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Mapping and Assessment of Activity Concentration of ^{222}Rn and its associated Exposures from the Drinking Water Sources of Taraba State, Nigeria

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Abstract

Radon is a colourless, odourless, and tasteless gas, which emanates from the natural disintegration of radioactive uranium in soils and rocks, dissolves, and accumulates in underground and surface water. ^{222}Rn is a natural and the most prevalent isotope of radon and poses the largest risk to humans. A Liquid Scintillation Counter (LSC) set-up at the Centre for Energy Research and Training (CERT), Ahmadu Bello University, Zaria, Nigeria, was used to determine the activity concentrations of ^{222}Rn in water samples from Taraba State. Water samples from Taraba had activity amounts of ^{222}Rn ranging from BDL to 11.5 Bq/L with an average value of $5.7 \pm 0.3\text{ Bq/L}$. This mean value was below the 10 Bq/L of WHO and 11.1 Bq/L of USEPA recommendations but was higher than 0.1 Bq/L acceptable limit for Nigerian Standard for Quality Drinking Water (NSQDW). The iso-concentration map of the state showed that Takum, Ussa, Bali, Sardauna, Gashaka, Yorro, Zing, Lau, Karim Lamido, and Jalingo LGAs of the state are hit with ^{222}Rn concentrations above 5.5 Bq/L , while Ardo-Kola, Ibi, Wukari, and Donga LGAs are hit with ^{222}Rn concentrations below 5.5 Bq/L . The yearly effective dosage (YED) resulting from ingesting radon-contaminated water in Taraba state were found in the range of *BDL to* $112.6\text{ }\mu\text{Sv/y}$ with a mean value of $55.7 \pm 2.8\text{ }\mu\text{Sv/y}$. The results of the statistical analysis showed a very strong positive correlation between the YED and activity concentration of ^{222}Rn in the state. An increase in the activity concentration of ^{222}Rn in the state relates to corresponding 9.82 mSv y^{-1} increase in YED in the state.

Keywords: ^{222}Rn , Activity concentration, Taraba State, Water, yearly effective dose

Introduction

^{222}Rn is a naturally occurring radionuclide, which builds up and dissolves in subterranean water sources. It is a decay product of uranium series, with half-life of 3.825 days, the most stable and prevalent isotope of radon. It comes from the alpha decay of alkaline earth element radium - ^{226}Ra , a radioactive decay chain and a by-product of ^{238}U 's disintegration (Akinagbe et al., 2018).

Depending on the soil's characteristics, such as porosity, moisture content, pore space ratio, etc, ^{222}Rn can enter the surrounding subterranean water at different rates (Manawi et al., 2024). For instance, ^{222}Rn travels more quickly through fractured rocks and permeable soils like gravel or coarse sand than through impermeable rocks and soils like clay soil.

About 42% of the public's exposure to ionizing radiation comes from radon, which is roughly 22% more than medical exposure, and 26% and 29% more than exposures from external terrestrial and cosmic sources, respectively (Darby, 2001).

Through drinking water, ^{222}Rn can reach the digestive system. Ingested ^{222}Rn can endanger delicate cells in the esophagus, stomach and intestines through irradiation (Hopke et al., 2000). Drinking water contaminated with ^{222}Rn poses a threat to public health, as it can cause cancer and other harmful health effects. Drinking water with elevated ^{222}Rn concentrations would increase the organ's effective dose and risk of digestive tract malignancies. Additionally, leukemia in children and acute leukemia in adults have been found to be highly associated with exposure to waterborne ^{222}Rn (Evrard et al. 2006). Also, exposure to waterborne ^{222}Rn was linked to prostate cancer, kidney cancer, childhood cancer and malignant melanoma (Henshaw et al., 1990; Little et al., 2018). Furthermore, exposure to waterborne ^{222}Rn has been connected to the mortality rate of stomach cancer (Manawi et al., 2024).

Tarabians primary source of drinking water is from subterranean sources, either as wells or boreholes, followed by surface water. Taraba State has been facing a lot of public health challenges ranging from disease outbreaks to cancer of various organs. Taraba State is among the states in northeast Nigeria with the highest cancer incidence, according to a 2022 Dugule report. Additionally, it was discovered that 84.4% of the 1090 Tarabians who were screened for cancer at the University of Maiduguri Teaching Hospital (UMTH) between 2004 and

2017 had cancer disease. Also, 488 of the 3237 patients screened in the Taraba State Teaching Hospital's histopathology department between 2011 and 2022 had cancer, according to Ezenkwa *et al.* (2023).

Taraba State has regions with high elevated terrestrial gamma radiation dose (TGRD) rates across almost the entire north-eastern part of the state (Odoh *et al.*, 2025), which may also contaminate the subsurface water in those areas above the recommended level. It is possible the ravaging public health issues in the state may be from the unverified effect of radon-concentration in the state, which, as far as the authors' knowledge; there is no epidemiological study that has verified this in Taraba. Hence, there is a need to assess the rate of ^{222}Rn concentrations in the state's water sources to see if it's related to high annual effective dose rates, a factor for cancer risk.

The aim of this paper is to assess the state's ^{222}Rn concentration in the state's water sources, evaluate the annual effective dose rates associated with it and represent ^{222}Rn concentrations in Taraba State on map.

Material and Methods

Study region

Taraba State, Nigeria was the region of this study, located within 5°N and 10°N and 7°E and 12°E. There are sixteen local government areas (LGAs) within it as shown in Figure 1. The state has four major rivers: Benue River, Donga River, Taraba River and Ibi River, which they all start from Cameroonian mountains and stretch almost through the entire state in the north and south directions and link up with Niger River. Taraba State is in the tropical zone of Nigeria with vegetation of sparse forest in the southern region and grassland in the northern region, with an undulated landscape with a few mountainous features.

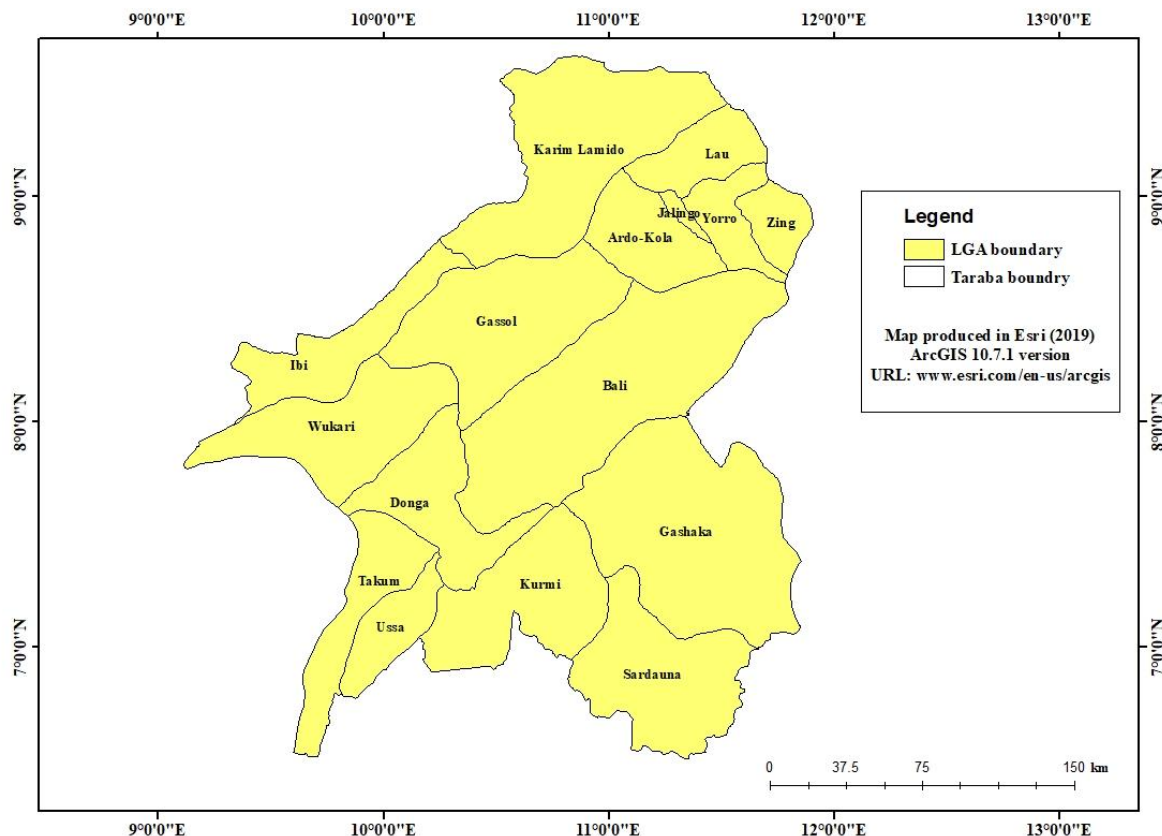


Figure 1: Map of Taraba State

Sample collection

A total of 71 water samples comprising 14 surface water (rivers and streams) and 57 groundwater (wells and boreholes) that were functioning as at the time of this study were gathered from the 16 LGAs of the state. The water samples were gathered randomly across the entire state in 500 ml bottles each, ensuring that all the water sources of the state were covered. In order to prevent the interference with the atmospheric radon, as described by Jorma (1993), the water was set to flow freely for some minutes while the sampling bottle was submerged beneath the water source's surface and securely recapped once filled before being removed from the water source.

The sample collection's time and date were noted. To minimize the impact of temperature on ^{222}Rn activity, the collected water samples were kept in a thermo-controlled container with constant temperature, and were taken to CERT, Zaria, for preparations and analysis.

Preparation of samples

Each liquid scintillation vial was filled with 10 ml of liquid scintillation solution using a syringe after being washed with distilled water in the lab. Ten millilitres of water were gently drawn using a hypodermic syringe after it had been immersed in the obtained sample. Through the vial's bottom, the water in the syringe was instantly moved into the liquid

scintillation vial holding the scintillation solution, and it was fastened firmly and shaken forcefully to extract ^{222}Rn from the water phase and transfer it to the organic scintillant solution.

Instrument Calibration

System calibration was done using the IAEA ^{226}Ra standard solution before counting. The solution was made by mixing 10 ml of organic scintillant solution with 10 ml of diluted ^{226}Ra standard solution in a vial. For 25 days, the vial was firmly sealed and stored in a dark location to enable ^{226}Ra and ^{222}Rn to reach secular equilibrium (Jorma, 1993). On the 26th day, the vial was shaking and was left for three more hours. After three hours, the vial was carefully placed inside the counting machine and counted for 60 minutes. A vial containing the organic scintillant solution was counted for the same 60 minutes in order to have a background count. Then, equation 1 was used to estimate the calibration factor (Jorma, 1993).

$$CF = \frac{VCR - BCR}{A_s} \quad 1$$

where VCR is the standard solution's count rate expressed in counts per minute (cpm), BCR is the blank counting vial's background count rate in cpm, and A_s is the activity of ^{226}Ra in the standard solution in Becquerel.

Counting

LSC was used to evaluate ^{222}Rn concentrations in the water samples. A cloth moistened with ethanol was used to carefully wipe the outside of each vial before each counting. After that, the vial was left for three hours to allow ^{222}Rn and its alpha-emitting decay products to attain secular equilibrium (Jorma, 1993). Following the three hours, each vial was directly put inside the liquid scintillation counter, and was countered for 60 minutes - a sufficient amount of time for low level measurement. For every sample, the time and date when counting started was recorded. To get the effective numbers for each sample, the background counts were subtracted from the total counts.

Evaluation of ^{222}Rn concentrations in the sample

The activity of ^{222}Rn concentrations in samples were evaluated using equation 2 (Jorma, 1993; Garba et al., 2013; Adams, 2017).

$$^{222}\text{Rn} = \frac{100 \times (SCR - BCR) e^{\lambda t}}{60 \times CF \times D} \quad 2$$

where ^{222}Rn is radon concentration in Bq/L , SCR is sample count rate in cpm, BCR is background count rate in cpm, λ is decay constant $= \frac{\ln 2}{T_{1/2}}$, and $T_{1/2} = 3.825$ days, t is the interval of time between sample collection and commencement of counting, the constants, 100 and 60 are conversion factors from per 10 ml to per litre and from min to sec, respectively, CF is the calibration factor of the counting system, D is the fraction of ^{222}Rn in the cocktail in a 22 ml total capacity vial, which is 0.964.

Risk assessment: Yearly effective dosage (YED)

A year's dose resulting from intake of radon-contaminated water was estimated using equation 3 (Garba et al., 2013; Adams, 2017; Kumar et al., 2022; Akinagbe et al., 2018; Opoku-Ntim et al., 2019):

$$D_{ing} (\text{mSvyr}^{-1}) = C_{water}^{Rn} (\text{Bql}^{-1}) \times CF_{ing} \times AWC \quad 3$$

where D_{ing} is the committed yearly effective dosage in mSv/yr , C_{water}^{Rn} is the ^{222}Rn concentration in water (Bq/l), CF_{ing} is the conversion factor for ingested of ^{222}Rn in water. According to UNSCEAR, this factor will be taken as $10^{-8} \text{ SvBq}^{-1}$ for an adult. AWC is the annual water consumption in litres. According to UNSCEAR, the amount of water an adult consumes each day is 2 litres, which amounts to 730 litres per year (SGV, 2024).

Results and Discussions

^{222}Rn concentration in the water samples

The evaluated ^{222}Rn concentrations in the water sources of Taraba State, using equation 2, ranged from below the detection limit (BDL) of the machine to 11.5 Bq/L with an average value of $5.7 \pm 0.3 \text{ Bq/L}$. In well, borehole and surface water sources, activities of ^{222}Rn concentration ranged from 2.5 to 11.5 Bq/L , 2.0 to 9.4 Bq/L and BDL to 10.6 Bq/L , in accordance, with average values of $5.8 \pm 0.4 \text{ Bq/L}$, $5.6 \pm 0.3 \text{ Bq/L}$ and $5.5 \pm 0.9 \text{ Bq/L}$, respectively. These mean values of the three water sources of the state were compared with the maximum contamination level of 11.1 Bq/L set by USEPA (1991, 1999, 2012, 2023) and the world average value of 10 Bq/L set by WHO (1993, 2004, 2008, 2017) in Figure 2. The pie chart revealed that the average activities of ^{222}Rn in the three water sources of the state are almost the same and are 11% and 14% below the WHO and USEPA recommendations, respectively.

However, almost all the water samples from the state, irrespective of the source, had an activity concentration of ^{222}Rn above 0.1 Bq/L acceptable limit set by the NSQDW (NIS, 2007), except in two surface water samples where ^{222}Rn was not detected.

The boxplots in Figure 3 compared the variability of the activities of ^{222}Rn in study area based on the water sources of the state. The plots revealed that the activities of ^{222}Rn in well and surface water sources in the state are right-skewed, indicating the presence of higher concentrations of ^{222}Rn pulling the average concentration towards the right, while for borehole water sources, activities of ^{222}Rn concentrations are left-skewed, indicating the presence of lower concentrations of ^{222}Rn in the water source.

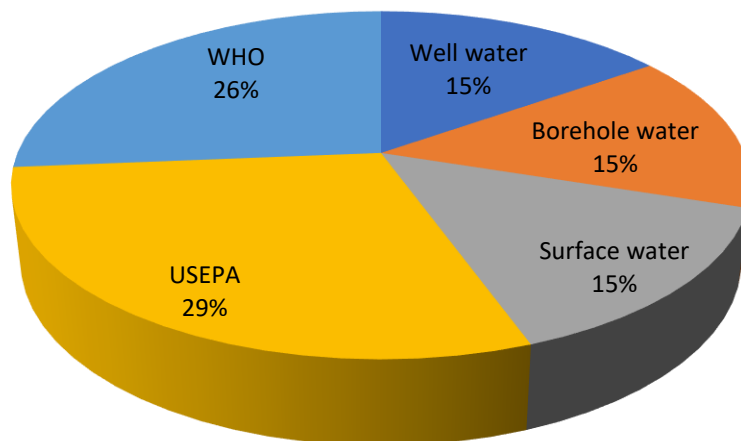


Figure 2: Comparing the mean activities of ^{222}Rn in Taraba Water sources with USEPA and WHO recommendations.

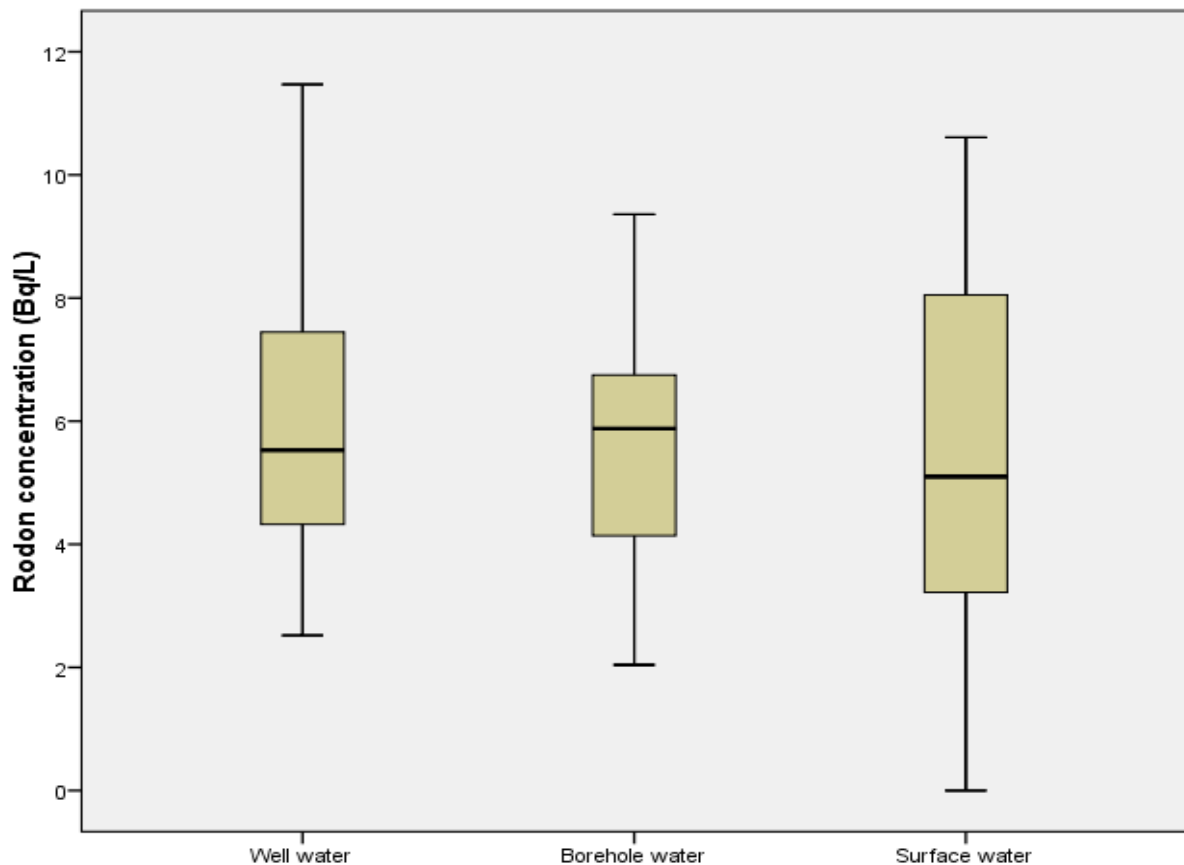


Figure 3: Box-and –whisker plots, comparing the activities of ^{222}Rn variations in water sources of Taraba State

Regions of equal concentration of ^{222}Rn in Taraba State

A map showing regions of equal concentrations of ^{222}Rn in the study area was drawn with Esri (2019) ArcGIS version 10.7.1 software, which is inbuilt with geospatial analytic tool, Inverse Distance Weighted (IDW). The IDW tool uses a linearly weighted combination of sample points to calculate cell values (Watson and Philip, 1985; Esri, 2019). The weight is determined by the inverse distance and is predicted on the assumptions that the activities of ^{222}Rn concentration being mapped decrease with distance from its sampled point. The tool calculated the magnitudes of ^{222}Rn concentration per unit area from each sampling point as shown in Figure 4. Figure 4 revealed the distributions of activities of ^{222}Rn concentration in the entire state. From the map, almost the entire state had a ^{222}Rn concentration within the range of 4.5 to 11.5 Bq/L, except in Ibi, Wukari, Donga and Ardo-Kola LGAs that recorded ^{222}Rn concentration within 0 to 6.5 Bq/L, they were designated as the regions of low concentration of ^{222}Rn in the state. High concentrations of ^{222}Rn within the range of 10.5 to 11.5 Bq/L were observed at Kasmbila town of Takum L.G.A. and Maisamari town of Sardauna L.G.A., designated as the regions of highest concentration of ^{222}Rn in the state. In

Gwagwala village at Wukari L.G.A., activities of Rn concentrations were recorded within the range of 0 to 1.5 Bq/L, representing the regions of lowest concentration of ^{222}Rn in the state.

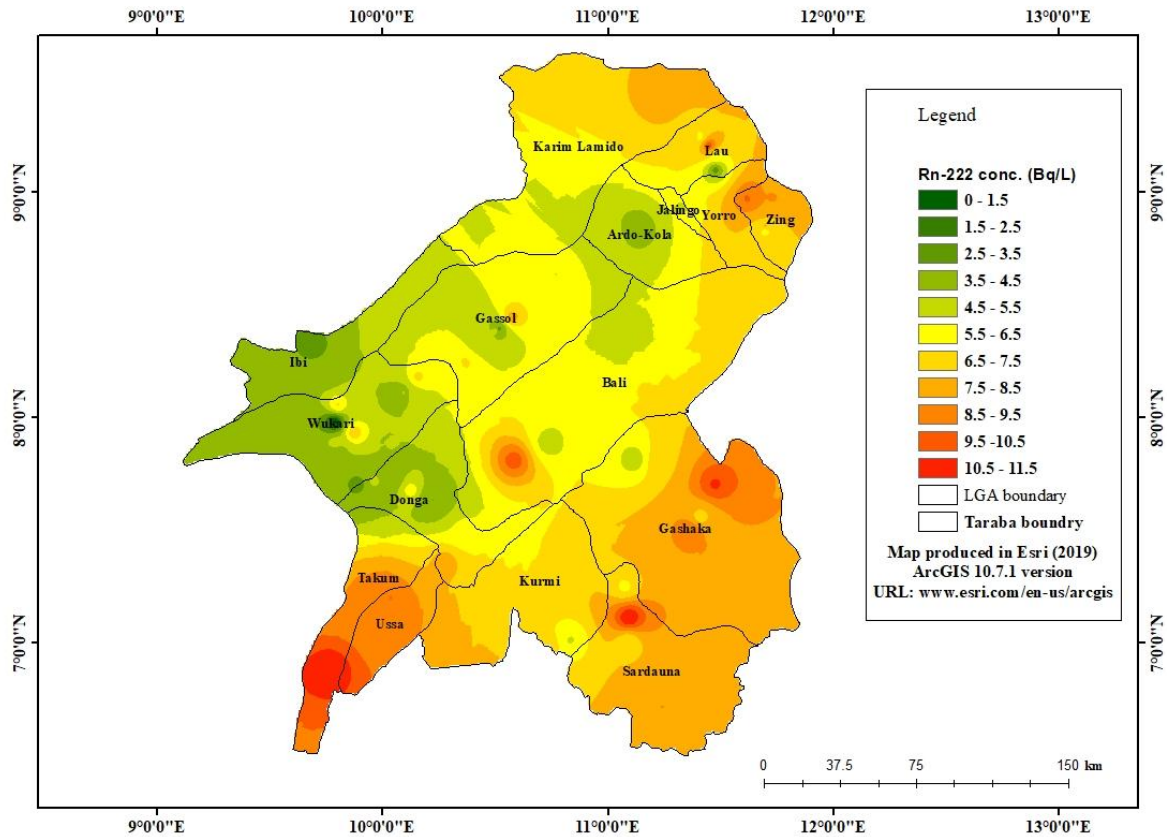


Figure 4: Map of Iso-concentration of ^{222}Rn in Taraba State

Yearly Effective Dosage (YED)

The YED resulting from ingesting of water contaminated with radon in Taraba State were presented in Table 1. According to the table, YED in surface, borehole and well water in the state ranged from BDL to $104.2 \mu\text{Svy}^{-1}$ with an average of $53.5 \pm 9.1 \mu\text{Svy}^{-1}$, 20.0 to $91.9 \mu\text{Svy}^{-1}$ with an average value of $55.1 \pm 4.0 \mu\text{Svy}^{-1}$ and 24.8 to $112.6 \mu\text{Svy}^{-1}$ with an average value of $57.2 \pm 3.9 \mu\text{Svy}^{-1}$. The evaluated mean values for all the water sources of the state were found below the recommended WHO value of $100 \mu\text{Svy}^{-1}$ (WHO, 2004, 2008 and 2017). Among the water samples, the highest YED of $112.6 \mu\text{Svy}^{-1}$ was observed in a well water sample collected from Sohon Gari village of Mika in Yorro L.G.A., which is 12.6% higher than the $100 \mu\text{Svy}^{-1}$ WHO recommendation. This means inhabitants of Sohon Gari village of Mika are slightly exposed to ^{222}Rn health-related issues if they consume well water from their location directly without proper treatment.

Table 1: YED for the adult population in Taraba State

Source of Water	Number of samples	Min YED (μSvy^{-1})	Max YED (μSvy^{-1})	Mean (μSvy^{-1})	
				Mean YED	Std. Error
Borehole water	26	20.0	91.9	55.1	4.0
Well water	31	24.8	112.6	57.2	3.9
Surface water	14	BDL	104.2	53.5	9.1

The mean YED of each of the water sources in Taraba was compared with the WHO-recommended value in Figure 5. The pie chart revealed that the mean value of the three water sources in Taraba were below the WHO recommended value of $100 \mu\text{Svy}^{-1}$ by 17%, 17% and 18% for borehole water, well water and surface water, respectively.

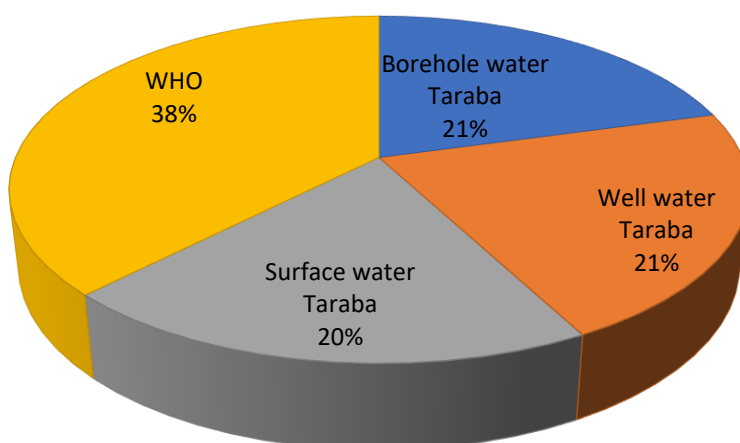


Figure 5: Comparing the mean YED of each of the water source in Taraba with WHO recommendation.

Statistical analysis

Linear regression analysis was carried out between the ^{222}Rn concentration and YED in the studied area to ascertain the kind of association between the variables. The analysis revealed there is a linear correlation between the YED and ^{222}Rn concentrations in the state as shown in Figure 6, with a correlation coefficient of 0.9999 as indicated in Table 2. The R-squared value of 0.9999 (Table 2) indicates that the variation between the YED and ^{222}Rn concentration in the studied area is 99.99%, meaning a very strong positive relationship between YED and ^{222}Rn concentration. Moreover, the unit value of R-squared in Figure 6

also indicated a very strong positive relationship between the two data sets. The standard error of 0.0269 in Table 2 shows the regression analysis is very accurate. This indicates that our data points, on average, drop 0.0269 units from the regression line. The significance value of 2.86×10^{-205} in the analysis, which is far less than 0.05 indicates the regression analysis, is statistically sound. Therefore, the ^{222}Rn concentrations have a statistically significant association with YED, which strongly indicates there is a correlation between ^{222}Rn concentrations and YED in Taraba State.

Table 2: Regression output summary

Regression Statistics						
Multiple R	0.999999369					
R Square	0.999998739					
Adjusted R Square	0.999998721					
Standard Error	0.026930856					
Observations	71					
ANOVA						
	Df	SS	MS	F	Significance F	
Regression	1	39683.8741	39683.8741	54715925	2.8595E-205	
Residual	69	0.0500437	0.00072527			
Total	70	39683.92414				
	Coefficients	Standard Error	t Stat	P-value	Lower 95%	Upper 95%
Intercept (YED)	0.006651312	0.008177885	0.81332916	0.4188271	-0.00966312	0.022966
²²² Rn concentration	9.819029747	0.00132743	7397.02135	2.86E-205	9.816381596	9.821678

The nature of the linear relationship existing between YED and ^{222}Rn concentrations in Taraba State was graphically viewed in Figure 6. The graph indicates that a unit increase in the concentration of ^{222}Rn in Taraba State significantly relates to correspond approximately

9.82 μSvy^{-1} increase in YED, as described in the linear regression output model in equation 4:

$$YED = 0.0067 + 9.819 (^{222}\text{Rn concentration}) \quad 4$$

From equation 4, the interceptor of 0.0067 indicates that 0.0067 μSvy^{-1} of YED represents the YED accounted by ^{222}Rn concentration from other sources like soil, rocks etc., other than waterborne radon in the state. Meanwhile, the coefficient of ^{222}Rn concentration in equation 4 (9.819) indicates that 9.819 μSvy^{-1} is the rate at which YED changes with per unit changes in ^{222}Rn concentration in Taraba State. That is, for each additional 1 BqL^{-1} of ^{222}Rn concentration in Taraba state, an average YED of 9.819 μSvy^{-1} is expected to increase, assuming ^{222}Rn contributions from other sources are constant.

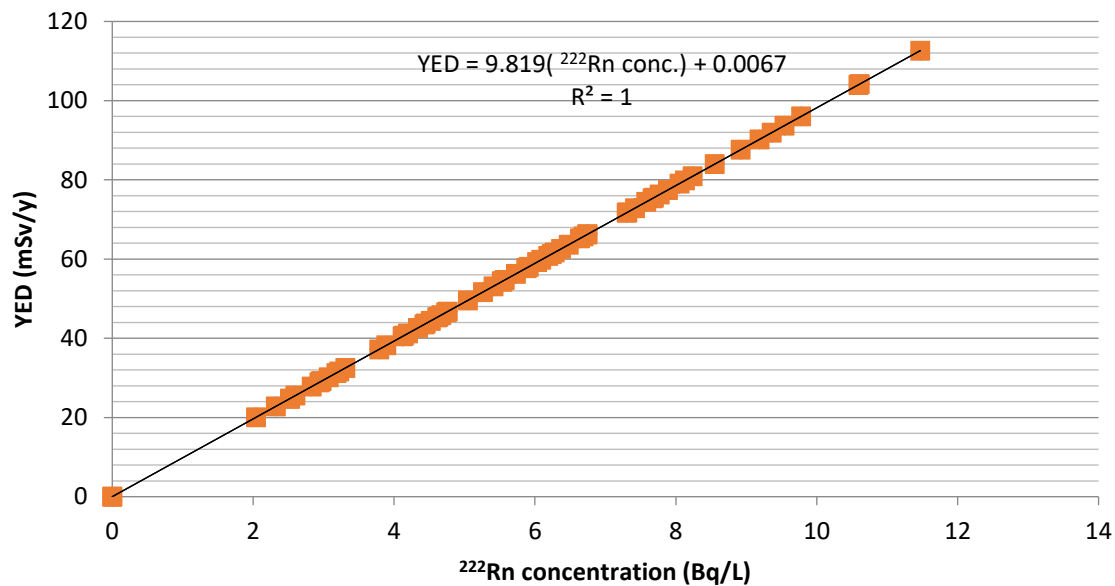


Figure 6: A graph of YED ($\mu\text{Sv/y}$) against ^{222}Rn concentration (Bq/L) in Taraba State

Conclusion

This study has investigated the activity concentrations of ^{222}Rn and its associated risks in the water sources of Taraba State. It has shown that the amount of ^{222}Rn present in the water sources of the state ranged from BDL to 11.5 Bq/L , with an average activity concentration of $5.7 \pm 0.3 \text{ Bq/L}$. This mean value, when compared with 10 Bq/L WHO and 11.1 Bq/L USEPA recommendations, was found to be 43% and 49%, respectively lower than their recommended limits. However, all the analysed water samples had activity concentrations of ^{222}Rn above 0.1 Bq/L acceptable limit of the NSQDW, except in two samples where activity

concentrations of ^{222}Rn were not detected. The iso-concentration map of the state showed that the LGAs hit with high activity concentrations of ^{222}Rn in Taraba are the southern part of Takum, Ussa, Bali, the northern part of Sardauna, and Gashaka LGAs of the state. The estimated YED due to the amount of ^{222}Rn in the state ranged from *BDL to* $112.6\ \mu\text{Sv/y}$, having a mean value of $55.7 \pm 2.8\ \mu\text{Sv/y}$. This mean value was 44.3% below the $100\ \mu\text{Sv/y}$ WHO recommendation.

The statistical analysis of the findings in Taraba State revealed that there is an extremely strong positive correlation between activity concentration of ^{222}Rn and YED in the state. A unit Bq/L increase in the activity concentration of ^{222}Rn in the state relates to $9.819\ \mu\text{Sv}^{-1}$ increase in YED.

Since the activity concentration of ^{222}Rn in Taraba State is above the $0.1\ \text{Bq/L}$ maximum allowable limit established by the NSQDW in almost all the water samples, it is advised that the inhabitants of Taraba State should not ingest water from the state directly without proper treatment irrespective of the water source, except in Gwagwala village of Wukari L.G.A., where the activity concentration of ^{222}Rn was not detected in their surface water sample.

It is recommended that future study in the state should look into the levels of ^{222}Rn in the air and soils in the state. This will help researchers get more complementary data to work with. It also recommends that a review of the public water system should be carried out in the state, and the inhabitants of the state should be made aware of ^{222}Rn , its health impacts, and the corrective measures required in reducing the amount of ^{222}Rn in water to the barest minimum.

Data availability statement

The datasets gathered and processed in this study are not publically available due to privacy issues in a small, closely connected group. Data are accessible from the corresponding author on a legitimate request.

Declarations

Funding: There was no external funding or scholarship to this study.

Ethics, Consent to Participate and Consent to Publish: Not applicable

Conflict of Interests: No potential conflicts of interest were declared by the authors.

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