Original Article

Natural radionuclides in rock and radiation exposure index from uranium mine sites in parts of Northern Nigeria

Samuel Odumu Ogana John[1], Iyabo T. Usman[2], Timothy C. Akpa[1,3], Sadiq Aliyu Abubakar[4], Godwin B. Ekong[1,3]

[1]Department of Physics, Nasarawa State University, Keffi, [4]Department of Physics, Federal University of Lafia, Lafia, Nasarawa State, [3]Radiological Safety Department, Nigerian Nuclear Regulatory Authority, Abuja, Nigeria, [2]Nuclear Structure Research Group (NSRG), School of Physics, University of the Witwatersrand, Johannesburg, South Africa

Abstract

Radiation exposure in humans can emanate from natural radionuclides through uranium and thorium decay series as well as ^{40}K due to emitted ionising radiations. It is important to estimate the exposure of humans to the diverse sources of radiation. In this study, the activity concentrations of natural radionuclides ^{238}U and ^{232}Th in granitic rock samples from uranium mine sites in parts of Northern Nigeria were measured, by inductively coupled plasma-mass spectrometry. The highest values of ^{238}U and ^{232}Th concentrations (924.56 ± 17.13 and 21.96 ± 1.28 Bq/kg, respectively) were observed at Mika-I and Riruwai. Furthermore, the radiological exposure parameters (i.e., absorbed dose rate in air [D], annual effective dose [external], radium equivalent activity (Ra_{eq}), external exposure index (H_{ex}), internal exposure index (H_{in}), and representative level index (I_{γ} and I_{α}) were estimated and compared to the international recommended values. In terms of terrestrial gamma radiation from granitic rock within the study area, it does not pose any significant radiation exposure to the workers and dwellers.

Keywords: Absorbed dose, inductively coupled plasma-mass spectrometry, radioactivity, radiological exposure, uranium ore

Address for correspondence: Dr. Samuel Odumu Ogana John, Department of Physics, Nasarawa State University, PMB 1022, Keffi, Nigeria.
E-mail: samjoh2014@gmail.com

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INTRODUCTION

In the universe, persons or individuals are exposed directly or indirectly to ionizing radiation. The sources of radiation exposure are the progenies of parent primordial nuclide decay series of uranium-238 (^{238}U) and thorium-232 (^{232}Th). Others include cosmic rays and artificial radioactivity from fallout in nuclear testing and industries as well as medical applications.[1,2]

Higher concentration and higher radiation levels are associated with igneous rocks, such as granite, and lower levels with sedimentary rocks. There are exceptions, however, as some shale and phosphate rocks have relatively high content of radionuclides.[3] Granitic rocks contain natural radioactivity which includes ^{238}U, ^{232}Th, and ^{40}K at various concentrations and contribute significantly to total radiation dose. The radiation dose emanates from gamma rays that are from the rocks, soils, and some building materials, and primarily depends on the geological and geographical conditions of the environment.[1,4] The concentrations of radionuclides in the rock, soil, or water–air; the time spent

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outdoors; and the shielding by buildings, have been the main factors that determine the exposure rate to a given person. It was estimated by the International Atomic Energy Agency (IAEA) that natural radionuclides contributed 80%, whereas 20% was from cosmic rays and nuclear processes, of the doses to the environment.[5,6]

Uranium is formed deep within the earth's surface through a series of geological processes, has heterogeneous distribution with widely varying concentrations in different rocks, and can be obtained through mining.[7] Upon mining and refining, it is useful as a fuel in nuclear reactor for power generation and research and has many industrial and medical applications. The mining and processing gives rise to external gamma radiation from the ores, inhalation of dusts containing long-lived alpha-emitting radionuclides, and inhalation of short-lived decay products of radon.[3] Radon decay progenies are inhaled in mine areas when poorly ventilated and can lead to exposures that exceed radiation exposure limits, resulting in the incidence of lung cancer in mine workers.[3]

It is important to estimate the exposures of humans to the different sources of radiation, both external and internal. Information obtained from such study is of interest in the area of environmental radioactivity monitoring, establishment of rules and regulation on radiation protection, and as biochemical and geochemical tracers in the environment.[8,9]

The main objective of the present study is to determine natural radioactivity concentration in uranium ore-mined rocks from some parts of Northern Nigeria, using inductively coupled plasma-mass spectrometry (ICP-MS). In addition, it also intended to evaluate the associated radiological exposure indexes such as exposure index, representative level index, absorbed dose rate, and annual effective dose (external), which can give an idea on the onsite occupational exposure of workers in this particular uranium mine areas.

MATERIALS AND METHODS

Geology of the study area

The study area covers accessible locations in parts of Northern Nigeria where mining activities of uranium ores are obtainable, which includes Adamawa (Michika), Kano (Riruwai), and Taraba (Mika) areas. Global Positioning System navigator (eTrex 10 GARMIN Garmin International, Inc., Olathe, Kansas, USA) was used to register the coordinates of each sample location, which are indicated in column 2, Table 1. The map of sampling area of this study is shown in Figure 1. Riruwai mine site is located at Kaffo, 10 km east of Riruwai, in Kano state, North-western Nigeria. The area was described as a ring complex, which is composed of elliptical multi-phase granitic massif bodies with related quartz veins that intruded into Pan-African basement. As Greisenized granite Precambrian (Age 541–4000 Ma) and part of Nigerian younger granite provinces that comprise mainly of granitoids and volcanic rocks, it is endowed with 28 valid minerals, including gold, pyrochlore, coffinite, thorite, and zircon. It is chemically distinct from the basement suites of per-alkaline and sub-alkaline.[10-12]. Mika and Michika mine sites are, however, located at Taraba and Adamawa states, respectively, both in North-eastern Nigeria. Uranium mineralization in the North-eastern Nigeria is of sandstone-hosted and vein-type mineralization. Mika and Michika are of vein-type mineralogy, which occurs in deformed migmatites and granitoid.[13] The mika area is more of granites with fine-grained granite, medium-grained granite, and porphyritic granite. Both primary and secondary uranium mineralization is rhyolite and occurs at depth for pitchblende, while near surface are mainly of meta-autunite and coffinite.[14,15] Michika is made up of mostly granitic rocks associated with rhyolite veins and brecciated, silicified, and mylonitized rocks. The area is underlain by crystalline basement rock of Precambrian age.[16,17] The vein-type (granite-related U-deposit) mineralization occurs in the ganitoids, with the major occurrences being Gubrunde, Kanawa, Ghumchi-Michika, Mika, and Monkin-Maza deposits.

Table 1: The activity concentrations of ^{232}Thorium and ^{238}Uranium (Bq/kg) of the uranium ore rock samples

Sample ID	Location coordinate	^{232}Th (Bq/kg)	^{238}U (Bq/kg)
UO01	N 10° 43.340'; E008° 47.007'	21.9±1.3	16.5±0.72
UO02	N 10° 39' 51"; E 13° 29' 20"	0.22±0.03	1.1±0.03
UO03	N8° 58' 46"; E 11° 36' 23"	0.65±0.01	924.6±17.1
UO04	N8° 58' 47"; E 11° 36' 23"	0.25±0.01	98.2±3.7

^{238}U: Uranium, ^{232}Th: Thorium

Sample collection and counting

Based on the geological features and mineralization of the sampling location, about 2.0 kg each of the uranium ore rock samples was collected at open pit-mining sites and immediately transferred into a zip-lock plastic bag to avoid contamination. Figure 1 illustrates the study area. ICP-MS analytical technique requires that the sample be introduced in liquid or aerosol form. Hence, the rock samples were crushed and milled into powder texture, using Dickie and Stockler TS-250, Johannesburg, crushing and pressurized milling machines. Digital Microwave Digestive System (Anton Paar Multiwave, PerkinElmer Germany) was used to dissolve the samples. About 0.5 g of the sample was

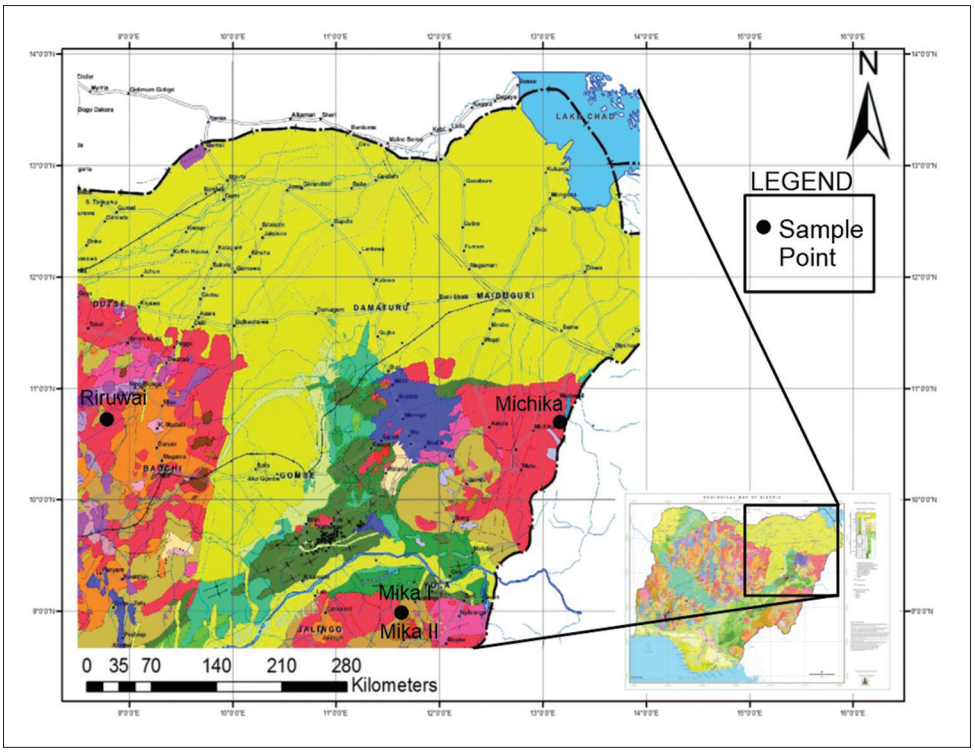

Figure 1: Map of Nigeria showing uranium ore sample mine sites (Riruwai, Mika-I, Mika-II and Michika) for this study

weighed into a microwave digester vial and 2.0 mL of distilled water was added, with 2.0 mL of concentrated HNO_3, 6.0 mL of concentrated HCl, and 0.5 mL H_2O_2. The vials containing the aliquot of sample are sealed into a rotor, placed into the microwave reactor system, and then heated at 180°C for a maximum period of 50 min. An aqueous solution in acidified matrix was then obtained. The dissolved samples were then filtered using polyvinylidene fluoride 0.22 µm. A diluent, composed of 5% (50 mL) HNO_3 and 1% (10 mL) HCl acids, was added to the filtrate in two sets by 10% and 50%. Both the prepared and blank samples, with the calibration standards (5 ppb, 20 ppb, 50 ppb, 100 ppb, 500 ppb, and 1000 ppb), and quality control samples (20 ppb and 100 ppb) were placed into the autosampler, for ICP-MS measurement and calibration [Figure 2]. Prior to the upload of the samples into the low-flow sample introduction system, nitric acid was used to clean the tubing, to avoid washing of residual memory. The cleaning and rinsing of the tubing was repeated after each sample to avoid cross contamination. The samples were introduced into a nebulizer containing plasma at atmospheric pressure, with an extremely high temperature of 10,000°C, which makes for easy transition of most elements from atoms to ions. Mostly, single-charged positive ions are formed and because argon plasma is used, it becomes relatively simple for mass spectra to be generated and interpreted. The plasma ionizes the samples and then through a

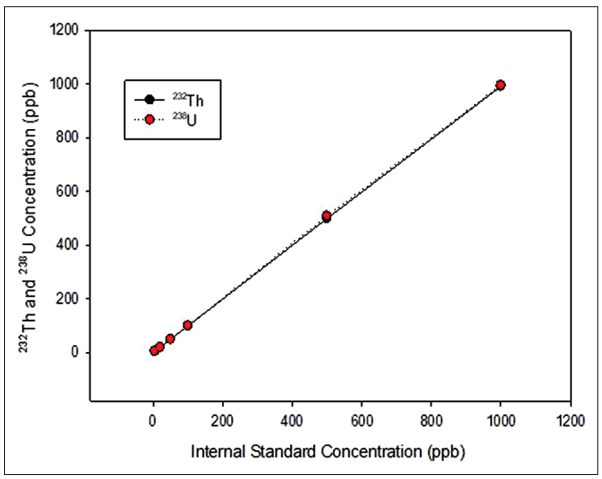

Figure 2: A graph of inductively coupled plasma-mass spectrometry calibration for thorium and uranium versus ppb of the samples in this study

vacuum interface, transfers to the mass analyzer – A hyperbolic quadrupole mass analyzer, which is sequential, thereby measures elements in sequence. Due to ICP-MS's abundant sensitivity, superior adjacent peak separation was achieved.[18] The separated ions are then detected by the ICP-MS detection system – a fast simultaneous dual-mode detector (of nine orders dynamic range), giving rise to data acquisition. Fifty elements were measured/identified including ^{238}U, ^{232}Th, trace elements, and rare earth elements, upon completion of measurement. The process is automatic and computerized. The control

is performed on the computer system via an installed software-ICP-MS (MassHunter Workstation software). ICP MS (The Agilent 7700 series, Agilent Technologies, Inc. USA) was used for sample analysis with rhodium internal standard (ISTD), at environmental analytical chemistry Laboratory, University of the Witwatersrand, Johannesburg, South Africa.

Determination of activity concentrations of ^{238}uranium and ^{232}thorium

The activity concentration of uranium (^{238}U) and thorium (^{232}Th) was obtained by converting concentration from ppb to ppm and then to Bq/kg, using equation (1),[19] which is consistent with the recommended conversion factors (CFs) from the IAEA.[20]

$$A = \left(\mathrm{Bq}/\mathrm{kg}\right) = \left(\frac{N_A \ln(2) C_I}{M_I t_{1/2}}\right) \times 10^{-6} \quad (1)$$

Where A is the specific activity concentration of the isotope in Bq/kg, N_A is Avogadro's number (6.023×10^{23} mol^{-1}), C_I is the measured isotope concentrations in ppm and $t_{1/2}$ is the half-life of the isotope in seconds, and M_I is atomic masses (kg/mol).

Absorbed dose rate and annual effective dose

The determination of the average absorbed dose rate (D) due to terrestrial sources of gamma radiations in air 1.0 m above the ground surface for uniform distribution of naturally occurring radionuclides (^{238}U and ^{232}Th) was calculated based on the guidelines from UNSCEAR, presented in equation (2).[1,3,6,21] However, the settings and ISTDs for the ICP-MS, in this work, used ^{40}K as correction isotope, therefore, A_K was subtracted from equation (2) and other ones applicable to ^{40}K. Therefore, the present work focused on the dose, which emanates from uranium and thorium as considered under the covered area of study.

$$D = 0.462 A_U + 0.604 A_{Th} + 0.042 A_K \quad (2)$$

Where D is the absorbed dose rate in nGy/h, and are the concentrations of uranium, thorium, and potassium in Bq/kg, respectively, the coefficients 0.462, 0.604, and 0.042 are the activity concentration to dose rate CFs of A_U, A_{TH} and A_K, respectively, in nGy/h per Bq/kg.[21] The absorbed dose rate has a global average value of 55 nGy/h.[3]

The net external gamma radiation dose rate (D) was used to estimate the annual effective dose (external), by taking into account the outdoor occupancy factor 0.2, and the CF from absorbed dose rate in air to effective dose 0.7 Sv/Gy for adults. Equation (3) is used to compute the E_{ext} as proposed by the UNSCEAR.[3,22]

$$E_{ext} = D \times 8760 \times 0.2 \times 0.7 \times 10^{-6} \quad (3)$$

Where E_{ext} is the external annual effective dose in mSv/a.

Determination of radiological index parameters

Radium equivalent dose

Due to nonuniform distribution of primordial radionuclides in rocks and soils, the radium equivalent activity (Ra_{eq}) in Bq/kg has been used as a single index that describes the gamma output from the mixture of ^{238}U, ^{232}Th, and ^{40}K from samples. The use of rocks and soils from mining sites to build houses resulting in the gamma radiation exposure to human can, therefore, be assessed.[6,9] The concentration of Ra_{eq} exceeding 370 may result in radiation exposure elevation. The exposure due to radiation in terms of radium equivalent is calculated using Equation (4).[3,22-24]

$$\mathrm{Ra}_{eq}\left(\mathrm{Bq/kg}\right) = A_{Ra} + 1.43 A_{Th} + 0.077 A_K \quad (4)$$

Where $A_{Ra,}$ $A_{Th,}$ and A_K represent the activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K, in Bq/kg, respectively.

Note also that daughter nuclide ^{226}Ra is replaced by the parent nuclide ^{238}U, even though there may be disequilibrium given as 1.03 between the parent and daughter nuclides, according to the UNSCEAR.[3,6]

External and internal exposure index

The external exposure index (H_{ex}) is used as a radiation exposure index to assess the indoor radiation dose rate with respect to external exposure to gamma radiation from the natural radionuclides. This is to limit such exposures from building materials in which rocks and soils are part. Internal exposure index (H_{in}), on the other hand, seeks to assess the internal exposure to radon and its short-lived progenies, which are hazardous to the respiratory organs.[6,9,23]

The values for the H_{ex} and H_{in} are calculated using equations (5) and (6), and must be less than unity for radiation hazard to be negligible.[3,23]

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (5)$$

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (6)$$

Where A_{Ra}, $A_{Th,}$ and A_K are the respective activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K, in (Bq/kg).

Representative gamma and alpha index

Inside the human body, radiation is being monitored using the representative gamma index (I_γ) and also to determine the level of risks as a result of human exposure to external

annual effective doses from gamma radiation of radioactive nuclides sources in rock samples, as proposed by the European Commission.[25,26] Equation (7) was applied to estimate the (I_γ) gamma, such that if (I_γ) ≤2, the annual effective dose increases by 0.3 mSv, but will increase by 1 mSv for 2≤ (I_γ) ≤6.[25] Hence, safe materials for the purpose of construction can be identified by I_γ.

$$I_\gamma = \frac{A_{Ra}}{300} + \frac{A_{Th}}{200} + \frac{A_K}{3000} \quad (7)$$

Where $A_{Ra,}$ $A_{Th,}$ A_K are the respective specific activities of ^{226}Ra, ^{232}Th, and ^{40}K, in Bq/kg.

The representative alpha index is the exposure index (I_α) used to estimate, in a given sample, the level of α-radiation exposure due to natural radionuclides and radon exhalation as presented in equation (8).[9,24]

$$I_\alpha = \frac{A_{Ra}}{200\ \text{Bq/kg}} \quad (8)$$

Where A_{Ra} is the specific activity of ^{226}Ra, in Bq/kg.

RESULTS AND DISCUSSION

The activity concentrations (Bq/kg) of the naturally occurring ^{232}Th and ^{238}U radionuclides were determined in the uranium-ore rock samples collected from some parts of Northern Nigeria, and the results are presented in Table 1. As shown in Table 1, the activity concentrations of the rock samples have highest values at 21.9 ± 1.3 Bq/kg for ^{232}Th and 924.6 ± 17.1 Bq/kg for ^{238}U. The lowest values were 0.22 ± 0.03 Bq/kg and 1.1 ± 0.03 Bq/kg, respectively. The results obtained are illustrated in Figure 3, which showed that the highest concentration for ^{232}Th radionuclide was detected from sample UO01, whereas for ^{238}U was from UO02, which were collected from Riruwai and Mika-I. The values from this study are below those reported by El-Taher and Uosif and Ademola *et al*.[1,6] and the world average concentration of 30 and 40 Bq/kg for ^{232}Th and ^{238}U,[3,21] except for ^{238}U value of sample UO03 that is extremely higher by a factor of 26.4 and sample UO04 higher by a factor of 2.8. The geological formations of the elements explain the reason for the variations in the natural radioactivity.[7]

The absorbed dose rates were estimated using Equation (2), and the result is presented in column 2 of Table 2 and displayed in Figure 4. The absorbed dose rate ranged from 0.62 ± 0.03 to 422.4 ± 7.8 nGy/h and was found to be below the world average value of 55 nGy/h,[3] except for sample U003 whose value is higher by a factor of 7.6. Figure 5a and b shows the correlation between the absorbed rate and activity concentrations of ^{232}Th, found to be least significant (R^2 = 0.999). This implied a major portion of dose contributed due to the uranium content of the rock. However, Rangaswamy *et al*.[9] had major portion of the dose contributed from thorium, in their studies, which could be attributed to variations in geological locations of the samples studied.

The Ra_{eq} was calculated using Equation (4), and the result is shown in column 3, Table 2, and illustrated in Figure 4. The Ra_{eq} from this study varies from 1.4 ± 0.06 to 914.4 ± 16.9 Bq/kg, with the highest value of 914.4 ± 16.9 Bq/kg reported from sample UO03,

Table 2: Results of the absorbed dose rate, radium equivalent activity, and annual effective dose estimated from uranium ore rock samples

Sample ID	D (nGy/h)	Raeq (Bq/kg)	Eext (mSv/a)
UO01	20.8±1.1	47.7±2.4	0.025±0.001
UO02	0.6±0.03	1.4±0.1	0.001±3.202E-05
UO03	422.4±7.8	914.4±16.9	0.518±0.009
UO04	44.9±1.7	97.4±3.6	0.055±0.002

D: Dose rate, E_{ext}: Annual effective dose, Ra_{eq}: Radium equivalent activity

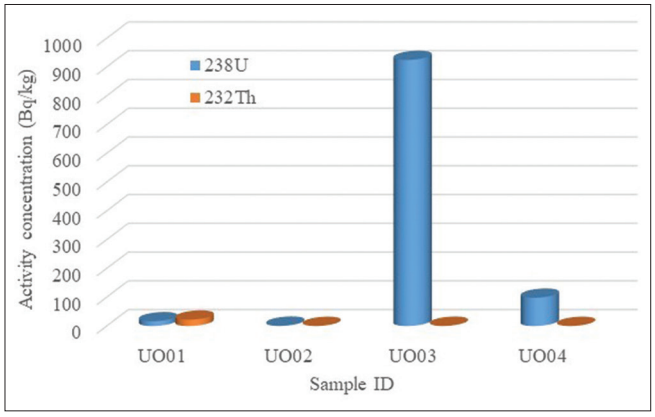

Figure 3: Comparison of the ^{238}Uranium activity concentration (Bq/kg) and ^{232}Thorium activity concentration (Bq/kg) present in the collected samples in the study area

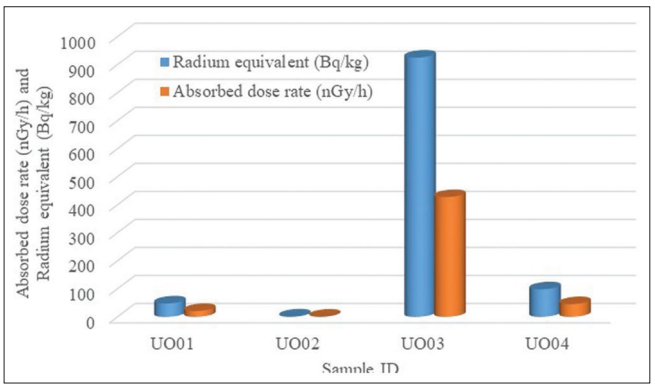

Figure 4: Absorbed dose rates (nGy/h) and radium equivalent activity (Bq/kg) due to ^{232}Thorium and ^{238}Uranium concentrations present in the study area

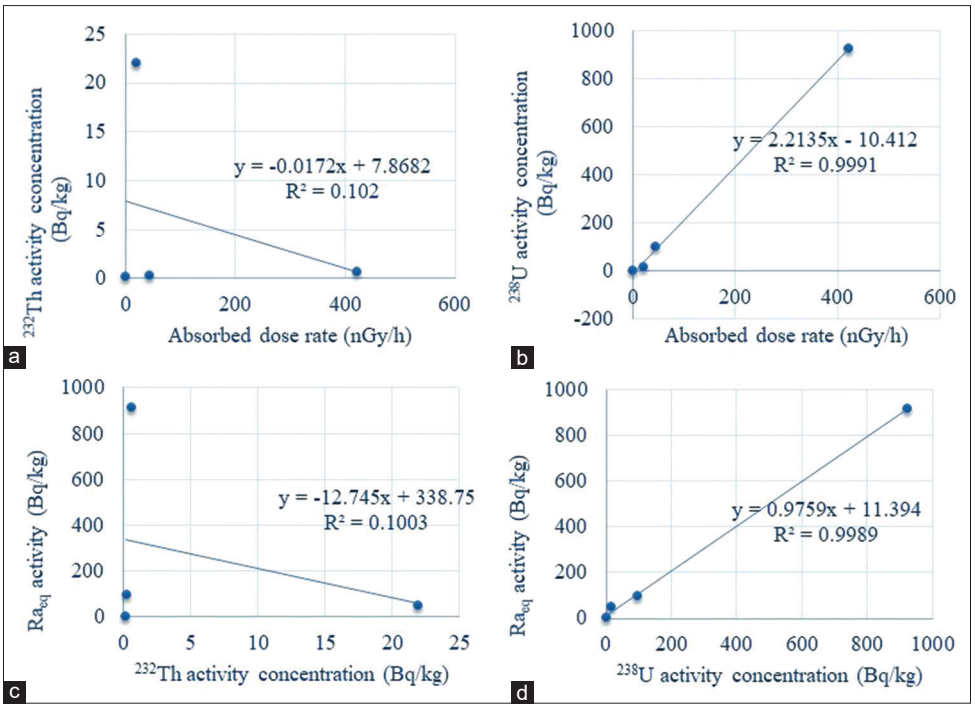

Figure 5: Correlation between (a) the absorbed dose rate and ^{232}Thorium activity concentration, (b) the absorbed dose rate and ^{238}Uranium activity concentration, (c) radium equivalent and ^{232}Thorium activity concentration, and (d) radium equivalent and ^{238}Uranium activity concentration of the rock samples in this study

whereas the lowest value was from sample UO02. It can be seen that the values of Ra_{eq} in all the measured samples are lower than the UNSCEAR-recommended value of 370 Bq/kg except the highest value. Figure 5c and d shows the correlation between Ra_{eq} with ^{232}Th and ^{238}U whose values are $R^2 = 0.1003$ and $R^2 = 0.9989$, respectively, which indicated good and strong correlation between Ra_{eq} and ^{238}U, implying that contribution is mostly from ^{238}U.

The annual effective dose (external) (E_{ext}) was determined using equations (3) and (4), to test the effect of the absorbed dose rates, and presented in column 4, Table 2. As shown in Table 2, the external annual effective dose ranges from 0.001 ± 3.202E-05 to 0.518 ± 0.009 mSv/a. The values are within the safety limit (1.0 mSv/a) proposed by the UNSCEAR[3,21] and lower than that reported by Ademola *et al.*[6]

Equations (5–8) were used to calculate H_{ex}, H_{in}, and representative gamma and alpha indexes (I_γ and I_α), and their respective results are presented in columns 2–5, in Table 3. It is shown for the study locations that H_{ex} and H_{in} have values that vary from 0.004 to 2.47 and 0.007 to 4.94, respectively, which were lower than unity as deserved, except for the highest value from UO03 that exceeded.

Both I_γ and I_α from Table 3 have values that ranged from 0.0046 to 3.05 and 0.0052 to 4.57, respectively. The criterion indicates that, for $I_\gamma \leq 2$ the annual effective dose increase

Table 3: External exposure, the internal exposure, gamma index, and alpha index for the study

Sample ID	H_{ex}	H_{in}	I_γ	I_α
UO01	0.13	0.17	0.16	0.08
UO02	0.004	0.007	0.0046	0.0052
UO03	2.47	4.94	3.05	4.57
UO04	0.26	0.52	0.33	0.49

H_{ex}: External exposure, H_{in}: Internal exposure, I_γ: Gamma index, I_α: Alpha index

by 0.3 mSv, but will increase by 1 mSv for $2 \leq I_\gamma \leq 6$[25] and applies for the samples in this study. For I_α it is recommended that the activity concentration of ^{226}Ra at upper limit be 200 Bq/kg. However, radon the progeny of ^{226}Ra has <200 Bq/m^3 whereby $I_\alpha < 1$ [9,25]. The values in this study are below the recommended limit, suggesting that radon exhalation from the rock samples will cause indoor concentration <200 Bq/m^3, except for sample UO03 that exceeded the limit.

CONCLUSIONS

The level of natural radioactivity in uranium-ore rock samples collected from some parts of Northern Nigeria was analyzed using ICP-MS. The results obtained showed the activity concentrations of ^{232}Th in the samples from the mining sites as 0.22 ± 0.03 to 21.9 ± 1.3 Bq/kg and ^{238}U as 1.05 ± 0.03 Bq/kg to 16.5 ± 0.72 Bq/kg to be lower than the world average values identified by the UNSCEAR.[3] However, activity concentration values of

^{238}U from sample locations Mika-II and Michika, 924.6 ± 17.1 and 98.2 ± 3.7 Bq/kg, respectively were much higher than the recommended value. The estimated values due to ^{232}Th and ^{238}U, of mean absorbed dose rate (D), annual effective dose (external), Raeq, Hex, Hin. Others include representative gamma and alpha indexes (I_γ and I_α), which were lower than the world average values except for Mika-II location. It can be concluded from the results in this study that the study areas investigated are radiologically safe for mining activity except for the sample location with higher values. The radiation dose from this study (maximum value being 0.52 ± 0.009 mSv/a, for external) does not generalize for the entire Nigeria, but deals particularly with the occupational radiation exposure for workers in the respective sample locations, where adequate protective measures should be emphasized. The values are far below the worldwide-allowed dose of 20 mSv/a for workers.[27]

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Conflicts of interest

There are no conflicts of interest.

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