Assessment of Radon Contamination in Drinking Water Sources Around Babban Tsauni Gold Mining Area, Federal Capital Territory, Nigeria

K. A., 1,2 Oladipo, M. O. A., 3 Onoja, M. A., 2 Musa, Y. and 4 Aremu, S. O.

1Department of Physics, Kaduna State University, Nigeria
2Centre for Energy Research and Training, A.B.U., Zaria, Nigeria
3Department of Physics, Ahmadu Bello University, Zaria, Nigeria
4Department of Science Laboratory Technology, Federal Polytechnic, Bauchi, Nigeria

*Corresponding author’s email: kafayatodelami@gmail.com; Phone: +2348030425668

ABSTRACT
Radon, a naturally occurring radioactive gas, is produced through the disintegration of radionuclides like uranium, thorium, or radium in the earth's crust. In this study, the radon levels and the annual effective dose from drinking water sources were assessed in Babban Tsauni, Gwagwalada, Nigeria, where artisanal gold mining is taking place. Water samples were collected and analyzed for $^{222}\text{Rn}$ using a liquid scintillation counter (Tri-Carb-LSA1000). The highest radon activity concentration was found in Dobi with a value of 3.215 Bq/L, while the lowest was found in Tsani 7; 0.025 Bq/L. Both values were significantly below the maximum contamination level of 11.1 Bq/L and the world average value of 10 Bq/L, as established by USEPA and WHO respectively. The annual effective dose from ingesting radon was also below the permissible limit of 0.1 mSv/y for all age groups, the values for adults, children, and infants ranged in mSv/y from 5.59E-07 to 2.35E-05, 8.39E-07 to 3.52E-05, 9.79E-07 to 4.11E-05 and 1.79E-07 to 2.55E-06, 2.68E-07 to 3.82E-06, 3.13E-07 to 4.46E-06 for ground and surface water sources respectively. This study concluded that the radon concentrations in the water sources remained within safe limits and that the anthropogenic activities of gold mining had no significant effect on the water sources.

Keywords:
Radon, Activity concentration, Effective Annual Dose, Water sources, Contamination.

INTRODUCTION
Good health begins with having access to potable water, free of pollutants and contaminants; potential contaminants may occur naturally or as a result of human activities. Water in its pure form is a transparent, tasteless, colorless, and odorless substance found all over the earth, the form it takes depends on the temperature. In liquid form, it can have its sources from rivers, streams, oceans, and wells. Water is a major resource for all living things to survive, it is an indispensable and essential commodity, hence its availability, accessibility, and safety. For it to be portable, fit for human consumption and general household use, it must not only contain a certain amount of dissolved solutes that are vital to the proper functioning of the body but also free from radiological, chemical, microbial, and other forms of contaminations as the case may be (Garba et al., 2008).
Radon is a naturally occurring radioactive gas, formed when uranium, thorium, or radium disintegrates within the earth’s crust, it can be found in rocks, soil, and water in concentrations that depend on the nature of the environment and anthropogenic activity in an area. As a natural gas, humans are exposed to radon through breathing air and drinking water containing dissolved radon gas (due to its solubility, mobility, and colorless and odorless properties in water). The groundwater more than any source contains high radon due to contact and proximity with geological formations of the environment (Saat et al., 2012) (or immersion in soil), technological processes (e.g., mining and agriculture), and the nature of its close system that allows accumulation of radon. Radionuclide concentration due to gold mining in any environment constitutes a major human health risk. $^{238}\text{U}$, 232$\text{Th}$, and 40$\text{K}$ and any of their decay products, constitute majorly naturally occurring radioactive materials (NORMs) in the environment which are accompanied by radiation emissions including radon gas. NORMs such as $^{238}\text{U}$, 230$\text{Th}$, and 40$\text{K}$ provide significant sources of human exposure to ionizing radiation, when released into the environment may subsequently disperse and get transported through the
same environment via various mechanisms (IAEA, 2003). Radon a by product of one of the NORMs ($^{238}$U), with a half-life of 3,842 days, emits heavy ionizing alpha particles alongside its short-lived decay products (radon progeny, also alpha emitters. When inhaled, deposits all its energies into the cells of the lungs only (due to its low penetrating power and long wavelength which makes it travel a very short distance), causing a great change in the DNA of the lung cells (initiating cancer). This is why radon concentration is believed to be higher in air than in water, consequently, the dose received through inhalation is greater compared to that received through ingestion and from other sources. Nevertheless, its concentration in water has been identified as a public health risk because of its ability to exude into the air when being used for household chores, bathing, and during the panning processes in goldmines. This accounts for one of the reasons “WHO, (2011) guidelines for drinking water quality” recommend that screening levels for radon in drinking water be set based on the national reference level for radon in air. In addition to exposure to naturally occurring radioactive substances, mining activities greatly enhance the radiation level in areas where mines are located, invariably, the low doses accumulate over a long time, which may result in lung or skin cancer or other radiation illnesses (UNSCER, 2000). Exposure to enhanced radiation from NORMs by small-scale miners can also constitute occupational exposure, as a result of prolonged exposure to the source, therefore radiation protection procedures should be strictly adhered to for this category of workers, to safeguard their health. Prolonged exposure could result in lung cancer, leukemia, or even death, as several studies have shown that the risk of developing lung cancer increases with exposure to radon (UNSCEAR, 2008; Bello et al., 2020). The majority of an ordinary person's annual radiation dose is caused by exposure to naturally occurring radiation, which is routinely unregulated in many industries and nations (World Nuclear Association, 2019).

Babban Tsauni like many other communities in developing countries like Nigeria, has no access to treated water supply and therefore depends largely on groundwater sources for consumption, household use, and for panning processes during the gold mining activities. Unregulated exposure to sources of radiation in mining areas, in addition to lack of awareness of the risk associated with drinking water with elevated radon concentrations resulting from enhanced radiation from NORMs, are a few other problems faced by inhabitants of these communities. Despite reports that high concentrations of radon in drinking water may increase the radiation dose to the stomach lining. Although, the link between stomach cancer and ingestion of drinking water with high radon levels is yet to be established (WHO, 2009). For this reason, the radon concentration in groundwater sources used for drinking and other household chores has received great attention from the scientific community due to its toxicity, and maximum contamination levels set by various international bodies including the United States Environmental Protection Agency (USEPA, 1999) with proposed value of 11.1 Bq/L and 100 Bq/L by World Health Organization (WHO, 2004; WHO, 2022). Radon concentration assessment has been carried out within and outside Nigeria, with some studies reporting values above maximum contamination level (MCL); (Akinnagbe et al., 2018; Kessongo et al., 2019; Bello et al., 2020; Jibril et al., 2021, Farai et al., 2023) especially from ground water sources. While some have reported lower values (Zaini et al., 2011, Zainal, 2012, Mohammed et al., 2016 and Umar et al., 2024), no literature has reported radon levels in this study area. Hence, this work aims to determine the radon activity concentrations in the water sources, and their contamination levels and to estimate the annual effective dose due to ingestion of water from these sources.

MATERIALS AND METHODS
Location and geology of the study area
Babban Tsauni community is located in Gwagwalada local government area, 58 km West of Abuja. Surrounded by other adjoining communities where gold mining is not taking place. The selection of the study area was based on the proximity of the mining site to the community and its possible effect on the artisans, inhabitants, and the general public. It can be assessed through the Kaduna-Lokoja road. It lies within latitude 9° 10′ 0″ N and 6° 58′ 0″ E. The Gold veins are available but not in commercial quantity compared to lead and zinc, according to the investigation by Geolank Exploration Concept in 2014 (GEC, 2014). The geology of the area as described by Okunlola et al., (2007) is underlain mainly by migmatised gneiss complex rocks such as; granite gneiss, migmatised amphibolite, pegmatites, biotite hornblende gneiss and migmatised as its major mineral composition. Discrete Pb-Zn sulfide mineralization of the galena and sphalerite varieties are noticed as embedded crystals within the pegmatite veins and quartz bodies, they are concentrated in regions of intense sharing and along fracture zones. (Okunlola et al., 2007)
Theoretical Description of Liquid Scintillation Technique

The principle of LSC as described in fundamentals of liquid scintillation counting (National Diagnostics, 2004) and Annunziata et al. (2020); 222Rn is extracted readily from the water sample in a plastic vial homogeneously mixed with an organic scintillant cocktail dissolved in a suitable solvent. The decay products of 222Rn remain in the water phase until 222Rn is extracted into the organic phase. The sample is stored for three hours until equilibrium is reached between 222Rn and its alpha-emitting decay products. Consequently, the alpha activity from 222Rn and its decay products is measured in a liquid scintillation counter. The Liquid Scintillation Counter detects radioactivity via the same type of light emission events that are used in solid scintillation. LSC takes place in a solution rather than in solid crystal, it also allows close contact between the isotope atoms and the scintillator, which is not possible in solid scintillation. Liquid scintillation cocktails absorb the energy emitted by radioisotopes and re-emit it as flashes of light. The cocktail may contain two components; the solvent and fluor which enable it to absorb and then re-emit light. The solvent carries out the bulk energy while dissolved in the solvent, molecules of fluor convert absorbed energy into light. It comprises 60-90% of the total solution and acts as an efficient collector of energy to the fluor molecules without quenching the scintillation of the fluor. The fluor (primary and secondary scintillators) provide the conversion of captured energy to the emission of light. Primary scintillators are excited to light emitting state by excited solvent molecules. The secondary scintillator captures the fluorescence energy of the excited primary scintillators and re-emits it as a longer wavelength signal. The wavelength of the photon is characteristic of the scintillator. The light is detected in two photomultiplier tubes operating in coincidence counting. The decay of radon and its short-lived decay products result in three alpha particles at 5.49 MeV (222Rn), 6.0 MeV (218Po), and 7.69 MeV (214Po). The decay of (214Bi) and (214Pb) yields two beta particles of endpoint energies ranging from 0.67-3.27 MeV. The electrical pulse's amplitude is transformed into a digital value, which is then transferred to an analyzer to be compared against digital values for every channel in the...
LSC. This digital value, which reflects the energy of the beta particle, is obtained. On a CRT, the quantity of pulses in every channel is displayed. The sample is analyzed in this way, and the spectrum can be plotted to reveal how much radon has been dissolved in the cocktail. Some of the advantages of this method are: small sample volume, minimal preparation time and automatic sample changing.

Sample Collection
Random sampling was adopted for the sample collection; this was because the water sources were closely located around the small community under study, and to allow for an unbiased conclusion. A total of fifteen plastic containers of 50 ml each of the water samples were collected (surface and ground), based on availability and proximity to the mining community. All containers were thoroughly washed and rinsed with distilled water before setting out for sample collection. The borehole water samples were evacuated for a few minutes before being collected into already rinsed containers. The well water was initially purged by drawing it out severally before pouring it into the pretreated containers, while for the surface water, containers were submerged in water each instance. The containers were filled to the brim to avoid CO₂ being trapped and nitric acid was immediately added for preservation purposes. These were transferred within 3 days to the Scintillation laboratory at the Centre for Energy Research and Training, A.B.U. Zaria.

Sample preparation
The sample preparation procedure reported by Bello et al., (2020) was adopted. Liquid Scintillation vials were produced containing 10 ml of liquid scintillation solution. This was followed by using a disposable syringe to draw 10 ml of each of the water samples and immediately adding to vials containing 10 ml of liquid scintillation solution. The vials were instantly air tight and vigorously shaken for three (3) minutes to extract ²²²Rn from the water phase to the organic scintillant solution due to its greater solubility in organic liquids. Leaving the vials for more than three (3) hours allowed for the ingrowth of the short-lived decay products of ²²²Rn, thus, attaining secular equilibrium and being ready for counting.

Sample Measurement
The prepared samples were analyzed using a liquid scintillation counter (Tri-Carb-LSA 1000) located at the Center for Energy Research and Training, Ahmadu Bello University, Zaria. The analysis was carried out after the prepared samples had been allowed to reach equilibrium. Calibration of the liquid scintillation counter was made before the commencement of counting using IAEA ²²⁶Rn standard solutions (IAEA-423 and IAEA-427). The background, calibration, and sample solutions were measured over the same spectral range and for the same counting period of 60 minutes and were then recorded. To determine the activity concentration, the sample volume, background count rate, decay time (time between sampling and counting), and efficiency of detection were considered. Equation 1 (Garba et al., 2013; Bello et al., 2020) was applied to determine the concentration of ²²²Rn in the water samples.

\[
R_n = \frac{100(N_f-N_b) \cdot \exp(\lambda t)}{60 \cdot P \cdot D}
\]

where Rn is the ²²²Rn concentration at the time of sampling (Bq/L), NS is the sample total count rate (count min⁻¹), NB is the background count rate (count min⁻¹), t is the elapsed time between sample collection and counting, λ is the ²²²Rn decay factor (1.26 min⁻¹), 100 is a conversion factor from per 10 ml to per liter, 60 is conversion factor from min. to sec., CF (13.47) is the calibration factor, D (0.964) is the fraction of ²²²Rn in the cocktail in vial of 22 ml total capacity vial (for 10 ml sample, 10 ml of cocktail and 2 ml of air).

2.6 Estimation of Annual Effective Dose from ²²²Rn in Drinking Water
The annual effective dose was estimated using Equation (2), taking into account the dose coefficient (mSv/y), the annual water consumption (L/Y), the “ICRP standard man” adopted, and the activity concentration of ²²²Rn obtained from Equation (1). This is very necessary because of the health risks associated with radon inhalation or ingestion.

\[
E = C_{Rn} \times D \times L
\]

where; \(C_{Rn}\) = Concentration of ²²²Rn in water, D = Ingestion dose conversion factor (10⁻⁸ Sv/Bq, 2×10⁻⁸ Sv/Bq, 7×10⁻⁸ Sv/Bq) for adults, children, and infants respectively (UNSCEAR, 2000), L = Annual water consumption by adults, children, and infants were 730, 547.5, and 182.5 (L/Y) (UNSCEAR, 1993; Bello et al., 2020).

RESULTS AND DISCUSSION
Table 1 presents the result of Radon activity concentrations and annual effective dose for water samples (ground and surface) collected from the Tsauni community with five other samples from adjoining communities free from gold mining activities. The overall arithmetic mean of radon activity concentrations in the drinking water was 0.839 Bq/L. The maximum value obtained was from the Dobi borehole with a value of 3.215 Bq/L and a minimum value was obtained from Tsauni 7 (one of the two streams available in the community) with a value of 0.025 Bq/L, both values were below the maximum contamination level (MCL) of 11.1 Bq/L and 100 Bq/L by USEPA and WHO.
The above results in Table 1 compare well with the reported literature in Table 2. Especially reports (Zainal, 2012; Mohammad et al., 2016 and Umar et al., 2024). A similar trend was also observed when compared with earlier publications that reported high radon concentrations in borehole sources (Oni et al., 2014; Akinnagbe et al., 2018; Farai et al., 2023). These studies reported high radon concentration values for water from boreholes but were not in agreement with the values obtained because their values exceeded the MCL. While this study has reported a high value in groundwater sources (borehole), but within permissible limits. It also was observed that despite the mining of gold in the community under study, which is believed could enhance the radon level that may result from elevated concentrations of NORMs and the geology of the rock type (granite-migmatite), (IAEA, 2003; USCEAR, 2008; Oni et al., 2014; Kessongo et al., 2020), no significant value was recorded compared to the highest value observed from the water sample collected from one of the control areas; borehole (3.215 Bq/L). Fig. 2 shows the variation in the activity concentrations of radon in the water samples. The overall radon concentrations in water samples varied in decreasing order; Borehole > well > surface water respectively. The lowest values observed for surface water sources can be attributed to the ability of radon gas to exude easily into the surrounding air.
The Annual Effective Dose (AED) values, in mSv/y estimated from the radon concentration obtained for both surface and groundwater sources range from 5.59E-07 to 2.35E-05, 8.39E-07 to 3.52E-05, 9.79E-07 to 4.11E-05, 2.68E-07 to 3.82E-06, 3.13E-07 to 4.46E-06 for ground and surface water sources respectively. It was observed that the annual effective dose varied in decreasing order; Infants > Children > Adults for all water sources as depicted in Fig. 3. The AED was highest for infants among the 3 age groups considered. This may not be unconnected with their low body mass which makes them more vulnerable, hence are at higher cancer risk compared to other age groups (children and adults). The results obtained corroborates results reported in previous studies as shown in Table 2. In this present study, AED for all the water sources considered were below the permissible limits of 0.1 and 0.2 mSv/y for adults and children, respectively (WHO, 2022; WHO; 2008).
CONCLUSION
Radon concentrations of some selected water samples around Babban Tsauni artisanal gold mine, Gwagwalada, Abuja, Nigeria, were collected from three different sources (well, borehole, and stream) for investigation. Data obtained for activity concentrations of $^{222}$Rn from the study ranged from 0.025 to 3.215 Bq/L, with an average value of 0.839 Bq/L. Results obtained from all the sources were below the maximum contamination level set by USEPA. However, these low values were unexpected due to the geology and anthropogenic activity in the area under study. The annual effective dose resulting from the ingestion of radon in the water sources for all ages is markedly below the permissible limit of 0.1mSv/y. It was, therefore, inferred that the radon concentration in the water sources of areas under study was within permissible limits set by international organizations and, hence safe for drinking. It was also established that anthropogenic activity (gold mining) did not affect all the sources of water, as compared to the control with higher values, but also within maximum contamination level.

REFERENCES


