ASSESSMENT OF RADIOLOGICAL DETRIMENTS IN ROCK SAMPLES FROM SOUTH WEST OF THE KERRI–KERRI FORMATION UPPER BENUE TROUGH, NORTHEAST NIGERIA

Supported by tetfund TERTIARY EDUCATION TRUST FUND

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Received: December 05, 2016 **Accepted:** February 08, 2017

Abstract: There was speculation that the Kerri-Kerri Formation is a potential host of uranium mineralization hence the need to assess the radiological hazard in the rocks of the area. Activity concentrations of radionuclides ^{238}U, ^{232}Th and ^{40}K were measured by laboratory gamma spectrometry for rock samples collected from the southwest of the Kerri-Kerri Formation, Northeastern Nigeria. The average activity concentrations of ^{238}Ra, ^{232}Th and ^{40}K were found to be 27.00±2.50Bqkg^{-1},61.91±3.33Bqkg^{-1} and 91.70±8.47Bq kg^{-1}, respectively. These values were used to evaluate the radiological hazard parameters in the study area. The mean values of the radiological parameters; absorbed dose rate (D), radium equivalent activity (Ra_{eq}), outdoor annual effective dose equivalent (AEDE), radioactivity level index (I_γ), external hazard index (H_{ex}) and internal hazard index (H_{in}) were found to be 53.69 nGyh^{-1}, 122.60 Bq kg^{-1}, 0.006 mSv y^{-1}, 0.86, 0.331 and 0.404, respectively. These results were compared with values obtained for different countries of the world and were found to be lower than the world average values. On the basis of low levels of natural radioactivity, the study area can be considered as a less natural background radiation hazard area.

Keywords: Activity concentrations, dose rate, gamma-ray spectrometry, hazard index,

Introduction

Nuclear radiation has become a huge public concern all over the world, even though nuclear radiation is an inevitable part of our natural environment(Rashed-Nizam*et al*, 2014). The authors maintained that apart from cosmic rays, the soil of our earth is an important source of nuclear radiation. A number of natural radionuclides, namely Uranium (^{238}U), Thorium (^{232}Th) and their decay products (^{226}Ra, ^{212}Pb, etc.) and Potassium isotope (^{40}K) are observed as inherent soil contents. These natural radionuclides contribute to the radiation exposure, externally through gamma ray emission and also internally through inhalation and the food chain (IAEA, 1989).

Ionizing radiations from natural radionuclides in the environment can disrupt life processes as a result of the natural radioactive materials that exist in rocks, soil, air, food and drinking water. According to the United Nations Scientific Committee on Effect of Atomic Radiation (UNSCEAR, 2000), all humans are exposed to radiations from cosmic sources and the earth's crust to an average level of 2.4 mSv radiation dose per year. It is therefore important to assess the effects of radiation exposure due to terrestrial and extraterrestrial sources in the environment. Radioactivity in the environment is mainly due to the presence of long-lived radionuclides; of ^{238}U, ^{235}U and ^{232}Th series and ^{40}K. Their distribution in the earth crust depends on the type of rock formation below the earth's crust (Mujahid and Hussain, 2010).

Naturally occurring radionuclides are known to be present in varying proportions in rocks and soil of different geological formations around the world. Due to weathering and other environmental processes, radionuclides in rocks and soils may accumulate in sediment and dissolve into drinking water, thereby leading to human exposure(Raymond *et al*., 2013). The knowledge of radionuclides distribution and radiation levels in the environment is important for assessing the effects of radiation exposure due to both terrestrial and extraterrestrial sources (Rajesh *et al*., 2013).Oshin (1986) suggested that the Kerri–Kerri and Bima Formations are potential hosts of uranium mineralization. The present study is aimed at estimating the levels of radiation exposure due to the natural radionuclides (^{238}U, ^{232}Th and ^{40}K)in rock samples collected from the southwest of Kerri-Kerri Formation, Northeastern Nigeria.

Materials and Methods

Description of study area

The area of investigation lies within the Kerri–Kerri Formation located southwest of Gombe town, Northeastern Nigeria. It is bounded by latitudes 10º 00' 47.2" N to 10º 01' 19.4" N and longitudes 11º 00' 57.5" E to 11º 01'29.7"E covering an area of 2.25 Km^2.Vegetation in the study area isGuinea Savannah grassland withconcentration ofwoodland. Theclimate condition exhibit two different seasons namely: a short wet season and a prolonged dry season. Temperatures during the day remain constantly high while humidity is relatively low throughout the year, with little or no cloud cover (OnlineNigeria.com.2015). The mean monthly temperatures during the day exceed 30 ºC while the mean temperature at night falls to 22ºC(OnlineNigeria.com. 2015). The topography of the study area is flat open plains. The ethnic groups in and arount the study area are Fulani and Tangale whose main occupation is farming.

The Kerri-Kerri Formation located in the Upper Benue Trough was laid down in a Continental environment ranging from Lacustrine to Deltaic (Carter *et al*,1963) which was derived from the weathering of the basement rocks as well as of Cretaceous sedimentary formations. The Kerri-Kerri Formation which overlies the gritty clay of the Gombe Sandstone unconformably consists of coarse grained cross bedded quartz arenite (Odedede and Adaikpoh, 2011). The stratigraphy of the Gongola Basin in the Upper Benue trough consists from the youngest to the oldest; Alluvium, Kerri-Kerri Formation, Gombe Sandstone, Pindiga Formation, Yolde Formation, Bima Group and the Basement Complex rocks (Odedede and Adaikpoh, 2011).

Collection of rock samples

Rock samples were collected at thirty six (36) locations within the study area. The samples are packed in plastic bags and covered as tight as possible. The rock samples were collected along grid points for the purpose of compositing. The study area was divided into nine cell units. For each cell, four discrete rock samples collected were mixed into a single homogenized sample, thus, nine (9) composite samples were formed for the laboratory gamma spectrometric analysis. The

composite samples were labeled 'CMP1' to 'CMP9'.The locations of the rock samples collected and composite samples formed are presented in Fig. 1.

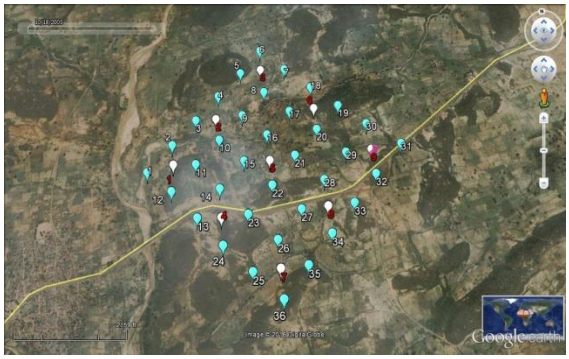

Fig. 1: Satellite view of the study area and measurement locations (Green spots are locations of rock samples collected while white spots are composite rock sample locations).

Laboratory procedure

The laboratory gamma ray spectrometric analysis of the rock samples was carried out at the Center for Energy Research and Training (CERT), Ahmadu Bello University Zaria, by using a coaxial high purity germanium (HPGe) detector (EG&G ORTEC® *p*-type, model GEM-70-S), having 60% relative efficiency with respect to 7.62 × 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 energyresolution for the ^{60}Co γ-ray line (1332 keV) at a source to detector top distance of 25 cm was1.85 keV (in terms of full width at half maximum, FWHM).

The dried samples were thoroughly grinded and distinctly packed in plastic containers measuring 8.0 cm in diameter by 6.5 cm in height and width made to fit on the high purity germanium detector. The packaging in each case was triply sealed. The sealing process included smearing of the inner rims of each container lid with Vaseline jelly, filling the lid assembly gap with candle wax to block the gaps between lid and container and tight sealing the lid container with masking adhesive tape. They were left for 21 days to allow radon and its short-lived progenies attain secular equilibriumas used in (Uosif, 2007). Spectrum of every sample was collected for 29,000s. The spectra acquired were analyzed using GammaVision®-32 software(EG&G ORTEC, v.6).

Calculation of Radiological Hazard Indices

To quantify the radiation detriment to members of the public and the environment as a result of the activity concentration of the three radionuclides in the rock samples, radiological parameters were calculated and used as radiological indicators to estimate the radiological implications of the use of sediments from the Kerri-Kerri Formation.

Radium equivalent activity

The activity concentrations of the radionuclides in Bq kg^{-1}presented in Table 1 were used to evaluate the radium equivalent (Ra_{eq}) index using equation (1) by Beretka& Mathew (1985),

$$Ra_{eq}(Bqkg^{-1}) = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad (1)$$

Where: A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bqkg^{-1}, respectively.

Table 1: Activity concentrations of ^{238}Ra, ^{232}Th and ^{40}Kof rock samples from the study area

Sample ID	Location	^{226}Ra (Bq Kg^{-1})	^{232}Th (Bq Kg^{-1})	^{40}K (Bq Kg^{-1})
CMP 1	10°00'48.60"N 11°01'03.00"E	29.18 ± 1.73	43.15 ± 9.89	127.68 ± 2.87
CMP 2	10° 01'01.20"N 11° 01'15.60"E	25.78 ± 1.51	45.23 ± 9.65	291.53 ± 28.66
CMP3	10° 01'15.60"N 11° 01'30.00"E	13.91 ± 0.63	69.10 ± 1.82	52.41 ± 6.53
CMP 4	10° 00'32.40"N 11° 01'15.60"E	11.06 ± 3.94	89.17 ± 1.12	64.85 ± 7.10
CMP 5	10° 00'46.80"N 11° 01'30.00"E	23.99 ± 2.32	82.33 ± 0.91	70.30 ± 9.08
CMP 6	10° 01'01.20"N 11° 01'44.40"E	39.55 ± 1.97	48.69 ± 2.78	88.49 ± 4.67
CMP 7	10° 00'18.00"N 11° 01'30.00"E	44.26 ± 3.13	67.16 ± 1.48	38.10 ± 3.73
CMP8	10° 00'32.40"N 11° 01'44.40"E	11.24 ± 3.38	72.29 ± 1.25	27.75 ± 7.79
CMP9	10° 00'46.80"N 11° 01'58.80"E	44.03 ± 3.86	40.10 ± 1.08	64.23 ± 5.75

Absorbed dose rate

The external exposure to radiation arising from naturally occurring radionuclides can be determined in terms of the absorbed dose rate in air at 1 m above the ground surface to assess the radiological risk. The mean activity concentrations of ^{238}U, ^{232}Th and ^{40}K (in Bqkg^{-1}) in the rock samples were used to compute the absorbed dose rate (D), using the conversion factor given by Beretka and Mathew (1985) and UNSCEAR (2000) as follows:

$$D(nGyh^{-1}) = 0.462A_{Ra} + 0.604A_{Th} + 0.0417A_K \quad (2)$$

Where: D is the absorbed dose rate in nGyh^{-1}.

External and internal hazard indices

The external hazard (H_{ex}) and internal Hazard (H_{in}) indices were calculated using equations (3) and (4) byBeretka and Mathew (1985). These are hazard indicators that predict the external and internal detriment of natural radiationfrom ^{238}U, ^{232}Th and ^{40}K. The H_{ex} equivalent upper limit of 1 mSvy^{-1} was calculated by Svoukisand Tsertos (2007). Internal hazard index (H_{in}) is used for consideration of the internal radiation from radon ^{222}Rn and its daughter in building materials (El-Taher, 2010).

$$H_{ex} = \left(\frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810}\right) \leq 1 \quad (3)$$

For safety, the value of H_{ex} should be below one, which corresponds to the upper limit of Ra_{eq} (370 Bqkg^{-1}).

$$H_{in} = \left(\frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810}\right) < 1 \quad (4)$$

Gamma radiation representative level index (I_γ)

An estimate of the gamma radiation hazard levels associated with natural radionuclides in the rock samples was calculated based on radiation hazard index I_γ (OECD, 1979)

$$I_\gamma = \left(\frac{A_{Ra}}{150} + \frac{A_{Th}}{100} + \frac{A_K}{1500}\right) \quad (5)$$

For radiation hazard to be insignificant, the value of these indices must be less than unity.

Annual effective dose equivalent (AEDE)

The measurement of the concentrations of radionuclides in the environment due to terrestrial gamma radiation from ^{238}U, ^{232}Th and ^{40}K can be estimated by the average outdoor conversion coefficient from absorbed dose rate in air and the average annual effective dose equivalent (AEDE). In the UNSCEAR (2000)reports, a value of 0.7 SvGy^{-1} was used for the conversion coefficient from absorbed dose in air to effective dose received by adults, and 0.2 SvGy^{-1}for the outdoor occupancy factor. The components of the annual effective dose in mSvy^{-1} are determined as follows:

$$AEDE_{out} = D(nGyh^{-1})x\ 0.7\ (SvGy^{-1})x\ 0.2\ x\ 8760\ (hy^{-1})x10^{-6} \quad (6)$$

$$AEDE_{in} = D(nGyh^{-1})x\ 0.7\ (SvGy^{-1})x\ 0.8\ x\ 8760\ (hy^{-1})x10^{-6} \quad (7)$$

Where: D is the dose rate and 8760 are hours in a year. The corresponding worldwide average values of $AEDE_{out}$ and $AEDE_{in}$ are 0.08 mSv, 0.42 mSv, respectively (UNSCEAR, 2000).

Results and Discussions

The activity concentration of ^{226}Ra in the rock samples varied from 11.06 ± 3.94 Bqkg^{-1} to 44.26 ± 3.13 Bq kg^{-1} with an average value of 27.00 ± 2.50 Bqkg^{-1}. The range of measured activity concentration of ^{232}Th varied from 40.10 ± 1.08 Bqkg^{-1} to 89.17 ± 1.12 Bq kg^{-1} with an average value of 61.91 ± 3.33 Bqkg^{-1}. The activity concentration of ^{40}K varied from 27.75 ± 7.79 Bqkg^{-1}to 291.53 ± 28.66 Bq kg^{-1} with an average value of 91.70 ± 8.47 Bqkg^{-1}. As presented in Table 2, the average activity concentration of ^{226}Ra and ^{232}Th obtained in this study fall within the range of values obtained in other countries as published by ICRP (1990) and UNSCEAR (2000). Also, we see from the Table 1 that, the activity concentration of the radionuclides in the present study area are in the order ^{238}Ra<^{232}Th<^{40}K.

Table 2: Activity concentrations of ^{238}Ra, ^{232}Th and ^{40}K of rock samples from the study area andthose obtained in other countries and the worldwide average (ICRP, 1990; UNSCEAR, 2000)

Country	Activity concentration (Bq kg^{-1})					
	^{238}Ra		^{232}Th		^{40}K	
	Range	Average	Range	Average	Range	Average
Egypt	5-64	17	2-96	18	29-650	320
USA	8-160	40	4-130	35	100-700	370
China	2-440	32	1-360	41	9-1800	440
Japan	6-98	33	2-88	25	15-990	310
Malaysia	38-94	67	63-110	82	170-430	310
India	7-81	29	14-160	64	38-760	400
Iran	8-55	28	5-42	22	250-980	640
Spain	6-250	32	2-210	33	25-1650	470
Greece	1-240	25	1-190	21	12-1570	360
Ghana**	5-9	7	6-8	7	219-453	480
Worldwide average	17-60	35	11-64	30	140-850	400
Present study area	11-44	37	40-89	62	28-292	92

** --- (Raymond *et al*., 2013)

Table 3: The radiological hazard indices for rock samples from the study area

Sample ID	Ra_{eq} (Bq Kg^{-1})	D (nGyh^{-1})	H_{ex}	H_{in}	I_γ	$AEDE_{out}$ (mSv y^{-1})	$AEDE_{in}$ (mSv y^{-1})
CMP 1	100.72	44.87	0.272	0.351	0.711	0.055	0.220
CMP 2	112.91	51.39	0.305	0.375	0.819	0.063	0.252
CMP 3	116.76	50.35	0.315	0.353	0.819	0.062	0.247
CMP 4	143.56	61.67	0.388	0.418	1.009	0.076	0.303
CMP 5	147.14	63.74	0.397	0.462	1.030	0.078	0.313
CMP 6	115.99	51.37	0.313	0.420	0.810	0.063	0.252
CMP 7	143.23	62.60	0.387	0.507	0.992	0.077	0.307
CMP 8	116.75	50.01	0.315	0.346	0.816	0.061	0.245
CMP 9	106.32	47.24	0.287	0.406	0.737	0.058	0.232
Minimum	100.72	44.87	0.272	0.346	0.711	0.055	0.220
Maximum	147.14	63.74	0.397	0.505	1.030	0.078	0.313
Average	122.60	53.69	0.331	0.404	0.860	0.066	0.263

Presented in Table 3 are theradiological hazard indices obtained in this study. The Ra_{eq} for rock samples in the study area was between 100.72 Bqkg^{-1} and 147.14 Bqkg^{-1} with an average value of 122.60 Bqkg^{-1}. It is observed that for all the rock samples analyzed, the radium equivalent activity value is well within the permissible limits of 370 Bqkg^{-1}.

The absorbed dose rate obtained varied from 44.87 to 63.74 nGyh^{-1}, with an average value of 53.69 nGyh^{-1}. The mean absorbed dose rate was lower than the World average value of 55 nGyh^{-1}(UNSCEAR, 1988). The calculated external hazard index (H_{ex}) varied from 0.272 to 0.397, with an average value of 0.331. The calculated average value was less than 1. The

internal exposure by radon and its progeny is controlled by the internal hazard index (H_{in}) which ranged between 0.346 and 0.505 with an average value of the 0.404. The average value was also less than 1. The calculated I_γ values for the rock samples varied from 0.711 to 1.030 with an average of 0.860. The calculated mean value was lower than the internationallyrecommended value ($I\gamma < 1$). The calculated indoor and outdoor (AEDE) effective dose values were also lower than the corresponding worldwide values of 0.08 and 0.42 $mSvy^{-1}$, respectively (UNSCEAR, 2000). A comparison of the radiological hazard indices obtained in this work with internationally recommended limits are presented in Table 4.

Table 4: Mean radiological hazard indices obtainedin thisstudy compared with internationally recommended limits

Radiological indices	Internationally recommended limits	Mean valuesobtained in this Study
Ra_{eq} (Bq Kg^{-1})	370	122.60
D ($nGyh^{-1}$)	55	53.69
H_{ex}	< 1	0.331
H_{in}	≤ 1	0.404
I_γ	< 1	0.860
$AEDE_{out}$ (mSv y^{-1})	0.08	0.066
$AEDE_{in}$ (mSv y^{-1})	0.42	0.263

Conclusion

The average concentrations for ^{226}Ra (^{238}U series), ^{232}Thand^{40}K obtained in this study were lower than the world average values. Also, the averages values of radiological hazard indices obtainedin this study were within the limits of internationally recommended values. It is therefore concluded that no harmful radiation effects were posed to the environment and population who live in the study area.

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