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Radiological Assay of Technologically Enhanced Naturally Occurring Radionuclides and Hazard Assessment in Soil Samples from Selected Towns in Kogi state, Nigeria

S. T. Yinusa[1,], F. S. Olise[1], S. T. Gbenu[2], M. I. Arowojolu[3], M. K. Ajekiigbe[1], S. A. Adeojo[1], H. B. Olaniyi[1]*

[1]Department of Physics and Engineering Physics, ObafemiAwolowo University, Ile – Ife, Nigeria
[2]Centre for Energy Research and Development (CERD), Obafemi Awolowo University, Ile – Ife, Nigeria.
[3]Department of Chemistry, University of Agriculture, Markudi, Benue State. Nigeria.

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Abstract: Extraction of certain minerals from the earth crust results in technologically enhanced naturally occurring radionuclides, which may pose danger to immediate environment. The activity concentrations of naturally occurring radionuclides ^{40}K, ^{238}U and ^{232}Th in soil samples from selected towns in Kogi State were measured using gamma spectrometry with NaI (Tl) detector. This was with a view to determine the radiological hazards due to these natural radionuclides. The total average activity concentrations of ^{40}K, ^{238}U and ^{232}Th determined in the study area were 974.7±6.8, 74.3±5.0 and 110.3±24.8 Bq/kg, respectively. The mean annual effective dose, the mean radium equivalent activity concentration, the mean external and internal hazard indices in the study area were less than the permissible limit set by international community. Therefore, mining activities in selected towns in Kogi State poses no radiological hazard to both the miners and populace of the study area.

Keywords: Radionuclides, Gamma Spectrometry, mining, Effective Dose.

1 Introduction

The so called "naturally occurring radioactive materials'' (NORM) refers to all the radionuclides that exist in the environment naturally. They are the primordial radionuclides ^{40}K, ^{232}Th and ^{238}U, and their decay products which are present within the earth crust.

Man is exposed to radiation from either natural or anthropogenic radioactive sources [1]. The natural sources of radiation are of terrestrial and cosmogenic origin. Terrestrial radiation is due to radioactive nuclides, ^{40}K, ^{232}Th and ^{238}U, present in varying amounts in the environment while cosmogenic radiation originates from the outer space as primary cosmic rays, [1]. Anthropogenic radioactive sources are from nuclear reactors and other radiation generators used for medical and industrial activities, [2].

The concentration of NORM in any substance in the environment is negligible in most of the cases and they are present everywhere. But the extraction of certain minerals from the earth crust results in the radionuclides become elevated. High level of NORM may be detected at a particular location due to the area's geological formation or if a radioactive hot spring flows through the area, [3].

Some human activities have led to increase in the relative concentration of radionuclides, for example mining. Higher exposures arise from such human activities because they involve extraction and disposal of large quantities of mineral containing ^{40}K and other radionuclides in the decay series of ^{232}Th and ^{238}U. Miners may be exposed to radiation during extraction, transportation and processing of the mineral ores, [2]. They may experience internal exposures from radon, and its short-lived decay products that are airborne or ingestible dust from their surroundings, [4].

Kogi state is located in north central Nigeria and it is known as one of the Nigeria's home of solid minerals. It is endowed with iron–ore, tin, tantalite, gold, coal and granite. Presence of these resources makes it to have hundreds of mining sites; mining is the second major activity of the

*Corresponding author e-mail: profftee85@gmail.com

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inhabitants apart from farming. With a lot of mining activities taking place within the state, the people of the state are likely to face radiation exposure risk due to the presence of naturally occurring radioactive materials (NORM) in the earth and in the mining by – products, and wastes derived from mining operations.

Environmental samples such as water, soil, vegetable and rock are materials found everywhere in man's environment, and as such constitute direct and indirect pathways to radiation exposure of man. The numerous uses of these materials further bring them, along the radionuclides they contain, in contact with man, both externally and internally (by ingestion, absorption, inhalation and injection). Thus a precise measurement of the radionuclides concentrations and subsequent calculation of certain radiologically important parameters are imperative to radiation protection of man against harmful effects of ionizing radiation exposure.

Several works have been reported on assessment of radioactivity concentrations in some mining areas in Nigeria, [5, 6, 7, 8]. However, studies in the area of natural radionuclides in the soils of North – Central Nigeria have been scanty. In order to contribute to this area, a number of radiological indices were determined based on samples taken from the mining field under consideration. The data yield may be used to assess the health effect on the populace and serve as a basis for further studies. It will also provide useful information for national and local authorities for decision making.

2 Experimental Sections

2.1 The Study Area

The study was carried out in six different towns in Kogi state where mining activity were prominent. Kogi state is located in the middle – belt along the Niger-Benue river basin. The state shares boundary with up to ten other states. It has two geological formation, basement complex and sedimentary rock. Half of the state is covered by crystalline basement complex while the other is covered by cretaceous to recent sediments, [9]. Nigeria basement and ancient hard rocks predominantly underline the western flank of the state. The eastern flank of the state is on the alluvium (young and most recent sedimentary rock). It coordinate are 7°45'0" N and 6°45'0" E. The towns where samples were collected include Odo – Ara, Egbe, Odo – Ere, Igbaruku, Okoloke and Ankpa, all in Kogi state.

2.2 Sample Collection and Preparation

A total of thirty 30 soil samples were collected from mining sites around six towns where mining activities were prominent in Kogi State. Five soil samples were collected from mining sites around each of the following towns; Odo – Ara, Egbe, Odo – Ere, Igbaruku, Okoloke and Ankpa, all in Kogi State. Tin, monazite and Tantalite are the major minerals mines in the selected towns. The soil samples were collected at a depth of about 0 – 15 cm using auger. Samples were put in separate polythene bag to avoid cross contamination. The collected samples were air dried; ground sieved using 2 mm mesh. Sieved samples were weighed and a mass of 200 g of each sample were placed in a non-radioactive plastic container and sealed with adhesive tape [10] for 30 days so as to achieve secular equilibrium.

2.3 Activity determination

The soil samples were analyzed using a 3" x 3" inch NaI (Tl) detector crystal optically coupled to a photomultiplier Tube (PMT). The assembly has an incorporate preamplifier and 0.7kVolt external source. The detector is enclosed in a 5mm thick lead shield. The stated arrangement is made in order to minimize the effects of background and scattered radiation. The sample were measured for a period of 25200 sec after which the net area under the corresponding gamma – ray peaks in the energy spectrum were used to compute the activity concentrations in the sample

The transition lines of 1460 keVof ^{40}K, 1120 keV of ^{214}Bi and 911 keV of ^{228}Ac were used to determine the concentration of ^{40}K, ^{238}U and ^{232}Th respectively. The detection limits of the NaI (Tl) detector system were calculated as 6.77, 11.40, and 12.85 Bq kg^{-1} for ^{40}K, ^{232}Th, and ^{238}U, respectively.

The specific activity (A_s) measured in Bq/kg of the soil were determined using the net area under the photo peak and calculated according to equation (1)

$$A_s = {C_n}/{\varepsilon_y M_s I_y} \quad (1)$$

Where,

A_s is the specific activity of radionuclide in the sample specified in Bq/kg,

C_n is the net count per second of the sample under the corresponding photo peak,

ε_y is the efficiency of the detector at the specific gamma ray energy of interest and

I_y is the intensity of gamma ray at a particular energy being counted,

M_s is the mass of the sample (kg).

2.4 Absorbed Dose Rate

In order to assess the radiation hazards due to the concentrations of natural occurring radionuclides ^{226}Ra, ^{232}Th and ^{40}K in the samples, the absorbed dose rate in air at 1m above the ground were calculated using equation (2) given by [11]

$$D = 0.662A_{Th} + 0.427A_{Ra} + 0.0432A_K \left({nGy}/{h}\right) \quad (2)$$

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Where; D is the absorbed dose rate in air 1m above the ground. A_{Ra}, A_{Th} and A_K are the specific activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K measured in Bq/kg. While 0.0427, 0.662, 0.043 are the conversion factor for ^{226}Ra, ^{232}Th and ^{40}K respectively.

2.5 Radium Equivalent

The output of gamma rays from a mixture of ^{226}Ra, ^{232}Th and ^{40}K in the sample is called Radium Equivalent Dose. This value measures in Bq/kg and it can be calculated by the equation (3)

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \qquad (3)$$

Where; A_{Ra} is the Activity of ^{226}Ra,A_{Th} is the Activity of ^{232}Th and A_K is the Activity of ^{40}K (all in Bq/kg) [11].

2.6 External and Internal Hazard Index

The External hazard index (H_{ex}) was used to evaluate a potential hazard which is associated with non – radiological and radiological effects. External hazard index was calculated using the equation given by [12].

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_k}{4810} \qquad (4)$$

Where; A_{Ra}, A_{Th} and A_K are the activities in Bq/kg of ^{226}Ra, ^{232}Th and ^{40}K. [13], submitted that the value of the external value index must be less than one in order for radiation hazard to be considered acceptable to the public.

2.7 Internal Hazard Index

This term quantified the internal exposure to radon and its daughter product, which is given by the equation:

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_k}{4810} \qquad (5)$$

H_{in} should be less than one for the radiation hazard to be insignificant [13].

2.8 Annual effective dose equivalent (AEDE)

The annual effective dose rate from outdoor gamma radiation can be estimated by taking into account the conversion coefficient from the absorbed dose in air to the effective dose (0.7Sv/Gy) and 0.3 was used as outdoor occupancy factor instead of 0.2 proposed by [11]. Outdoor occupancy factor depends on the living style of the people which is not the same in rural and urban area [14]. Since the selected towns are rural area 0.3 which represent about 8hours out of 24hours of the day was used as outdoor occupancy factor.

$$AEDE\left(\frac{mSv}{y}\right) = D\left(\frac{nGy}{h}\right) x\, 8766\left(\frac{h}{y}\right) x\, 0.3\, x\, 0.7\left(\frac{Sv}{Gy}\right) x\, 10^{-6} \quad (6)$$

3 Results and Discussion

3.1 Activty Concentrations

The value of specific activities of naturally occurring radionuclides detected in the thirty (30) soil samples collected from the six (6) selected towns in Kogi State are presented in Table 1.

All radionuclides detected and quantified came from the naturally occurring ^{238}U and ^{232}Th decay series except ^{40}K. ^{238}U has a specific activity range of (48.9±4.1 – 124.8±8.4) Bq/kg with the lowest value in Odo Ara and highest in Egbe respectively. The highest activity concentration of ^{232}Th was detected in Egbe (316.8±63.8 Bq/kg) and the highest activity concentration of ^{40}K (2191.6±12.5) Bq/kg was detected in Egbe. From Table 1, the mean value of the result for each selected towns revealed that Egbe has the highest mean activity of (1493.2±9.2) Bq/kg, (91.7±4.0) Bq/kg, (304.0±61.3) Bq/kg. The highest activity of ^{40}K may be attributed to the fact that the study area are moving towards the guinea savanna, there is need to enhance the soil nutrient by the use of inorganic fertilizer. Generally, the activity concentration of thorium and uranium in the sample are higher than maximum permissible activity concentration of 30 and 35 Bq/kg for the general public [11].

Table 1 – Activity concentrations of radionuclides in the six selected towns

Town	K-40 (Bqkg^{-1})	U-238 (Bqkg^{-1})	Th-232 (Bqkg^{-1})
Ankpa	980.2 ± 7.0	72.2±5.4	50.7±13.5
Egbe	1493.2±9.2	91.7± 4.0	304.0 ± 61.3
OdoAra	871.6 ± 6.3	61.8± 4.8	83.8 ± 20.0
Odo Ere	886.1±6.1	85.8± 6.1	89.5 ± 20.9
Okoloke	800.2±6.0	58.6±4.0	61.6±15.6
Igbaruku	816.7±5.8	75.9±5.9	72.3±17.7
Average	974.7±6.8	74.3±5.0	110.3±24.8

3.2 Radiation dose and hazard index

The absorbed dose rate was calculated and presented in Table 2. The annual effective dose and radium equivalent activity was calculated and their average in each selected towns is presented in Table 2. Table 3 presents the calculated radiation hazards in the representative soil samples. Using equation 5 and 6, external hazard index and internal hazard index were calculated and their averages were equally presented in Table 3.

The average absorbed dose delivered by these radionuclides range from 66.4 ± 10.3 in Okoloke to 375.6 ± 46.4 nGy/h in Egbe, with an average value of 146.9 ± 18.9 nGy/h. The average value obtained is higher than the world average of

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51 nGy/h [11]. The annual effective dose for the study area due to soil activity was estimated and the average annual effective dose range from 0.2 to 0.6 mSv/y, with total average of 0.3 mSv/y. This value is less than 1mSv/y the acceptable limit for the general public.

Table 2 – Mean values of Absorbed Dose, Annual Effective Dose, Radium Equivalent.

Town	Absorbed dose rate (nGy/h)	Annual Effective Dose (mSv/y)	Radium Equivalent (Bq/kg)
Ankpa	106.5 ± 11.5	0.2	220.3 ± 25.2
Egbe	304.9 ± 42.7	0.6	641.5 ± 92.3
OdoAra	119.5 ± 15.6	0.2	248.8 ± 33.9
Odo Ere	134.2 ± 16.8	0.3	282.0 ± 36.6
Okoloke	100.4 ± 12.3	0.2	208.4 ± 26.7
Igbaruku	115.6 ± 14.3	0.2	242.2 ± 31.3
Average	146.9 ± 18.9	0.3	307.2 ± 41.0

Table 3 - Mean values of External and Internal Hazard Indices.

Town	External Hazard Index	Internal Hazard Index
Ankpa	0.6 ± 0.1	0.8 ± 0.1
Egbe	1.7 ± 0.3	2.0 ± 0.3
OdoAra	0.7 ± 0.1	0.8 ± 0.1
Odo Ere	0.8 ± 0.1	1.0 ± 0.1
Okoloke	0.6 ± 0.1	0.7 ± 0.1
Igbaruku	0.7 ± 0.1	0.9 ± 0.1
Average	0.9 ± 0.1	1.0 ± 0.1

The average radium equivalent activity ranged from 208.4 ± 26.7 in Okoloke to 641.5 ± 92.3 Bq/kg in Egbe, with total mean radium equivalent activity of 307.2 ± 41.0, which is lower than the recommended safety limit of 370 Bq/kg [11].

The average internal hazard index ranged from 0.7 ± 0.1 in Okoloke to 2.0 ± 0.3 in Egbe, with total average internal hazard index of 1.0 ± 0.1. The obtained value is within the unity recommended by [11]. This indicates that the soils in the study areas are save for either building or agricultural purpose. The average external hazard index ranged from 0.6 ± 0.1 to 1.7 ± 0.3, with total average of 0.9 ± 0.1 which is lower than the unity.

3.3 Comparison of the mean activity with those obtained from other country

A Comparison of the Mean activity concentrations values obtained in this work with values obtained in other part of the country and the world average were presented in Table 4. Lower activity concentration was determined by [7, 15, 6] in Nigeria. [16] in Ghana and [17] in Egypt for ^{238}U compared to this study but lower than the concentration obtained in China [18], in Turkey [19] and in Nigeria [8]. The average activity concentration of ^{238}U estimated in this study is higher than the world average [11].

The average activity concentration of ^{232}Th obtained in this study is higher than that obtained by [7, 15, 6] in Nigeria. [16] in Ghana, [17] in Egypt and [18] in China but lower than that obtained by [8] in Nigeria and [19] in Turkey. The average activity concentration of ^{232}Th is higher than the world average [11]. In line with what was obtained from other studies as shown in Table 4, the activities concentrations of ^{40}K in soil samples are significantly higher than the concentration of ^{238}U and ^{232}Th. The average activity concentration of ^{40}K in this study is higher than the world average.

Table 4- Comparison of the activity concentrations obtained with those obtained from other country

Reference	Country	^{238}U (Bqkg^{-1})	^{232}Th (Bqkg^{-1})	^{40}K (Bqkg^{-1})
[15]	Nigeria	32.52	56.32	403.96
[6]	Nigeria	12.1	60.1	426.5
[8]	Nigeria	512.2	2635.78	-
[7]	Nigeria	39.8	17.7	384.2
[16]	Ghana	13.6	24.2	162.1
[17]	Egypt	22.12	10.27	180.04
[18]	China	112	71.5	-
[19]	Turkey	115	192	-
Present study	Kogi	74.3 ± 5.0	110.3 ± 24.8	974.7 ± 6.8
UNSCEAR, 2000	World Average	35	30	400

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4 Conclusion

The activity concentration, radium equivalent activity, gamma dose rate, annual effective dose, internal and external hazard indices in soil samples from selected towns in Kogi State have been carried out using gamma ray spectroscopy with NaI(Tl) detector. The obtained values are compared to the world average. The result of the mean activity concentrations of ^{238}U, ^{232}Th and ^{40}K are higher than the world average. The annual effective dose in the soil samples from selected towns was less than 1 mSv/y, the acceptable limit for the member of the public.

It can be concluded that there is no potential danger to the health of both miners and populace of the selected towns in Kogi State due to radiological hazards. Hence, this is an indication that the mining activities in selected towns appear to have low impact on the radiation burden of the environment. The result of this study may serve as basis for further studies and the data obtained in this study may provide useful information for local and international authorities for reference.

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