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Evaluation of Naturally Occurring Radionuclide Materials in Soil Samples Collected From Some Mining Sites in Zamfara State, Nigeria

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Authors' contributions

This study was carried out in collaboration among all the authors. Author AJI collected and prepared the field samples, participated in the laboratory procedures, performed the statistical analysis and wrote the first draft of the manuscript. Author MYO designed the study and contributed to the statistical analysis. Author SAJ supervised the analyses of the study. All authors read and approved the final manuscript.

Research Article

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ABSTRACT

Aims: To identify and quantify prominent gamma emitting-NORMs associated with mining activities in Zamfara State, Nigeria.
Study Design: Soil samples were collected from 10 different mine sites in Zamfara State, northwest-Nigeria for gamma spectroscopy analysis.
Place and Duration of Study: Department of Physics, Nigerian Defence Academy Kaduna, Nigeria, between June 2012 and October 2012.
Methodology: A laboratory based γ-ray spectrometry NaI(Tl) at the Centre for Energy Research and Training (CERT), Ahmadu Bello University Zaria, Nigeria was employed to carry out the analysis of the soil samples.
Results: The values of activity concentration found for ^{40}K, ^{238}U and ^{232}Th range from 227.10±7.54 – 590.44±10.57, 4.68±3.52 – 18.98±0.84 and 40.58±1.85 – 94.92±2.75 Bq.kg^{-1} respectively, while absorbed dose rate in the soil samples for ^{40}K, ^{238}U and ^{232}Th range from 9.47±0.31 – 24.62±0.44, 2.16±1.62 – 8.77±0.39 and 24.51±1.12 – 57.33±1.66 nGy.h^{-1} respectively. The total average absorbed dose rate of the soil samples collected is 59.70 nGy.h^{-1} and the estimated annual effective dose for the study areas range from 52 – 106 μSv.y^{-1}, with an average annual effective dose of 73 μSv.y^{-1}.

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Conclusion: The radiation exposure level for members of the public in the study areas is within the safety limit.

Keywords: Radionuclides; soil; mining; activity concentration; absorbed dose; Zamfara.

1. INTRODUCTION

The world is naturally radioactive and about 90% of human radiation exposures arise from natural sources such as cosmic radiations, exposure to radon gas, and terrestrial radiations. Significant naturally occurring radionuclides present in the soil are ^{238}U, ^{232}Th and ^{40}K [4]. Since these radionuclides are not uniformly distributed, the knowledge of their distribution in soils and rocks play an important role in radiation protection and measurement. Some of the exposures are fairly constant and uniform for all individual persons everywhere, for example, the dose acquire from ingestion of ^{40}K in foods. However, other exposures vary widely depending on location. Cosmic rays, for example, are more intense at higher altitudes and concentrations of uranium and thorium in soils are elevated in localized areas. High levels of uranium and its decay products in rock and soil, and thorium in monazite sands are the main sources of high natural background radiations that have been identified in several areas of the world [13].

Radiation exposures arise in the mining and mineral processing industries through three principal pathways. These are external gamma radiation from ores, inhalation of dusts containing long-lived alpha-emitting radionuclides and inhalation of the short-lived decay products of radon. Inhalation of radon decay products in poorly ventilated underground mines can lead to exposures in excess of current radiation exposure limits, and this could cause high incidence of lung cancer in the mine workers. There is consequently a need to adopt careful radiological control measures in mining and mineral processing operations involving radioactive ores, in order to protect those involved and to meet dose limits.

Exposure of workers to naturally occurring radioactive material (NORM) continued to be an emerging industrial issue, for example in the extraction of rare earths, the zircon and zirconium industries, coal-fired electricity generation and the phosphate industry [5]. According to United Nations Scientific Committee on the Effects of Atomic Radiation, about 13 million workers are exposed to natural sources of radiation [14]. Historically, such industries have not always been subjected to the same strict regulation regime as is the case with artificial sources. Environmental problems associated with NORM in solid minerals mines and processing occur in the process of drilling, leaching, handling, storage, transportation of mineral ores and the use of contaminated equipment or waste media without controls. These usually lead to the spread of NORM contaminating the environment, resulting in potential radiation exposure of members of the public. An example of negative effect of contaminated environment on humans, as a result of mining activities was that reported by [12]. According to the source, in March 2010, an unusual high number of deaths, primarily among children under age 5 in Bukkuyum and Anka local government areas of Zamfara State, north-western Nigeria, was reported by Médecins Sans Frontières (MSF-Holland) to state health authorities. Further study on blood samples taken by MSF revealed that the increased mortality was the result of acute lead poisoning, determined to be caused by massive environmental contamination from artisanal mining and processing of gold found in lead-rich ore. The grinding of the ore into fine particles by local miners resulted in extensive dispersal of lead dust in the villages, including their compounds. Ingestion and inhalation of the fine lead particles was determined to be the major reason for high blood

lead levels in victims' bodies. Blood lead levels were found to be unprecedented for human being.

Since radiation is neither visible nor can it be felt by the human sense organs, it is important that the total amount of radiation emitting-NORMs in the area is accurately known and kept to a level as low as reasonably achievable in order to safeguard the live of the people and ensure radiation-pollution free environment. Therefore, this study seeks to provide the public with the knowledge of NORM level associated with solid mineral mining activities in Zamfara, with the hope that results of the study will serve as baseline for future assessment of gamma emitting-NORMs level in the study area. The main objective of this study is to identify and quantify prominent gamma emitting-NORMs associated with mining activities in Zamfara State, Nigeria using gamma spectroscopy system at Centre for Energy Research and Training, Ahmadu Bello University, Zaria Nigeria.

The results obtained in this study might compliment the data base of some related studies carried out in other locations within Nigeria. Such studies are determination of terrestrial gamma dose rates and physical-chemical properties of farm soils from ex-tin mining locations in Jos [7]; Characterization of distribution of gamma-emitting radionuclides in soils around the Centre for Energy Research and Training (CERT) Ahmadu Bello University, Zaria [9]; 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 [3]; Characterization of distribution of some natural gamma emitting radionuclides in the soils of the coastal areas of Nigeria [2]; Estimation of natural radionuclides and elemental composition of chemical fertilizers used in Nigeria [8]; baseline measurement of natural radioactivity in soil, vegetation and water in the industrial district of the Federal Capital Territory (FCT) Abuja, Nigeria [11]; Measurement of activity concentration of ^{40}K, ^{226}Ra and ^{232}Th for assessment of radiation hazards from soils of southern region of Nigeria [1].

2. MATERIALS AND METHODS

2.1 Sample Collection and Preparation

The study site is located in Zamfara State, northwest Nigeria between $006^{0}30'11"$ – $007^{0}00'03"$ E of the longitude and $12^{0}39'18"$ – $12^{0}46'01"$ N of the latitude. The types of soil samples used for the investigation were soils samples collected from solid mineral processing site at Danba-Gusau and mine sites at other locations within Zamfara state, Nigeria. 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.

In the laboratory, the soil samples were put in an oven and set to a temperature of 105ºC to allow for drying overnight in order to remove any available moisture. After drying, the samples were crushed and sieved with a mesh having holes each of diameter of 2 mm in order to remove organic materials, stones and lumps. Afterwards, the homogenized samples were packed to fill cylindrical plastic beakers of height 7cm by 6cm diameter which is the same as geometry of the counting detector. This satisfies the selected optimal sample container height [6]. The samples were carefully sealed using vaseline, candle wax and masking tape in order to prevent trapped radon gas from escape. They were weighed on a digital weighing balance with a precision of ± 0.01g, each plastic beaker accommodated

approximately 300g of the soil sample. 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 [10] before gamma counting.

2.2 The Experimental Set-Up and Procedure

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 [11]. In addition, a computer based Multichannel Analyser (MCA) Maestro Programme from ORTEC was used for the data acquisition and analysis of gamma spectra.

The soil samples were placed on the detector surface and each counted for a lifetime of about 29,000 seconds (8 hrs, 3mins) 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 1764 KeV γ-line of ^{214}Bi for ^{226}Ra was used in the assessment of the activity concentration of ^{238}U while 2614.5 KeV γ-line of ^{208}Tl was used for investigating the activity concentration of ^{232}Th. The single 1460 KeV γ-line of ^{40}K was used in its content evaluation.

3. RESULTS AND DISCUSSION

3.1 Net Count per Second (cps)

The gross area count G_c is related to the net area count N_c by [10]:

$$N_c = G_c - B_c \quad (1)$$

Where B_c is the background area count, (the area count recorded by the detector in the absence of samples). Table 1 presents the background area counts corresponding to ^{40}K, ^{238}U and ^{232}Th as obtained from the measurements.

Table 1. Area under the photo peaks due to background count

Nuclide	Bgrd count (B_c)	Stat. error
^{40}K	3816	±338
^{238}U	902	±307
^{232}Th	3103	±227

Using equation (1), the net area counts N_c was calculated from the gross area counts G_c generated by the gamma spectroscopy system. Consequently, the net count per second (cps) was also calculated for all the three radionuclides.

3.2 Activity Concentrations

The activity concentrations for the natural radionuclides in the measured samples were computed using the following relation [10].

$$A_c = \frac{N_c}{L_t}\sigma^{-1} \quad (2)$$

Where L_t is the lifetime of counting. And σ is a conversion factor, it is constant for each radionuclide at constant geometry and it is a characteristic of efficiency of the NaI(Tl) detector assembly used. Table 2 presents the values of the conversion factor (σ) for ^{40}K, ^{238}U and ^{232}Th [11] used for converting the raw data into conventional unit data.

Table 2. Conversion constants and gamma ray line for each nuclide

Nuclide	Cps /Bq.kg^{-1})	Gamma ray line (KeV)
^{40}K	0.000643	1460
^{238}U	0.000863	1764
^{232}Th	0.000877	2614.5

All the raw data obtained from the detector were converted to conventional units using calibration factors to determine the activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th respectively. Using equation (2), the activity concentrations were calculated and the results are as presented in Table 3 and Fig. 1.

Table 3. Activity concentrations of the soil samples

	Activity concentrations (Bq.kg^{-1})			
Sample ID	^{40}K	^{238}U	^{232}Th	Total
ZM.G1	561.32 ± 10.57	12.39 ± 2.36	94.916 ± 2.75	668.63±15.67
ZM.G11	590.44 ± 10.57	18.26 ± 0.48	73.448 ± 1.61	682.15±12.66
ZM.G12	528.66 ± 09.49	11.07 ± 2.20	52.058 ± 1.81	591.79±13.50
ZM.H1	370.25 ± 10.94	07.87 ± 2.16	40.577 ± 1.85	418.70±14.95
ZM.L1	425.86 ± 10.94	05.79 ± 1.28	52.805 ± 1.77	484.46±13.99
ZM.L2	465.49 ± 09.33	14.35 ± 1.60	44.627 ± 2.91	524.46±13.84
ZM.H2	338.29 ± 07.72	18.98 ± 0.84	54.221 ± 1.57	411.49±10.13
ZM.L12	227.10 ± 07.54	17.41 ± -1.27	41.698 ± 0.67	286.20±06.94
ZM.L11	244.97 ± 08.15	04.68 ± 3.52	56.501 ± 2.60	306.15±14.26
ZM.G2	512.80 ± 11.45	10.42 ± 0.53	90.315 ± 2.25	613.54±14.23
Mean	426.51 ± 9.67	12.12 ± 1.37	60.117 ± 1.98	498.76±13.02

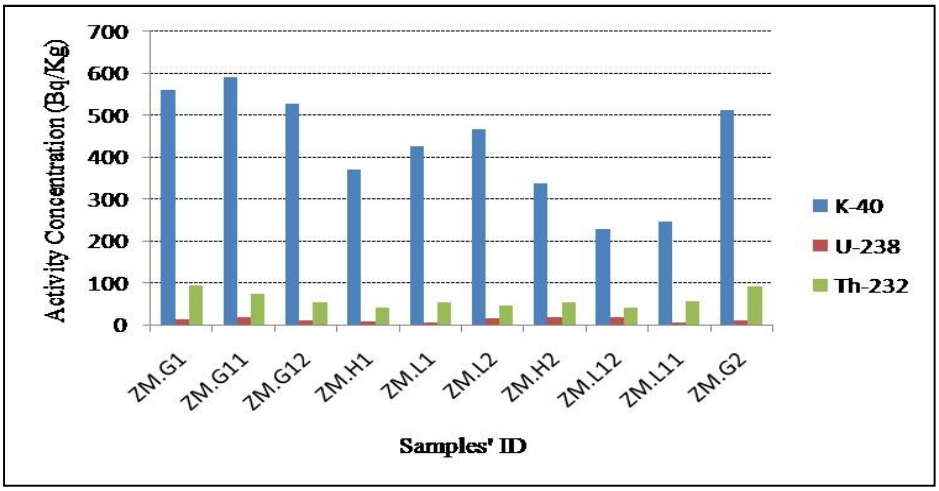

Fig. 1. Activity concentration for the samples

3.3 Absorbed Dose Rates (D)

The external absorbed dose rate, $D(nGy.h^{-1})$ in air at 1 m above the ground level due to activity concentrations of ^{40}K, ^{238}U and ^{232}Th for the 10 samples were calculated using equation (3) [13]:

$$D(nGy.h^{-1}) = 0.0417A_K + 0.462A_U + 0.604A_{Th} \quad (3)$$

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}.

The conversion factors 0.0417, 0.462, 0.604 are expressed in $nGy.h^{-1}/Bq.kg^{-1}$. Table 4 presents the results of the absorbed dose rates calculated. The absorbed dose rates in air are usually related to human absorbed dose in order to assess radiological implications.

Table 4. Absorbed dose rates of the soil samples

Sample ID	^{40}K ($nGy.hr^{-1}$)	^{238}U ($nGy.hr^{-1}$)	^{232}Th ($nGy.hr^{-1}$)	Total ($nGy.hr^{-1}$)
ZM.G1	23.41 ± 0.44	5.72 ± 1.09	57.33 ± 1.66	86.46 ± 3.19
ZM.G11	24.62 ± 0.44	8.44 ± 0.22	44.36 ± 0.79	77.42 ± 1.64
ZM.G12	22.05 ± 0.40	5.12 ± 0.02	31.44 ± 1.09	58.60 ± 2.50
ZM.H1	15.44 ± 0.46	3.64 ± 1.00	24.51 ± 1.12	43.59 ± 2.57
ZM.L1	17.76 ± 0.46	2.68 ± 0.59	31.89 ± 1.07	52.33 ± 2.12
ZM.L2	19.41 ± 0.39	6.63 ± 0.74	26.96 ± 1.76	52.99 ± 2.89
ZM.H2	14.11 ± 0.32	8.77 ± 0.39	32.75 ± 0.95	55.62 ± 1.66
ZM.L12	09.47 ± 0.31	8.04 ± -0.59	25.19 ± 0.40	42.70 ± 0.13
ZM.L11	10.22 ± 0.34	2.16 ± 1.62	34.13 ± 1.57	46.50 ± 3.53
ZM.G2	21.38 ± 0.48	4.82 ± 0.24	54.55 ± 1.36	80.75 ± 2.08
Total	177.86 ± 4.03	56.00 ± 6.32	363.11 ± 11.95	596.96 ± 22.30
Mean	17.79 ± 0.40	5.60 ± 0.63	36.31 ± 1.20	59.70 ± 2.23

3.4 Annual Effective Dose (E_d)

To estimate the annual effective dose rates, the conversion coefficient from absorbed dose in air to effective dose (0.7Sv.Gy^{-1}) and outdoor occupancy factor (0.2) proposed by [13] were used. Therefore, the annual effective dose rates (mSv.yr^{-1}) were calculated from the following formula [4]:

$$E_d = D(nGy.hr^{-1})\ x\ 8760\ (hr.y^{-1})\ x\ 0.2\ x\ (0.7\ x\ 10^3 mSv)\ x\ (10^9 nGy)^{-1} \quad (4)$$

Equation (4) simplifies into

$$E_d = D\ x\ 1.2264\ x\ 10^{-3} \quad (5)$$

where E_d is the annual effective dose rate in $(mSv.y^{-1})$ and D is the value of absorbed dose rate earlier calculated from equation (3). Table 5 presents the calculated annual effective dose for the entire samples.

Table 5. Annual effective dose rates

Sample ID	^{40}K ($\frac{}{mSv.y^{-1}}$)	^{238}U ($\frac{}{mSv.y^{-1}}$)	^{232}Th ($\frac{}{mSv.y^{-1}}$)	Total ($\frac{}{mSv.y^{-1}}$)
ZM.G1	0.029	0.0070	0.070	0.106
ZM.G11	0.030	0.0103	0.054	0.095
ZM.G12	0.027	0.0063	0.039	0.072
ZM.H1	0.019	0.0045	0.030	0.053
ZM.L1	0.022	0.0033	0.039	0.064
ZM.L2	0.024	0.0081	0.033	0.065
ZM.H2	0.017	0.0108	0.040	0.068
ZM.L12	0.012	0.0099	0.031	0.052
ZM.L11	0.013	0.0026	0.042	0.057
ZM.G2	0.026	0.0059	0.067	0.099
Total	0.220	0.0687	0.445	0.732
Mean	0.022	0.0069	0.045	0.073

3.5 Discussion

The highest radioactivity concentration of ^{40}K was found in soil sample ZM.G11 (Table 3, Fig. 1). This high value could be due to presence of abundant radioactive minerals such as smectite, kaolinite and feldspars in the sample. The least radioactivity concentration of ^{40}K was found in soil sample ZM.L12. The highest radioactive concentration of ^{238}U was found in soil sample ZM.H2, it could be due to high presence of uranium minerals such as uraninite, zircon, monazite and so on. Soil sample ZM.L11 had the lowest radioactivity concentration of ^{238}U. Also, Table 3 shows that the highest radioactive 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 [10]. The smallest radioactive concentration of ^{232}Th was also found in soil sample ZM.L12. As expected, the radioactivity concentrations due to ^{40}K is highest compared with ^{238}U and ^{232}Th.

From Table 4, it was shown that the absorbed dose rate due to the three radionuclides is highest for soil sample ZM.G1. This might be due to accumulation of mineral sands from different mine sites at the processing plant. According to Table 4, ^{232}Th had the highest

value of total absorbed dose rate among the three radionuclides detected in the soil samples collected, thus it had the highest dose level in the study areas while ^{238}U had the least total of 56.00 $nGy.h^{-1}$.

4. CONCLUSION

The method of gamma spectrometry had been used to measure the radioactivity concentration of 10 soil samples collected from mine sites in Zamfara State, Nigeria. The result shows that the total concentration of ^{232}Th is much higher than that of ^{238}U by approximately a factor of 5 while ^{40}K leads the table of radioactivity concentrations.

The annual effective dose rates in air at the study area range from 0.052 - 0.106 $mSv.y^{-1}$. Thus, the exposure level for the members of general public is within the recommended value of 1 $mSv.y^{-1}$ [13]. Therefore, this is an indication that the mining activities in the study areas do not appear to have any impact on the radiation burden of the environment. However, it is recommended that proper radiation monitoring exercise be conducted on the processing site (ZM.G1) from time to time in other to checkmate the possible rise in radiation level due to accumulation of mineral sands from various mine sites.

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COMPETING INTERESTS

There is no competing interest whatsoever that could have influenced the results of this study in any manner.

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