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Radon-222 in Drinking Water: Concentration and Radiological Risk in Nigerian Boreholes

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Abstract. *The consumption of contaminated water, especially those polluted by radioactive elements, heavy metals, effluents, etc., has posed a significant health risk in most developing economies. Therefore, researchers nowadays strive to ameliorate this menace. Hence, this study analyzed the concentration of Radon-222 in drinking water samples from some boreholes in Lagos and Ogun states, Nigeria to provide insight into the distribution of Radon-222 in the southwestern states of Nigeria. The Radon-222 concentrations in borehole water samples collected from Lagos and Ogun states were measured using a liquid scintillation counter at the Center for Energy Research and Training (CERT) in Ahmadu Bello University, Zaria. Radon concentrations for borehole water collected from 10 locations in Lagos State range between 51.78 Bq/L and 118.88 Bq/L, while those obtained from 10 selected locations in Ogun State range between 12.92 Bq/L and 110.26 Bq/L. The recorded mean values of radon concentrations obtained in this study for the borehole water in the two States were found to be above the world average Maximum Contamination Level (MCL) of 10 Bq/L set by the World Health Organization and the MCL of 11.1 Bq/L set by the United States Environmental Protection Agency (USEPA).*

Keywords: *Measurement, radon, concentration, drinking water*

1. Introduction

Water is used for various reasons such as transportation, power generation, agriculture and other domestic activities hence its availability and quality as regards radiological, microbiological, chemical and any other form of contamination are delicate and vital issues (Garba, 2018). Water is said to be life because no human can survive without the use of water. In the production and preparation of human food, water plays a very vital role. Drinking water is also essential as the human body is about 60% water. Many other household chores are done using water. So, a good potable and drinkable water source is required to be used in every home. Usable water must be odourless, tasteless and colourless (Garba, 2018). The importance of safe and drinkable water for mankind in Nigeria cannot be overemphasized. Nigeria has an estimated 267 billion cubic metres of surface water and 92 billion cubic metres of ground water per annum, with over 200 dams with a combined storage capacity of 34 billion cubic metres (UNDP, 2022). However, despite the huge water potential, the country is still classified as 'water short' because of its inability to meet the challenges of supplying potable water to meet the domestic needs of the populace.

The consequences of unavailability of potable water supply in rural and urban communities in Nigeria have been quite grievous. The World Health Organization has estimated that about 361,900 people die yearly due to poor water and sanitation conditions in

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Nigeria. According to Water Aid (2017), 60,000 children under the age of five die from diarrhoea, a disease caused by poor water. This crisis severely affects people's health, well-being and economic productivity. The lack of safe water leads to waterborne diseases, such as cholera, diarrhoea and typhoid fever which cause millions of deaths yearly especially among children (World Health Organisation, 2022).

One of the major problems to safe drinking water is water source. There are two major sources of water used domestically or industrially. These two sources are surface water such as streams, rivers, ponds, and lakes, and ground waters such as wells and, boreholes. Both types are used domestically and industrially, but it was reported that groundwater is safer for domestic use and less susceptible to contamination (Okeola, Kolawole, & Ameen, 2023). As a long-term solution to water inaccessibility in Nigeria, the public, private and individuals develop the use of boreholes.

The Radon concentration in water at any particular place can be affected by the human activities in the area. A more dense and industrial area water will most likely be found well-fortified with radioactive sample of Radon-222 (^{222}Rn) than scarcely populated and none industrialized area of which Lagos and Ogun States are case study. In other words, human activities in Lagos and Ogun States may also contribute to the contamination of the water sources found in these areas and both radioactive and non-radioactive impurities may have been added by man. It was also reported in the study of Ball et al. (2019) that radon content in surface water may be affected by several factors, viz. the geology of the area, bottom sediments, inputs from streams, degree of water turbulence and temperature. Several surveys have been made of radon concentrations in ground waters (and drinking water derived from these sources) where it was suspected that levels of activity were high enough to constitute a potential health hazard (Burgess et al., 2019) but there are fewer data on "normal" waters. However, that does not appear to have been a significant study of the natural radioactivity of water in these areas, some of which are used to supply both the local population and some major conurbations. It is therefore important to investigate the presence of Radon-222 concentration in drinking water samples from some boreholes in Lagos and Ogun States so as to determine its' quality and goodness for human consumption.

The exposure of human beings to Radon is dual. These are through inhalation of radon released into the ambient air and through ingestion when water is used for drinking. Randon-222 activity concentration in water is due to the decay of Radium-226 associated with the rock and soil. Apparently, the radon gas percolates through the soil and rock, and dissolves in the water. Therefore, the concentration of Radon in water is higher than one would expect (Xinwei, 2006).

Normally, the activity concentration of radon in groundwater is higher than that in surface water. This is because radon is a gas and when water that contains radon is exposed to the air the radon tends to be released into the air. However, surface water polluted with natural occurring radioactive materials (NORM) from human activities such as mining and fertilizer production are prone to high activity concentration of radon. People who are exposed to radon in drinking water may have increased risk of getting Cancer over the course of their lifetime, especially Stomach and Lung cancer (UNSCEAR, 2019). Aransiola *et al* (2016) in their work defined radiation as the outflow of particles, rays or waves produced from breaking down of radioactive isotopes. As water flow beneath ground surface, it comes into contact with natural occurring radioactive materials as it moves on its part, get contaminated by the radioactive elements which may eventually decay to the well-known Radon, which is commonly associated with water. The radioactive substance might not be noticed, since it does not change the physical look of water and none of its characteristics is being affected by the presence of radiation. Aruwa *et al.* (2017), stated that Radon is the number one cause of lung cancer among non-smokers and in overall, radon is the second leading cause of lung cancer and is responsible for about 21,000 lung cancer deaths every year. This is a very high number of casualty, which can actually be avoided if the water sample is tested and treated before its consumption.

Groundwater from wells and boreholes usually contain higher radon concentrations than surface water, because surface water is exposed to the air and since radon is a gas it tends to escape into the air. Also, the concentration of Radium-226 in the aquifer matrix is the basis for the high activity concentration of radon in groundwater (Vinson et al., 2009). Therefore, in this research, we intend to analyze the concentration of Radon-222 in borehole water used by households in Lagos and Ogun states, with the possibility of providing recommendations that could help corp this menace, especially in developing countries with similar environmental decay.

2. Methods

2.1. Sample Collection

A total of twenty (20) borehole water samples, of one (1) litre each, (which included ten (10) borehole water samples from Lagos State and ten (10) borehole water samples from Ogun State) were collected in glass bottles for analysis. The samples were taken to the Centre for Energy Research and Training, Ahmadu Bello University, Zaria, Kaduna State Nigeria, where Radiometric analysis were conducted using Liquid Scintillation Counter. The bottles were tightly sealed so as to avoid escape of radon-222 to the surroundings. Each sample was tagged for ease of identification before transmission to the laboratory for analysis.

2.2. Sample preparation and experimentation

The procedure for the determination of radon-222 by liquid scintillation counting is quite specific for this radionuclide. In view of this, we shall first discuss the general method required for the measurement of ^{222}Rn. Radon-222 is extracted readily from the water sample by an organic scintillant. The decay products of radon-222 remains in the water phase whilst radon-222 is extracted into the organic phase. Before measurement, the sample was stored for three hours until equilibrium is reached between radon-222 and its alpha emitting decay products. The alpha activity from radon-222 and its decay products were measured in a liquid scintillation counter. The basic apparatus used in this measurement are:

2.3. Liquid scintillation counter

We used low background liquid scintillation vials with a capacity of at least 20 ml. The use of polyethylene scintillation vials is not recommended because of potential losses of radon through the walls of the vial. The use of teflon or viton seals in the caps of the vials is recommended to ensure that there is no escape of radon from the vial. A disposable hypodermic syringe, with capacity 10-15 ml was also used.

2.4. Reagents

We used distilled or deionized water, stored in a glass bottle for at least a month, and a Scintillation solution. Commercially available toluene-based scintillation cocktails could be used, however, it should be tested for suitability prior to routine use.

2.5. Sampling

Prior to sampling, liquid scintillation vials were prepared containing 10 ml of liquid scintillation solution. Where the water was supplied by a tap, the water was allowed to flow freely for several minutes, without aeration, prior to sampling. Then 10 ml of the sample was carefully drawn from the source of the water into a disposable syringe ensuring that there was no aeration of the sample. Special care was taken when collecting samples to prevent significant loss of radon-222 by out-gassing.

The sample in the hypodermic syringe was immediately, in situ, transferred to the liquid scintillation vial containing the scintillation solution. The sample was now injected at the bottom of the vial, below the immiscible scintillation solution. After the injection, the syringe with its needle was carefully withdrawn from the vial and immediately the cap of the vial was tightly secured. The vial was then vigorously shaken so as to extract Radon-222 from the water phase to the organic scintillant solution due to its greater solubility in organic liquids, such as toluene. After the collection of the sample, the sample vials were transferred to the laboratory at the Centre for Energy Research and Training (CERT), Ahmadu Bello University, Zaria, Nigeria, for measurement. In this work, we used the materials and procedure as recommended by the American Society for Testing and Materials (ASTM, 1999) given in a publication titled "Standard Test Method for radon in Water".

2.6. Counting procedure

When the sample vials were returned to the laboratory for measurement, the outside of each vial was carefully wiped with a cloth dampened with ethanol or similar solvent and then left for a minimum of 3 hours to allow for ingrowth of the short-lived decay products of radon-222. Radon-222 and its short-lived daughters will be in equilibrium after 3 hours. The calibration of the counting system as described will take into account any degree of disequilibria between radon-222 and its daughters in the organic phase and will be consistent with measurements on samples. The counting vials were then placed in the liquid scintillation counter and each one counted for a pre-set time of 60 minutes using a calibrated window for alpha counting. While doing this, effort was made to ensure that when transferring vials from storage to the counter after the 3-hour ingrowth, there was no shaking as this will greatly disturb the state of equilibrium between radon-222 and its short-lived daughters in the organic scintillant. The time and date for the counting were noted. The background was counted using a vial with 10 ml of the organic scintillant solution and 10 ml of deionized or distilled water only. The background was measured over the same spectral range as the samples and for the same counting period.

2.7. Determination of Efficiency

To determine the efficiency of counting, in situation where the sample vial contains equal volumes of water sample and the organic scintillant, the Ra-226 standard solution was diluted with distilled water and a set of calibration solutions in liquid scintillation vials to cover the anticipated activity range of the samples to be analysed was prepared. The total volume of aqueous solution in each vial should be 10 ml. To this, 10 ml of the organic scintillant solution was added and the cap of the vial tightly sealed. The vials were then left aside in a dark place for at least 25 days to allow for secular equilibrium to be reached between radium-226 and radon-222. At the end of the build-up period, the calibration vials were shaken and left for a further period of 3 hours before counting. The radium-226 remains in the aqueous phase and does not contribute significantly to the count rate from the organic scintillant. Each vial was then counted and the liquid scintillation counting systems was calibrated as follows:

For a single calibration solution, the efficiency can be calculated using the expression (ASTM 1999)

$$E = \frac{R - R_o}{A_s} \dots \dots \dots 1$$

where: E is the counting efficiency for the system, in number of pulses per second counted per becquerel of radium-226 in the calibration solution, R is the counting rate of the calibration solution (i.e. radon-222 and its decay products), in pulses per second, R_o is the counting rate of the blank counting vial, in pulses per second and A_s is the activity of radium-226 in the calibration solution, in becquerel.

2.8. Sample Concentration

The concentration of Radon-222 in a sample was determined using the formula (ASTM 1999)

$$Rn = \frac{100 \text{ x } (SC - BC)e^{\lambda t}}{60 \text{ x } 5 \text{ x } 0.964} \dots \dots \dots 2$$

where Rn is Radon level in Bq/l, SC is sample count rate (Count/min), BC is background count rate (Count/min), λ is decay constant, t is time elapsed between sampling to count, 100 is a conversion factor from per 10 ml to per liter, 60 is a conversion factor from min to sec, 5 is number of emission per disintegration of Radon-222 (3α and 2β) assuming 100% detection efficiency for each, 0.964 is the fraction of Radon-222 in cocktail in 22 ml total capacity vial.

The Annual effective dose in a sample was determined using the formula (ASTM 1999)

$$E_{Rn} = DF_{Rn} \times I_w \times R_n \dots \dots \dots 3$$

where E_{Rn} is effective dose, DF_{Rn} is the coefficient (for adult it is 10^{-8}Sv/Bq), I_w is annual water consumption (for adult it is assumed 2 liters per day from WHO), R_n is radon concentration.

3. Results and Discussion

A total of ten (10) samples were collected from ten (10) boreholes in Lagos metropolis and analysed using Liquid Scintillation Counter. The results of the analysis are shown in Table 1.

Table 1. Results of Radon Concentrations in (Bq/L) and their corresponding Annual Effective Doses (AED) in (mSv/y) for Borehole Samples from Lagos

S/N	Sample Location	Coordinates	R_n (*Bq/L*)	AED (*mSv/y*)
1.	LG1	Lat ($6^0\ 31^1\ 24^{11}$ N) Long ($3^0\ 19^1\ 24^{11}$ E)	90.26	6.68
2.	LG2	Lat ($6^0\ 27^1\ 7^{11}$ N) Long ($3^0\ 18^1\ 59^{11}$ E)	102.19	9.01
3.	LG3	Lat ($6^0\ 35^1\ 18^{11}$ N) Long ($3^0\ 20^1\ 41^{11}$ E)	118.88	14.24
4.	LG4	Lat ($6^0\ 34^1\ 43^{11}$N) Long ($3^0\ 30^1\ 14^{11}$E)	51.78	2.37
5.	LG5	Lat ($6^0\ 26^1\ 0^{11}$N) Long ($3^0\ 27^1\ 40^{11}$E)	66.88	3.56
6.	LG6	Lat ($6^0\ 32^1\ 0^{11}$N) Long ($3^0\ 21^1\ 15^{11}$E)	80.48	7.26
7.	LG7	Lat ($6^0\ 30^1\ 25^{11}$N) Long ($3^0\ 19^1\ 35^{11}$E)	63.27	3.33
8.	LG8	Lat ($6^0\ 31^1\ 0^{11}$N) Long ($3^0\ 23^1\ 45^{11}$E)	56.48	2.71
9.	LG9	Lat ($6^0\ 26^1\ 10^{11}$N) Long ($3^0\ 23^1\ 5^{11}$E)	62.32	3.08
10.	LG10	Lat ($6^0\ 30^1\ 30^{11}$N) Long ($3^0\ 28^1\ 30^{11}$E)	58.35	2.98
	Mean Value		75.09	5.52

The result of the analysis of radon concentrations for the ten (10) borehole water samples collected at different locations of Lagos metropolis (Table 4.1) revealed that the concentrations of Radon-222 varied from 51.78 Bq/L to 118.88 Bq/L. The maximum concentration was obtained from LG3 (118.88 *Bq/L)* while the minimum concentration was found at LG4 as shown in Figure 1. All the values obtained from these samples were found to be above the maximum contaminant levels of 11.1 Bq/L set by USEPA and the world average value of 10 Bq/L set by World Health Organization (WHO, 1993). However, 20% of the analysed borehole water samples which include samples from LG2 and LG3 locations were found to be above 100 $Bq/$L. These high concentrations of Radon-222 in LG2 and LG3 water samples, which can pose a threat on the health of the inhabitants of these areas, could be due to the geology, high level of industrialization of the area and human activities. LG2 is a location in Lagos State with the largest population of about 3,082,900 (Population, 2019) while LG3 is the most industrious location in Lagos State. The geology, advent of industrialization and human activities in these two areas might have led to pollution and a lowering of water quality. Leachate from domestic and industrial waste also pose a high risk linked to radon in water in these two areas.

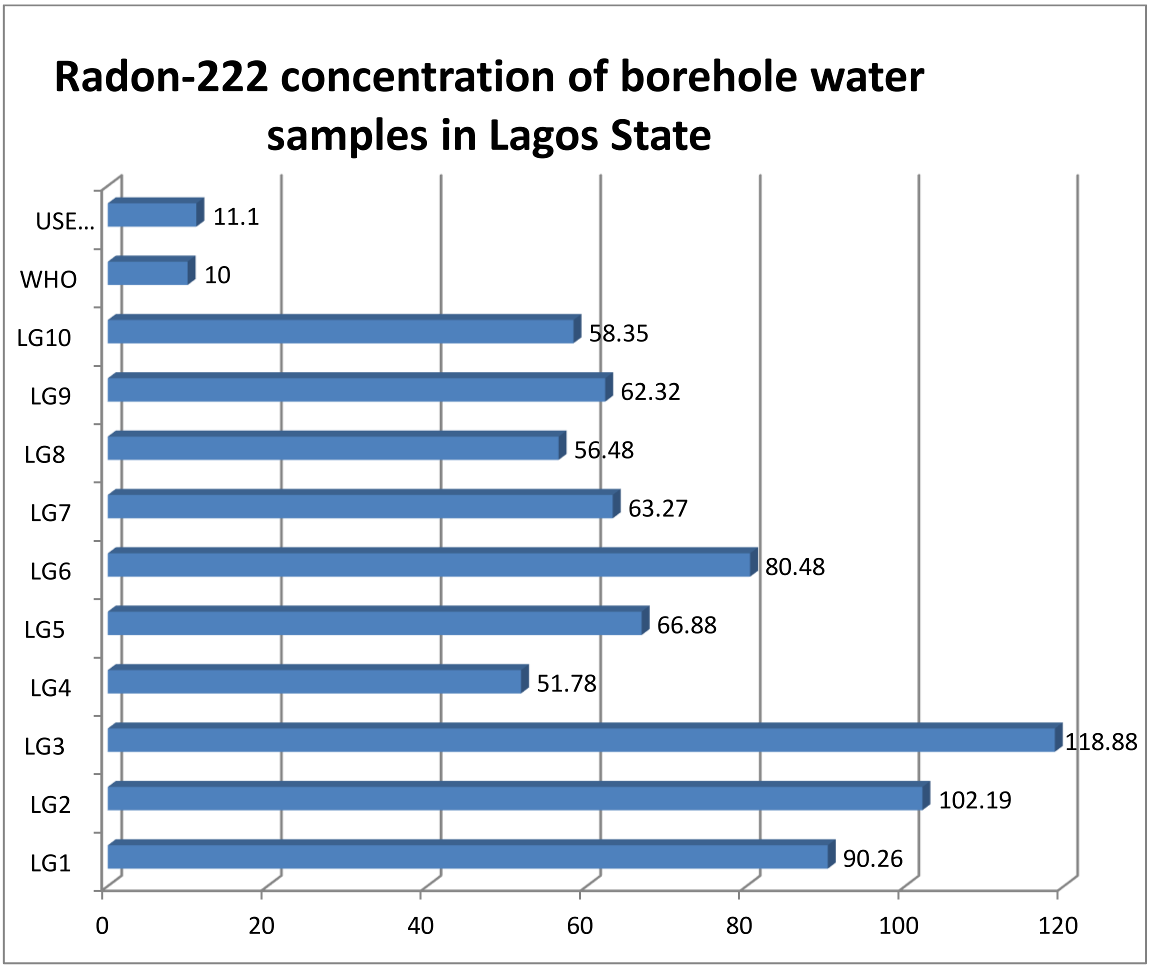

Figure 1. Bar Chart showing Radon-222 concentration (Bq/L) of borehole water samples in Lagos State

Table 2. Results of Radon Concentrations in (Bq/L) and their corresponding Annual Effective Doses (AED) in (mSv/y) for Water Samples from Ogun State

S/N	**Sample Location**	**Coordinate**	**R_n (*Bq/L*)**	**AED (*mSv/y*)**
1.	OG1	Lat (7^0 9^1 26^{11} N) Long (3^0 20^1 45^{11} E)	110.26	8.25
2.	OG2	Lat (7^0 8^1 0^{11} N) Long (3^0 20^1 0^{11} E)	101.88	6.01
3.	OG3	Lat (6^0 4^1 0^{11} N) Long (3^0 1^1 0^{11} E)	12.92	0.12
4.	OG4	Lat (6^0 47^1 0^{11} N) Long (4^0 1^1 0^{11} E)	23.81	0.34
5.	OG5	Lat (6^0 56^1 0^{11} N) Long (4^0 5^1 0^{11} E)	72.82	1.09
6.	OG6	Lat (6^0 49^1 28^{11} N) Long (3^0 55^1 24^{11} E)	40.03	0.53
7.	OG7	Lat (6^0 58^1 0^{11} N) Long (3^0 32^1 0^{11} E)	52.90	0.60

S/N	Sample Location	Coordinate	R_n *(Bq/L)*	AED *(mSv/y)*
8.	OG8	Lat (7^0 14^1 0^{11} N) Long (3^0 32^1 0^{11} E)	37.49	0.49
9.	OG9	Lat (6^0 44^1 0^{11} N) Long (3^0 20^1 45^{11} E)	50.14	0.58
10.	OG10	Lat (6^0 49^1 54^{11} N) Long (3^0 38^1 8^{11} E)	21.10	0.30
	Mean Value		52.34	1.83

The result of the analysis of radon concentrations for the ten (10) borehole water samples collected at different locations of Ogun State and which are presented in Table 4.2 revealed that the concentrations of Radon-222 varied from 12.92 Bq/L to 110.26 Bq/L. The maximum concentration was obtained from OG1 (110.26 *Bq/L)* while the minimum concentration was found at OG3 (12.92Bq/L) as shown in Figure 4.2. All the values obtained from these samples were found to be above maximum contaminant levels of 11.1 Bq/L set by USEPA and the world average value of 10 Bq/L set by World Health Organization (WHO, 1993). However, 20% of the analysed borehole water samples which include samples from OG1 and OG2 locations were found to be above 100 Bq/L. These high concentrations of Radon-222 in OG1 and OG2 water samples which can pose a threat on the health of the inhabitants of these areas could be due to the geology of the area which is basically made up of granites and the concentration of Radium-226 from the parent rock in the aquifer matrix.

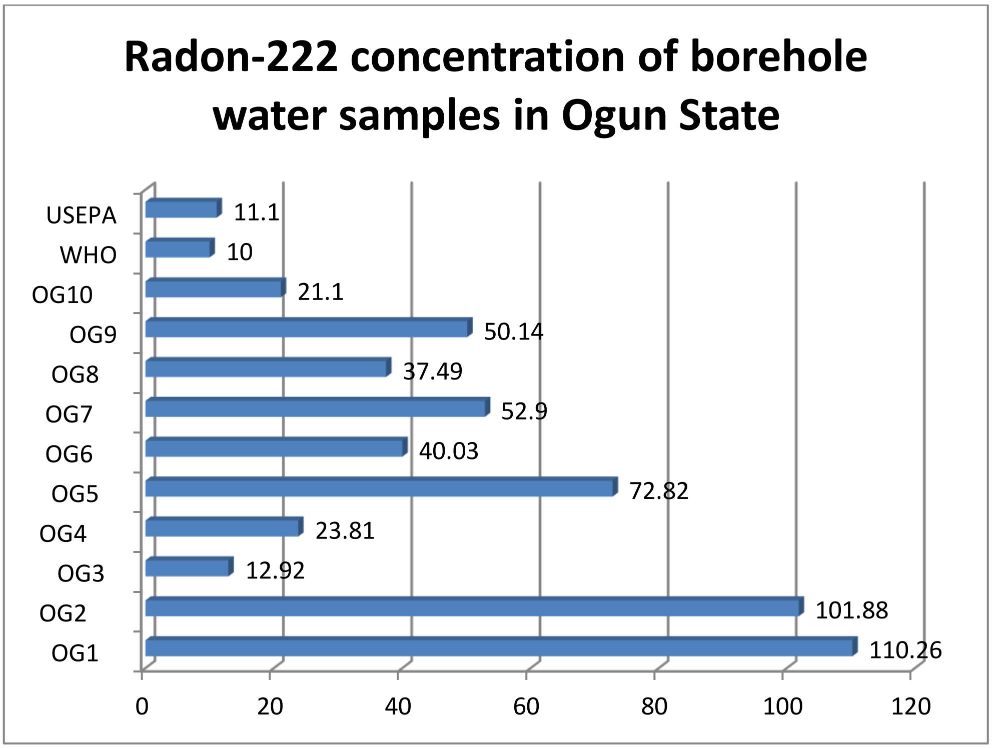

Figure 2. Bar Chart showing Radon-222 concentration (Bq/L) of borehole water samples in Ogun State

3.1. Comparison of Radon Concentration in the two Locations

In order to compare the levels of Radon-222 in the two States sampled for the study, the average values of Radon concentration in Lagos and Ogun States were calculated to be 75.09B/L and 52.34B/L respectively and plotted as shown Figure 4.3. It is important to mention here that since Lagos State and Ogun State, each has a larger area, the concept of average used here is simply to represent the divergence or otherwise of the values and for ease of comparison. Samples from boreholes in Lagos State have higher concentration of Radon-222 than samples from boreholes in Ogun State. All the values from the two states exceeded the maximum contaminant levels of 11.1Bq/L set by USEPA (1991) and the world average value of 10 Bq/L set by WHO (1993). The higher value of Radon-222 concentration in Lagos State as compared to radon concentrations of Boreholes in Ogun State could be linked to the geology of Lagos State and also to the fact that radon readily dissolves in water under pressure which led to the radon accumulation in groundwater as reported by Cho et al., (2004).

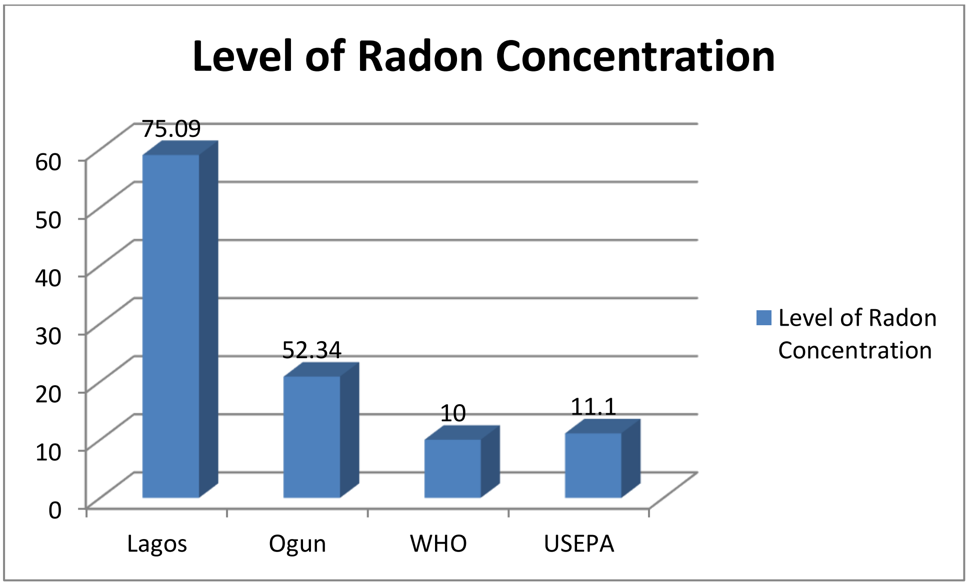

Figure 3. Bar Chart showing Radon-222 concentration of borehole water sample in Lagos and Ogun States

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Conclusion

Results obtained from the measurement of the activity concentrations of Radon-222 in water samples collected from boreholes in Lagos and Ogun States at different locations, revealed that 100% of the recorded values of radon concentrations in the present study were above the world average Maximum Contamination Level (MCL) of 10 Bq/L set by World Health Organization and the MCL of 11.1 Bq/L set by United States Environmental Protection Agency (USEPA, 1991). These significantly high values of radon concentration can be ascribed to the nature of the basement rock and soil type in Ogun State and high level of industrialization in Lagos State. Therefore, these water sources pose a threat to the health of the inhabitants of Lagos and Ogun State if continually ingested without proper treatment. The likelihood of these threats to health (which could be stomach or lung cancer) is more on dwellers of Lagos State than Ogun State as evident from the estimated Annual Effective doses of the corresponding radon concentrations in water in which most of the estimated annual effective doses were found to be above the reference level of 0.1 mSv/y set by World Health Organization (WHO, 2004) for intake of radionuclide in water.

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Conflicts of Interest

The authors declare no conflict of interest.

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