

NORM, radon emanation kinetics and analysis of rocks-associated radiological hazards

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Abstract Analysis of naturally occurring radioactive material (NORM) and environmental (radiological) hazard associated with radionuclides in rocks from northeastern Nigeria was carried out and the results are presented. NORMs analysis on rock samples was undertaken with gamma-ray spectroscopy, while radon isotopes were analyzed using an electric radon measurement device (RAD 7). Activity concentration (Bq kg⁻¹) of ²²⁶Ra, ²³²Th and ⁴⁰K ranges from 11.4 ± 2.6 to 74.6 ± 4.8, 26.6 ± 0.8 to 183.4 ± 3 and 30.1 ± 4.6 to 1120.3 ± 16.4, respectively. Ranges for annual effective dose (mSv) are 0.03–0.20; radium equivalent (Bq kg⁻¹) is 5.7.33–358.36; external hazard index is 0.16–0.99; ²²²Rn and ²²⁰Rn exhalation rates (Bqm⁻³ h⁻¹) are 2.60–6.25 and 0.50–7.10, respectively. For radon emanation coefficient, the working month level and related dosages ranges are 0.012–0.058, 0.25–0.00 and 1.38–3.30 mSv, respectively. The results reveal that the possible radionuclides enrichment in the rocks is potentially an environmental (radiation) hazard to the inhabitants of the area.

Keywords Limit · Activity · Gamma ray · Emanation · Radiation hazard · Nigeria

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Introduction

The main source of external radiation to the human body is the background terrestrial radiation from naturally occurring radioactive isotopes such as ⁴⁰K, ²³⁸U and ²³²Th series (USNRC 1991). These exist in all geologic materials. The study area is geologically dominated by Precambrian granite and gneiss which are known to have higher radionuclide compositions compared to other rock types (Tzortzis and Tsertos 2004). This implies that higher radiation can conveniently be associated with these rock types except in some cases where some other rock type such as sandstones might have high radionuclide content through mobilization and precipitation by groundwater in a favorable environment (Arabi et al. 2013a, b); some shale and phosphate rocks were reported to have relatively high radionuclides content (UNSCEAR 2000a Report). Apart from the exposure from direct cosmic rays and cosmogenic radionuclides, natural exposures to naturally occurring radioactive material (NORM) arise mainly from the primordial radionuclides which are spread widely and are present in almost all geological materials in the earth's environment (Wilson 1994; NCRPM 1975).

Radon (²²²Rn), an inert radioactive gas, is formed through alpha decay of radium (²²⁶Ra) in the uranium series. It emanates from rock materials and can migrate easily and enter from the subsurface into groundwater bodies through fractures. When inhaled, these solid radionuclides are deposited in the long tissue and can damage the cells that line the lung. Long-term exposure to radon can lead to lung cancer.

Since the exit of the Nigeria Uranium Mining Company (NUMCO) from the study area (Benue trough in northeastern Nigeria; Fig. 1) in the early 1980s (unpublished archive of the Nigeria Geological Survey Agency), many

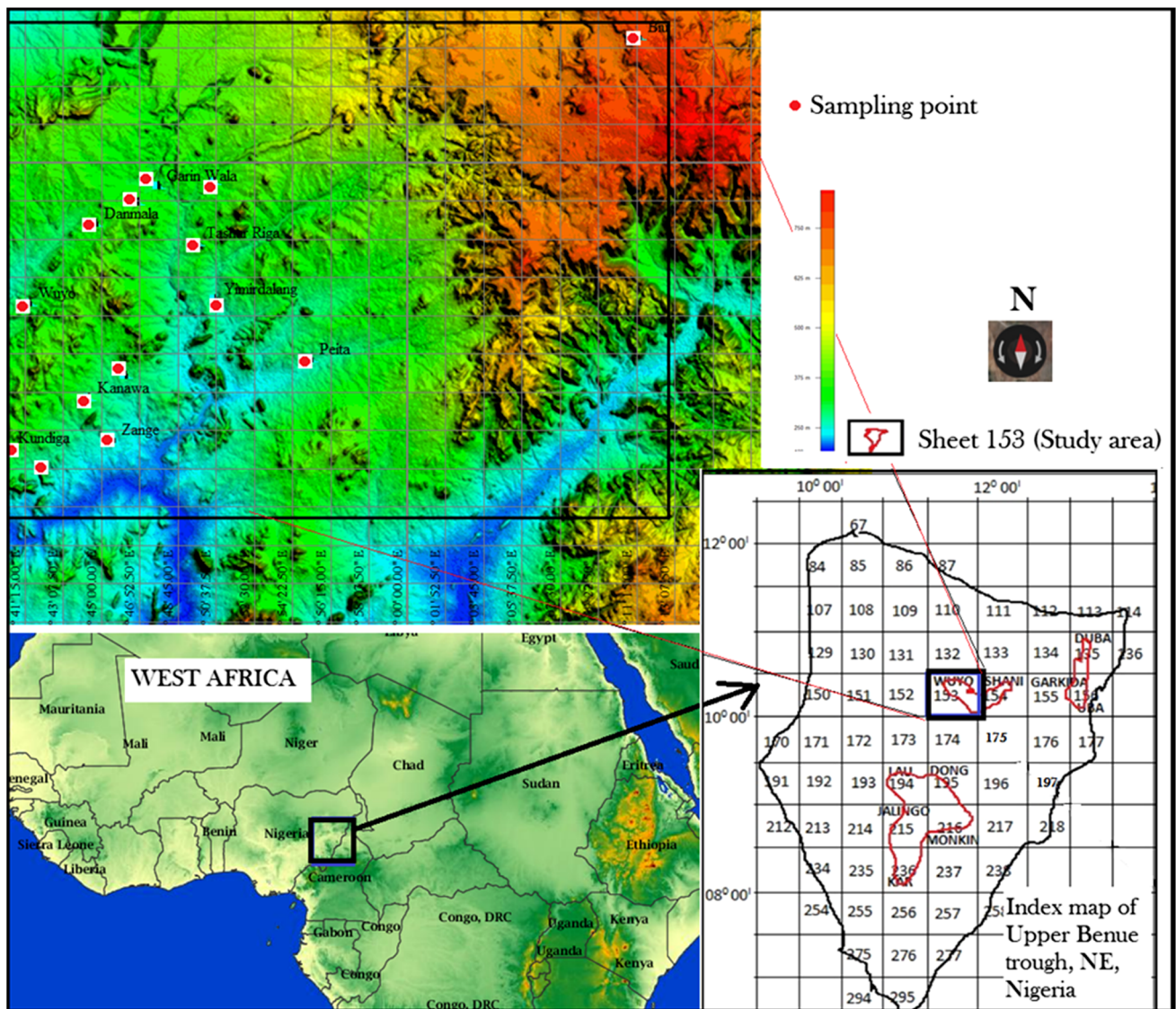


Fig. 1 Digital elevation model (DEM) of the study area showing sampling locations, its location on the index map of the Upper Benue trough and map of West Africa (*red demarcation on the index map shows areas with uranium amplitude >4μRh⁻¹*)

efforts were made on individual worker basis, from academic institutions, to look at the different aspects of radionuclide occurrence and associated environmental issues in the area. These efforts were hampered by resources that mainly impacted on mobility for detailed work, but the wide scope nature of such activities involved has made it impossible to date to complete a comprehensive study.

This study, therefore, is aimed at contributing and exploring techniques not utilized in the previous studies by Okujeni (1987), Okujeni et al. (1990), Okujeni and Funtua (1994), Funtua et al. (1997) and Elegba et al. (1993) which may serve as baseline data sets in terms of resources evaluation and environmental realities that prevail in the area. There are sketchy data on environmental radiation

from the area. Our objective in this study also focused on NORM activity concentration, variational trend and rock characteristics based on a radiological risk index.

Materials and methods

The study commenced with demarcation of areas of high uranium amplitude from over 60 aero-radiometric maps that covered the entire Upper Benue trough (Fig. 1b). Rock samples from sheet 153 (WUYO), which is the study area, were chipped from fresh rock outcrop using a geologic hammer and brought to the laboratory for analysis of naturally occurring radionuclides (²²⁶Ra, ²³²Th and ⁴⁰K) using gamma spectrometry, while another portion was utilized in

the analysis of radon exhalation and emission using electric radon detector (RAD 7) (Fig. 2).

Gamma spectrometry

For the gamma spectrometry, 10 g of sample was ground in a ball mill, oven dried and sealed in a vial using paraffin wax and taken for the analysis. The γ -ray spectrometer used (Fig. 2a) consisted of a coaxial HPGe detector, having 60 % relative efficiency with respect to 7.62 \times 7.62 cm NaI (Tl) crystal at an operating voltage of 4000 V. The detector peak to Compton ratio was 78:1 and the measured energy resolution for the ^{60}Co γ -ray line (1332 keV) at a source to detector distance of 25 cm was 1.85 keV.

The validation of the full energy peak efficiency (FEPE) for optimizing the measurement of ^{40}K , ^{238}U , and ^{232}Th in a geological matrix was achieved using the standard calibration process. The procedure adopted ensures direct conversion of the net count rate of a γ -ray peak of a given energy, E, from a specific radionuclide to the activity of that radionuclide; it eliminates the need for corrections and is well suitable for the measurement of a few radionuclides. A detailed description of gamma-ray spectrometry measurement and analysis techniques can be found in Tzortzis et al. 2003 and Hamby and Tynybekov (2000).

Radon measurement

There have been a number of developments in the study of exhalation of radon from the earth’s surface since the 2003 WMO/IAEA/CNRS meeting at Gif sur Yvette and the resulting report WMO/GAW (Allegrini et al. 1994). Comprehensive, ground-validated radon flux density maps for China (Marcazzan and Testa 1995) and Europe

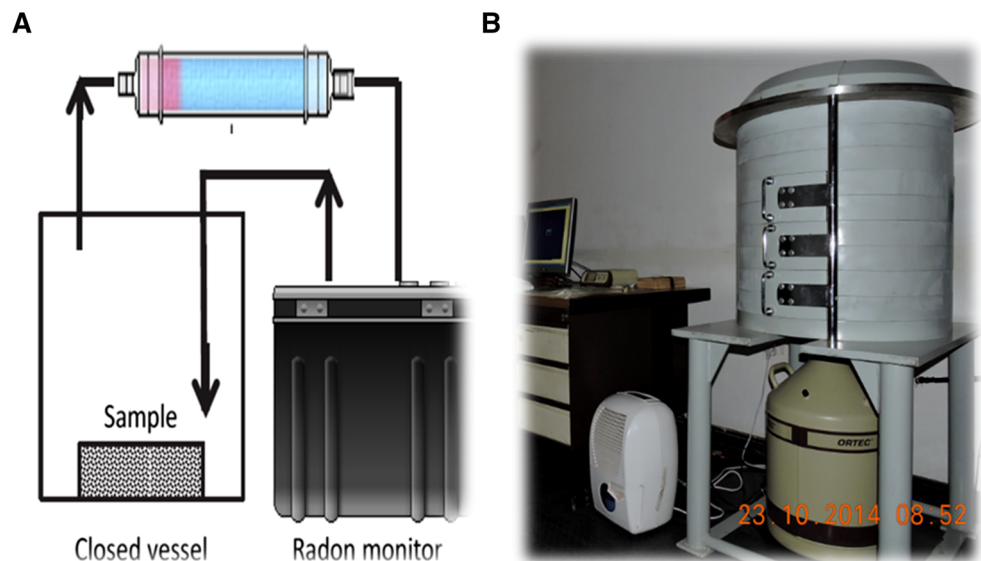
(Sesana et al. 1998) have been published. The production of these maps involved new algorithms relating radon flux density to closely associated variables such as soil radium or surface gamma dose rate. Perrino et al. (2001) published the first global maps estimating the spatial distribution of monthly averaged radon flux density from the oceans.

In this study, radon and thoron exhalation rates were determined using a vessel as a sample container in which air from the sample is passed into a Rad 7 chamber and back to the sample vessel via a closed loop as described in Fig. 2a and also in Ishmori et al. (2013). The internal sample cell of the RAD 7 detector is a 0.7-L hemisphere coated on the inside with an electrical conductor, with a solid-state ion-implanted planar silicon α -particle detector arranged at the center of the hemisphere. Radon emission/exhalation measurement using RAD 7 has been described in detail in Arabi et al. (2015).

The emanation coefficient was evaluated by the combination of radium and radon measurements and γ -ray spectrometry under different conditions.

The sample material was packaged in a cylindrical plastic container and held in a sealed vessel for more than 4 weeks to ensure the establishment of radioactive equilibrium between ^{222}Rn and ^{226}Ra . After that, the total activity of radon released into the air from the sample material was evaluated by measurement of the radon concentration and effective volume of the measurement system (Fig. 2). The effective volume was determined by considering the volume of the closed vessel, all material inside the closed vessel, the loop system for radon measurement, and the radon monitor. The total activity of radium in the sample material was determined by γ -ray spectrometry (Arabi et al. 2015).

Fig. 2 Setup for a radon exhalation measurement using RAD 7; **b** gamma spectrometry for radionuclides analysis



Parameter calculations

Radon emanation coefficient (E), radium equivalent activity (Ra_{eq}), absorbed dose rates, external hazard index (H_{ex}) and annual effective dose were calculated as follows.

Radon emanation coefficient (E)

The Radon emanation coefficient (E) of the samples was calculated from the relation (Arabi et al. 2015):

$$E = \frac{VC}{MR}, \quad (1)$$

where E is the emanation coefficient; V is the effective volume of the sample holder (m^3); C is the radon concentration in the sample (Bqm^{-3}); M is the total mass of the sample (kg); and R is the radium activity concentration in the sample ($Bq\ kg^{-1}$).

For other details on radon emanation fraction of soil, rock and mineral, refer to Sakodaa et al. (2011).

Radium equivalent activity (Ra_{eq})

Uniformity with respect to exposure to radiation has been defined in terms of radium equivalent activity (Ra_{eq}) in $Bq\ kg^{-1}$ assuming that $370\ Bq\ kg^{-1}$, of ^{226}Ra or $259\ Bq\ kg^{-1}$, of ^{232}Th or $4810\ Bq\ kg^{-1}$, of ^{40}K will give the same gamma dose rate (Alamgir et al. 2012).

Ra_{eq} was calculated according to the relation (Arabi et al. 2015):

$$Ra_{eq} = cRa + 1.43cTh + 0.07cK, \quad (2)$$

where cRa , cTh and cK are the activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in $Bq\ kg^{-1}$, respectively.

Calculation of absorbed dose rates

Absorbed dose rates (outdoor) due to terrestrial gamma rays at a distance of 1 m above ground level can be determined from the ^{226}Ra , ^{232}Th and ^{40}K concentration values in geological materials assuming other radionuclides have a negligible dose from the environmental background (Kocher and Sjoreen 1985; Jacob et al. 1986; Leung et al. 1990). The conversion factors used to calculate the absorbed dose rates are given in Eq. (3) assuming that all the decay products of ^{226}Ra and ^{232}Th are in radioactive equilibrium with their parent isotope (UNSCEAR 1993).

$$D = 0.461cRa + 0.623cTh + 0.0414cK. \quad (3)$$

External hazard index (H_{ex})

The values of the external hazard indices (H_{ex}) must be less than unity (Eq. 4) for radiation hazard to be negligible (Diab 2008; Beretka and Mathew 1995).

Mathematically, an external hazard index was calculated as

$$H_{ex} = \frac{cRa}{370} + \frac{cTh}{259} + \frac{cK}{4810} \leq 1, \quad (4)$$

where cRa , cTh and cK are the activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in $Bq\ kg^{-1}$, respectively.

Calculation of annual effective dose

Using calculated absorbed doses as detailed in UNSCEAR (2000), the annual effective dose from background gamma radiation was estimated as follows:

$$E = (D_{out} \times OF_{out} + D_{in} \times OF_{in}) \times T \times CC,$$

where E (nSv) is the annual effective dose, D_{out} and D_{in} ($nSvh^{-1}$) are mean outdoor and indoor absorbed dose rates, T (h) is the time to convert from year to hour (8760 h), OF_{out} and OF_{in} are outdoor and indoor occupancy factors (20 and 80 % for outdoor and indoor, respectively) and CC is conversion coefficient (0.7 for adults) reported by UNSCEAR to convert absorbed dose in air to the effective dose in humans

Results and discussion

The results of NORM activity concentration ($Bq\ kg^{-1}$), radon emanation, radium equivalent, external hazard index and annual effective dose for all the samples are presented in Table 1. These results show that the activity concentration ($Bq\ kg^{-1}$) of ^{226}Ra , ^{232}Th and ^{40}K in the rock samples ranges from 11.4 ± 2.6 to 74.6 ± 4.8 , 26.6 ± 0.8 to 183.4 ± 3 and 30.1 ± 4.6 to 1120.3 ± 16.4 , respectively. This implies that 23.7 % of the samples analyzed have ^{226}Ra values above the world average of $35\ Bq\ kg^{-1}$; 76.9 % have ^{232}Th values above the world average of $30\ Bq\ kg^{-1}$; and 46.15 % have ^{40}K values greater than the world average of $400\ Bq\ kg^{-1}$.

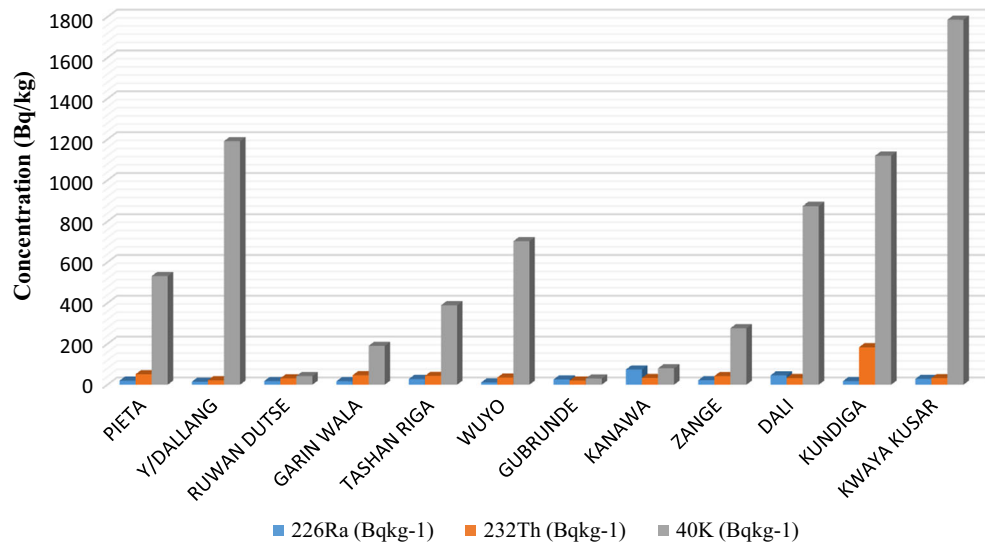
Working month level and radiation dose (Table 3) associated with the use of the studied material for construction exceeded the set limits. These values range from 0.28 to 0.66 and 1.38 to 3.30 mSv for the WLM and dose, respectively.

Samples from Kanawa, Dammala and Dali have the highest activity concentration values of 74.6 ± 4.8 , 46.8 ± 6.2 and

Table 1 Results of radon emanation, activity concentration of NORM and associated radium equivalent, external hazard index and annual effective dose for the entire sample

S/N	Sample ID	Radon emanation coefficient	Gamma spectrometry method			Ra _{eq} (Bq kg ⁻¹)	External hazard index	Annual effective dose (10 ⁻⁵ Sv)
			²²⁶ Ra (Bq kg ⁻¹)	²³² Th (Bq kg ⁻¹)	⁴⁰ K (Bq kg ⁻¹)			
1	Kundiga	0.012	17.7 ± 4.	183.4 ± 3.0	1120.3 ± 16.4	130.45	0.36	7.26
2	Ruwan dutse	0.020	17.9 ± 0.3	31.1 ± 0.3	42.3 ± 1.7	129.35	0.37	7.46
3	Y/dallang	0.014	15 ± 0.4	21.7 ± 3.0	1191.6 ± 5.0	65.33	0.18	3.63
4	Zange	0.012	22 ± 2.3	42.4 ± 0.3	276.7 ± 1.9	96.90	0.27	5.45
5	Dali	0.034	45.9 ± 1.9	32.7 ± 1.5	874.5 ± 10.4	117.24	0.32	6.46
6	Kanawa	0.057	74.6 ± 4.8	33.1 ± 1.3	80.5 ± 9.4	110.26	0.31	6.25
7	Wuyo	0.015	11.4 ± 2.6	34.8 ± 0.8	703 ± 3.0	57.23	0.16	3.23
8	Gubrunde	0.014	25.6 ± 2.0	20.6 ± 0.8	30.1 ± 4.5	127.49	0.35	7.06
9	Pieta	0.022	19.8 ± 1.7	51.3 ± 0.7	532.2 ± 7.5	102.03	0.28	5.65
10	Garin wala	0.014	17.3 ± 2.1	46.4 ± 1.0	190.7 ± 5.1	153.80	0.43	8.67
11	Tashan riga	0.024	28 ± 1.0	43.3 ± 0.3	389.1 ± 3.0	358.36	0.99	19.97
12	Kwaya kusar	0.028	28.3 ± 2.8	33.1 ± 0.4	80.5 ± 7.0	81.21	0.22	4.44
13	Dammala	0.058	46.8 ± 6.2	25.1 ± 0.1	421.2 ± 4.6	112.22	0.31	6.25

Fig. 3 Concentration levels of radium, thorium and potassium in the studied samples

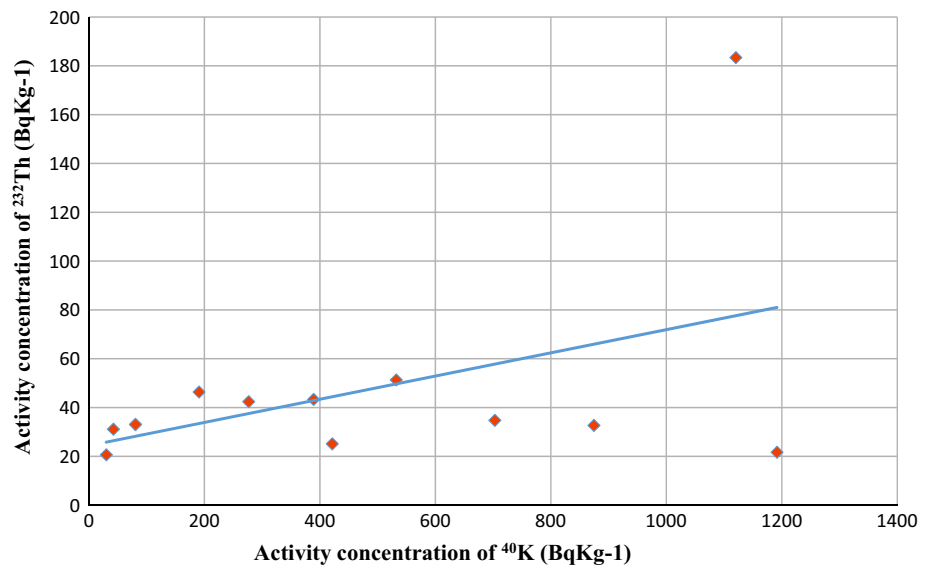


45.9 ± 1.9 Bq kg⁻¹ for ²²⁶Ra and ²³²Th, respectively (Fig. 3). Also, samples from Kwaya kusar, Yimirdalang and Kundiga recorded ⁴⁰K values >1000 Bq kg⁻¹ (Fig. 3) This high value recorded for ⁴⁰K might not be unconnected to its very long half-life of 1.251 × 10⁹ years. These areas which recorded higher activity concentration values confirmed reports by earlier workers of possible uranium mineralization in rocks around Kanawa and in groundwater samples around Kundiga and Dali as reported by Funtua (1992) and Arabi et al. (2013a, b), respectively.

A plot of activity concentration level for ²³²Th against ⁴⁰K (Fig. 4) revealed a linear relationship indicating a constant of proportionality between the two. It is interesting that in this study, high ²²⁶Ra activity concentration

values come from sandstone samples and not the Precambrian gneiss and granites as expected. The conductivity, pH, phosphate and iron content of rocks and groundwater in the area might have played a role in uranium inhibition in groundwater through precipitation in case of iron and formation of other complexes in the case of phosphate. In terms of ²³²Th, the sample from the Precambrian granite around Kundiga has the highest activity concentration values of 183.4 ± 3.0 Bq kg⁻¹ and also the highest ⁴⁰K values (>1000 Bq kg⁻¹) from the same Precambrian granite around Kundiga and Yimirdalang.

The analysis of radon and thoron (results in Table 2) exhalation rates, emanation coefficient and radium concentration revealed that the radon emanation coefficient

Fig. 4 ^{40}K and ^{232}Th activity concentration in rock samples**Table 2** Results of radon and thoron emission and exhalation rates from samples

S/ N	Location name	Geology	$^{222}\text{Rn}/^{226}\text{Ra}$ activity ratio	Radon emission from rock (Bq/m^3)	Radon exhalation rate ($\text{Bq}/\text{m}^3/\text{h}$)	Thoron emission from rock (Bq/m^3)	Thoron exhalation rate ($\text{Bq}/\text{m}^3/\text{h}$)
1	Kundiga	Granite	0.15	119 ± 8	4.96	1250 ± 38	52.08
2	Ruwan dutse	Granite	0.25	70.4 ± 7	2.93	170.4 ± 15	7.10
3	Yimirdalang	Gneiss	0.18	84.7 ± 7	3.53	81.9 ± 10	3.41
4	Zange	Sandstone	0.15	144 ± 9	6.00	58.6 ± 8	2.44
5	Dali	Sandstone	0.43	107 ± 5	4.46	73.4 ± 9	3.06
6	Kanawa	Sandstone	0.71	105 ± 8	4.38	52 ± 8	2.17
7	Wuyo	Sandstone	0.18	62.5 ± 6	2.60	115 ± 12	4.79
8	Gubrunde	Granite	0.17	150 ± 10	6.25	12 ± 4	0.50
9	Peita	Gneiss	0.27	73.5 ± 6	3.06	69.7 ± 9	2.90
10	Garin wala	Granite	0.18	96.7 ± 8	4.03	38.6 ± 7	1.61
11	Tashar riga	Gneiss	0.30	94.3 ± 7	3.93	161 ± 13	6.71
12	Kwaya kusal	Gneiss	0.35	79.8 ± 7	3.33	40.9 ± 7	1.70
13	Danmala	Sandstone	0.73	64.3 ± 7	2.68	56 ± 13	2.33

and exhalation rates increase with the ^{226}Ra content of the sample from Dali and Kanawa (Fig. 5). Also, the radon emanation coefficient recorded seems to increase with an increase in ^{226}Ra concentration as can be observed from Fig. 6; it is also dependent on the porosity and moisture content of the sample material.

Radon emanation coefficients of the different rock samples, which range from 0.012 to 0.058, is far below values (emanation fractions of 0.22 for the phospho-uraninite ore and 0.31 for the meta-autunite ore) reported by Funtua et al. (1985), though his values vary for different grain sizes of the samples (Table 3). Values reported in this work are for bulk rock samples in the same form as brought

in from the field. Radon emanation of rock samples are to some extent dependent on rock porosity, moisture content, radium content and the techniques used during measurement.

Austin (1973), Barretto (1973) and Rokotoson et al. (1983) reported that emanation of radon from rock sample has been established to be dependent on not only the mineral ore type, but also the uranium content. The distribution of $^{222}\text{Rn}/^{226}\text{Ra}$ activity ratio are generally not too consistent between rock types in this case, but the trend is such that higher values were somewhat consistent with the sandstones, suggesting, as reported in Vinson et al. (2009), to be consistent with the rock type. $^{222}\text{Rn}/^{226}\text{Ra}$ provides

Fig. 5 Plot of ^{226}Ra concentration against radon exhalation rates

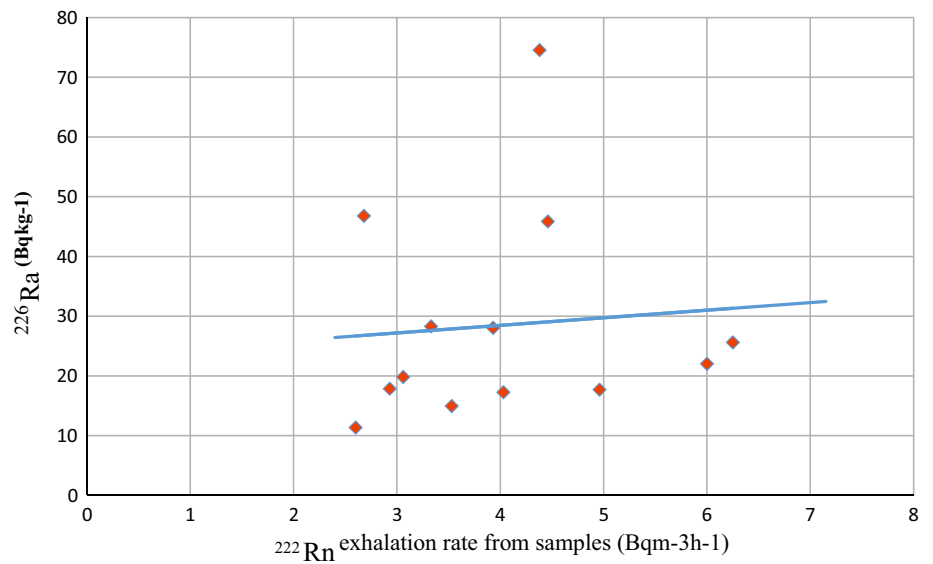
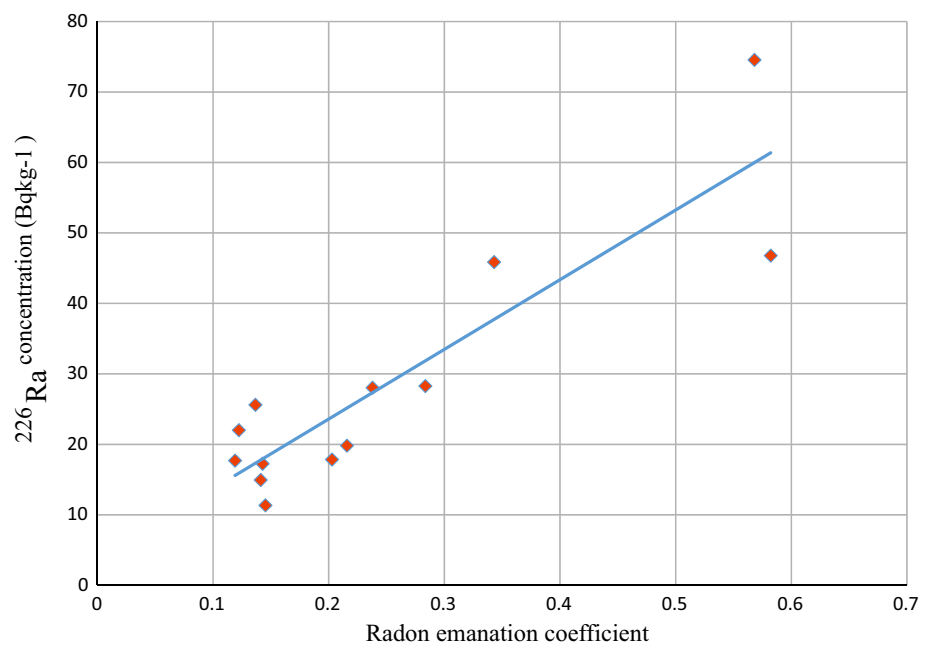


Fig. 6 Plot of ^{226}Ra concentration against radon emanation coefficient



information on radium adsorption in crystalline rock fractures regardless of the radium and radon sources in each rock type (Vinson et al. 2009).

From the comparison of values obtained in this work and those reported by other workers elsewhere (Table 4), the values we reported in this work are still within the ranges reported in the IAEA, Technical report 419 (IAEA 2003), but at the same time our values for some locations within the study area are above those reported by the IAEA (2003) for countries such as Australia and Germany and for ^{226}Ra (Romania).

Conclusion

This study set out to investigate the levels of naturally occurring radioactive materials (NORM), radon emanation kinetics in rocks and radiological hazards associated with them. Radiological risk evaluation suggests that inhabitants of the area might be at risk from radiation emitted by rocks in the area. High ^{226}Ra and ^{232}Th values in some locations in the study area might be as a result of uranium mineralization as reported by earlier workers. Working level month and radiation dose recor-

Table 3 Working level month and dose associated with the studied rock samples

S/N	Location name	Geology	$^{222}\text{Rn}/^{226}\text{Ra}$ activity ratio	Working level month (WLM)	Dose (MSV)
1	Kundiga	Granite	0.15	0.52	2.62
2	Ruwan dutse	Granite	0.25	0.31	1.55
3	Yimirdalang	Gneiss	0.18	0.37	1.86
4	Zange	Sandstone	0.15	0.63	3.17
5	Dali	Sandstone	0.43	0.47	2.35
6	Kanawa	Sandstone	0.71	0.46	2.31
7	Wuyo	Sandstone	0.18	0.28	1.38
8	Gubrunde	Granite	0.17	0.66	3.30
9	Peita	Gneiss	0.27	0.32	1.62
10	Garin wala	Granite	0.18	0.43	2.13
11	Tashar riga	Gneiss	0.3	0.41	2.07
12	Kwaya kusar	Gneiss	0.35	0.35	1.76
13	Danmala	Sandstone	0.73	0.28	1.41

Table 4 Comparison of NORM, Ra_{eq} , H_{ex} and annual effective dose obtained in this study and those elsewhere

Worker	Locality	^{226}Ra (Bq kg ⁻¹)	^{232}Th (Bq kg ⁻¹)	^{40}K (Bq kg ⁻¹)	Ra_{eq} activity concentration (Bq kg ⁻¹)	External hazard	Annual Effective dose (10 ⁻⁵ Sv)
IAEA (2003) ^b	World average (sandstone)	18–415	10–959	5–700	a	a	a
	World average (building stone)	1–500	1–310	76–7011	a	a	a
This work	Benue trough, Northeastern, Nigeria	11.33–74.56	26.64–153.38	30.06–1126.34	57.33–358.36	0.16–0.99	3.23–19.97
Singh et al. (2005)	Punjab and Himachal Pradesh, India	8.40–29.88	21.68–77.67	3.33–7.51	74.29–281.90	0.20–0.76	4.1–15.5
Arabi et al. (2015)	Zaria, Northwestern, Nigeria	20–56.7	8.4–187.9	51–127.3	32–267	a	a
IAEA (2003) NORM VII (China) ^b	Australia	19–24	11–69	23–100	a	a	a
	Germany	10–145	10–63	10–700	a	a	a
	Romania	Up to 557	Up to 170	a	a	a	a

^a Values not reported

^b Source: World Nuclear Association (updated in May, 2015) <http://www.world-nuclear.org/info/Safety-and-Security/Radiation-and-Health/Naturally-Occurring-Radioactive-Materials-NORM/>

ded for all the samples exceeded the set limit by NCRP, USNRC, WHO and poses great hazard; therefore, it is not suitable for use as building aggregate. Some measured activity concentration values for ^{226}Ra , ^{232}Th and ^{40}K are much higher than the world average values, which is a source of concern in terms of their radiological effect for dwellers of the area. The dispersion trend is observed to be consistent with an earlier trend, which reported higher concentrations of uranium in groundwater around Kundiga, Kanawa and Zona, some few kilometers

from Yimirdalang. It is also observed that the ^{226}Ra and ^{232}Th content of the rock samples studied is the primary control on observed ^{222}Rn and ^{220}Rn exhalation and emanation.

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