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Research article

Pilot groundwater radon mapping and the assessment of health risk from heavy metals in drinking water of southwest, Nigeria

Yinka Ajiboye [a], Matthew Omoniyi Isinkaye [b], Ganiyu Olabode Badmus [a,*], Oluwaseun Temitope Faloye [c,e], Vincent Atoiki [d]

[a] Department of Mathematical and Physical Sciences, Afe Babalola University, Ado-Ekiti, Nigeria
[b] Department of Physics, Ekiti State University, Ekiti State, Nigeria
[c] Department of Food and Biosystems Engineering, Afe Babalola University, Ado Ekiti, Nigeria
[d] Department of Science Technology, Federal Polytechnic, Ado-Ekiti, Nigeria
[e] Department of Water Resources Management and Agrometeorology, Federal University, Oye-Ekiti, Nigeria

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ABSTRACT

Radon and heavy metals are sources of groundwater pollution and are identified as potential carcinogens. Southwest Nigeria's populace mostly relies on groundwater source for drinking. This study aims to map radon distribution in groundwater of southwest Nigeria and to determine the health risk of radon and heavy metal in drinking water. Radon concentrations of 145 groundwater samples were measured using RAD7 electronic radon detector and heavy metal concentrations of 52 groundwater samples were measured using atomic absorption spectrophotometer. Radon concentration distributions were delineated using geographical information system. Radon concentration of water samples ranges between 1.6 Bq l^{-1} and 271 Bq l^{-1} with an average value of 35.9 ± 38.4 Bq l^{-1}. The average groundwater radon concentration is higher than US-EPA recommended level of 11.1 Bq l^{-1} but lower than the WHO recommended limit of 100 Bq l^{-1}. The estimated average annual effective radiation doses to infants, children, and adults are 29 $\mu Sv\, y^{-1}$, 41 $\mu Sv\, y^{-1}$ and 92 $\mu Sv\, y^{-1}$ respectively. The radon distribution map of the study area reveals regions of high, medium, and low groundwater radon concentrations. The average concentration values of heavy metals in groundwater samples are of the order Mn > Zn > Pb > Cu > Cr > Ni > Cd. 84% of groundwater exhibits good to excellent quality in terms of heavy metal pollution. However, about 16% of the samples which lie in the sedimentary regions of Ogun and Lagos States exhibit poor to very poor quality. Overall, ingestion of groundwater in the study area may not pose a serious health hazards from radon ingestion and heavy metal toxicity.

1. Introduction

Radon and heavy metals are considered major pollutants which pose a serious threat to water quality. These two water pollutants are well known for their carcinogenic potentials (Marković et al., 2020; Ahmad et al., 2020). Due to the rapid increase in world population, exploitation of water resources has also increased to cope with the rising demand from domestic usage, particularly for drinking purposes (Díaz-Alcaide and Martínez-Santos, 2019). Nigeria is the most populated country in Africa with a population of over 200 million. The majority of Nigerian residents rely on groundwater as the main source of drinking water due to inadequate potable water distribution (Abubakar, 2018). Groundwater contributes about 60% of the drinking water source to the Nigerian populace (Omole, 2013). Residents in most villages drink directly from water fetched from hand-dug wells. Whereas in many households in towns and cities, water from wells is pumped through the use of motorized pumps into overhead tanks from where the water is channeled through pipes into the houses for domestic utilization such as showering, dishwashing, and drinking. Pollution of the underground water sources with radon and heavy metal is, therefore, a threat to the health of the population.

Radon is a colourless and odourless radioactive gas that exists naturally in soil, air, and groundwater. It exists in three different isotopic forms as: ^{222}Rn ($t_{1/2} = 3.8\ d$), ^{220}Rn ($t_{1/2} = 55.6\ s$) and ^{219}Rn ($t_{1/2} = 4\ s$) in the ^{238}U, ^{232}Th and ^{235}U decay series respectively. Due to the relatively short half-lives of the latter two radon isotopes, ^{222}Rn is mostly given

* Corresponding author.
E-mail address: ogbadmus@abuad.edu.ng (G.O. Badmus).

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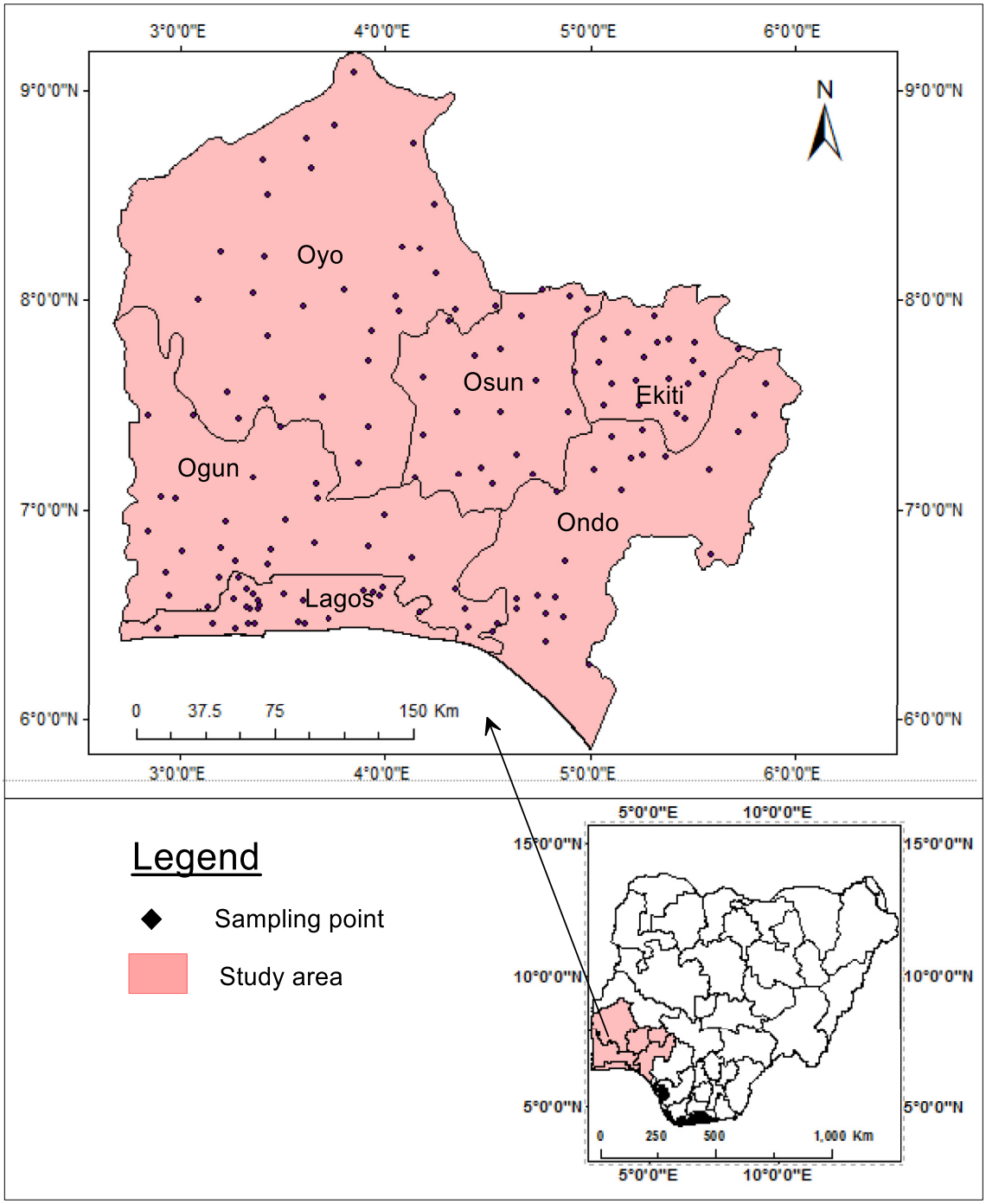

Figure 1. The Southwest Nigeria map showing sampling locations (Inset: Map of Nigeria).

significant consideration from a radiological health hazard perspective. Since ^{238}U is present in all rocks and soil, its only gaseous decay product, ^{222}Rn, is produced from the rocks and soils and then moves through diffusion into pore spaces within soil grains (Szabó et al., 2013). The movement could either be into the soil gas within the pore spaces, dissolution into groundwater occupying the pore spaces, or re-adsorption into the soil grains (Nazaroff, 1992). The concentration of radon within the soil pore spaces depends on the level of radium content of the soil (Khan et al., 2012). The radon gas produced at depths within a few meters to the surface soil tends to make it to the surface before it completely decays. The concentration of radon within the soil pore spaces may accumulate and build up to higher pressure than the atmospheric pressure, thereby leading to radon influx into the atmosphere and indoor environments (Iakovleva and Ryzhakova, 2003).

Radon has several economic uses such as therapeutic applications in spas and mines. Thousands of people especially from Europe and America seek radon therapy in spas scattered across the continents despite regulatory agencies' warnings due to the risk of radon exposure as a potential carcinogen (Erickson, 2007). Its application is also found in geological fault prediction, earthquake precursor, and groundwater-freshwater mixing quantification (Dimova and Burnett, 2011; Hamada, 1999; Reddy and Nagabhushanam, 2011). Despite the economic importance of radon, it is also a source of health hazards to humans. Studies on underground miners of uranium exposed to high levels of radon revealed

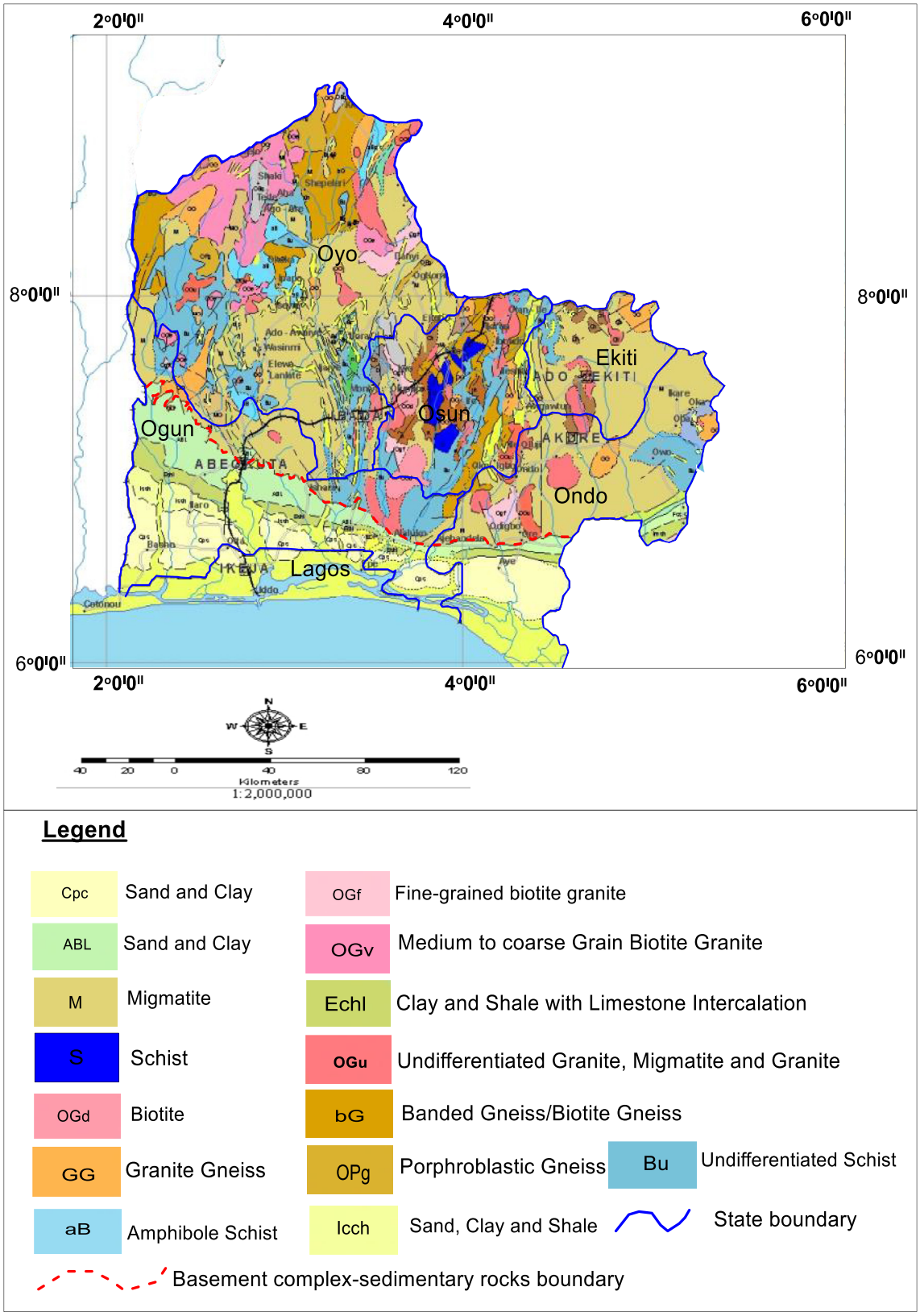

Figure 2. Geology of southwest Nigeria.

that inhalation of radon present risks of incidence of lung cancer (Tomášek et al., 1994). Several studies have also investigated the risks associated with low-level exposure in indoor environments (Samet, 2006; Sandler et al., 2006; Thumvijit et al., 2020). Over half of the total radiation dose from natural sources is attributable to radon (UNSCEAR, 2000). Health risks due to inhalation of radon in the air have been well-studied, however, the risk posed to health from ingestion of radon-borne water has not been studied so much. Groundwater

Table 1. Radon concentration in groundwater of the six southwestern states of Nigeria.

State	Number of samples	Population ($\times 10^6$)	Radon Concentration (Bql^{-1})			
			Minimum	Maximum	Mean	Std. Dev.
Oyo	30	7.8	1.8	125.9	23.8	28.1
Ondo	20	4.7	26.0	64.5	38.3	11.3
Ekiti	20	3.3	9.5	271.0	81.7	69.3
Osun	20	4.7	5.8	109.5	47.5	36.2
Ogun	30	5.2	1.6	52.6	26.0	15.2
Lagos	25	17.5	5.1	21.2	14.3	4.6
Southwest Nigeria (Total)	145	43.2	1.6	271.0	35.9	38.4

employed for drinking purposes with a high radon concentration increases the risk of the development of stomach and gastrointestinal cancers (Messier and Serre, 2017).

Radiological hazard from ingestion of groundwater-borne radon has two major pathways – gastrointestinal exposure from drinking of water which poses a potential risk of stomach cancer and respiratory tract exposure which may arise from inhalation of radon released from water to air which can lead to the occurrence of lung cancer. Potential radiation dose deposition to other body organs which may occur from the uptake and redistribution of water through the bloodstream has also been investigated (Chen et al., 2018a, b; Salih et al., 2016). The radon in the indoor air pathways is brought about by domestic utilization of water such as showering, dishwashing etc. The ingestion pathway of radon exposure is also significant. Typically, the radon concentration in wells is several orders of magnitude higher than radon concentration in surface water (Mittal et al., 2016). Radon concentration in groundwater has also been found to exhibit dependency on types of the well (Isinkaye and Ajiboye, 2017; Isinkaye et al., 2021).

To limit the exposure of the public to radon, guidelines in radon exposure on drinking water are set by international and local agencies to protect people from adverse health effects of the radioactive gas. The World Health Organization in its 2008 guidelines recommended that groundwater employed for public consumption should not have radon concentration above 100 $Bq\,l^{-1}$ (WHO, 2008). The United States Environmental Protection Agency (USEPA) recommended a radon level limit of 11.1 $Bq\,l^{-1}$ in drinking water for States without enhanced indoor policy (Hopke et al., 2000).

Heavy metals in the context of this research refer to potentially toxic metals with relatively high specific gravity and atomic weight. These metals are toxic in the sense that they are poisonous even at low concentrations. They include lead (Pb), arsenic (As), cadmium (Cd), zinc (Zn), gold (Ag), copper (Cu), iron (Fe), chromium (Cr), nickel (Ni), etc. Heavy metals are naturally occurring in the environment. Some are biologically essential, that is, they play important roles in metabolic processes. They include zinc (Zn), iron (Fe), nickel (Ni), and copper (Cu). Bioaccumulation of these metals can disrupt the function of vital organs such as the liver, kidney, heart, etc. The transport of heavy metals in the environment may be through natural (Chen et al., 2018a, b; Eid et al., 2018) or anthropogenic media (Zhang et al., 2019) such as those released from mining activities, industrial wastes, automobile exhaust, and agricultural activities. Many studies across the world have also determined the carcinogenicity of heavy metals (Taiwo et al., 2018; Mohammadi et al., 2019).

Different studies have been carried out in Nigeria to determine the distribution of radon in drinking water (Ajiboye et al., 2018; Isinkaye and Ajiboye, 2020). The studies were however limited geographically. Data from such studies are not robust enough to be employed by regulatory agencies for purpose of decision-making. Due to this reason, the national regulatory agencies do not have sufficient data on which regulatory limits could be set. In this study, an attempt has been made to investigate the distribution of radon in groundwater in the six southwest states of Nigeria. The radiation risk to infants, children, and adults population in Southwest Nigeria from radon in groundwater has been evaluated. Geographical Information System has been used to map the distribution of radon concentration in groundwater. The groundwater samples obtained from wells employed for drinking purposes were investigated for seven important heavy metals (Pb, Cd, Ni, Cr, Zn, Cu, and Mn) from a toxicological perspective. Associated toxicological indices from the heavy metals were also determined.

2. Study area

Southwest Nigeria is one of the six major geopolitical zones of Nigeria. It comprises six States and has a latitudinal span which stretches from $5^o49'$ to $9^o15'$ North, while that of longitude is between $2^o40'$ and $6^o1'$ East (Figure 1). The area has a landmass of 78,000 km^2 and a population of about 43 million which is greater than the populations of over 80% of African countries. The area is majorly dominated by basement complex rocks, with sedimentary basins existing around the coastal region of Lagos, Ogun, and Ondo. The annual average rainfall in the area is between 400 and 2100 mm.

2.1. Geology and hydrogeology of the area

The geology of Nigeria is generally defined with three lithopetrological components namely: Basement complex, Younger granite, and Sedimentary Basins (Oli et al., 2019). The study area is underlain by both the Precambrian Basement Complex rocks and the Sedimentary rocks of the broad Dahomey Basin which spans from Nigeria to Ghana (Figure 2).

The Basement complex rocks dominate the northern part of the study area covering Oyo State, Osun State, Ekiti State, and the northern part of Ogun State. It is characterized by granite gneiss, undifferentiated schist, migmatite, porphyritic granite, biotite granite, granodiorite, and older granite. This complex formation impacts permeability and porosity essentially needed for a robust groundwater occurrence. However, the deformation of the Basement Complex has resulted in the formation of joints and fractures which significantly enhance the groundwater occurrence in the basin.

The sedimentary rocks which cover Lagos, and part of Ondo and Ogun State begin with the Abeokuta Group (sand, clay, and shale). Other Tertiary strata that overlie the Abeokuta Group includes Ewekoro Formation (shale and limestone), Akinbo Formation (shale and clay), Oshosun Formation (clay, shale, and sandstone), Ilaro Formation (sandstone and limestone), and the Benin Formation (sand, silt, and clay). The prevailing sedimentary formation in the study area is the Benin Formation which is considered permeable as it consists of an alternation of sands and clay. Generally, the occurrence, accumulation, and movement of groundwater in the study area are primarily controlled by geology (Joel et al., 2020).

The hydrogeology of Southwest Nigeria varies across the six states of the region. Lagos and southern regions of Ondo and Ogun States are sedimentary terrains having different aquifer types including unconfined, semi-confined, and confined aquifers (Yusuf and Abiye, 2019).

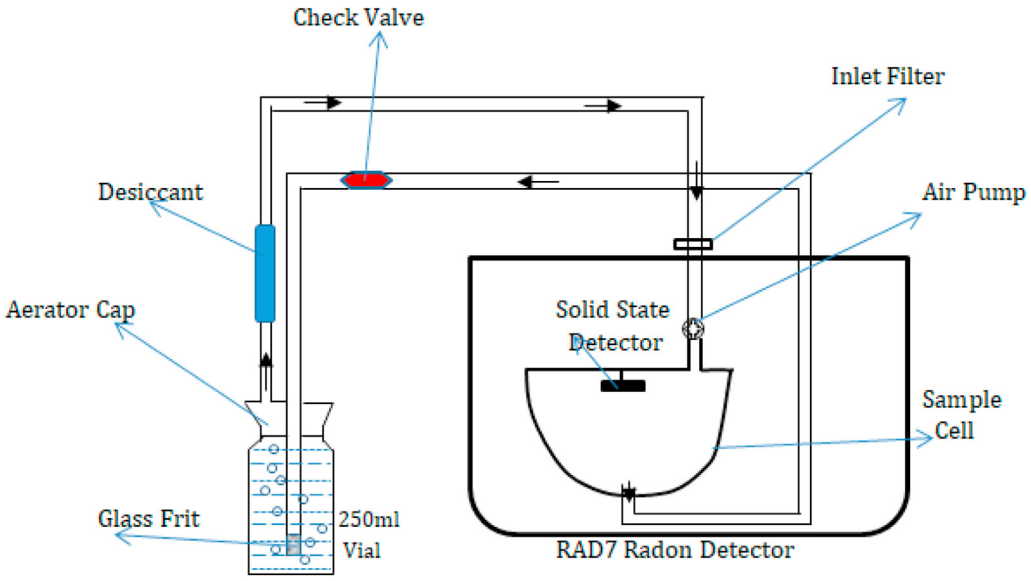

Figure 3. Schematic diagram showing set-up for radon-in-water measurement.

Other states of the southwest region comprising of Oyo, Osun, Ekiti, and Northern parts of Ondo and Ogun all lie in the basement complex of Southwest Nigeria. The main aquifers in these states have been reported to be weathered basement and fractured aquifers (Oyedele et al., 2019; Badmus and Olatinsu, 2010). The majority of the drinking water wells in Southwest Nigeria are hand-dug wells that tap into shallow aquifers (Egbinola and Amanambu, 2014).

3. Materials and methods

3.1. Sampling method

The water sample collection method was determined by the well types and water retrieval systems in the different localities where samples were collected. The majority of the wells in the area are open wells with an average diameter of 1.2 m and depths ranging from 15 m to 35 m. Water samples were collected spatially across the six Southwest States of Nigeria as indicated in Table 1. All sampled well are within the unconfined aquifer. The sample collection method was carried out in the same way members of the public fetch water from the wells to correctly estimate their radiation exposure to radon from the water sources. Water samples were collected from the wells with the aid of bailers and carefully fed into a 250 ml volume sample vial according to the EPA drinking water sample collection guide (USEPA, 2016). Water samples were collected from manual pump wells by pumping carefully directly into the 200 ml sample vial while ensuring minimal aeration of the sample. Due to the high volatility of radon gas (Freyer et al., 1997), water samples were sealed with the provided air-tight sample vial caps and thereafter transported to the laboratory for analysis. Groundwater samples collection across the six southwest states of Nigeria was determined spatially. Samples collection per state was based on the landmass and also the population density. Also, at selected sites, 250 ml water samples were collected in plastic bottled to determine the heavy metal composition and concentration.

3.2. Radon measurement techniques

Measurement of ^{222}Rn in groundwater samples was carried out using a RAD7 electronic radon detector with RadH_2O accessories. The RAD7 system which comprises of RAD7 unit, aerator assembly, and desiccant (Figure 3) is a versatile system that enables radon measurement within minutes in the concentration range between 0.004 and 750 Bq L^{-1} with ±5% absolute accuracy. Before each measurement, the RAD7 sample cell was thoroughly dried out using the supplied laboratory drying unit (bigger desiccant), since high humidity in the sample cell of the RAD7 reduces the efficiency of collection of ^{218}Po (^{222}Rn decay product). The air passes through an inlet filter with a pore size of 1 mm which serves the purpose of blocking fine dust particles and all radon daughters from entering the sample cell. The measurement setup is a closed-loop aeration scheme in which both water and air volume remain constant and independent of the flow rate. Wat-250 protocol was used for the measurement which allows such that air recirculates through the water sample and extracts the dissolved radon until a state of equilibrium is achieved. For the Wat-250 protocol, the extraction efficiency is typically 94%. The extracted radon is collected into a hemispheric sample cell of RAD7 unit which has a volume of 0.7 L. The cell is coated with an electrical conductor. There is a solid state ion-implanted planar silicon alpha detector at the centre of the sample cell. A very high potential difference (2,000 to 2,500 V) created by the electronics of the RAD7 is applied between the detector and the surface of the sample cell. The created electric field propels the positive ^{218}Po ion which results from the decay of the ^{222}Rn nucleus within the cell toward the detector. The ^{218}Po ions which are electrostatically drawn to the detector decay upon the active surface of the detector by emitting an alpha particle which has a 50% probability of entering the detector and producing an electrical signal which is proportional to the energy of the alpha particle. The electrical signal is amplified and the RAD7 determines the ^{222}Rn concentration using the ^{218}Po signal.

Measurement of radon concentration in groundwater samples at a later time in the laboratory does not truly represent the original concentration. It is therefore important to correct measured concentration to concentration at the time of sampling. The measured concentration is corrected using the decay correction factor (DCF). The relationship between activity A of any radionuclide at any time (t) and original activity A_o is:

$$A = A_o\left(e^{-\lambda t}\right) \tag{1}$$

where

$$\lambda\ (\text{decay}\ \ \text{constant}) = \frac{0.693}{t_{1/2}} \tag{2}$$

where $t_{1/2} = 3.82\ d$ is the half-life of radon. The factor $e^{-\lambda t}$ is the decay correction factor, where λ is the decay constant. The measured radon

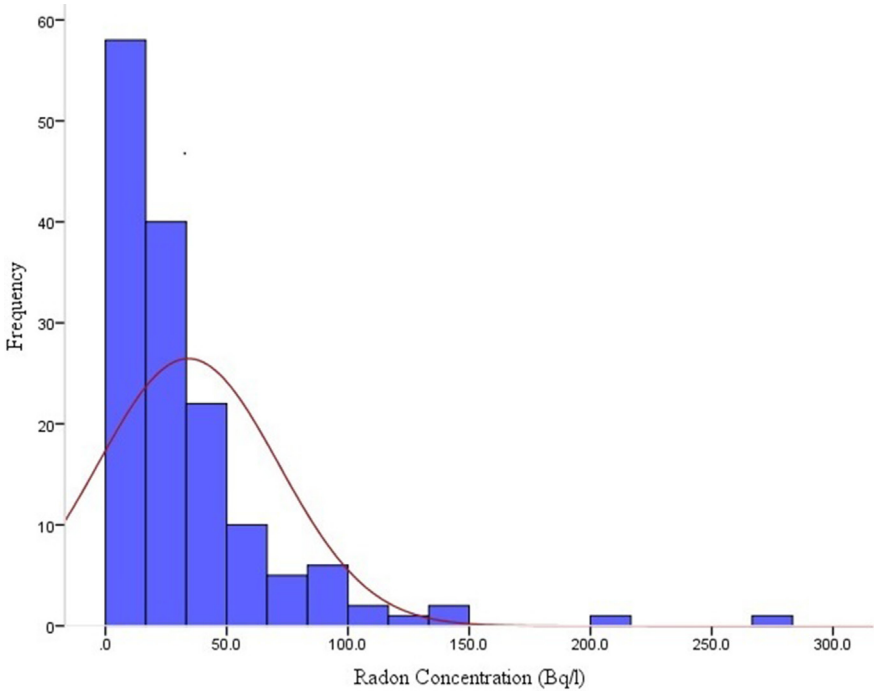

Figure 4. Radon concentration distribution in groundwater.

concentration is multiplied by the DCF to obtain the corrected radon concentration.

3.3. Heavy metals determination techniques

The materials employed for the measurement of heavy metals in water include 100 ml sample bottles, stock standard solution of the element, 100 ml standard flask, pipettes, and distilled/deionized water. Before measurement, all glasswares were washed with distilled water to guide against sample contamination. 10 ml of the stock standard solution was pipetted into a clean 100 ml standard flask and then made up with distilled water. This solution is 100 ppm of the standard. From the 100 ppm, 0.5, 1.0, 1.5, 2.0, and 2.5 ml were pipetted to different 100 ml standard flasks each and were made up to 100 ml with distilled water. Each sample bottle was filled with a specific standard and thereafter appropriately labeled. An atomic absorption spectrophotometer (AAS), model 211 VGP manufactured by Buck Scientific was used for the metal concentration analysis. The AAS works on the principle that atoms in a ground state absorb light of wavelengths which are characteristic to each element when light is made to pass through the atoms in a vaporized state. For each element to be analysed, the instrument was auto-zeroed using distilled water. The standards were thereafter aspirated into the flame from the highest to the lowest concentration. The corresponding absorbance was obtained by the instrument and the graph of absorbance against concentration was plotted. The samples were analysed with the concentrations of displayed metals in parts per million (ppm) after extrapolation from the standard curve.

3.4. Estimation of radiation doses

The annual effective dose from ingestion of water, D_{Ing}, is estimated using Eq. (3) as given in UNSCEAR (2000).

$$D_{Ing} = Rn \times V \times F \tag{3}$$

where Rn is the experimentally determined radon concentration in water in $Bq\,l^{-1}$, V is water consumption per year $(l\,y^{-1})$, and F is the dose conversion factor in $Sv\,Bq^{-1}$. For the computation of D_{Ing} for infants, children, and adults, water consumption rates of 230, 330, and 730 $l\,y^{-1}$ (Grandjean, 2005) were used respectively. The respective radon F values used are 23, 5.9, and 3.5 nSv Bq^{-1}.

Estimation of the annual effective dose from inhalation of radon released from water, D_{Inh}, was done using Eq. (4) (UNSCEAR, 2000):

$$D_{Inh} = Rn \times CR \times O \times E_q \times DCF \tag{4}$$

where CR is the air-water concentration ratio (10^{-4}) arising from the assumption that a radon concentration of 1000 Bql^{-1} in drinking-water will increase indoor radon concentration by 100 Bqm^{-3} (1 Bql^{-1}), O is the occupancy factor (7000 hy^{-1}), E_q is the equilibrium factor given as 0.4, and DCF is the dose conversion factor given as 9 nSv$(Bqhm^{-3})^{-1}$.

The population-weighted average dose D_{Pop} for the six southwest states of Nigeria was estimated using Eq. (5):

$$D_{Pop} = \frac{\sum_{i=1}^{n} D_i \times P_i}{P_{Tot}} \tag{5}$$

where D_i is the average dose for states, P_i is the population of states, and P_{Tot} is the total population of the six southwest states.

3.5. Estimation of excess lifetime cancer risk

The excess lifetime cancer risk (ELCR) is a probabilistic parameter that has been used to determine the proportion of a population which may likely develop cancer over a lifetime period. The ELCR is estimated using the following expression:

$$ELCR = D \times LE \times F \tag{6}$$

where D is the estimated radiation dose, LE is the life expectancy (70 y) and F is the risk factor (0.05 Sv^{-1}).

3.6. Estimation of water quality index

The water quality index (WQI) is a tool widely used to determine the degree of pollution of drinking water. Different indices have been used by

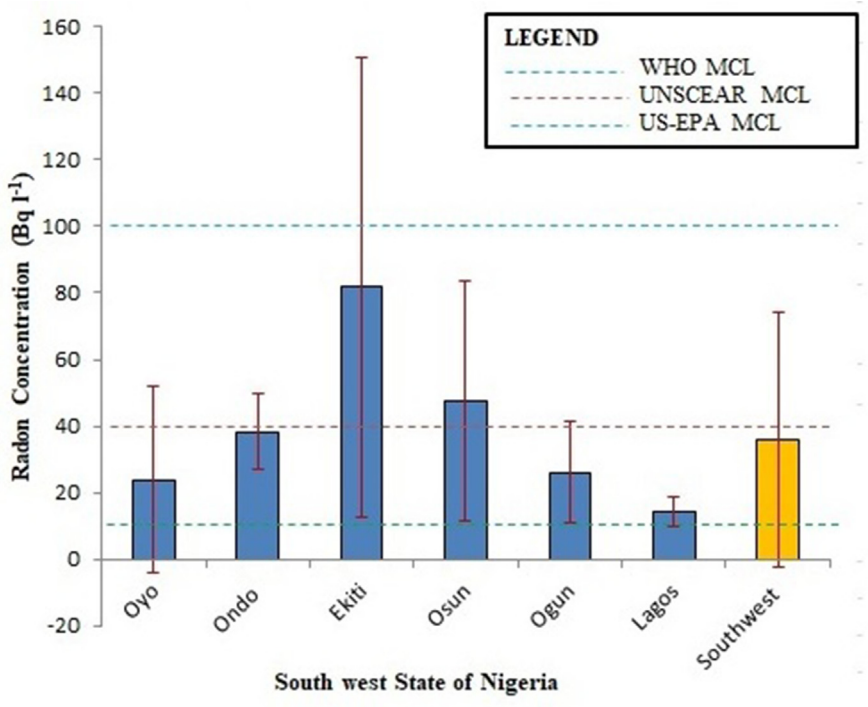

Figure 5. Distribution of radon concentration in groundwater of six southwest states.

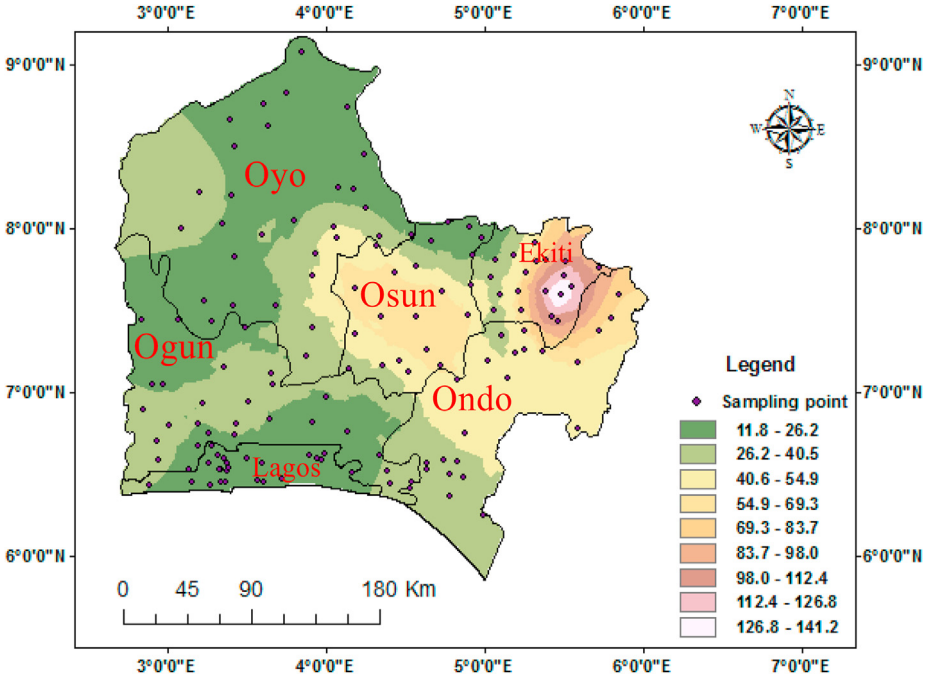

Figure 6. Radon concentration distribution map for southwest Nigeria.

researchers to characterise water quality with respect to heavy metals (Rezaei et al., 2019; Saleh et al., 2019). Such indices include heavy metal evaluation index, degree of contamination, heavy metal pollution index, and modified heavy metal pollution index. The WQI index is evaluated using the following equations:

$$WQI = \sum_{i=1}^{n} SI_i \tag{7}$$

where SI_i is a sub-index estimated using Eq. (8):

$$SI_i = W_i \times Q_i \tag{8}$$

where W_i is the relative weightage factor for each of the analysed parameter which can be computed with Eq. (9):

$$W_i = \frac{w_i}{\sum_{i=1}^{n} w_i} \tag{9}$$

w_i is the unit weightage factor and it has an inverse relationship with the maximum allowable concentration (MAC), and Q_i is a quality rating estimated using Eq. (10):

$$Q_i = \left(\frac{C_i}{S_i}\right) \times 100 \tag{10}$$

C_i is the measured concentration of heavy metal in the i^{th} parameters, and S_i is the standard value proposed in Nigerian Standard for Drinking Water Quality (NSDWQ, 2007) guidelines.

3.7. Heavy metal pollution index

Heavy metal pollution index (HPI) is a technique for evaluating the composite influence of individual heavy metals on overall water quality through the assignment of weight (w_i) to each selected parameter (Rezaei et al., 2019). The HPI was calculated using the following equations:

$$W_i = \frac{w_i}{\sum_{i=1}^{n} w_i} \tag{11}$$

where W_i is the relative weight of the i^{th} parameter and w_i is the unit weight of the i^{th} parameter.

$$Q_i = \left(\frac{|C_i - I_i|}{S_i - I_i}\right) \times 100 \tag{12}$$

where Q_i is the sub-index of the i^{th} parameter, C_i is the measured concentration of the parameter, S_i is the standard value of the parameter, and I_i is the ideal value of the parameter.

$$HPI = \sum_{i=1}^{n} W_i Q_i \tag{13}$$

where n is the number of parameters considered.

3.8. Statistical analysis

Statistical analysis was carried out using Minitab software (version 17.0) to separate the means of the measured radon across southwest region of Nigeria. One-Way Analysis of Variance (ANOVA) was performed and the means were separated at a 5% level of significance using Tukey's test.

4. Results and discussion

4.1. Radon activity

^{222}Rn activity concentrations have been measured in 145 groundwater samples collected across six southwest states of Nigeria. The results of the activity concentrations are presented in Table 1. The studied groundwater samples show variations in the range from 1.6 Bq $l^{-1}$to 271 Bq l^{-1} with an overall mean value of 35.9 ± 38.4 Bq l^{-1}. The distribution of activity concentration of radon in groundwater across southwest Nigeria shows a high standard deviation which indicates a high spread of the concentration value from low to high. As shown in Figure 4, the distribution is highly skewed to the right with a skewness value of 2.91. The activity concentration distribution across the six southwest states shows that Lagos State has the lowest mean radon concentration of 14.3 ± 4.6 Bq l^{-1} whereas, Ekiti State has the highest value of 81.7 ± 61.3 Bq l^{-1}. The maximum contaminant levels (MCL) of radon in water set by WHO, UNSCEAR, and US-EPA are 100 Bq l^{-1}, 40 Bq l^{-1}, and 11.1 Bq l^{-1} respectively. 6% of the water samples have concentrations exceeding the WHO MCL. 30% of the water samples have concentrations above the UNSCEAR reference level and 82% have concentrations above the US-EPA reference level. As presented in Figure 5, the average ^{222}Rn activity concentrations in groundwater of all the states are above the US-EPA limit, whereas Ekiti and Osun States have average activity concentrations exceeding the UNSCEAR limit. However, the average ^{222}Rn activity concentrations in groundwater of all the six southwest states are below the WHO limit.

Table 2. Comparison of radon concentration in groundwater in the present study with other countries.

s/n	Region/Country	^{222}Rn concentration in groundwater (Bq l^{-1})		Reference
		Range	Mean	
1	Southwest, Nigeria	1.6–271.0	35.9	Present Study
2	Rajasthan, India	0.8–15.0	4.8	Mittal et al. (2016)
3	Punjab, India	1.4–5.3	3.5	Jakhu et al. (2020)
4	Kurdistan, Iraq	1.1–10.3	6.8	Ezzulddin and Mansour (2020)
5	Kericho County, Kenya	4.6–22.5	12.4	Rotich et al. (2020)
6	Southern Greater Poland	0.4–10.5	1.9	Bem et al. (2014)
7	Chihuahua, Mexico	1.8–39.8	16.1	Villalba et al. (2005)
8	Saudi Arabia	0.01–67.4	6.8	Alabdula'aly, 2014
9	South Korea	1.1–2393.5	86.6	Cho et al. (2019)
10	Canary Island, Spain	0.3–76.9	12.9	Alonso et al. (2015)
11	Rajasthan, India	12.5–862.0	113.0	Duggal et al. (2020)
12	Covilhã's county, Portugal	2.0–1690	352.8	Inácio et al. (2017)
13	Jerba Island, Tunisia	0–2860	867.0	Telahigue et al. (2018)
14	Nablus City, Palestine	2.9–23.4	9.5	Al Zabadi et al. (2012)
15	Zarand City, Iran	5.2–14.4	9.7	Fard et al., 2020

4.2. Radon concentration distribution map

The radon concentration distribution map across southwest Nigeria is as shown in Figure 6. The map delineates the basement complex-sedimentary boundary. Groundwater radon concentrations within the basement complex are generally higher than that of the sedimentary terrain. The average concentration of 41.6 ± 42.5 Bq l^{-1} was obtained in the basement complex. This value is more than double the average value of 19.8 ± 12.8 Bq l^{-1} obtained for sedimentary terrain. The highest groundwater radon concentrations were obtained within Ekiti State (around the Northeast region of the study area) with concentrations ranging between 26 Bq l^{-1} and 141 Bq l^{-1}. These areas lie within the basement complex formation of southwest Nigeria. The map clearly delineates three regions of groundwater radon concentration: high ($C_{Rn} > 83\ Bq\ l^{-1}$), medium ($26\ Bq\ l^{-1} < C_{Rn} < 83\ Bq\ l^{-1}$) and low ($< 26\ Bq\ l^{-1}$). The sedimentary regions comprising of Lagos State and southern parts of Ogun and Ondo States lie within low groundwater radon concentration areas, whereas, radon concentration ranges from low to high in the basement complex regions. The eastern portion of the basement complex generally exhibits medium to high concentration while the western portion exhibits low to medium concentration.

Comparison of the groundwater radon concentration in this study with similar studies in different parts of the world revealed that the average radon concentration in groundwater in Southwest Nigeria is higher than the average concentration values reported for India (Jakhu et al., 2020; Mittal et al., 2016), Iraq (Ezzulddin and Mansour, 2020), Kenya (Rotich et al., 2020), Poland (Bem et al., 2014), Mexico (Villalba et al., 2005), Saudi Arabia (Alabdula'aly, 2014), Spain (Alonso et al., 2015), Palestine (Al Zabadi et al., 2012), and Iran (Fard et al., 2020) as presented in Table 2. However, the average groundwater radon concentration in Southwest Nigeria is lower in comparison to those reported in South Korea (Cho et al., 2019), Khetri Copper Belt of Rajasthan, India (Duggal et al., 2020), Portugal (Inácio et al., 2017), and Tunisia (Telahigue et al., 2018).

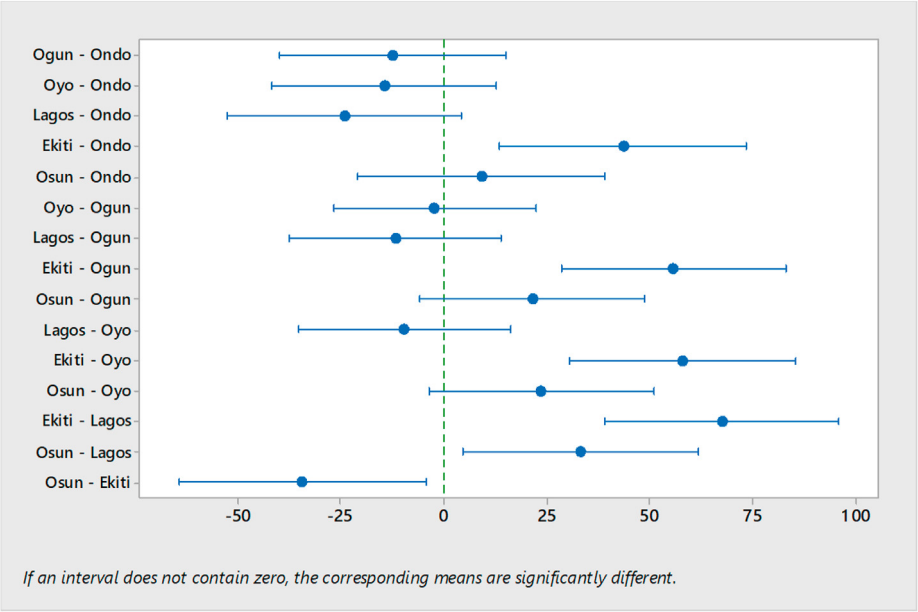

Figure 7. Graphical illustration of means separation for radon concentration in southwest Nigeria. (Note: if an interval does not contain zero, the corresponding means are significantly different).

4.3. Variation in the means of measured radon concentration

The variation in the means of the measured radon concentration across the south-western region of Nigeria is as illustrated in Figure 7. The result of the analysis showed that there was a significant difference in their means at a 5% level of significance ($P < 0.05$). This difference might be attributed to different anthropogenic activities being carried out at locations where the water samples were collected. For instance, OECD, 2009 reported that residues from anthropogenic activities such as mining of uranium ores and extraction industries could cause the release of radon isotopes into the atmosphere, which could contaminate the groundwater. As a result of possible differences in anthropogenic activities in the areas where the water samples were taken, coupled with differences in proximity to where these anthropogenic activities are taken place, we observed significant differences in our analysis (Figure 6; $p < 0.05$). This suggests the need for the prediction of radon in water bodies with respect to distance from mining sites or chemical extraction industries, where radon is being released for further studies. Similarly, another possible explanation for differences in the means of the radon measurements ($P < 0.05$) could be attributed to varied geological formations in the locations. This is in agreement with the report of some researchers (Singh et al., 2006; Choubey et al., 2007) who reported that soil geological formation and climate have an effect on the concentration of radon concentration in groundwater.

4.4. Annual effective doses due to ingestion and inhalation of radon and excess lifetime cancer risk

The annual effective doses due to ingestion of groundwater for infants, children, and adults range from 0.001 to 0.218, 0.002 to 0.313, and 0.004 to 0.692 mSv y^{-1} respectively. The mean values are 0.029 ± 0.031, 0.041 ± 0.044, and 0.092 ± 0.098 mSv y^{-1} respectively. The estimated dose due to inhalation of radon released from water varies from 0.004 to 0.683 mSv y^{-1} with average value of 0.090 ± 0.097 mSv y^{-1}. These values are less than the recommended limit of 0.1 mSv y^{-1} set by the World Health Organization (WHO, 2008), although dose estimation from radon concentration in water shows that 15% of the water samples have ingestion dose above the recommended limit. As presented in Table 3, the ingestion dose for adults and the inhalation dose for Ekiti and Osun State are above the recommended dose limit. The population-weighted average dose D_{Pop} for ingestion is 0.074 mSv y^{-1}. Also, the D_{Pop} value for inhalation is 0.073 mSv y^{-1}.

Using a life expectancy of 70 y and a risk factor or 0.05 Sv^{-1} (UNSCEAR, 2000), the excess lifetime cancer risks due to ingestion of groundwater and inhalation of radon gas released from water are 0.503×10^{-3} and 0.497×10^{-3} respectively. These values indicate the probability of 503 persons and 497 persons developing stomach cancer and lung cancer respectively from a population of one million.

Table 3. Groundwater radon concentration and estimated annual effective dose in six States in Southwest Nigeria.

State	Population ($\times 10^6$)	Ingestion Dose ($mSvy^{-1}$)			Inhalation Dose (μSvy^{-1})
		Infants	Children	Adults	
Oyo	7.8	0.019	0.027	0.061	0.060
Ondo	4.7	0.031	0.044	0.098	0.097
Ekiti	3.3	0.066	0.094	0.209	0.206
Osun	4.7	0.038	0.055	0.122	0.120
Ogun	5.2	0.021	0.030	0.066	0.066
Lagos	17.5	0.012	0.017	0.037	0.036
SW (Total)	43.2	0.029 ± 0.031 (0.001–0.218)	0.041 ± 0.044 (0.002–0.313)	0.092 ± 0.098 (0.004–0.692)	0.090 ± 0.097 (0.004–0.683)

Table 4. Concentrations of heavy metals (Pb, Cd, Ni, Cr Zn, Cu, and Mn) in selected regions of Southwest Nigeria.

S/N	State	Pb (ppb)	Cd (ppb)	Ni (ppb)	Cr (ppb)	Zn (ppb)	Cu (ppb)	Mn (ppb)
1	Lagos	10.0	0.0	0.0	6.0	70.0	9.0	43.0
2	Lagos	378.0	0.0	0.0	0.0	1.0	25.0	5.0
3	Lagos	80.0	1.0	0.0	0.0	7.0	8.0	0.0
4	Lagos	9.0	4.0	0.0	0.0	3.0	0.0	13.0
5	Lagos	9.0	4.0	0.0	0.0	12.0	10.0	94.0
6	Lagos	0.0	0.0	0.0	167.0	5.0	0.0	103.0
7	Lagos	0.0	0.0	0.0	64.0	0.0	49.0	79.0
8	Lagos	0.0	0.0	0.0	0.0	14.0	0.0	112.0
9	Lagos	0.0	0.0	0.0	79.0	0.0	17.0	90.0
10	Lagos	0.0	0.0	0.0	3.0	0.0	7.0	1.0
11	Lagos	0.0	0.0	0.0	0.0	0.0	37.0	20.0
12	Lagos	0.0	0.0	0.0	195.0	0.0	11.0	35.0
13	Lagos	0.0	10.0	0.0	0.0	0.0	0.0	0.0
14	Lagos	91.0	0.0	4.0	0.0	0.0	0.0	35.0
15	Lagos	0.0	0.0	0.0	98.0	0.0	17.0	21.0
16	Lagos	101.0	0.0	0.0	0.0	2.0	9.0	0.0
17	Lagos	213.0	2.0	0.0	0.0	2.0	43.0	29.0
18	Ogun	0.0	0.0	0.0	0.0	0.0	0.0	22.0
19	Ogun	55.0	1.0	0.0	0.0	0.0	0.0	33.0
20	Ogun	29.0	0.0	0.0	0.0	3.0	0.0	2.0
21	Ogun	0.0	0.0	0.0	68.0	29.0	43.0	0.0
22	Ogun	131.0	0.0	0.0	0.0	2.0	24.0	61.0
23	Ogun	0.0	0.0	0.0	0.0	7.0	0.0	17.0
24	Ogun	0.0	0.0	25.0	0.0	3.0	62.0	26.0
25	Ogun	25.0	2.0	0.0	0.0	2.0	5.0	0.0
26	Ogun	131.0	0.0	0.0	45.0	9.0	32.0	50.0
27	Ekiti	30.0	0.0	11.0	40.0	88.0	7.0	20.0
28	Ekiti	47.0	2.0	5.0	21.0	121.0	13.0	76.0
29	Ekiti	10.0	0.0	0.0	16.0	87.0	50.0	10.0
30	Ekiti	60.0	0.0	0.0	2.0	14.0	182.0	26.0
31	Ekiti	14.0	0.0	0.0	3.0	74.0	27.0	78.0
32	Ekiti	26.0	1.0	0.0	2.0	131.0	32.0	122.0
33	Ekiti	18.0	1.0	0.0	1.0	26.0	92.0	137.0
34	Ekiti	10.0	2.0	1.0	15.0	63.0	34.0	94.0
35	Ekiti	5.0	1.0	0.0	2.0	44.0	125.0	218.0
36	Ekiti	13.0	0.0	0.0	97.0	152.0	170.0	105.0
37	Osun	4.0	1.0	25.0	22.0	200.0	6.0	110.0
38	Osun	7.0	1.0	18.0	28.0	95.0	2.0	84.0
39	Osun	15.0	1.0	10.0	17.0	12.0	5.0	42.0
40	Osun	0.0	0.0	0.0	26.0	10.0	3.0	17.0
41	Osun	3.0	0.0	0.0	10.0	8.0	7.0	20.0
42	Osun	6.0	0.0	2.0	5.0	2.0	14.0	15.0
43	Oyo	2.0	0.0	1.0	7.0	0.0	12.0	8.0
44	Oyo	18.0	2.0	12.0	8.0	0.0	10.0	70.0
45	Oyo	4.0	1.0	1.0	14.0	80.0	5.0	85.0
46	Oyo	3.0	0.0	0.0	9.0	126.0	3.0	22.0
47	Oyo	15.0	2.0	0.0	4.0	85.0	7.0	56.0
48	Oyo	5.0	1.0	0.0	3.0	33.0	3.0	15.0
49	Oyo	2.0	1.0	6.0	14.0	92.0	2.0	18.0
50	Oyo	1.0	0.0	3.0	6.0	6.0	7.0	2.0
51	Oyo	8.0	0.0	0.0	10.0	34.0	2.0	15.0
52	Oyo	2.0	0.0	0.0	5.0	16.0	9.0	12.0
Average		30.6	0.8	2.4	21.4	34.0	23.8	45.5
WHO Standard		10.0	3.0	70.0	70.0	N/A	2000.0	400.0

Table 5. Water Quality Index (WQI) and Heavy Metal Pollution Index (HPI) of groundwater in the study area were below the critical limit of 100 proposed by Mohan et al. (1996) indicating that the water from this region is suitable for consumption while the HPI values for samples taken from some part of Lagos (S-2, S-6, S-12, S14, S-16, S-17) and Ogun (S-22 and S-26) indicate signs of impairment as they were found to be above the proposed permissible index value.

Sample no.	State	WQI	Water type	HPI	Degree of Pollution
S-1	Lagos	15.91	Excellent	40.20	Low
S-2	Lagos	461.87	V. Poor	611.38	V. High
S-3	Lagos	97.94	Good	156.03	Medium
S-4	Lagos	11.73	Excellent	50.40	Low
S-5	Lagos	11.77	Excellent	50.34	Low
S-6	Lagos	102.03	Poor	228.02	High
S-7	Lagos	39.17	Excellent	70.66	Low
S-8	Lagos	0.03	Excellent	64.63	Low
S-9	Lagos	48.30	Excellent	93.61	Medium
S-10	Lagos	1.84	Excellent	60.08	Low
S-11	Lagos	0.05	Excellent	64.60	Low
S-12	Lagos	119.13	Poor	270.78	High
S-13	Lagos	1.83	Excellent	65.72	Low
S-14	Lagos	111.46	Poor	172.72	High
S-15	Lagos	59.89	Good	122.66	Medium
S-16	Lagos	123.41	Poor	188.36	High
S-17	Lagos	260.67	V. Poor	358.82	V. High
S-18	Ogun	0.01	Excellent	64.66	Low
S-19	Ogun	67.39	Good	117.85	Medium
S-20	Ogun	35.43	Excellent	78.42	Low
S-21	Ogun	41.59	Excellent	76.81	Low
S-22	Ogun	160.10	Poor	234.13	High
S-23	Ogun	0.01	Excellent	64.67	Low
S-24	Ogun	1.72	Excellent	63.17	Low
S-25	Ogun	30.92	Excellent	71.78	Low
S-26	Ogun	187.60	Poor	211.21	High
S-27	Ekiti	61.82	Good	48.37	Low
S-28	Ekiti	70.98	Good	72.80	Low
S-29	Ekiti	22.05	Excellent	24.87	V. Low
S-30	Ekiti	74.76	Good	122.36	Medium
S-31	Ekiti	18.99	Excellent	50.84	Low
S-32	Ekiti	33.25	Excellent	70.43	Low
S-33	Ekiti	22.94	Excellent	59.63	Low
S-34	Ekiti	21.88	Excellent	25.78	V. Low
S-35	Ekiti	7.73	Excellent	53.50	Low
S-36	Ekiti	75.37	Good	110.13	Medium
S-37	Osun	20.19	Excellent	23.28	V. Low
S-38	Osun	27.05	Excellent	9.27	V. Low
S-39	Osun	29.57	Excellent	29.87	V. Low
S-40	Osun	15.89	Excellent	24.96	V. Low
S-41	Osun	9.79	Excellent	44.80	Low
S-42	Osun	10.54	Excellent	47.66	Low
S-43	Oyo	6.80	Excellent	50.81	Low
S-44	Oyo	28.06	Excellent	47.73	Low
S-45	Oyo	13.72	Excellent	36.78	V. Low
S-46	Oyo	9.17	Excellent	46.33	Low
S-47	Oyo	21.16	Excellent	50.37	Low
S-48	Oyo	8.13	Excellent	52.18	Low
S-49	Oyo	11.58	Excellent	39.42	V. Low
S-50	Oyo	5.09	Excellent	53.69	Low
S-51	Oyo	15.89	Excellent	37.17	V. Low
S-52	Oyo	5.51	Excellent	53.96	Low

Table 6. Water quality index range/water type (Badmus et al., 2021).

Water quality index range	Water type	% of sample
<50	Excellent	71%
50–100	Good	13%
100–200	Poor	12%
>200	V. poor	4%

4.5. Heavy metal concentration in groundwater

The concentrations of heavy metals in this study are presented in Table 4 with their average values and WHO standard values presented in the last two rows of the table. The average concentration values in part per billion (ppb) of the heavy metals in the assayed groundwater are such that Mn > Zn > Pb > Cu > Cr > Ni > Cd. The average concentration of Zn is higher than the maximum permissible level (MPL) set by the World Health Organization (WHO, 2008) for drinking water. Although zinc is essential to human health as it is essential for growth, hormonal production, and boosting of the immune system, bioaccumulation of zinc can have detrimental effects on human health. Such health effects include nausea, anaemia, skin inflammation, and pancreatic disorders (Jamshaid et al., 2018). 38.5% of water samples have Pb concentrations above the MPL of 10 ppb and 3% of samples have Cd concentrations above the MPL of 3 ppb. Also, 7% of samples have Cr concentrations above the MPL value of 50 ppb. All water samples have Ni, Zn, Cu, and Mn concentrations below their respective MPL.

4.6. Water quality index (WQI) and heavy metal pollution index (HPI) of groundwater

The WQI results for groundwater in the study area are presented in Table 5. The WQI values in all the water samples vary from 0.01 to 461.87. WQI values were classified as excellent, good, poor, and very poor as presented in Table 6. 71% of the groundwater samples were identified to be excellent, 13% in the good category, 12% in the poor category, and 4% fell in the very poor category. Drinking water in the very poor category is considered not suitable for drinking. The WQI table revealed that all samples taken from Ekiti, Osun, and Oyo were suitable for drinking as they fall in the good to excellent water type category while samples taken from some part of Lagos (S-2, S-6, S-12, S-14, S-16, S-17) and Ogun (S-22 and S-26) show sign of deterioration as it ranges from poor to very poor water category. This may be attributable to the fact that Lagos and Ogun State are in the sedimentary terrain of Southwest Nigeria. These areas are more susceptible to groundwater pollution infiltration (Ukah et al., 2019; Xiao et al., 2022). Also, these areas have a greater density of industrialization in Southwest Nigeria.

The HPI results for groundwater in the study area are presented in Table 5. The values for the HPI vary from 9.27 to 611.38. The HPI values were classified as very low, low, medium, high, and very high as presented in Table 7. 17% of the samples were identified to fall within the very low pollution risk level, 55% were within the low pollution risk level, 12% were within medium risk level, 12% within high risk level and 4% were identified within very high risk level. The HPI values for all samples taken in Ekiti, Osun, and Oyo.

Table 7. Heavy metal pollution index range/degree of pollution (modified after Rezaei et al., 2019).

Heavy metal pollution index range	Degree of pollution	% of sample
<40	V. low	17%
40–80	Low	55%
80–160	Medium	12%
160–300	High	12%
>300	V. high	4%

5. Conclusion

Assessment of radon concentration in groundwater used for drinking purposes in southwest Nigeria revealed an average value above the recommended limit of 11.1 Bq l^{-1}. Groundwater radon concentrations in areas underlain by basement complex are generally higher than in sedimentary terrain. The concentration distributions have been well delineated on a map using Arc GIS software. The map will serve as a useful tool for planning and regulation purposes. Water quality index assessment of groundwater in terms of heavy metal pollution in the study area showed that 71% of water samples exhibit excellent quality. Although 38.5% of water samples have Pb concentrations above the maximum permissible level of 10 ppb, 3% of samples have Cd concentrations above the maximum permissible level of 3 ppb, and 7% of samples have Cr concentrations above the maximum permissible level of 50 ppb. However, deterioration of water quality due to heavy metal pollution was observed in some water samples of more industrialised areas of Ogun and Lagos States. A stricter regulation should be put in place on disposal mechanisms of industrial waste in the regions.

Declarations

Author contribution statement

Yinka Ajiboye & Ganiyu Olabode Badmus: Conceived and designed the experiments; Performed the experiments; Analyzed and interpreted the data; Wrote the paper.

Matthew Omoniyi Isinkaye: Conceived and designed the experiments; Analyzed and interpreted the data; Wrote the paper.

Oluwaseun Temitope Faloye: Analyzed and interpreted the data; Wrote the paper.

Vincent Atoiki: Performed the experiments; Wrote the paper.

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Data availability statement

The authors do not have permission to share data.

Declaration of interests statement

The authors declare no conflict of interest.

Additional information

No additional information is available for this paper.

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