

RADIOCHEMICAL EVALUATION OF GROUNDWATER AROUND MIKA URANIUM MINERALIZATION AND ENVIRONS.

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Abstract: Naturally occurring radon gas (^{222}Rn) exists in groundwater and previous study of the problem of natural radioactivity in drinking water from wells drilled in rock types rich in uranium has shown tendency to have high radon concentration. It has been widely reported that radon is contributing the largest component of human exposures to natural radiation and it is considered as the second major cause of lung cancer, after cigarette smoking. Seventeen (17) groundwater samples around Mika Uranium mineralization were collected and ^{222}Rn concentrations were measured using the DURRIDGE RAD7 electronic radon detector. Radon (^{222}Rn) concentrations in the groundwater ranged from 2350 to 46,200 Bq/m³ with an average of 29,400 Bq/m³. The results show that 76.5% of the groundwater samples collected have ^{222}Rn concentrations far above the U.S.E.P.A maximum contaminant level (MCL) of 11.1 Bq/l (for states without radon monitoring policy and enhanced indoor air programs), while 23.5% has values lower. Meanwhile, all ^{222}Rn values were below the MCL of 100 Bq/l recommended by European Union (EU) for measurements that warrant consideration of possible remedial actions. Borehole samples show relatively higher ^{222}Rn concentrations than samples collected from open-wells. The annual effective dose due to ingestion (H_{ing}) of ^{222}Rn in the groundwater ranged from 0.05 to 0.92 mSv, with an average of 0.59 mSv. These values are within the ICRP recommended reference level of 1 mSv/yr for the intake of radionuclide in water by the general public for a prolonged exposure.

Keywords: Groundwater: Radon: Nigeria: Hazard: External Hazard

1.0 INTRODUCTION

Access to safe drinking water is essential to health and it is a basic human right [1]. For over two decades, groundwater has been the dominant source of drinking water in Nigeria [2]. It is often thought to be cleaner and easier to treat as compared to surface water and as a result many wells have either been sunk or drilled [3]. A groundwater source can potentially contain several naturally occurring chemical elements, many of which are not tested routinely as indices of water quality despite their known toxicity [4].

In general, radon exists in air, water and soil as gas [5]. If radon and radon daughters are ingested or inhaled and decay inside the human lungs, the radiation has the potential to split water molecules and produce free radicals (e.g. OH). The free radicals are very reactive and may damage the DNA of the cells in the lungs, thus causing cancer. It is a radioactive noble gas which is formed through decay of radium. It occurs as three different isotopes in nature, ^{219}Rn , ^{220}Rn and ^{222}Rn , of which ^{222}Rn with a half-life of 3.82 days is the most important, this is because, other isotopes are very short-lived (^{220}Rn : 55.6 sec; ^{219}Rn : 3.96 sec), and will not be transported very far before they decay. ^{222}Rn is formed through decay of ^{226}Ra in the decay chain of ^{238}U [6].

Concentration measurements of ^{222}Rn and its progeny for the determination of radiation doses to

occupationally exposed individuals and members of the public living in proximity to supervised radiation area are standard practice. The International Commission on Radiological Protection [7] recommendations for limits to ionizing radiation from man-made sources are 20 mSv.yr⁻¹, effective dose for occupationally exposed workers and 1 mSv.yr⁻¹ for members of the public; this does not include medical as a patient or natural background.

1.2 STUDY AREA

1.2.1 Location, Extent and Accessibility

The study was conducted in parts of Zing and Yorro Local Government Areas, of Taraba State, Nigeria. The study area can be located on a topographic map (Monkin Sheet 216 and part of Dong Sheet 195). It lies between latitudes 08°48' to 09°4'N and longitudes 11°30' to 11°48'E. It is located within the Upper Benue trough and covers an area of about 1004 km². The area is bounded to the east and north by Mayo-Belwa and Jada Local Government Areas of Adamawa state and to the west and south by Lau and Jalingo Local Government Areas of Taraba State, respectively.

It can be accessed through intra/interstate roads of Jalingo – Zing road and Mayo-Belwa – Zing road respectively. Footpaths and many rural road networks also link the study area from nearby communities (Fig. 1).

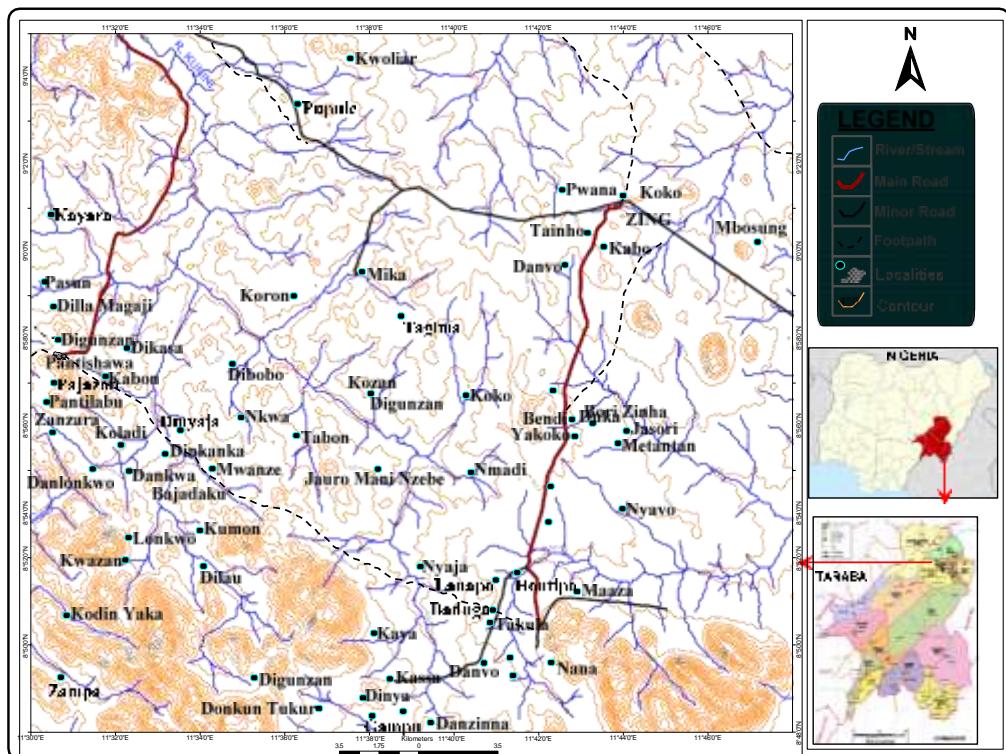


Fig. 1: Topographic map of the study area

1.2.2 Relief and Drainage.

The relief configuration of the study area can be categorized into two zones, highlands mountain range and lowlands (Fig.2). The highlands occupy the southern region stretching from west to south in chains of mountain with elevation ranging from 1800

— 2400 meters high (asl), forming the Atlantica/Shebshi and Adamawa massifs ranges. The lowland which occupies about 60% of the region hosts most of the settlements. The area is drained by River Kunini one of the tributaries of the Lau River, an off-shoot of the major Benue River.

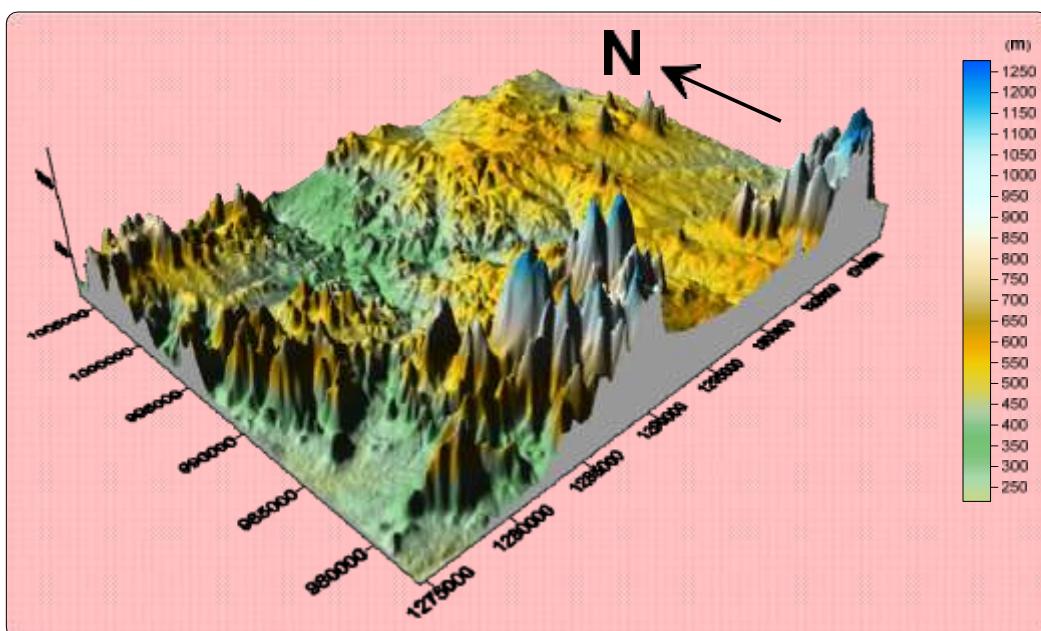


Fig 2: Digital Elevation Map of the study area.

1.2.3 Climate and Vegetation.

The climate of the area is typically a tropical climate marked by dry and rainy seasons. The mean annual rainfall of the area ranges from 819 – 1761mm. It is spread over seven months (April to October). The onset of the rains is April, with low amount but increases gradually reaching a maximum in August, the amount drops gradually with cessation in October [8]. Mean monthly temperature ranges between 20°C – 25°C while the relative humidity is lowest (26%) in March and reaches 98% in August [9]. The study area is within the savannah grassland belt, particularly in the guinea Savannah sub-region, characterized by scattered, deciduous tall trees with broad leaves and tall grasses.

1.2.4 People and Land-use

Zing and Yorro Local Government areas, predominantly accommodates the Mumuye ethnic tribe in Taraba State. Their unique tribal marks and a large opening on the ear, mostly seen on elderly women, makes them stand-out from other tribes within and outside the State. Other tribes like the Fulani, mostly engaged in cattle rearing, are found settled around the Mumuye communities. Mumuye people are predominantly farmers, producing crops

like yam, cassava, maize and beans. They also engage in civil service jobs, politics, small scale livestock, hunting as well as petty trading. Bush burning is a common practice during the dry season (mostly for hunting purposes, e.g. Bush rats and Squirrels), even though, this practice affects the environment negatively as it increases the rate of erosion during the rainy season.

2.0 GEOLOGICAL SETTING

The Study area is located within the basement (granitoids) of the area in Northern part of Adamawa Massif (Fig. 3) which is dominated by porphyritic granites. Other rock units include medium-grained biotite granite, fine-grained granites, sheared granites, Olivine basalt, brecciated rhyolite and siliceous veins, with pegmatites as the only minor rock unit [10]. Textural varieties exist within the medium-grained granite but are difficult to demarcate. It is greyish to pinkish in color with some orientation of feldspar crystals [10]. These rocks form prominent rocky hills in the study area and exhibit considerable textural variation and contact relationships. The area is characterized by extensive fractures and shear zones.

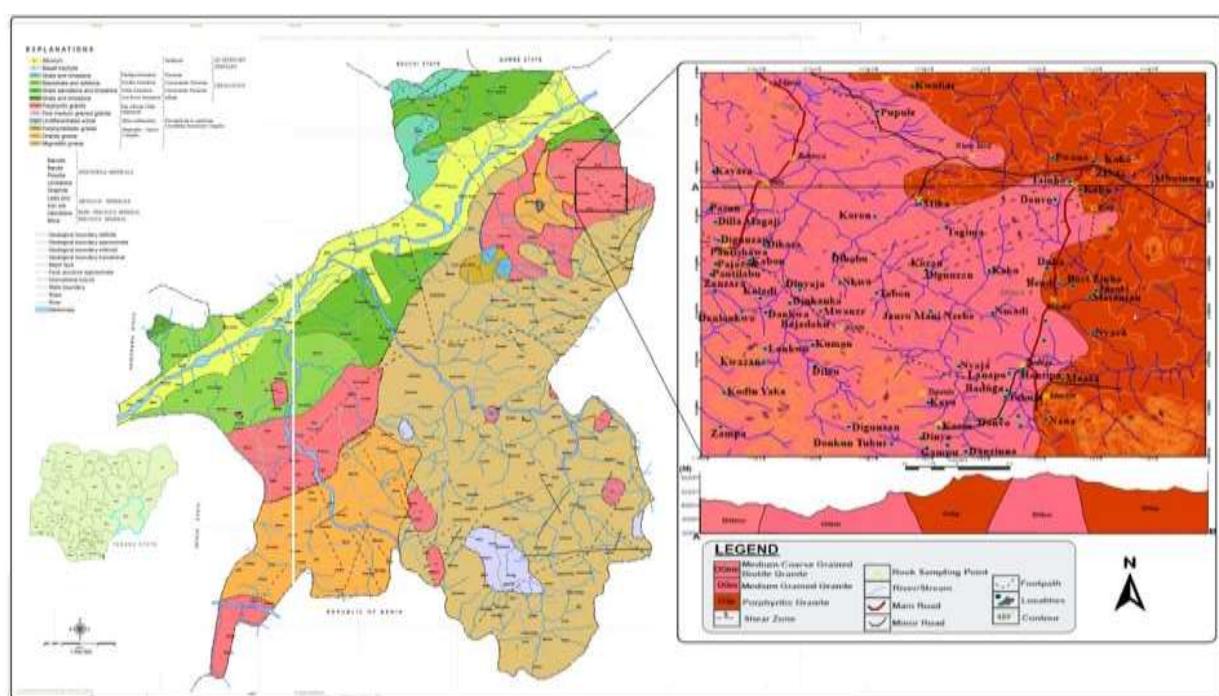


Fig 3: Geologic and mineral Resources Map of Taraba State showing the study area (GSNA, 2006)

The mineralogy of the granites of the study area are generally characterized by three major minerals; plagioclase, biotite, and microcline, even though

hornblende was also reported around the south-eastern part of the area [10; 11].

3.0 MATERIALS AND METHODS

3.1 Water sampling/measurement

Groundwater samples were collected, mostly from boreholes equipped with hand pumps and a few hand-dug wells. The sampling was carried-out based on sample availability and effort to ensure uniform spread, so that results may be a fair representation of the study area.

Boreholes were pumped continuously for at-least 5 minutes before sampling and the hand-dug wells

chosen were in constant use. This is to ensure that “fresh” water samples were collected directly from the aquifer. Seventeen (17) groundwater samples were collected from 17 communities/settlements and measurements were carried-out insitu using the DURRIDGE RAD7 electronic radon detector (Fig.4), in accordance with the standard procedure described in [12].



Fig 4: Typical set-up of RAD7 electronic radon detector

3.2 Radon Dose Calculation

In order to estimate the annual effective dose rate received by the population, one has to take into account the conversion co-efficient from the absorbed dose and the indoor occupancy factor. According to the [13] report, the committee proposed 9.0×10^{-6} mSv/h per Bq/m^3 to be used as a conversion factor, 0.4 for the equilibrium factor of ^{222}Rn indoors and 0.8 for the indoor occupancy factor. Occupancy factor of 0.5 was assumed and used as the time spent indoors by an average person in the study area (for the purpose of this present work). Calculating the annual effective dose to the population, the equation below was used [14]. At a certain radon concentration C_{Rn} in Bq/m^3 , the annual absorbed dose, D_{Rn} is usually expressed in the unit of mSv from the following relation below:

$$D_{\text{Rn}} (\text{mSv/yr}) = C_{\text{Rn}} \cdot D \cdot H \cdot F \cdot T$$

Where;

C_{Rn} = the measured Rn-222 concentration (Bq/m^3),

F = the Rn-222 equilibrium factor indoors (0.4),

T = the indoor occupancy time $24 \text{ h} \times 365 = 8760 \text{ h/yr}$

H = the indoor occupancy factor (0.5), and

D = the dose conversion factor (9×10^{-6} mSv/hr per Bq/m^3).

The annual effective doses due to the ingestion of radon (H_{ing}) in water were calculated from the mean activity concentration using the following equation;

$$H_{\text{ing}} (\text{w}) = D F_{\text{Rn}} \times I_w \times A_{222\text{Rn}}$$

Where;

$D F_{\text{Rn}}$ is the dose conversion factor by ingestion of ^{222}Rn in water by adult members of the public living in the study area, given as $10^{-8} \text{ SvBq}^{-1}$ [15], $A_{222\text{Rn}}$ is the activity concentration of ^{222}Rn in water samples and I_w is the daily water consumption rate (L/a), considered to be 2 L/day [16].

To calculate the annual equivalent dose and effective dose, one has to apply a tissue and radiation weighting factors according to [17]. The equivalent dose is the radiation-weighted absorbed dose. The radiation weighting (WR) factor for alpha particles is 20 as recommended by [7]. With the effective dose, a tissue weighting (WT) factor is applied. According to

ICRP, the tissue weighting factor for lung is 0.12. The annual effective dose is then calculated

$$H_E (\text{mSv/yr}) = D_{Rn} \cdot W_R \cdot W_T$$

Where,

D_{Rn} = Annual Absorbed dose

W_R = Radiation Weighting Factor for Alpha Particles, 20

W_T = Tissue Weighting Factor for the Lung 0.12

according to the equation below:

4.0 RESULTS AND DISCUSSIONS

A total of seventeen (17) groundwater samples were analyzed for radon, 15 of which were sampled from hand-pump boreholes while two (2) of the samples were from hand-dug wells. The results obtained from analysis using the RAD7 Electronic Radon detector (A product of DURRIDGE Company) showed that radon (^{222}Rn) concentration measured in Becquerel per cubic metres (Bq/m^3) range from 2340 ± 300 to $46,200 \pm 1300$ with an average of about $29,400 \pm 1000$. The mean value is far above the upper value of the ICRP reference level of $1500 \text{ Bq}/\text{m}^3$, while thoron (^{220}Rn) concentration also measured in Bq/m^3 range from 65.3 ± 70 to 586 ± 210 , with an average value of about 240 ± 130 . The calculated potential dose by the degassing of radon from the groundwater shows an average annual absorbed dose of 463.59 mSv and average annual effective dose value of 1112.62 mSv , respectively. The 4th schedule in the Nigeria Basic Ionization Radiation Regulations of 2003, under the heading "Dose limits" specified classes of persons to who dose limits apply, to include persons of 18 years or above and 18 years or below [17]. 76.5% of the calculated doses are above the highest regulatory limit of 500 mSv yr^{-1} which represents limit on equivalent dose for the skin, hands, forearms, feet and ankle while about 23.5% are below. The annual effective dose due to ingestion (H_{ing}) of ^{222}Rn in groundwater varied from 0.05 to 0.92 mSv, with an average value of 0.59 mSv yr^{-1} . These are within the ICRP recommended reference level of 1 mSv yr^{-1} for the intake of radionuclide in water by the general public for a prolonged exposure. 11.77% representing two (2) groundwater samples are within WHO recommended reference level of 0.1 mSv yr^{-1} while 88.23% of the sampled groundwater showed an annual effective dose higher than the recommended level. The open-well at Kan-Iyaka and the borehole at Kakulu recorded the lowest H_{ing} while boreholes at Kpantisawa and Nyaja recorded the highest. Detail of the results is presented in the Table 1.

In the environment, the concentration of ^{222}Rn in waters is highly variable because of the variations in

the rock characteristics and flow distribution. It is noteworthy that groundwater from fractured igneous and high-grade metamorphic rocks frequently exhibit high activity of dissolved Rn [18], this is evident from the result presented in Table 1 above, as samples were collected from wells drilled within the uranium-mineralized granitic rocks. 76.5% of the samples in the area under study have ^{222}Rn concentrations far above the United States Environmental Protection Agency (USEPA) maximum contaminant level (MCL) of 11.1 Bq/l (for States without radon monitoring policy and enhanced indoor air programs) [19] while 23.5% have values lower. All radon values are below the MCL recommended by European Union (EU), 100.0 Bq/l (measurement that warrants consideration of possible remedial actions) and 1000.0 Bq/l (upper bound above which remedial action is definitely required) [20]. But on the other hand, all the samples have radon values far above the Standards Organization of Nigeria (SON) recommended an MCL of 0.1 Bq/l (for radionuclide concentration in drinking water in Nigeria) [21]. The highest ^{222}Rn concentration of $46,200 \pm 1300 \text{ Bq}/\text{m}^3$ was measured from a borehole at Kpantisawa town while the lowest concentration $2350 \pm 300 \text{ Bq}/\text{m}^3$ was recorded from an open-well at Kan-Iyaka community. It is worthy to note here that, higher radon concentrations were recorded in borehole waters.

Low radon value recorded at Kan-Iyaka may be due to degassing of ^{222}Rn from the open-well before sampling/measurement because of direct contact with the atmosphere (Sampling and measurements from open-well was deliberately carried-out for comparison purpose) but ^{222}Rn concentration from Tapenla (open-well) show a high value of about $41,300 \pm 1300 \text{ Bq}/\text{m}^3$, this might be because as at the time of sampling/measurement of water from Tapenla, the open-well was in continual use, it may mean that "fresh" water sample (from the aquifer) was collected and immediately measured, therefore, did not allow time for much radon degassing.

Table 1: Measured Radon and Thoron Concentrations, Calculated Radon Annual Absorbed Dose, Annual Effective Dose and Annual effective dose due Ingestion by Drinking.

| S/N | Sample ID | Coordinates | Location Name | ^{222}Rn (Bq/m ³) | ^{220}Rn (Bq/m ³) | Annual Absorbed Dose (DRn) (mSvyr ⁻¹) | Annual Effective Dose (HE) (mSvyr ⁻¹) | $H_{\text{ing}}(w)$ (mSvyr ⁻¹) | Well Type |
|-----|-----------|----------------------------|---------------|---|---|--|--|---|-----------|
| 1 | GW01ZN | 08°59.353'N 11°44.905'E | ZING | 18,200 ± 900 | 142 ± 110 | 286.98 | 688.71 | 0.36 | Borehole |
| 2 | GW02AB | 08°55.947'N 11°42.596'E | ABUJA | 30,100 ± 1100 | 201 ± 130 | 474.62 | 1139.09 | 0.60 | Borehole |
| 3 | GW03MN | 08°50.652'N 11°41.939'E | MONK IN | 6,740 ± 520 | 114 ± 100 | 106.28 | 255.07 | 0.14 | Borehole |
| 4 | GW04KK | 08°59.028'N 11°43.165'E | KAKU LU | 3,540 ± 370 | 109 ± 90 | 55.82 | 133.97 | 0.07 | Borehole |
| 5 | GW05KI | 08°59.963'N 11°39.718'E | KAN-I YAKA | 2,350 ± 300 | 65.3 ± 70 | 37.06 | 88.94 | 0.05 | Open Well |
| 6 | GW06MK | 08°58.718'N 11°37.653'E | MIKA | 7,580 ± 550 | 66.9 ± 70 | 119.52 | 286.85 | 0.15 | Borehole |
| 7 | GW07WY | 09°00.511'N 11°39.588'E | WURO -YAYA | 31,700 ± 1100 | 76.3 ± 80 | 499.85 | 1199.64 | 0.63 | Borehole |
| 8 | GW08MZ | 09°03.695'N 11°36.293'E | MAN ALANG | 39,900 ± 1200 | 109 ± 90 | 629.14 | 1509.94 | 0.80 | Borehole |
| 9 | GW09KJ | 09°03.558'N 10°34.238'E | KWOJI | 44,300 ± 1300 | 193 ± 120 | 698.52 | 1676.45 | 0.89 | Borehole |
| 10 | GW10MY | 09°00.477'N 11°31.826'E | M/YOR RO | 42,600 ± 1300 | 450 ± 190 | 671.72 | 1612.13 | 0.85 | Borehole |
| 11 | GW11BK | 08°59.500'N 11°33.166'E | BAKIN YA | 34,100 ± 1200 | 382 ± 170 | 537.69 | 1290.46 | 0.68 | Borehole |
| 12 | GW12DL | 08°56.723'N 11°32.417'E | DILA | 38,200 ± 1200 | 586 ± 210 | 602.34 | 1445.62 | 0.76 | Borehole |
| 13 | GW13KPS | 08°53.458'N 11°30.927'E | KPANT ISAWA | 46,200 ± 1300 | 322 ± 160 | 728.48 | 1748.35 | 0.92 | Borehole |
| 14 | GW14NJ | 08°51.007'N 11°34.694'E | NYAJA | 45,900 ± 1300 | 510 ± 200 | 723.75 | 1737.00 | 0.92 | Borehole |
| 15 | GW15TP | 08°49.428'N 11°38.022'E | TAPEN LA | 41,300 ± 1300 | 348 ± 160 | 651.22 | 1562.93 | 0.83 | Open Well |
| 16 | GW16KS | 08°51.520'N 11°38.394'E | KASSA | 37,100 ± 1200 | 203 ± 130 | 584.99 | 1403.98 | 0.74 | Borehole |
| 17 | GW17BD | 08°51.119'N 11°41.119'E | BODU GA | 30,000 ± 1100 | 213 ± 130 | 473.04 | 1135.30 | 0.60 | Borehole |

Kakulu recorded the lowest ^{222}Rn concentration obtained from borehole measurements; this may mean that a borehole showing high radon concentration relative to others may have numerous fractures intersecting the borehole, with each fracture contributing to the total radon concentration in the borehole or well. This is because radon content of fluids is expected to respond to variations in ^{222}Rn flux as well as the extent and length of water/rock

contact, which in-turn depends on the fracture surface [22]. The radon concentration results presented in the table 1 above, coincides with radon concentration ranges in Swedish groundwater from Uranium-rich granites [23]. It is generally accepted that high radon levels will be found in terrain of high grade metamorphic rock and granites [24], most especially Uranium-rich ones.

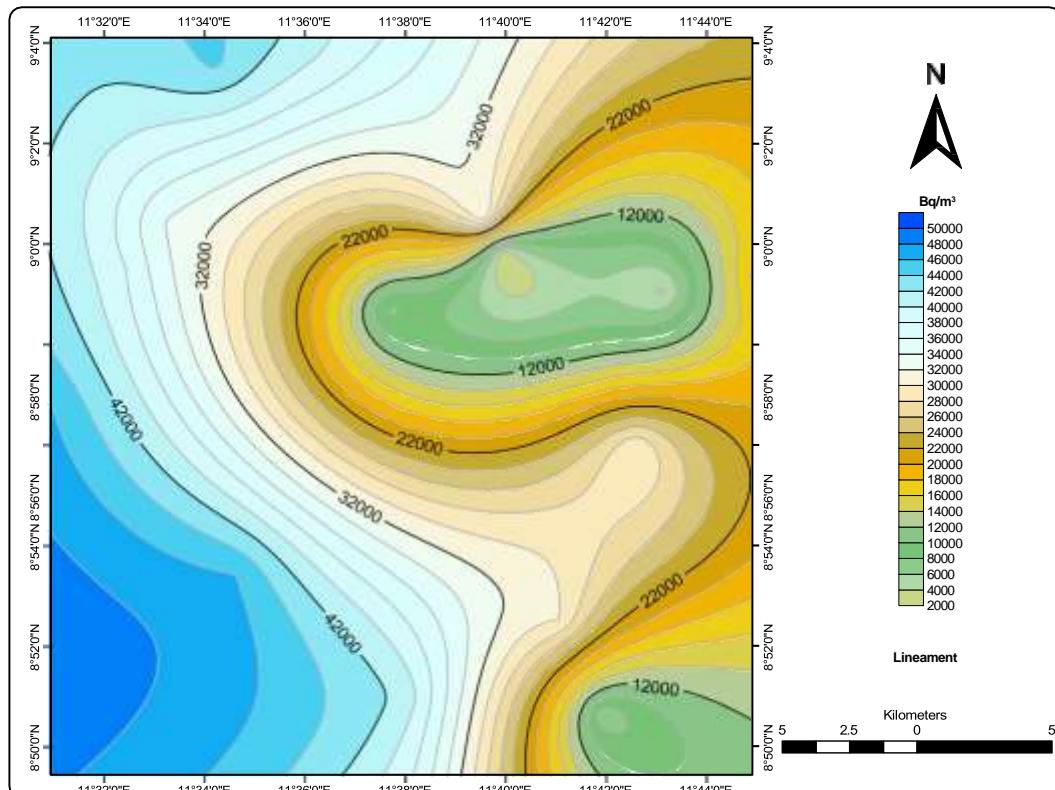


Fig. 5: Radon (in groundwater) contour map of the study area.

A superimposed contour maps of radon concentration (Fig. 5) and geologic map of the study area (Fig. 3), show that radon concentrations in groundwater are

higher within the medium-grained granites than within the coarse-grained granite (Fig. 6)

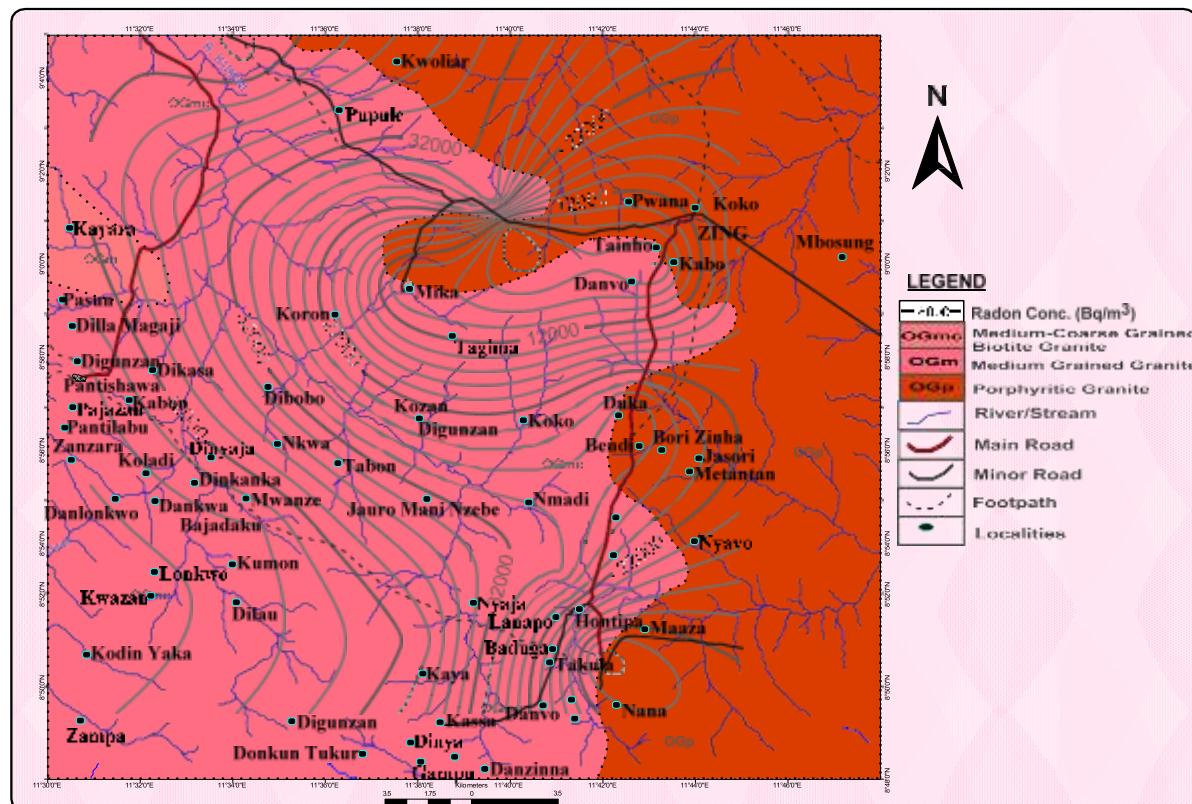


Fig. 6: Superimposed Radon contour map on the Geologic map of the study area.

5.0 SUMMARY AND CONCLUSION

This study was set-out to evaluate radon concentration in groundwater around Mika uranium mineralization.

Radon concentrations increases towards the southern and western parts of the study area mostly within the medium-grained granites, ranging from 2340 to 46,200 Bq/m^3 with an arithmetic mean value of 29,400 Bq/m^3 which is far above the ICRP upper reference level of 1500 Bq/m^3 . Thoron (^{220}Rn) concentration measured simultaneously with radon, has an average value of 240 Bq/m^3 from a range of 65.3 to 586 Bq/m^3 . The short half-life of thoron makes it less significant for this work therefore, was not considered further.

Potential dose rates from the degassing of radon in groundwater was calculated and the result obtained show an average annual absorbtion dose of 463.59 mSv/yr , average annual effective dose of 1112.62 mSv/yr and an average annual effective dose due to ingestion of groundwater was calculated to be 0.59 mSv/yr . These values are above the regulatory limits specified in the Nigeria Basic Ionization Radiation Regulation. While the doses due to ingestion were all within ICRP recommended limit of 1 mSv/yr for radionuclides ingestion through water, only 11.77% were found to be within WHO recommended reference level of 0.1 mSv/yr , the remaining 88.23% are above this limit.

Due to the high radon concentrations of the groundwater samples collected within the Mika uranium mineralization and environs, the groundwater can be said to pose a significant health risk even though the values do not warrant an immediate remedial actions, continuous monitoring of the groundwater and indoor air for radon concentration, is recommended.

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