

RESEARCH ARTICLE

Evaluation of environmental natural radioactivity levels in soil and ground water of Barkin Ladi, Plateau state, Nigeria.

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Graphical abstract



Abstract

Gamma spectrometry was used to measure gamma dose rate in air and to determine the activity concentration of naturally occurring radionuclides viz. 226 Ra 232 Th and 40 K in soil samples collected across the geological formations of Barkin Ladi. Dose rates measured in-situ ranged from 5 nGy h^{-1} to 1265 nGy h^{-1} with a mean value of 325 nGy h^{-1}. The activity concentrations of 226 Ra ranged from 27 to 327 Bq kg-1, 34 to 457 Bq kg-1 for 232 Th and 43 to 1055 Bq kg-1 for 40 K. Their mean values are significantly higher than their corresponding global average values. Annual effective dose due to exposure to gamma dose is calculated at 0.34 mSv y-1. Mass concentrations of uranium (238 U) and thorium (232 Th) determined in groundwater samples using Inductive Coupled Plasma Mass Spectrometry (ICP-MS) ranged from 2.5 to 35 μ g I^-1 for 238 U and 0.5 to 15 μ g I-1 for 232 Th. Ingestion effective dose varies between 10.5 and 142 mSv y-1 for 238 U and between 0.34 and 10.2 mSv y-1 for 232 Th. Mass concentration of three water samples exceed the toxicity limit of U in groundwater provided by WHO. The result revealed that human risk due to ingestion of groundwater is from chemical toxicity rather than radiological effects. The results of this work will be useful for radio-geochemical investigation and groundwater resources management.

Keywords: Radioactivity, ²²⁶Ra, ²³²Th and ⁴⁰K, geological unit, ICP-MS, Barkin Ladi © 2018 Penerbit UTM Press. All rights reserved

INTRODUCTION

The occurrence of natural radioactivity in the environment depends on the types of rock formations, soils (Mangset *et al.*, 2014) as well human activities in the area. Human activities include tin mining and use of fertilizers in agriculture. They are mainly release by primordial radionuclides such as the decay series of ²³⁸U and ²³²Th; and non-decay series ⁴⁰K contained in the earth crust most often referred to terrestrial radionuclides (Langmuir, 1978). Higher activity concentration of these radionuclides are associated with rocks that comprised of intrusive materials (UNSCEAR, 2000), such as granitic type of igneous rocks and pegmatites due the presence of uranium and thorium in considerable amount compared to low grade metamorphic and some sedimentary rocks (Aieta *et al.*, 1987; Zapecza and Szabo, 1986).

Natural radionuclides exist in groundwater as a result of interaction between water and the rocks hosting the aquifer. Consumption of groundwater with elevated amounts of natural radionuclides may increase the radiotoxicity risk to human and internal exposure to radiation caused by the decay of the natural radionuclides taken into the body through ingestion as well as inhalation. The decay process leads to the release of several alpha and beta particles which are responsible for the total radiation dose received from natural radioactivity as well as artificial (Karahan G. *et al.*, 2000). Relationship between radium in groundwater, local geology, and the total dissolved solids content (TDS) was investigated for North Carolina (USA) by Michel (1991). In Nigeria, information on natural environmental radioactivity is sparse and limited; and no effort has been made to carry out an extensive field survey to cover its entire communities (Jibiri, 2001).

Barkin Ladi hosts a quite number of old tin mining locations which has drawn the attention of many researchers to investigate the levels of natural of radioactivity in the area as result of past tin mining activities for decades. Extraction and processing of tin minerals may cause the incorporation of radionuclides into the hydrosphere through surface or ground water (Pujol Li and Sanechez-Cebeza, 2000). Studies on natural radioactivity have been conducted previously in this area (Babalola, 1984; Oresegun and Babalola, 1990; Oresegun and Babalola, 1993; Sanni *et al.*, 1985) but none them considered the incorporation geological settings of the area for this type of study. Mangset, *et al.* (2014) reported elevated level of gross alpha and beta activity concentration in groundwater for the same area. Recently, similar studies in the same region reported relatively higher dose rates (Abba *et al.*, 2017a; Abba *et al.*, 2017c) compared to other part of Nigeria (Jibiri and Farai, 1998).

Therefore, this study aims to determine the natural radioactivity levels in soil and groundwater based on geological formations in Barkin Ladi. This study will screen the radioactive background of the area which will enable the future estimate of radiological impact on the environment if any.

The study area

Barkin Ladi local government area (LGA) is geographically located between the latitudes $9^{0}25'08' - 9^{0}42'48''$ north, and the longitudes $8^{0}50'22'' - 8^{0}58'22''$ east, in Plateau state of Nigeria. It has a total land area of 1,032 km² and a population of 175,267 (NPC, 2006). It is bordered on the north by Jos South and Jos East LGAs, on the east by Mangu LGA, on the south by Bokkos LGA and to the west by Riyom LGA. The study area is characterised as tropical with temperature between 20°C to 22°C (Macleod *et al.*, 1971).

Barkin Ladi is underlain by five bedrock formations broadly classified under three geological groups; namely basement complex of Precambrian age, Younger granites of Jos Plateau and Volcanic rocks of quaternary and tertiary ages as given in Table 1 and shown in Fig. 1. The basement complex is made-up of migmatite-gneiss-quartzite complexes which predominantly underlies the study area and in some places intruded by older granites, diorites, charnockite etc. The younger granites of Jos Plateau is of late Palaeozoic age (mostly Biotitegranites) are unconformably overlain by Cretaceous and younger sediments which are now partly exposed by erosion. The volcanic rocks is of tertiary origin which is associated with older basalts, quaternary newer basalts, pumice, lava flows and ash deposits. Minerals of economic important such as tin and columbite are also present in the area (Macleod, et al., 1971). Heavy radioactive minerals such as ilmenite, monazite and zircon are commonly found in reasonable quantities in tin mineral. These minerals contain radioactive elements, thorium and uranium (Hewson, 1996). Thus, appropriate radiation protection measures is timely required wherever these minerals are found, mined, processed and refined.In view of its geology and the decades of mining activities that took place in Barkin Ladi, measurement of natural radioactivity is timely required.

Table 1 Geological formations in Barkin Ladi.

Label	Geological unit	Composition
G1	Basalt, Trachyte and Rhyolite	Fine-grained, igneous rock composed mainly of plagioclase and pyroxene minerals overlaid by a thick cap of lateritic ironstone.
G2	Older Basalts	Fine-grained, igneous rock mixed with Posstasium Felspar.
G3	Older Granites	Composed of felsic and felspatic acidic intrusive igneous rock.
G4	Younger Granites	Composed of grainy biotite microgranites and some basic rocks.

G5 Basement complex Sedimentary basins that are metamorphic or igneous in origin.



Fig. 1 Geological units of Barkin Ladi.

Direct measurement of gamma dose rate

Gamma dose rates measurement was conducted (in-situ) 1 m above ground surface using a portable NaI(Tl) survey meter manufactured by Ludlum measurement (Olise *et al.*, 2014). Measurement were conducted at 192 different location across the five geological formation of the area. Coordinates of measurement locations were recorded with the aid of Global Positioning System (GPS), Garmin eTrex model 10. Gamma dose rate readings were recorded in in μ R h⁻¹ which was later transformed to nGy h⁻¹ using a conversion factor 8.7 (Abba, *et al.*, 2017c). The gamma dose rates obtained by the field survey is from both cosmic and terrestrial sources.

Gamma ray spectrometry of soil samples

A total of thirty seven top (at 0-30 cm depth) soils samples were collected across the geological units and far away from public structure. Coordinates of the sampling locations were also recorded as shown in Figure 2. At the laboratory, soil samples were cleared of stones, organic matters and other debris, dried to constant weight in an oven at 105° C and then homogenized. The prepared samples were filled in airtight 500 ml standard Marinelli beakers to prevent escape of radon gas and kept for 30 days before radiometric analysis to achieve secular equilibrium between ²³⁸U and ²³²Th and their daughter products.



Fig. 2 Soil and groundwater samples locations.

Gamma emission from ²²⁶Ra, ²³²Th and ⁴⁰K were measured using with a coaxial high purity germanium detector (GC2018-7500 SL) with a relative efficiency of 20 %, and resolution of 1.8 keV at 1,332 keV gamma ray emission of 60Co. The detector is coupled to 8192 multichannel analyzer (MCA) with a high voltage bias of 4,000 V (DC) through preamplifier. Genie 2000 (VI.3) software from Canberra was used to collect and analysed the spectra. Point sources were used for energy calibration whereas a mixed source was used for the efficiency calibration in the same geometry as the soil samples. IAEA reference materials (RG-U, RG-Th and RG-K) in the same size and volume as the soil samples were used as standards for comparison. To strip out emission from background radiation sources, an empty standard beaker was counted for the same length of time (six hours). The value of Minimum Detectable Activity (MDA) was computed to be 13 Bq kg⁻¹ for ⁴⁰K, 1 Bq kg⁻¹ for ²²⁶Ra and 2 Bq kg⁻¹ for ²³²Th. The experiment was conducted at Nuclear Laboratory University Teknologi Malaysia.

At equilibrium, the energy peaks considered in the present analysis of the measured gamma-ray spectra are: 214 Pb (352 keV) and 214 Bi (609 keV), for 226 Ra and 208 Tl (583.1 keV) and 228 Ac (911.2 keV), for 232 Th and direct energy emission of 1461.8 keV by 40 K was used to determine its activity concentration. Total errors calculated due to activity measurement was typically in the range of 5-12%, these include counting statistical errors and other systematic errors such as uncertainties in calibrations.

Following the spectrum analysis, specific activity of each radionuclide in the soil samples was computed using equation 1(Ademola and Farai, 2006; Isinkaye, 2013).

$$C(Bqkg^{-1}) = \frac{C_n}{\varepsilon p_{\nu} M_s} \tag{1}$$

Since in-situ gamma dose rate measurement includes both cosmic and terrestrial gamma radiations, therefore, contribution of terrestrial

radionuclides to the background gamma dose from activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K was obtained by equation 2. The conversion factors used to estimate the absorbed dose rate is given by UNSCEAR (2000).

$$D_{R} = (0.462A_{Ra} + 0.604A_{Th} + 0.0417A_{K})nGyh^{-1}$$
⁽²⁾

where, 0.462, 0.604 and 0.0417 are the activity concentration conversion factors of 226 Ra, 232 Th and 40 K, respectively to dose rate in air (UNSCEAR, 1998).

Annual effective dose (AED) outdoor due exposure to background gamma radiation dose for an individual in the area was calculated by the equation below:

$$AED (mSv y^{-1}) = D_R x D_E x F_{occ} x 24 x 365 x 10^{-6}$$
(3)

where D_R is the dose rate in nGy h⁻¹, D_E is the is the conversion coefficient from absorbed dose in air to effective dose received by an adult given as 0.7 (Sv Gy⁻¹) and F_{occ} is the the outdoor occupancy factor given by 0.2 (UNSCEAR, 2000).

Measurement of uranium and thorium in groundwater

A total of 15 groundwater samples from boreholes and hand dug wells were collected across the geological units for the analysis of uranium (²³⁸U) and thorium (²³²Th) mass concentrations. Samples were collected in clean plastic containers (100 ml) at the sites and were stabilized with 5 ml of nitric acid per litre and transported to laboratory for preparation and analysis. Locations of the sampling were recorded using GPS as shown in Fig. 2. Samples preparation and measurement were performed at Industrial Research Laboratory Universiti Teknologi Malaysia.

Inductive Coupled Plasma Mass spectrometry (ICP-MS) was used to determine the mass concentration of 238 U and 232 Th. The instrument is manufactured by Perkin Elmer Corporation with model number NeXION 350 X. For reliability, each sample was measured in triplicate to check the reproducibility of the results. The samples and the calibration standards were measured for 55 min. Syngistix software was used for data collection and intensity analysis. The lowest detection concentration of the instrument was 0.285 ng l⁻¹ for 232 Th and 0.323 ng l⁻¹ for 238 U.

Annual ingestion effective dose equivalent due groundwater consumption is evaluated using conversion factors and annual water consumption rate of an adult proposed by US Environmental Protection Agency (US-EPA). The equation used is as follows:

$$ADR_{w} = A_{R} \times IDF \times I_{w} \tag{4}$$

where ADR_w is the annual effective dose equivalent, A_R is the activity concentration of ²³⁸U or ²³²Th, I_w annual water consumption rate (730 litres per annum) and IDF is the ingestion effective dose equivalent factor (4.5×10⁻¹ mSv Bq⁻¹ and 2.3×10⁻¹ mSv Bq⁻¹ for ²³⁸U and ²³²Th).

RESULTS AND DISCUSSION

Measurements of gamma dose rates

The gamma dose rates varies between 55 nGy h^{-1} measured over geological unit basement complex (G5) and 1265 nGy h^{-1} noted on younger granites (G4) with a gross mean value of 325 nGy h^{-1} , which is approximately six times the world average of 59 nGy h^{-1} (UNSCEAR, 2000). The mean is also higher than the value reported by Abba, *et al.* (2017c) for the same region. Dose rates distribution for this study is presented as frequency distribution curve in Fig. 3.



The activity concentration of the radionuclides were determined to be in the range of 27 to 327 Bq kg⁻¹ for ²²⁶Ra, 34 to 457 Bq kg⁻¹ for ²³²Th and from 43 to 1055 Bq kg⁻¹ for ⁴⁰K. Descriptive statistics of the activity concentration and the calculated dose rate is presented in Table 2. The mean values were found to be significantly higher than their corresponding world average values of 35 Bq kg⁻¹, 45 Bq kg⁻¹ and 420 Bq kg⁻¹ for ²²⁶Ra, ²³²Th and ⁴⁰K, respectively (UNSCEAR, 2000) and distinctly lower than values of 4200 Bq kg⁻¹ for ²²⁶Ra and 18000 Bq kg⁻¹ for ²³²Th reported by Arogunjo *et al.* (2009) for the same region. The variation could be due to the fact that samples were directly collected from tin mining locations for analysis in the earlier study.

Table 2 Descriptive statistics for the activity concentration of 226 Ra, 232 Th and 40 K and the calculated dose rate.

Statistic	²²⁶ Ra (Bq kg−¹)	²³² Th (Bq kg−¹)	⁴⁰K (Bq kg−¹)	Dose rate (nGy h−¹)
Mean	153	345	453	298
Range	27-327	34-457	43-1055	35-471
Std. Error	6	13	33	11
Std. Dev.	46	90	234	76
95% conf.	131-173	318-374	385-523	281-328
interval for				
mean				

The mean activity concentration of ²²⁶Ra, ²³²Th and ⁴⁰K for each geological unit is given in Fig. 6(a) and the calculated absorbed dose rate in Fig. 6(b). The highets activity concentration of ²²⁶Ra was measured in soil sample collected from areas of geological unit basement complex (G5) and lowest in soil sample from areas of older granites (G3). The higher concentration of ²²⁶Ra in basement complex could be linked to the substantial amount of U and Th contained in igneous rocks (UNSCEAR, 2000), while its lower concentration may be linked to the little layer of basaltic formation associated with the older granites in the region (Abba et al., 2017b). This results is in consistent with similar studies (Ramli et al., 2001; Saleh et al., 2013a). Highest mean activity concentration of ²³²Th was oberved in soil sample from areas of older granites and the lowest was in areas of younger granites (G4). The basic rocks mixed with the younger granites could be the reason for lower activity concentration of ²³²Th. For ⁴⁰K, the highest activity concentration was measured in soil sample from areas of older basalt (G2) which could be attributed to the mineral potassium felspar that is mixed with this formation. The lowest mean of ⁴⁰K is measured in soil sample from areas of younger granites.



Fig. 6. Activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K and dose rate for each geological unit presented in left and right places, respectively.

The activity concentration was used to calculate the terrestrial gamma radiation absorbed dose rate in air for the area and was found to ranged between 35 and 471 nGy h^{-1} with a mean value of 298 nGy h^{-1} . The dose rate in Barkin Ladi was found to be largely contributed by ²³²Th (69%) followed by ²²⁶Ra and ⁴⁰K contributing 25% and 6%, respectively. Figue 4 presents dose rate distribution map for the area. This estimates is comparatively higher than that reported by Obed *et al.* (2005) for eighteen majors cities in Nigeria. Their results of gamma dose rate ranged from 19 to 88 nGy h^{-1} with a gross mean of 42 nGy h^{-1} . The calculated mean dose rate of the current work is found to be 7% lower than that of the measured dose rate which represent the contributions from the cosmic and other sources.



Fig. 4 Isodose of gamma dose rate for the area.

A positive correlation (R = 0.923) was observed between the measured and the calculated dose rates as shown in Fig. 5. The non-zero intercept is assumed to a contribution from cosmic and other terrestrial radionuclides (Saleh, *et al.*, 2013a). The measured dose rates were corrected by multiplying with a factor of 1/0.851.



Fig. 5 Correlation between measured and calculated dose rates.

Mass concentration of ²²⁸U and ²³²Th in groundwater

Fig. 7 presents the mass concentration for the 15 water samples collected from various sources across the geological formations in Barkin Ladi. The mass concentration of 238 U and 232 Th in the groundwater samples ranged from 2.5 µg l⁻¹ (W1) measured in a water sample from areas of geological formation basalts, trachyte and rhyolite (G1) which consist of lateritic ironstone to 35 µg l⁻¹ (W11) noted in an aquifer confined by older granites (G3), this rock composed of acidic intrusive materials which contained minerals hosting U and Th in reasonable quantity. Three water samples were found to exceed reference level of 30 µg l⁻¹ set by World health organisation (WHO, 2011) and US Environmental Protection Agency (USEPA, 2008).

Mass concentration of 232 Th was found to ranged between 0.5 µg l⁻¹ and 15 µg l⁻¹ with highest measured in areas of younger granites (G4) and lowest in an aquifer hosted by basement complex (G5). Mass concentration of 238 U was observed to be higher than 232 Th in all the samples except W4 and W5, this could be linked to its high mobility in

all environmental conditions while stable nature of ²³²Th making it insoluble in aqueous solution (Saleh *et al.*, 2015).

The elemental concentrations of 238 U and 232 Th were converted to activity concentrations using conversion factors (1 µg I⁻¹ =12.35 mBq I⁻¹) for 238 U and (1 µg I⁻¹ = 4.06 mBq I⁻¹) for 232 Th (IAEA, 1989). The activity concentrations ranged from 31 mBq I⁻¹ to 432 mBq I⁻¹ for 238 U and ranged between 2 mBq I⁻¹ and 61 mBq I⁻¹ for 232 Th. Activity concentration levels in all the groundwater samples are generally lower than the refence value of 1 Bq I⁻¹ and 0.5 Bq I⁻¹ for 238 U and 232 Th, respectively.



Fig. 7 Mass concentrations of ²³⁸U and ²³²²Th in groundwater samples.

Annual mean effective doses due to exposure to gamma radiation dose was calculated at 0.34 mSv y^{-1} (equation 3). The annual effective dose due ingestion of groundwater in Barkin Ladi ranged between $10.5 - 142 \text{ mSv y}^{-1}$ for ²³⁸U and between $0.34 - 10.2 \text{ mSv y}^{-1}$ for ²³²Th (equation 4). Water samples from three sources have annual effective dose exceeding the referce limit of 0.1 mSv y⁻¹ provided by WHO (2011).

CONCLUSION

Gamma spectrometry was used to measured the external gamma dose rates and to determined the activity concentration 226 Ra, 232 Th and 40 K is soil across the geological formations of Barkin Ladi. Dose rates measured in-situ was found to ranged from 5 nGy h⁻¹ to 1265 nGy h⁻¹ with a mean value of 325 nGy h⁻¹. Activity concentration of 226 Ra, 232 Th and 40 K ranged from 27 to 327 Bq kg⁻¹, from 34 to 457 Bq kg⁻¹ and from 43 to 1055 Bq kg⁻¹, for 226 Ra, 232 Th and 40 K, respectively, while the calculated dose rates was found to ranged between 35 to 471 nGy h⁻¹ with a mean value of 298 nGy h⁻¹. Gamma dose rates was found to be largely (69%) contributed by 232 Th. Higher concentration of 226 Ra was meassured in areas of basement complex (G5), whereas that of 232 Th and 40 K were recorded in soil samples from areas of older granites (G3) and older basalt (G2), respectively.

Mass concentrations of ³⁸U and ²³²Th in groundwater samples were determined using Inductive Coupled Plasma Mass Spectrometry and the concentration ranged betwenn 2.5 and 35 μ g l⁻¹ for ²³⁸U and 0.5 μ g l⁻¹ and 15 μ g l⁻¹ for ²³²Th. Their corresponding activity concentration in the water samples ranged from 31- 432 mBq l⁻¹ for ²³⁸U and 2 mBq l⁻¹ to 61 mBq l⁻¹ for ²³²Th. Mass concentration of three water samples were found to exceed the reference limit of of 30 μ g l⁻¹ in drinking water recommended by WHO, but radiologically safe. Annual effective dose due to exposure to gamma dose was calculated at 0.34 mSv y⁻¹ and annual ingestion effective dose equivalent due to consumption of groundwater ranged between 10.5 and 142 mSv y⁻¹ for ²³⁸U and 10.2 mSv y⁻¹ for ²³²Th.

The results presented, will be useful for future radio-geochemical investigations in the area and also useful for water resources management and protection of groundwater resources.

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