Radionuclides and radon levels in soil and ground water from solid minerals-hosted area, south-western Nigeria

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Abstract: In this study, we determine the activity concentrations of natural radionuclides, 238U, 232Th and 40K, in soil from solid mineral-hosted site. The dose rates were calculated and radon concentration in the ground water system was measured. The radionuclides were determined using the HPGe detector-driven gamma spectrometric technique and the RAD7/RAD H2O system was used to determine the radon concentrations. Some radionuclides in the samples including the mineral, Mica, and some of the ground water samples, especially from well and borehole, had high activity and radon concentrations. We concluded that the two mica-granite geological structure and the depth of the source is a predominant factor for high radon concentration and definitely contributed to the level observed in the borehole sources. The relatively high levels of the radionuclides and radon indicate certain level of health risk. Though the effective dose seemed low, effects of prolonged exposure to radiation is still possible.

Subjects: Environment & Health; Environmental Health & Safety; Environmental Studies & Management

Keywords: artisanal mining; TENORM; RAD7/RAD H2O; thorium; radon

ABOUT THE AUTHORS
Felix S. Olise has a PhD in Engineering Physics. In general, his research work has been on the development and applications of atomic and nuclear physics based analytical techniques: X-ray fluorescence, particle induced X-ray emission, instrumental neutron activation analysis and gamma spectrometry. Research papers, which are by either experimental or theoretical approaches, are aimed towards an improved understanding of the behaviour, transfer and distribution of elemental signatures and/or radionuclides in natural systems. For radioactivity studies, applications range from using environmental radionuclides as mechanistic or timescale tracers of natural processes to assessments of the radio-ecological or radiological effects of environmental radioactivity.

Deborah M. Akinnagbe and Olugbenro S. Olasogba have MSc in radiation Physics. While Deborah study was on the radon level in ground water of an area rich in certain minerals, Olugbenro emphasised on the activity levels of the primordial radionuclides associated with certain minerals in the same area.

PUBLIC INTEREST STATEMENT
Minerals are naturally associated with radiation emitting substances (radionuclides) and toxic metals. Mining the minerals, especially in an unprofessional (artisanal) way, brings these radionuclides and toxic metals into direct contact with the miners, surface soils, water bodies, etc., and even at elevated levels. The impacted environmental media, therefore, becomes a route of exposure (vulnerability) of the public to the radionuclides and metals through inhalation of the radon gas, contaminated tiny particles, ingestion through water, plants and animals, etc. Soil and water samples from the areas hosting the minerals have been analysed using relevant radiation detection facilities. The detected and relatively high levels of the radionuclides and radon indicate certain level of health risk. Though the effective dose seemed low, effects of prolonged exposure to radiation is still possible.
1. Introduction

Continuous exposure to ionising radiation from natural sources is certain and this may exceed those from all man-made sources combined. The natural terrestrial gamma radiation dose rate is an important contribution to the average dose rate received by the world’s population. In assessing the health risk to a population, it is important to estimate the radiation dose distribution. This serves as reference in documenting changes to environmental radioactivity in soil due to anthropogenic activities. Exposure to radiation is either directly or from the accumulation of radionuclides in the body through inhalation or ingestion. Therefore, investigating the levels of radionuclides and their distribution in the environment provides essential radiological information.

Radionuclides in soil and rock are not evenly distributed; hence natural radioactivity depends primarily on the geological formations and geographical conditions, giving rise to the various levels of the radionuclides in such media (Avwiri, 2005; UNSCEAR, 2000). Mining and other industrial activities result in large volumes of materials containing naturally occurring radioactive materials (NORM). Mining and processing of the ores generates waste that is richer in the radionuclides (Olise et al., 2011), and as such called technologically enhanced naturally occurring radioactive materials (TENORM). The natural background radiation, which may consequently increase to significant levels, is of great concern to radiation protection regulatory bodies.

The abandoned mining sites had played host to large-scale mining but now hosts artisanal small-scale mining. These sites witness manual excavation of soil and sediment that contains naturally occurring radionuclides in significant amounts. This, which is an external radiation exposure pathway to the population, has been a subject of study in recent times (Abdulkarim & Umar, 2013; Ademola & Obed, 2012; Girigisu et al., 2014; Ibrahim et al., 2013; Innocent et al., 2013; Jibiri & Esen, 2011; Olise, Owoade, Olaniyi, & Obiajunwa, 2010; Tubosun et al., 2014). The use of the huge generated wastes, especially in the radionuclide-rich cassiterite-hosted Jos Plateau area, as building materials and other developmental projects is a common practice (Olise et al., 2010, 2011). This is particularly dangerous since detailed studies on indoor radiation doses and effects on the inhabitants of prolonged exposures are not likely to have been carried out (Olise et al., 2011). There are various efficient ways of radionuclide measurements in water resources to obtain reliable data. The distribution of the radionuclides like uranium in surface and ground water resources has also received some attention (Waseem, Ullah, Rauf, & Ahmad, 2015). This is in view of their toxicity in their various chemical forms and their contribution to water-related diseases. The measurements of radon have been utilised in some other application. Zhang, Zhang, Wu et al. (2014) have investigated mining-induced fractures using the on-site radon detection method. The application of the geophysical-chemical properties of radon in mining engineering was explored as a potential solution to the shortcomings of existing surveying methods. The presented method could be used as an indirect technical support to increase the safety of coal mining by acting as a simple, fast and reliable method of detecting mining-induced fractures in overlying strata. Xue et al. (2008) carried out detailed investigation in applying the surface-based $^{222}$Rn technique to locate subsurface coal heatings in Australia. The exact location of the heatings that occur in usually inaccessible deep locations is a key to allowing effective control measures to be taken over significant problems posed by subsurface coal heatings in many coal mines. Also, the separation process of radon based on its geophysical-chemical properties has been presented (Zhang, Zhang, Wang et al., 2014). Considering the geological conditions of mining, the authors established the mathematical model of radon migration in underground multilayer strata to investigate the distribution law of radon concentration in such strata. The authors reported that the distribution of radon concentration in the strata is affected by both the properties of the strata and the depth of cover and that the radon concentration law varies at different depths even within the same layer stratum.

Presently, mining in Nigeria including the study area, is carried out by illiterate artisans across all the age range. The crude methods of obtaining the minerals expose the miners, immediate and adjoining environments to dust with high levels of radionuclides and heavy metals. High level of lead poison and eventual death of miners have been reported in two to three towns in Northern
Nigeria. The miners brought soil and stones rich in certain minerals home for further processing. As an immediate consequence, this method of mining exposes the community, including farmlands and water bodies to contamination. Also, the tailings (waste) is exposed to wind and the prevailing weather conditions resulting in further transportation of particles containing radionuclides and heavy metals to once uncontaminated areas. The clay waste from some parts of the mines is used by the locals in pottery.

The populace still rely on untreated ground water (shallow wells and boreholes) for drinking and other household activities. Ground water, particularly from areas underlain by uranium bearing rocks, has been reported to contain high radon concentrations (UNSCEAR, 2006). Radon has been described to be the second most frequent cause of lung cancer after cigarette smoking. Exposure to radon in water or air over a long period of time is believed to increase one’s life time risk of developing cancer. When radon-rich ground water is used as drinking water, people are exposed both through water consumption and inhalation.

Prior to the commencement of full industrial mining operation, it would be necessary to establish the levels of the naturally occurring radionuclides. This work reports an assessment of the level of radionuclides in the soil, radon in the ground water and the evaluation of the possible radiological health hazards from the impact of the present artisanal mining activity in the study area.

2. Materials and methods

2.1. The study area

The study area, Ijero, is within the south western part of the Nigerian Precambrian basement complex (Ale et al., 2014; Jimoh, 2011) and within latitude 7°42.0611’ N and longitude 5°17.009’ E. It has the pegmatite rock as the major type of rock, occurring as ridges and lowland. The area has an elevation ranging from 1,218 to 1,469 m, above the sea level and has an average elevation of 1,343 m (Odeyemi, 2014).

The present mining activity in the study area is similar to that in the Jos Plateau, which is purely artisanal in nature. Unlike the occasional movement of the ore-containing soil to other areas in Jos, the ore-containing soils are processed on the sites to obtain the minerals. The minerals mined from the mountain, called Oke-kusa in Ijero, include columbite-tantalite, tantalite ore (containing tantalum, iron and manganese), kaolin, migmamite, gneiss, quartz, schist, muscovite (mica), feldspar and gemstone (Ale et al., 2014; Okunlola & Akinlola, 2010). The geological map of Ijero-Ekiti is shown in Figure 1.

2.2. Soil sample preparation and gamma spectrometric analysis

2.2.1. Soil sample preparation

A total of 17 soil samples were collected by random sampling method from the mine sites. It included the tailings and the mined minerals. The soil samples collected were packed and labelled reflecting activity and location. The samples, obtained at 10-cm depth, were taken to the laboratory and allowed to dry to constant weight. The sample were subsequently sieved and sealed for 28 days to allow for radioactive secular equilibrium between the heads of the uranium and thorium decay series and their short-lived progenies (Veiga & Baker, 2004) before gamma counting.

2.2.2. Gamma spectrometric analysis

The gamma ray spectrometry analysis was carried out at the National Institute of Radiation Protection and Research (NIRPR), University of Ibadan, Nigeria. This was done using a Canberra vertical HPGe detector of length 10 cm and diameter 10 cm with a relative efficiency of 20.2%, enclosed in a lead shield of thickness 10 cm. The complete electronic instrumentation was connected to a PC-based multichannel analyser for gamma spectrum evaluation. The energy and efficiency calibration of the detector was carried out using the 1.33 MeV gamma line of Co-60 resulting to energy
resolution of 2.3 keV (FWHM) with a relative yield of 1.73%. Each sample was counted for a period of 18,000 s (5 h) and the gamma spectrum peak area analysis and quantification was carried out using the Genie 2000 software.

The activity concentrations of the radionuclides were determined using the equation:

$$A_C = \frac{C_n}{P \gamma M \varepsilon}$$  \hspace{1cm} (1)

where $A_C$ is the activity concentration of the radionuclide in the sample in Bq/kg, $C_n$ is the net count under the corresponding peak, $P \gamma$ is the absolute transition probability of the specific gamma ray, $M$ is the mass of the sample (kg) and $\varepsilon$ is the detector efficiency at the specific gamma ray energy.

### 2.2.3. Radiometric measurements

To ascertain the level of radiological hazard, the exposure to radiation from radionuclides present in the soil has to be determined. From the results of the activity concentration of the radionuclides, the average absorbed dose rate (nGy h⁻¹) in air, 1 m above ground, can be estimated. The mean activity concentrations of $^{238}U$, $^{232}Th$ and $^{40}K$ (Bq/kg) in the soil samples were used to calculate the absorbed dose rate, $D$ (UNSCEAR, 2000):

$$D = 0.429 A_U + 0.666 A_{Th} + 0.042 A_K$$  \hspace{1cm} (2)

where $A_U$, $A_{Th}$ and $A_K$ are the activity concentrations of $^{238}U$, $^{232}Th$ and $^{40}K$, respectively.
The outdoor absorbed dose rates in the air due to terrestrial gamma rays, 1 m above the ground, were calculated using the 40K, 232Th and 238U concentration values assuming that the contribution to the total dose from other radionuclides like 137Cs, 90Sr and 235U decay series can be neglected (Jacob et al., 1986). The radiological hazard to an individual from exposure is not directly accounted for by the absorbed dose in air at 1 metre above ground. Thus, considering the annual effective dose, the annual effective dose was calculated by taking into consideration the conversion coefficient from the absorbed dose in air to effective dose and indoor occupancy factor. The residents in Ekiti, Nigeria spend about 60% of their time outdoors. The annual effective dose was computed using the following components: the outdoor effective dose rate (OEDR in mSv y\(^{-1}\)):

\[
OEDR = D \times 24 \text{ h d}^{-1} \times 365.25 \text{ d y}^{-1} \times 0.6 (\text{OCPF}) \times 0.7 \text{ Sv Gy}^{-1} (\text{CONVC}) \times 10^{-6} \tag{3}
\]

where OCPF and CONVC are the occupancy factor and conversion coefficient, respectively. Since the indoor and outdoor occupancy factors need to sum up to 1.0, in other to make up 100% of the group’s exposure time, we used an indoor factor of 1 - 0.6 = 0.4 to calculate the indoor effective dose rate (IEDR in mSv y\(^{-1}\)):

\[
IEDR = D \times 24 \text{ h d}^{-1} \times 365.25 \text{ d y}^{-1} \times 0.4 (\text{OCPF}) \times 0.7 \text{ Sv Gy}^{-1} (\text{CONVC}) \times 10^{-6} \tag{4}
\]

### 2.3. Sample preparation and analysis for radon

#### 2.3.1. Sample preparation

A total number of 40 water samples from boreholes, wells and streams were collected and analysed for radon concentration. Samples of 500 mL were collected in pre-cleaned plastic pet bottles. The containers were cleaned with water and detergent and rinsed with distilled water to avoid contamination. For borehole sources, the samples were collected after turning on and allowing runoff for about 5–10 min. This was to allow the water temperature to stabilise and also for purging of trapped-in air. Also, the stream of flow was reduced to about 1/8 inch in diameter, bringing air bubbling to a minimum. Samples previously stored in tanks were not collected. The well samples were collected with the aid of bailers. For the stream samples, the bottles were carefully dipped in to collect the samples. The bottles were filled to the brim to prevent the formation of air pockets. The samples were marked and carefully labelled to reflecting the time of collection and location.

The analysis of the samples for \(^{222}\text{Rn}\) does not need any preservation. Since the maximum holding time is just 3 days, the samples were taken to the laboratory almost immediately in order to reduce the decay coefficient. Since the samples would give up radon readily at higher temperatures, they were transported to the laboratory in a bag containing ice in order to ensure low temperature condition of the samples.

#### 2.3.2. Sample analysis

The samples were analysed using the Durridge Inc. RAD7. It is a sophisticated, active radon detector that uses a computer-driven electronic detector with pre-programmed set-ups. The real-time monitoring device uses the alpha spectrometric technique. To analyse radon in water, an accessory, RAD H\(_2\)O is connected to RAD7. The analysis result is obtained in about 30 min, making it a relatively fast technique. The RAD7/RAD H\(_2\)O system is well documented (Durridge, 2013).

The collection, sealing and counting of the samples took 28 h. This led to the reduction of the radon concentration in the samples due to radioactive decay. To account for this reduction, the measured concentrations were corrected using the decay correction factor (DCF), from the time of sampling to the time of counting. DCF, a simple exponential function of a time constant, \(T_c\) is given by (Durridge, 2013):

\[
T_c = \frac{3.825 \text{ days} \times (\text{half-life of Rn - 222}) \times 24 (\text{hours day})}{\ln 2} = 132.4 \text{ hours} \tag{5}
\]
For a decay time, $T$ (h), the results were corrected back to the sampling time by multiplying with the DCF:

$$DFC = \exp \left( \frac{T}{T_c} \right) = \exp \left( \frac{T}{132.4 \text{ hours}} \right)$$  \hspace{1cm} (6)

### 2.3.3. Committed annual effective dose

Assessment of drinking water for radon is extremely important because of the recognised health risks, primarily as a cause of lung and stomach cancer (Crawford-Brown, 1990; Hurlburt, 1989; United States Environmental Protection Agency [USEPA], 1993). The committed annual effective dose (CAED) from ingestion was calculated thus (UNSCEAR, 1993):

$$CAED (\text{Sv}) = K \times G \times C \times t$$  \hspace{1cm} (7)

where $K$ is the ingesting dose conversion factor of $^{222}\text{Rn}$ (Sv Bq$^{-1}$), $G$ is the water consumption per day, $C$ is the concentration of $^{222}\text{Rn}$ (Bq L$^{-1}$) and $t$ is the duration of consumption. To calculate CAED, we assumed that an adult and a child drink directly from the source and consumes an average of 2 and 1 L of water per day, respectively (UNSCEAR, 1993). For adults, committed effective dose per unit intake from the ingestion of radon in water ($K$) is $10^{-8}$ Sv Bq$^{-1}$, and for children it is $2 \times 10^{-8}$ Sv Bq$^{-1}$ (Binesh, Mohammadi, Mowlavi, & Parvaresh, 2010; UNSCEAR, 1993).

### 3. Results and discussion

#### 3.1. Activity concentrations of $^{238}\text{U}$, $^{232}\text{Th}$ and $^{40}\text{K}$

The activity concentration of $^{238}\text{U}$, $^{232}\text{Th}$ and $^{40}\text{K}$ were determined in seventeen samples which include fourteen soil and three mineral samples (Table 1). The activity concentrations of $^{238}\text{U}$, $^{232}\text{Th}$ and $^{40}\text{K}$ vary from 12.90 ± 0.02 to 250.00 ± 0.02 Bq/kg, 1.00 ± 0.05 to 115.37 ± 0.02 Bq/kg and 45.56 ± 0.01 to 2610.27 ± 0.01 Bq/kg, respectively. The mean activity concentrations of both $^{238}\text{U}$ (70.57 ± 0.08 Bq/kg) and $^{232}\text{Th}$ (19.56 ± 0.03 Bq/kg) were also determined.

**Table 1. Activity concentrations of radionuclides in the samples (Bq/kg)**

| Sample | Longitude | Latitude | $^{238}\text{U}$ | $^{232}\text{Th}$ | $^{40}\text{K}$ |
|--------|-----------|----------|------------------|------------------|-----------------|
| 1      | E005°04'200" | N07°49'622' | 39.80 ± 0.04 | 0.99 ± 0.05 | 106.23 ± 0.01 |
| 2      | E005°04'207' | N07°49'626' | 58.63 ± 0.03 | 6.13 ± 0.03 | 162.36 ± 0.01 |
| 3      | E005°04'203' | N07°49'622' | 28.54 ± 0.02 | 115.37 ± 0.02 | 542.73 ± 0.01 |
| 4      | E005°04'206' | N07°49'636' | 226.21 ± 0.01 | 2.92 ± 0.04 | 575.2 ± 0.01 |
| 5      | E005°04'210' | N07°49'618' | 31.10 ± 0.03 | 9.57 ± 0.03 | 126.87 ± 0.01 |
| 6      | E005°04'214' | N07°49'619' | 247.46 ± 0.02 | 1.92 ± 0.05 | 621.59 ± 0.01 |
| 7      | E005°04'211' | N07°49'611' | 37.20 ± 0.02 | 8.30 ± 0.03 | 211.53 ± 0.01 |
| 8      | E005°04'211' | N07°49'615' | 54.42 ± 0.02 | 7.59 ± 0.04 | 1,287.24 ± 0.01 |
| 9      | E005°04'207' | N07°49'645' | 34.57 ± 0.02 | 26.67 ± 0.02 | 182.82 ± 0.01 |
| 10     | E005°04'214' | N07°49'632' | 54.18 ± 0.03 | 3.49 ± 0.05 | 220.34 ± 0.01 |
| 11     | E005°03'443' | N07°49'804' | 47.39 ± 0.04 | BDL | 2,609.27 ± 0.01 |
| 12     | E005°03'444' | N07°49'815' | 59.85 ± 0.04 | 17.55 ± 0.03 | 865.12 ± 0.01 |
| 13     | E005°03'444' | N07°49'815' | 12.92 ± 0.02 | BDL | 1,417.13 ± 0.01 |
| 14     | E005°04'125' | N07°49'649' | 55.66 ± 0.02 | 73.20 ± 0.02 | 299.62 ± 0.01 |
| Feldspar |            |          | 21.54 ± 0.99 | 5.15 ± 0.04 | 445.32 ± 0.01 |
| Quartz  |            |          | BDL | 5.76 ± 0.03 | 45.56 ± 0.01 |
| Mica    |            |          | 172.05 ± 0.01 | 27.60 ± 0.01 | 635.42 ± 0.01 |
| Mean (1–14) |      |          | 70.57 ± 0.08 | 19.56 ± 0.03 | 659.15 ± 0.01 |
kg) and ⁴⁰K (659.15 ± 0.01 Bq/kg) in the soil samples were observed to be, respectively, higher than the 40 (for ²³⁸U and ²³⁷Th) and 370 (for ⁴⁰K) Bq/kg world average values (UNSCEAR, 2000). The high level of the radionuclides poses a risk to the health of the miners and the populace through inhalation of dust containing the progenies of the radioactive elements. Another possible root of impact is through soil-to-plant transfer and surface runoff to the ground water system. However, ²³²Th had activity concentrations lower than the worldwide average of 40 Bq/kg in all the samples, including the minerals. The ²³⁸U activity concentration in mica was 172.05 ± 0.01 Bq/kg while quartz had relatively low activity concentrations of ⁴⁰K, ²³²Th and ²³⁸U. Mica and feldspar had relatively high-activity concentrations of ⁴⁰K, which were 635.42 ± 0.01 Bq/kg and 445.32 ± 0.01 Bq/kg, respectively. The further proper processing of the minerals should eliminate the radionuclides.

3.2. Outdoor absorbed and annual effective dose rates

The average outdoor absorbed dose rates and also the outdoor, indoor and total effective dose rates due to the measured radionuclides are presented in Table 2. The outdoor absorbed dose rates, as a result of the presence of ⁴⁰K, ²³²Th and ²³⁸U in the soil samples, ranged from 5.75 ± 0.02 to 133.55 ± 0.04 nGy h⁻¹ with a mean of 70.98 ± 0.05 nGy h⁻¹. The mean dose rate was observed to be higher than the worldwide average of 60 nGy h⁻¹ (UNSCEAR, 2000). This shows that the value of the calculated absorbed dose rate in air, outdoors at the site, is high. As a result of this, the residents within and around the mining site expose the radiosensitive tissues of the body to a considerable level of risk. Mica, amidst the three minerals, has a relatively high outdoor absorbed dose rate of 118.88 ± 0.01 nGy h⁻¹; and hence the major mineral contributing to the absorbed dose rate.

The average annual outdoor effective dose (Table 2) ranged from 0.02 to 0.49 mSv with a mean of 0.26 mSv. The mean is found to be four orders of magnitude higher than the worldwide average of 0.06 mSv for outdoor exposures (UNSCEAR, 2000). Also, the annual indoor effective doses from

| Sample | Outdoor absorbed dose rate (nGy h⁻¹) | Effective dose rate (mSv) | Total annual effective dose (mSv) |
|--------|--------------------------------------|--------------------------|---------------------------------|
|        |                                      | Outdoor | Indoor |                          |
| 1      | 22.20 ± 0.05                         | 0.08    | 0.05   | 0.13                      |
| 2      | 36.05 ± 0.03                         | 0.13    | 0.09   | 0.22                      |
| 3      | 111.87 ± 0.02                        | 0.41    | 0.27   | 0.68                      |
| 4      | 123.15 ± 0.03                        | 0.45    | 0.30   | 0.75                      |
| 5      | 25.04 ± 0.03                         | 0.09    | 0.06   | 0.15                      |
| 6      | 133.55 ± 0.04                        | 0.49    | 0.33   | 0.82                      |
| 7      | 30.37 ± 0.02                         | 0.11    | 0.07   | 0.18                      |
| 8      | 82.47 ± 0.03                         | 0.30    | 0.20   | 0.50                      |
| 9      | 40.27 ± 0.02                         | 0.15    | 0.10   | 0.25                      |
| 10     | 34.82 ± 0.04                         | 0.13    | 0.09   | 0.22                      |
| 11     | 129.92 ± 0.02                        | 0.48    | 0.32   | 0.80                      |
| 12     | 73.70 ± 0.03                         | 0.27    | 0.18   | 0.45                      |
| 13     | 65.06 ± 0.01                         | 0.24    | 0.16   | 0.40                      |
| 14     | 85.21 ± 0.02                         | 0.31    | 0.21   | 0.52                      |
| Feldspar | 31.37 ± 0.45                         | 0.12    | 0.08   | 0.20                      |
| Quartz | 5.75 ± 0.02                          | 0.02    | 0.01   | 0.03                      |
| Mica   | 118.88 ± 0.01                        | 0.44    | 0.29   | 0.73                      |
| Mean (1–14) | 70.98 ± 0.05              | 0.26    | 0.18   | 0.44                      |
| Worldwide average | 60 | 0.06 | 0.41 | 0.3–0.6 |
external exposure to gamma rays emitted from the radionuclides in the soil samples, if used as building materials range from 0.01 to 0.33 mSv with a mean of 0.18 mSv. This mean value is two times lower than the worldwide average of 0.41 mSv for indoor exposures (UNSCEAR, 2000). The sum of the outdoor and indoor annual effective doses, which gives the total annual effective dose (Table 2), ranges from 0.03 to 0.82 mSv with a mean of 0.44 mSv. This average is, also, lower than the worldwide average of 0.48 mSv (UNSCEAR, 2000). This situation notwithstanding, there is still the need for proper monitoring and control of materials used for construction, especially when it is from mineral-rich environments, in order to ensure the protection of workers and the public from radiological hazard.

3.3. Radon concentrations in the water samples
Table 3 presents the radon concentration values in the samples, with the minimum and maximum concentrations being 0.168 and 78.509 Bq/L from the stream and borehole samples, respectively. Irrespective of the sources, 18 (45%) out of the 40 samples had radon concentrations higher than the maximum permissible limit of 11.1 Bq/L (USEPA, 2003) and the world average value of 10 Bq/L (UNSCEAR, 1993; World Health Organisation [WHO], 2004). For the five stream samples, the concentration ranged from 0.168 to 10.237 Bq/L. However, samples 5 (from Egbeku) and 4 (from Arokodara) have respective radon concentrations of 10.237 and 8.442 Bq/L, which are considerably high when compared with samples from other streams. This observation is at variance with the reported works that have indicated surface water to generally contain a very low concentration of radon-222 because of the diffusional losses to the atmosphere (Binesh et al., 2010; USEPA, 2003; United States Geological Survey [USGS], 2008). Due to aeration, streams are expected to have low radon concentration, typically ranging from 0.001 to 0.5 Bq/L (USEPA, 2003; WHO, 2004). The radon concentrations observed in the two samples in question, are very close to the maximum contaminant limit, 11.1 Bq/L. The two samples, in question, are from the streams traversing the binary granites terrain, which are among those aquifer materials that have high radon risks (Kosh et al., 1988). The two-mica or binary granite is the major type of granite in Ijero (Ale et al., 2014; Okunlola & Akinlola, 2010). Although, uranium concentration in two-mica granite is low, uranium within this type of granite is labile; hence, they get dissolved in ground water during weathering. A significant number of the populace in Ijero Ekiti rely heavily on streams particularly in the dry season resulting in exposure and possible impact.

In the 20 well samples, the radon concentrations ranged from 4.511 to 26.542 Bq/L. Seven out of these samples had radon concentration greater than the maximum permissible limit of 11.1 Bq/L. It was observed that some samples have unusually high concentration of radon, especially with the very shallow nature of the wells. The sampled wells have depths ranging from 2 to 4 m and are observed to contain shiny materials at the base, which were identified to be mica. This observation is in agreement with high absorbed dose rate presented by the mica sample discussed above and also consistent with radon concentration in wells from other areas with high background radiation in south-western Nigeria (Oni et al., 2014).

For borehole samples, 11 (73.33%) out of the 15 samples analysed had radon concentrations greater than 11.1 Bq/L with the lowest and highest concentrations 2.698 and 78.501 Bq/L, respectively. The arithmetic mean radon concentration of borehole samples was 23.036 Bq/L. This observation showed a similar trend when compared with the reported high radon concentrations in borehole sources (Binesh et al., 2010; Somlai et al., 2007). Comparing borehole and well sources from high background radiation areas in south-western Nigeria, Oni et al. (2014) reported that borehole source has the highest concentration of radon. It has been reported that the geological structure of an area is a predominant factor for high radon concentration (Vaupotič, Kobal, & Planinič, 1998) and the saturated zone of the earth crust contain high radon (USGS, 2008). This implies that the depth of the source is also a predominant factor for high radon concentration. The two mica-granite structure of the study area have definitely contributed to the high radon concentrations observed in borehole sources.
Table 3. Mean and corrected mean $^{222}$Rn concentrations in the samples

| Sample ID | Sample type | Decay coefficient | Mean concentration (Bq/L) | Corrected mean concentration (Bq/L) |
|-----------|-------------|-------------------|---------------------------|-----------------------------------|
| 1         | Stream      | 1.103             | 0.152                     | 0.168                             |
| 2         | Stream      | 1.112             | 0.525                     | 0.584                             |
| 3         | Stream      | 1.128             | 1.170                     | 1.320                             |
| 4         | Stream      | 1.181             | 7.150                     | 8.444                             |
| 5         | Stream      | 1.162             | 8.810                     | 10.237                            |
| 6         | Well        | 1.199             | 0.996                     | 1.194                             |
| 7         | Well        | 1.190             | 2.280                     | 2.713                             |
| 8         | Well        | 1.128             | 2.530                     | 2.854                             |
| 9         | Well        | 1.120             | 3.170                     | 3.550                             |
| 10        | Well        | 1.190             | 3.320                     | 3.951                             |
| 11        | Well        | 1.103             | 4.090                     | 4.511                             |
| 12        | Well        | 1.128             | 4.510                     | 5.087                             |
| 13        | Well        | 1.199             | 4.460                     | 5.348                             |
| 14        | Well        | 1.154             | 5.280                     | 6.093                             |
| 15        | Well        | 1.208             | 5.300                     | 6.402                             |
| 16        | Well        | 1.217             | 6.720                     | 8.178                             |
| 17        | Well        | 1.236             | 7.000                     | 8.652                             |
| 18        | Well        | 1.217             | 8.710                     | 10.600                            |
| 19        | Well        | 1.181             | 10.500                    | 12.401                            |
| 20        | Well        | 1.154             | 11.500                    | 13.271                            |
| 21        | Well        | 1.172             | 14.000                    | 16.408                            |
| 22        | Well        | 1.226             | 13.700                    | 16.796                            |
| 23        | Well        | 1.217             | 17.300                    | 21.054                            |
| 24        | Well        | 1.172             | 18.900                    | 22.151                            |
| 25        | Well        | 1.154             | 23.000                    | 26.542                            |
| 26        | Borehole    | 1.199             | 2.250                     | 2.698                             |
| 27        | Borehole    | 1.112             | 4.870                     | 5.415                             |
| 28        | Borehole    | 1.128             | 6.840                     | 7.716                             |
| 29        | Borehole    | 1.120             | 8.810                     | 9.867                             |
| 30        | Borehole    | 1.163             | 12.400                    | 14.421                            |
| 31        | Borehole    | 1.226             | 12.300                    | 15.080                            |
| 32        | Borehole    | 1.181             | 14.700                    | 17.361                            |
| 33        | Borehole    | 1.226             | 14.700                    | 18.022                            |
| 34        | Borehole    | 1.162             | 17.300                    | 20.103                            |
| 35        | Borehole    | 1.120             | 18.700                    | 20.944                            |
| 36        | Borehole    | 1.137             | 21.200                    | 24.104                            |
| 37        | Borehole    | 1.217             | 23.700                    | 28.843                            |
| 38        | Borehole    | 1.137             | 25.900                    | 29.448                            |
| 39        | Borehole    | 1.128             | 47.000                    | 53.016                            |
| 40        | Borehole    | 1.128             | 69.600                    | 78.509                            |
3.4. CAED assessment

The summary of the annual effective dose from ingestion of radon-222 in water by both adults and children is presented in Table 4. The minimum and maximum annual effective doses from the samples were 0.000003 and 0.001570 mSv/y for adults; and 0.000007 and 0.003140 mSv/y for children, respectively. The arithmetic mean for adults and children were 0.000282 and 0.000563 mSv/y, respectively. It was observed that none of these sources had annual effective dose higher than the maximum permissible limit of 0.2 mSv/y, if consumed by children and 0.1 mSv/y, if consumed by adults (WHO, 2004). It was also observed that the dose receivable by children is quite low relative to the dose receivable by adults.

Cottage or full-scale industrial mining activities, without adequate planning to take care of all possible impacts, would result in increased level of radionuclides and radon in ground water. Technologically enhanced naturally occurring radionuclides would get leached into the ground water, increasing the risk to radiation exposure. Although, none of the investigated samples exceeded the maximum permissible limit if consumed by adults and children, the present level of radon can increase to unsafe levels if uncoordinated mining activities continue. There is therefore the need to pay urgent attention to this situation in order to protect the populace from further radiological hazards that might arise from ingestion and inhalation of radon.

4. Conclusion

The activity concentrations of $^{238}$U, $^{232}$Th and $^{40}$K in the soil samples hosting mineral ores have been obtained using gamma-ray spectrometry. The RAD7/RAD H$_2$O system has also been used to obtain radon concentration data of the ground water system from the study area. While $^{232}$Th was observed to have low activity concentrations, $^{238}$U and $^{40}$K had concentrations higher than the worldwide average. Mica was noted to have the highest activity level among the minerals just like we had from some of the ground water samples, especially the well and borehole samples. We noted that the geological structure and the depth of the source is a predominant factor for the high radon concentrations. The two mica-granite structure of the study area has definitely contributed to the high radon concentrations observed in borehole sources. The relatively high levels of the radionuclides in the soil samples and radon in the ground water system presents some level of health risk from the inhalation of the radionuclides-rich dust particles as well as the inhalation and ingestion of radon. Though the effective dose seemed lower than the worldwide average, effects of prolonged exposure to radiation which could lead to severe health hazards are still possible. The public water system should be revisited by relevant government agencies and efforts should be made to educate the public on the effects of radon.

| Sample Type | No. of Samples | Adults | Children |
|-------------|----------------|--------|----------|
|             | Minimum mSv/y | Maximum mSv/y | >0.1 mSv/y | Minimum mSv/y | Maximum mSv/y | >0.2 mSv/y |
| Borehole    | 0.000054      | 0.001570 | –         | 0.000108      | 0.003140 | –         |
| Well        | 0.000024      | 0.000531 | –         | 0.000048      | 0.001062 | –         |
| Stream      | 0.000003      | 0.000205 | –         | 0.000007      | 0.000409 | –         |
| Total       | 0.000003      | 0.000205 | –         | 0.000007      | 0.000409 | –         |

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