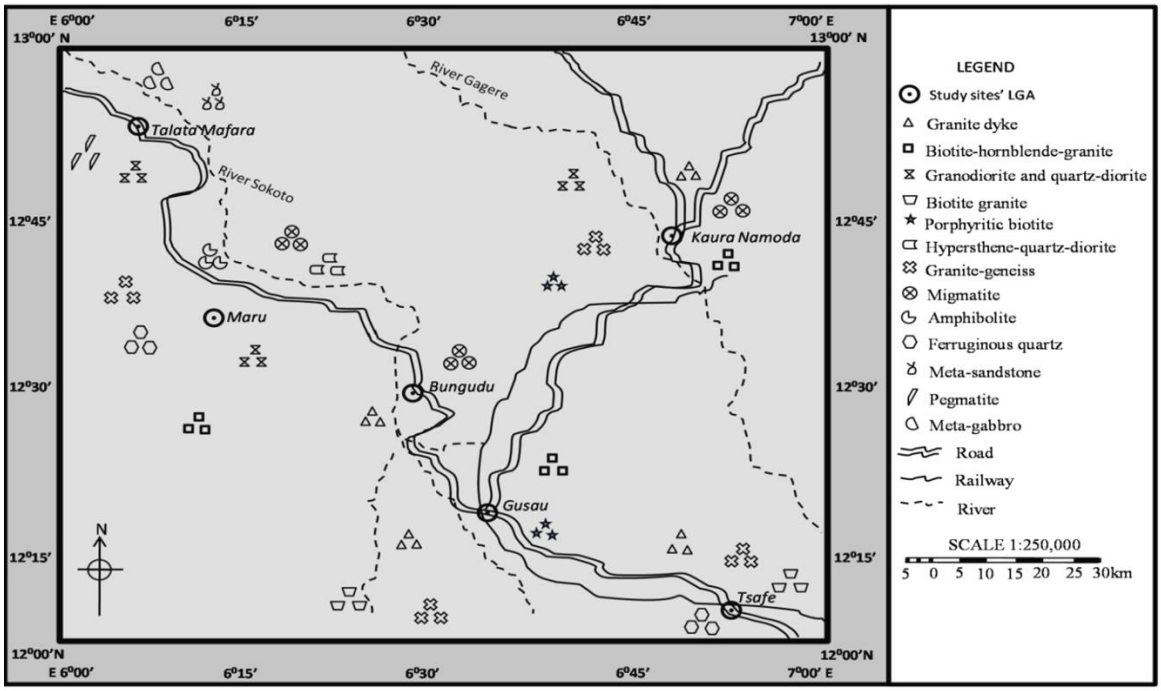

Figure 1. Geological map of the study area. Source: Parker et al. (1965).

add to the data base of some related studies carried out in other parts of the world. Similar studies aimed at investigating the environmental natural radioactivity in Nigeria are: determination of terrestrial gamma dose rates and physical-chemical properties of farm soils from ex-tin mining locations in Jos (Jibiri et al., 2011); Characterization of distribution of gamma-emitting radionuclides in soils around the Centre for Energy Research and Training (CERT) Ahmadu Bello University, Zaria (Muhammad et al., 2010); determination of radiological safety assessment and occurrence of heavy metals in soil from designated waste dumpsites used for building and composting in south-western Nigeria (Faweya and Babalola, 2010); characterization of distribution of some natural gamma emitting radionuclides in the soils of the coastal areas of Nigeria (Alatise et al., 2008); estimation of natural radionuclides and elemental composition of chemical fertilizers used in Nigeria (Jonah et al., 2002); baseline measurement of natural radioactivity in soil, vegetation and water in the industrial district of the Federal Capital Territory (FCT) Abuja, Nigeria (Umar et al., 2012).

MATERIALS AND METHODS

Geology of the study area

The study sites are located in 6 local government areas of Zamfara State, northwest Nigeria between 6°00′ – 7°00′ E of the longitude and 12°00′ – 13°00′ N of the latitude. Figure 1 presents the geological map of the study area. The number of abandoned mine sites visited are 10 and they include 2 in Tsafe local government area, 1 in Gusau local government area, 2 in Bungudu local government area, 1 in Maru local government area, 2 in Talata-Mafara local government area and 2 in Kaura-Namoda local government area of Zamfara state.

Geologically, the study sites are characterised by very old igneous rocks, formed during the precambrian-paleozoic era (Parker et al., 1965). Granite, gneisses and migmatites are lively resistant to erosion, but when weathered, they result into poor soils. The meta-sediments, on the other hand consist of phyllites, quartzites and meta-conglomerates. Although, meta-sediments are also resistant to erosion, when weathered they give rise to more fertile soils on account of the fact that the schists are rich in magnesium minerals. Many mineral deposits abound in the study area. For examples, there are smectite, kaolinite, feldspars, granite, pegmatite, quartz, migmatites, amphibolites, monazite, zircon, thorianite, ferruginous quartz, meta-sandstone, porphyritic biotite and many others.

Materials

The materials employed for this research work include cylindrical plastic containers, petroleum jelly, candle wax, matchbox, masking tape, wooden block, sieve, digital

weighing balance, oven, NaI(Tl) detector, lead shielding, PC-based data acquisition system and printer.

Sampling procedure

The soil samples were collected by random sampling method from the mine sites at selected locations in five local government areas of the state. The soil samples were packed in plastic containers from the areas of surveillance, properly sealed and labelled for easy identification and then transported to the environmental laboratory at Centre for Energy Research and Training (CERT), Ahmadu Bello University, Zaria, Nigeria.

Sample preparation

In the laboratory at CERT Zaria, the soil samples were put in an oven and set to a temperature of 105°C to allow for drying overnight in order to remove any available moisture. The dried samples were gently crushed and sieved with a sieve of mesh size 2 mm to remove organic materials. Thereafter, the samples were homogenized and packed to fill cylindrical plastic containers of height 7 cm by 6 cm diameter to fit adequately into the lead shield housing the counting detector. This satisfies the selected optimal sample container height for the NaI(Tl) analytical instrument (Ibeanu et al., 2000). The samples were carefully sealed using petroleum jelly, candle wax and masking tape in order to prevent trapped radon gas from escape. The sealed samples were kept for a minimum period of 30 days so as to allow for ^{226}Ra and its short-lived progenies to reach secular radioactive equilibrium before gamma counting.

Experimental set-up

The gamma-ray spectrometry set-up consists of a 7.62 cm by 7.62 cm NaI (Tl) detector housed in a 6 cm thick lead shield and lined with cadmium and copper sheets in order to assist in the reduction of background radiation (Umar et al., 2012). In addition, a computer based multichannel analyser (MCA) Maestro Programme from ORTEC was used for the data acquisition and analysis of gamma spectra.

Sample counting

The soil samples were placed on the detector surface and each counted for a lifetime of 29,000 s (8 h, 3 min) in reproducible sample detector geometry. The configuration and geometry were maintained throughout the analysis, as previously characterized based on well established protocol of the laboratory at the Centre for Energy Research and Training, Zaria. The background count was also measured for the same period of lifetime. The 1.764 MeV γ-line of ^{214}Bi for ^{226}Ra was used in the assessment of the activity concentration of ^{238}U while 2.615 MeV γ-line of ^{208}Tl was used for investigating the activity concentration of ^{232}Th while the single 1.46 MeV γ-line of ^{40}K was used for its content evaluation.

Radiological calculations

Activity concentrations

The activity concentrations for the natural radionuclides in the measured samples were computed using the relation (Ibeanu, 1999).

$$A_c = \frac{N_c}{L_t}\sigma^{-1} \quad (1)$$

where L_t is the lifetime of counting, σ is a conversion factor which is constant for each radionuclide at constant geometry and is a characteristic of efficiency of the NaI(Tl) detector assembly used at CERT Zaria. All the raw data obtained from the detector were converted to conventional units using the calibration factors to determine the activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th, respectively. Table 1 presents the values of activity concentrations obtained.

Radium equivalent activity

The significance of ^{40}K, ^{226}Ra and ^{232}Th concentrations, with respect to radiation exposure is expressed in terms of radium equivalent activity (Ra_{eq}), which was evaluated using Equation 2 (Beretka and Matthew, 1985). According to (OECD, 1979), the maximum value of Ra_{eq} must be less than 370 Bq.kg^{-1} for the radiological effect to be considered negligible.

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad (2)$$

where A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively. Equation 2 is based on the estimation that 1 Bq.kg^{-1} of ^{226}Ra, 0.7 Bq.kg^{-1} of ^{232}Th and 13 Bq.kg^{-1} of ^{40}Kgenerate the same gamma-ray dose rate (Siak et al., 2009).

External hazard index

External radiation hazard index (H_{ex}) is a widely used hazard index which reflects the external exposure level due to gamma radiation. It is estimated from the relation

Table 1. Activity concentrations of the samples.

Sample	^{40}K (Bq.kg^{-1})	^{238}U (Bq.kg^{-1})	^{232}Th (Bq.kg^{-1})	Mean (Bq.kg^{-1})
ZM.G1	561.32 ± 10.56	12.39 ± 2.36	94.92 ± 2.75	222.88 ± 5.22
ZM.G11	590.44 ± 10.56	18.26 ± 0.48	73.45 ± 1.61	227.38 ± 4.22
ZM.G12	528.66 ± 09.49	11.07 ± 2.20	52.06 ± 1.81	197.26 ± 5.50
ZM.H1	370.25 ± 10.94	07.87 ± 2.16	40.58 ± 1.85	139.57 ± 4.98
ZM.L1	425.86 ± 10.94	05.79 ± 1.28	52.81 ± 1.77	161.49 ± 4.66
ZM.L2	465.49 ± 09.33	14.35 ± 1.60	44.63 ± 2.91	174.82 ± 4.61
ZM.H2	338.29 ± 07.72	18.98 ± 0.84	54.22 ± 1.57	137.16 ± 3.38
ZM.L12	227.10 ± 07.54	17.41 ± 1.27	41.70 ± 0.67	095.40 ± 2.31
ZM.L11	244.97 ± 08.15	04.68 ± 3.52	56.50 ± 2.60	102.05 ± 4.75
ZM.G2	512.80 ± 11.45	10.42 ± 0.53	90.32 ± 2.25	204.51 ± 4.74

Table 2. Radium equivalent, external index, internal index and gamma index.

Sample ID	$Ra_{(eq)}$ (Bq.kg^{-1})	H_{ex}	H_{in}	I
ZM.G1	191.34 ± 7.11	0.517	0.550	0.703
ZM.G11	168.75 ± 3.60	0.456	0.505	0.625
ZM.G12	126.22 ± 5.51	0.341	0.371	0.473
ZM.H1	094.41 ± 5.64	0.255	0.276	0.353
ZM.L1	114.10 ± 4.65	0.308	0.324	0.425
ZM.L2	114.00 ± 6.48	0.308	0.347	0.426
ZM.H2	122.56 ± 3.68	0.331	0.382	0.447
ZM.L12	094.52 ± 0.26	0.255	0.302	0.342
ZM.L11	104.33 ± 7.85	0.282	0.294	0.380
ZM.G2	179.06 ± 4.63	0.483	0.512	0.657
Mean	130.93 ± 4.94	0.354	0.386	0.483

(UNSCEAR, 2000):

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

Internal hazard index

In addition to H_{ex}, the internal exposure to radiation is quantified by the internal hazard index (H_{in}) as defined by (UNSCEAR, 2000):

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad (4)$$

The representative level of gamma index is (I) was calculated using the following equation (UNSCEAR, 2000):

$$I = \frac{A_{Ra}}{300} + \frac{A_{Th}}{200} + \frac{A_K}{3000} \quad (5)$$

According to UNSCEAR (2000), for radiological effects to be considered negligible, the values of each of H_{ex}, H_{in} and I must be less than 1. Table 2 presents the calculated values of radiation indices H_{ex}, H_{in}, I, and Ra_{eq} for the entire samples.

Absorbed dose rates

The external absorbed dose rates were calculated using the equation (UNSCEAR, 2000):

$$D = 0.0417A_K + 0.462A_U + 0.604A_{Th} \quad (6)$$

where A_K, A_U and A_{Th} are the activity concentrations of ^{40}K, ^{238}U and ^{232}Th respectively in Bq.kg^{-1}. Table 2 presents the results of the absorbed dose rates calculated.

Annual effective dose

To estimate the annual effective dose, the conversion coefficient (0.7Sv/Gy) and outdoor occupancy factor (0.2) as proposed by (UNSCEAR, 2000) were used. Therefore, the annual effective dose were calculated from the relation (Harb et al., 2010):

Table 3. The absorbed dose and annual effective dose.

Sample ID	D (nGy.h^{-1})	E_d mean (mSv.y^{-1})	E_d range (mSv.y^{-1})
ZM.G1	86.46 ± 3.19	0.035	0.007 – 0.070
ZM.G11	77.42 ± 1.64	0.032	0.010 – 0.054
ZM.G12	58.60 ± 2.50	0.024	0.006 – 0.039
ZM.H1	43.59 ± 2.57	0.018	0.005 – 0.030
ZM.L1	52.33 ± 2.12	0.021	0.003 – 0.039
ZM.L2	52.99 ± 2.89	0.022	0.008 – 0.033
ZM.H2	55.62 ± 1.66	0.023	0.011 – 0.040
ZM.L12	42.70 ± 0.13	0.017	0.010 – 0.031
ZM.L11	46.50 ± 3.53	0.019	0.003 – 0.042
ZM.G2	80.75 ± 2.08	0.033	0.006 – 0.067
Mean	59.70 ± 2.23	**0.073**	

$$E_d = D(nGy.hr^{-1}) \, x \, 8760 \, (hr.y^{-1}) \, x \, 0.2 \, x \, (0.7 \, x \, 10^3 mSv) \, x \, (10^9 nGy)^{-1} \quad (7)$$

where E_d is the annual effective dose rate in $(mSv.y^{-1})$and D is the value of absorbed dose rate earlier calculated. Table 3 presents the calculated values of absorbed dose rate and annual effective dose for the entire samples.

RESULTS AND DISCUSSION

The mean activity concentration due to contribution from the three radionuclides per sample ranged from 95.40 ± 2.31 to 227.38 ± 4.22 Bqkg^{-1} with sample ZM.G1 leading the activity concentration table. The highest activity concentration of ^{238}U was found in sample ZM.H2 with 18.98 Bqkg^{-1}. Though, this value is low compare to the worldwide average of 33 Bqkg^{-1} (UNSCEAR, 2000). Table 1 shows that the highest activity concentration of ^{232}Th is found in soil sample ZM.G1, which could be due to presence of abundant radioactive thorium minerals such as monazite, zircon and thorianite (Okeyode and Akanni, 2009).

From Table 2, the values of the radiological hazard indices show that radium equivalent activity ranged from 94.41 ± 5.64 to 191.34 ± 7.11 Bq.kg^{-1} with mean value of 130.93 ± 4.94 Bq.kg^{-1}. The mean values of gamma index level, external and internal hazard indices are 0.48, 0.35 and 0.39, respectively. Furthermore, the mean absorbed dose rate of the samples was calculated to be 59.7 nGy.h^{-1} while the mean annual effective dose was estimated at 73 *μSvy*$^{-1}$ (Table 3).

CONCLUSION

The radiation hazard indices were estimated for soil samples collected from the abandoned mine sites. The result shows that all the hazard indices (H_{ex}, H_{in} and I) calculated have value less than unity. In conformity with the values of hazard indices, the values of annual effective dose ranged from 0.052 to 0.106 mSv.y^{-1}. Thus, the values of radiological hazard indices are within the safety limits.

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