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Radon Measurement in Commercial Borehole Water from Some Selected Areas of Kaduna Metropolis Using Liquid Scintillation Counter

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Abstract: **Current level of Radon gas in groundwater is of great interest in the context of current level of cancer cases in Nigeria. The Rn-222 activity concentration in ground water from 12 boreholes and the corresponding purified drinking water in Kaduna North metropolis were investigated in the present study. The liquid scintillation counter used for sample analysis provides reliable measurement of Rn-222 activity concentrations which ranges over 3.34–17.70 kBq/m^3 resulting in large spatial differences in Rn-222 concentration. The results obtained show that all the boreholes monitored had radon water concentration within international standard limits such as the U. S EPA Alternative Maximum Contamination level (AMCL) which is 146 kBq/m^3 and the European Union reference level of 1000 kBq/m^3. However a small fraction of the boreholes monitored exceeded the recent proposed limit of U.S Environmental Protection Agency of 11kBq/m^3 for Maximum Contaminant Level (MCL). The estimated doses due to the consumption of drinking water containing radon vary significantly with the quantity of drinking water consumed and the degree to which the water has been processed prior to consumption. However dose estimates based on measurements made in this study demonstrate that Rn-222 in drinking water may not pose a significant health risk, in the short term, to consumers who depend on drinking water supplies derived from the boreholes monitored.**

Keywords: **Radon, groundwater, cancer, activity, concentration, boreholes.**

1. INTRODUCTION

Radon is an odorless, tasteless and invisible radioactive noble gas produced from radioactive decay of uranium and radium that are commonly present throughout the earth crust. Naturally occurring radon emanates constantly from rock and is dissolved in, and transported by, groundwater. Radon is also generated from the decay of radium in the radioactive waste. Some types of rocks such as granite, phosphates, shale and pitchblende are characterized with relatively high uranium concentration and so store natural deposits of radon. Consequently, there is elevated radon concentration in soils and weathered bedrock in areas where those rocks are located (A.I. Veeger, 1998). Areas with sediments surfaces or ground water from rock units with high concentration of uranium also have increased chances of elevated radon concentration.

The most abundant form of radon is Rn-222, with a half-life of 3.82 days. As such, this is the radon isotope that is considered in this analysis. Rn-222 is a decay product of radium (Ra-226), and a member of the U-238 decay chain (the most common isotope of uranium). It occurs at low levels in virtually all rock, soil, water, plants, and animals.

Radon is classified as Group 1 carcinogen by International Agency for Research on Cancer (IARC), a part of the World health Organization (WHO) as a result of direct evidence from human exposure studies that support the link between exposure to radon and induction of lung cancer. In line with this position, United Nation Scientific Committee on the Effect of Atomic Radiation (UNCEAR, 2000) reported a population weighted average radon concentration in homes of 39 Bq/m^3 to provide the basis for arriving at a reference level for radon exposure. It should be noted that large variation in indoor radon concentration are commonly found within Countries and these are not adequately described by a single average value. Indoor radon has been reported to be 2 to 3 orders of magnitude higher than the average value in many Countries. The observed high variability can be explained in terms of the fact that indoor radon usually follow log-normal distribution. This fact also provides additional input for arriving at reference level of radon exposure.

United States of America Environmental Protection Agency (EPA) and the World Health Organization (WHO) also declared radon as the second leading cause of lung cancer after smoking going by data reported from several epidemiological studies that indicated that exposure to indoor radon causes lung cancer, according to Handbook on Indoor Radon (WHO, 2009). In light of the latest scientific data on radon epidemiology, WHO proposed a reference level or action level of 2.7 pCi/L to minimize the health hazard due to indoor exposure of Rn-222. Data by radon professionals and mitigators, suggest that one in every 10 homes may have radon concentration levels above the EPA and WHO action level.

Radon is also important in the context of potable groundwater due to its high solubility in water (Smetanov´a *et al*, 2010). The greatest exposure to waterborne radon occurs in homes that are located in areas with high levels of Rn-222 in the groundwater consumed by inhabitants of the area. Many water supply systems have little or no treatment, minimal natural aeration, and a short time interval between pumping and consumption; thereby limiting dissipation of radon (Clapham C and Horan, 1996; Frengstad *et al*, 2003).

Today the increasing interest on presence of Rn-222 in our environment was due to health risks poses by its associated radiation exposure. WHO (2009) guidelines for drinking-water quality recommend a treatment of the water source, if the radon concentration exceeds a limit of 100 Bq/m^3. The logic behind the proposed action level is that it would broadly correspond to the risk posed to an individual from exposure to radon (T.P Ryan *et al*, 2003). It has been estimated that 1000 Bq/l of radon in water will, on average, increase the radon concentration in indoor air by 100 Bq/m^3 (EC, 2001). In line with this, EPA has also set an action level of 0.15 $Bq\ L^{-1}$ (4 $pCi\ L^{-1}$) for indoor air (EPA, 1986). On the basis of these findings, exposure to Rn-222 through drinking water should be subjected to continuous monitoring.

This therefore demonstrated that measurement of Rn-222 in commercial drinking water is necessary especially in our community which depends largely on private ground water supplies as their primary source of drinking water. This compelling need to monitor radon levels in drinking water sources from boreholes in Kaduna and environments will provide additional data on Rn-222 exposure levels in the general population.

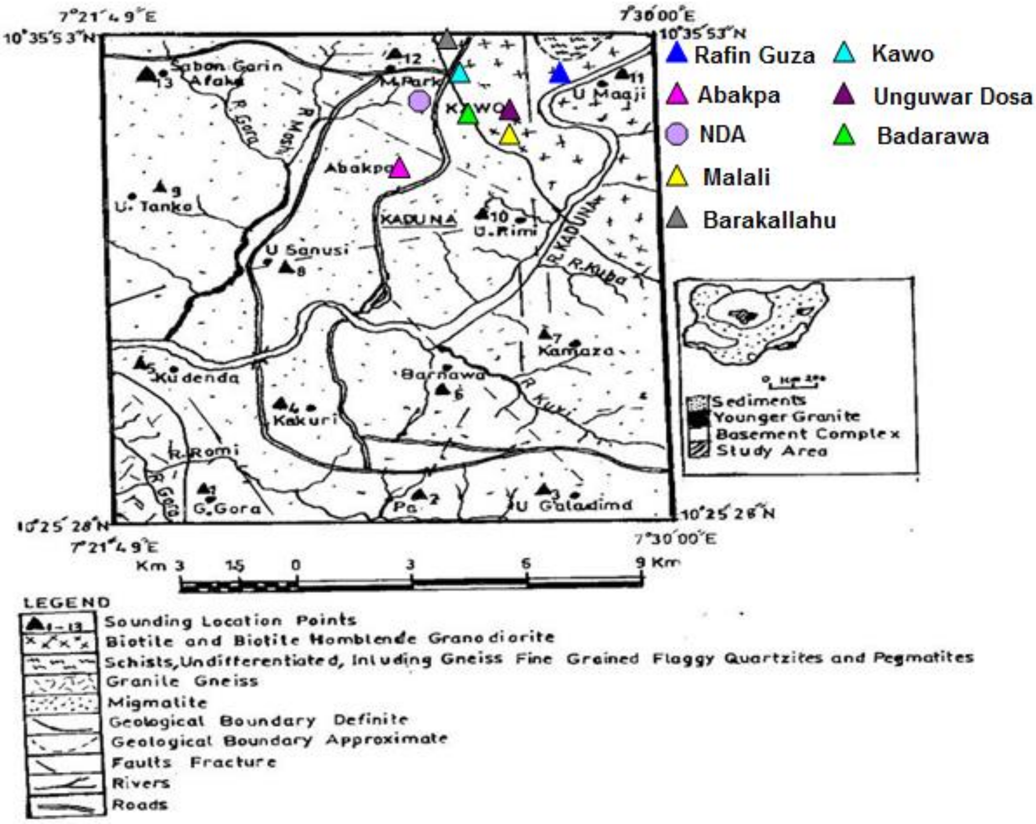
2. GEOLOGY OF THE STUDY AREA

Kaduna State lies between Latitudes 9^o 00"N and 11^o 40"N and longitudes 6^o 20"E and 80^o 80"E covering an area of 43,898sq km (Akintola, 2006).There are two distinct seasons, the rainy season extends from April to October and the dry season, between November and April. The mean annual temperature ranges between 24^o C to 30^o and the annual rainfall ranges between 112mm to 150mm (Akintola, 2006).

The selected areas where samples were collected comprise Abakfa, Badarawa, Barakallahu, Kawo, Malali Rafin Guza and Unguwar Dosa of Kaduna North Local Government Area with 425425 populations (Ajibade, 2009) in the state capital.

Figure 1. Map of Kaduna State showing the location of the study site.

It is generally accepted that high radon levels will be found in terrain of high grade metamorphic rock and granites (Brutsaert *et al*, 1981). This is primarily due to high concentrations of uranium in some of the minerals. Radon concentrations have been identified by rock type with typical concentrations of ground water in granites. The Basement Complex rocks of the study area (Kaduna) are mostly migmatite, granite, gneiss, undifferentiated schists, porphyritic, biotite and granitites (figure 2).



Source: B. S. Jatau et al, 2013.

Figure 2. Geological map of the study area (Extracted and modified from GSN, 2644)

3. EXPERIMENTAL PROCEEDURE

3.1 Calibration of the Liquid Scintillation Counter:

The Liquid Scintillation Counter (Tri-Carb-LSA1000 LSC) located at the Centre for Energy Research and Training Zaria was calibrated using Ra-226 secondary calibration standard. The calibration consists of 10ml of the Ra-226 dissolved in distilled water. The solutions were stored for more than one month for secular equilibration to be attained. Thereafter, 10 ml of the solution was mixed with 10 ml of Instal-gel scintillation cocktail and counted with a window setting of 25-900 in region C of the LSC for 60 minutes. The calibration factor was determined using equation 1 below.

$$CF = \frac{(I_{Std} - I_{BG})}{C_{std} \times V_{std}} \qquad (1)$$

Where:

I_{Std} = Ra-226 intensity (count/min),

I_{BG} = Background count (count/min),

C_{std} = Concentration of Ra-226 (Bq/L) and,

V = Volume of Ra-226 solution (L)

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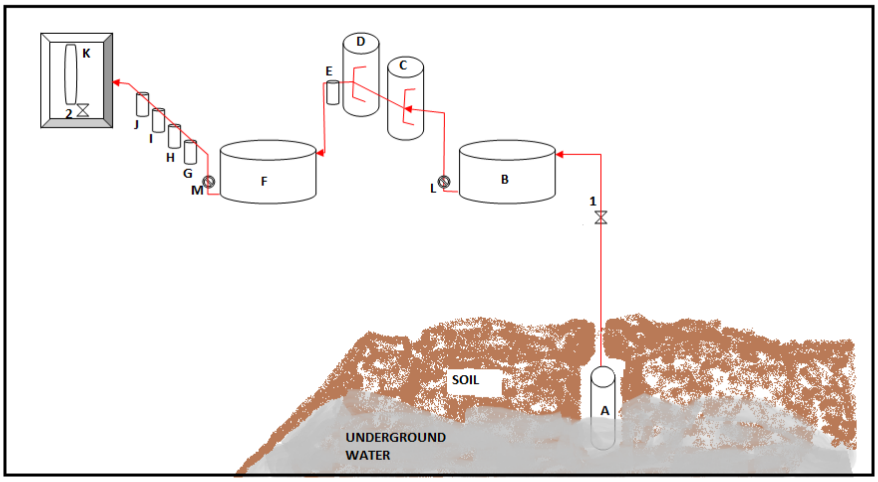
3.2 Sample Collection:

Water samples were collected from commercial boreholes used for the production of Table (Sachet) water and Bottled water. Water samples from the borehole were collected directly by sucking the water with short tubing connected to the pipe that connects to the submersible pump inside the borehole. Before sample collection, the borehole was allowed to operate for at least three minutes in order to ensure collection of fresh water samples (Nuraddeen *et al*, 2012). This is shown in Figure 3.

Figure 3. Device and pipe connection arrangement

During sample collection the water flow speed was first reduced with a faucet attached to the constructed device and a 10 ml plastic vial (sample collection bottle) which was tilted at roughly angle 45^{o} inside a clean 500ml beaker placed appropriately to receive the water flowing from the tubing. Water sample was then allowed to flow in the 10ml plastic vial slowly to avoid aeration until the plastic vial and the beaker is filled up and overflows. The plastic vial was then secured using TFE cap while it is still submerged in the beaker. The vial was then removed and immediately inverted to check for bubbles. The process is repeated when bubble(s) are observed or otherwise the sealed vial is labeled appropriately with necessary information such as sample ID, sample collection time and geographical location.

For the purposes of standardization and assessment of Rn-222 ingestion, commercial water samples supplied to consumers were also collected from the last outlet at the sealing machine unit (Figure 4). These samples were collected by first reducing the speed of the water flow from the machine following the same procedure as in the former case.



→ PVC pipe

A: Submersible pump	D: Sand filter	G: 1 micro filter	J: Carbon filter	M: Pump 2.
B: Water reservoir 1	E: Filter	H: 0.5 micro filter	K: Sealing machine	1: sample collection point 1
C: Carbon filter	F: Water reservoir 2	I: Sand filter	L: Pump 1	2: sample collection point 2

Figure 4. Schematic diagram of table water filtration system showing two sample collection points (1 & 2)

The samples were analysed for radon as soon as possible after collection to minimise radioactive decay of Rn-222 and reduce uncertainties in the analysis. All measurements were decay corrected to the time of sampling. Linear regression analysis of the duplicate samples for each supply demonstrated excellent agreement (R^2=0.999). All collected samples were transported to CERT Zaria (about 80km from the study area) in cooler boxes within 3 days to avoid decay of Rn-222. Table 2 gives the geographical details of sampling location of the various drinking water samples collected.

3.3 Sample preparation:

Ten (10) mls of the water sample was carefully drawn with 10 ml syringe and added to a scintillation vial containing 10 ml of insta-gel scintillation cocktail. The vial was then sealed tightly and shaken for more than two minutes to extract Rn-222 in water phase into the organic scintillator. All the water samples prepared this way were orderly arranged for analysis of radon.

3.4 Theoretical Background:

Liquid scintillation is the detection of beta radiation in a scintillation cocktail, which occurs when a neutron is converted to a proton, and a beta particle (electron) and antineutrino are ejected. For example; the nucleus of tritium contains one proton and two neutrons. When tritium undergoes beta decay, one of the neutrons decays to a proton, and a beta particle and antineutrino are ejected. As a result, tritium is converted to helium, as shown in Equation 3.1.

$$^{3}H_{1} \rightarrow {}^{3}He_{2} + {}^{0}e_{-1} \qquad (2)$$

The scintillation cocktail captures the beta emission energy and transforms it into a photon which can be detected via a photocathode, amplified by a photomultiplier tube, and converted to Counts Per Minute (CPM).

3.5 Decay correction factor:

Corrections are made in the decay of Rn-222 to augment some loses (decay) in the process of transporting the sample from the collection point to the mid time (30 min) of sample measurement in liquid scintillation counter. This is represented by the decay correction factor (D) given by

$$D = \exp(-T\lambda) = \exp\left(-\frac{0.693T}{t_{1/2}}\right) \qquad (3)$$

Where:

D= Decay correction factor

T= delay time between sample collection and the middle time of sample measurement (minutes)

λ = decay constant

4. SAMPLE ANALYSIS

The sample is dark-adapted and equilibrated, and then counted in a liquid scintillation counter using a region or window of the energy spectrum optimal for radon alpha particles i.e. region C. Results are reported as CPM. Prepared samples were allowed to stand for at least 60 minutes after preparation in order to establish radioactive equilibrium between ^{222}Rn and its daughter progeny (Nuraddeen *et al*, 2012). They were then loaded to the LSC and counted for 60 minutes.

The adopted counting time was a compromise between the sensitivity of the method and the time available for the analysis of all collected samples. However, the resulting limit of detection (LDD) of 0.04Bq/l is considered to be sufficient for the low Rn-222 concentrations that are expected. Working standards were counted alongside the groundwater water samples and one background sample was included for every 10 samples. Table 4 shows the intensity data obtained from measurement of radon in water.

4.1 Data Processing:

The activity concentration of ^{222}Rn (A) as provided by the LSC were given in Counts Per Minute (CPM). These results can be converted to specific activity in Bql^{-1} using the following equations:

$$A(Bql^{-1}) = \frac{100(S-B)\exp(\lambda T)}{60 \times CF \times D} \qquad (4)$$

Where:

A =Radon activity in Bql^{-1}

S =Sample count rate (count min^{-1})

B =Background count rate (Count min^{-1})

T =Delay time between sampling and mid time of counting (minutes)

CF=Calibration factor

Note that 1000 Liter = m^3 and Bq/L=1000Bq/m^3.

5. RESULTS AND DISCUSSION

The Rn-222 activity associated with water samples collected in each of the 12 boreholes and the corresponding purified commercial water samples in Kaduna North is displayed in Table 5 and Table 6 respectively. The measured activities in boreholes ranged approximately between 3.34±0.02–17.70±1.04 kBq/m^3 with a mean value of 7.002±0.33 kBq/m^3, while the measured activity concentration of Rn-222 in the purified water samples ranges between 3.71±0.02 and 10.28±0.27 kBq/m^3 with a mean value of 6.446±0.36 kBq/m^3. Thus large differences occur in radon activity concentration amongst most of the boreholes monitored as well as in the commercial water samples produced from the boreholes. The observed variability in the borehole water and purified water samples are also depicted in Figure 5 and 6 respectively. The observed variability of Rn-222 activity concentration is not limited to this work alone but similar situation occurred in water samples from similar studies (Nuraddeen *et al*, 2012; Smetanov´a *et al*, 2010). Further investigation is required to fully quantify this variability and its resultant implications. Such high variability tends to indicate that the collection of one sample for radon risk determination may be insufficient for radon risk determination.

In KBW1 and KBW6 Rn-222 activity concentration in water reached up to 18 and 11 kBq/m^3 respectively, while the other boreholes have Rn-222 activity concentration that is less than 10kBq/m^3. The relatively high Rn-222 concentrations in some of the boreholes monitored can be explained in terms of expected Rn-222 emanation from high grade metamorphic rock and granites (Brutsaert *et al*. 1981). This granitic nature forms the basement full of relicts of high grade granitic and metamorphic rocks which contains higher concentrations of Ra-226 and other radionuclide belonging to the uranium and thorium series.

Table 1. Sampling data for Borehole water in Kaduna North Metropolis

Sample ID	Borehole depth(m)	Longitude (^{o})	Latitude(^{o})
KBW1	33	10.3455	7.2721
KBW2	48	10.3590	7.2832
KBW3	72	10.3512	7.2827
KBW4	33	10.3457	7.2648
KBW5	66	10.3760	7.2750
KBW6	38	10.3716	7.2743
KBW7	66	10.3720	7.2734
KBW8	27	10.3328	7.2550
KBW9	36	10.3339	7.2758
KBW10	42	10.3320	7.2614
KBW11	30	10.3331	7.2644
KBW12	42	10.3430	7.2752

Table 2. Sampling data for drinking (treated) water samples from Kaduna North Metropolis.

Sample ID	Borehole depth(m)	Longitude (^{o})	Latitude(^{o})
KDW1	33	10.3455	7.2721
KDW2	48	10.3590	7.2832
KDW3	72	10.3512	7.2827
KDW4	33	10.3457	7.2648
KDW5	66	10.3760	7.2750
KDW6	38	10.3716	7.2743
KDW7	66	10.3720	7.2734
KDW8	27	10.3328	7.2550
KDW9	36	10.3339	7.2758
KDW10	42	10.3320	7.2614
KDW11	30	10.3331	7.2644
KDW12	42	10.3430	7.2752

Table 3. Instrument settings for radon analysis with Tri-Carb-LSA1000 LSC

Radionuclide	Rn-222
Measurement time (min)	60
Background rate (CPM)	18.97
Alpha window	2000
Lower limit (Bq l^{-1})	0.02
Upper limit	0.04

Table 4. Rn-222 count-rate from the various borehole water samples

Sample	Net Counts per min
KBW1	593.43
KBW2	276.88
KBW3	118.23
KBW4	164.42
KBW5	121.28
KBW6	185.88
KBW7	127.20
KBW8	309.56
KBW9	218.68
KBW10	167.68
KBW11	327.87
KBW12	259.20
KBW13	142.47
KBW14	177.93
KBW15	120.92
KBW16	122.87
KBW17	280.07
KBW18	208.07
KBW19	234.08
KBW20	191.32
KBW21	167.85
KBW22	113.45
KBW23	135.07
KBW24	125.63

Table 5: Sampling data for Borehole in Kaduna North Metropolis

Sample ID	Borehole depth(m)	Mid Time Interval T (min)	^{222}Rn Conc. (kBq/m^3)
KBW1	33	212	17.70±1.04
KBW2	48	2608	4.14±0.02
KBW3	72	2760	4.34±0.03
KBW4	33	227	3.34±0.02
KBW5	66	1418	7.16±0.63
KBW6	38	1577	11.30±0.35
KBW7	66	2863	5.31±0.34
KBW8	27	1292	3.60±0.03
KBW9	36	1425	9.37±0.33
KBW10	42	154	6.58±0.54
KBW11	30	2684	6.26±0.47
KBW12	42	2750	4.92±0.10

The results obtained show that all the boreholes monitored had Rn-222 water concentration within international standard limits such as the alternative maximum contamination level (AMCL) which is 146 kBq/m3 and the European Union reference level of 1000 kBq/m^3. However about 16.7% of the boreholes monitored exceeded the recent proposed limit of U.S Environment Protection of 11 kBq/m3. Low Rn-222 activity concentration as observed in some of the samples is probably due to the fact that these boreholes are not directly connected by fractures with the underlying bedrock.

Comparison of Table 5 and 6 also reveals that the purification process of the borehole water led to reduction of activity concentration of Rn-222 in the commercial water samples.

Table 6: Sampling data for treated water in Kaduna North Metropolis

Sample ID	Mid Time Interval T (min)	^{222}Rn Conc. (kBq/m^3)
KDW1	260	7.99±0.64
KDW2	2742	6.16±0.37
KDW3	2828	7.15±0.58
KDW4	283	9.03±0.61
KDW5	1527	5.41±0.19
KDW6	2819	10.28±0.27
KDW7	2924	6.89±0.52
KDW8	1377	3.71±0.08
KDW9	1482	6.84±0.59
KDW10	222	5.32±0.44
KDW11	2745	4.00±0.03
KDW12	2858	4.59±0.02

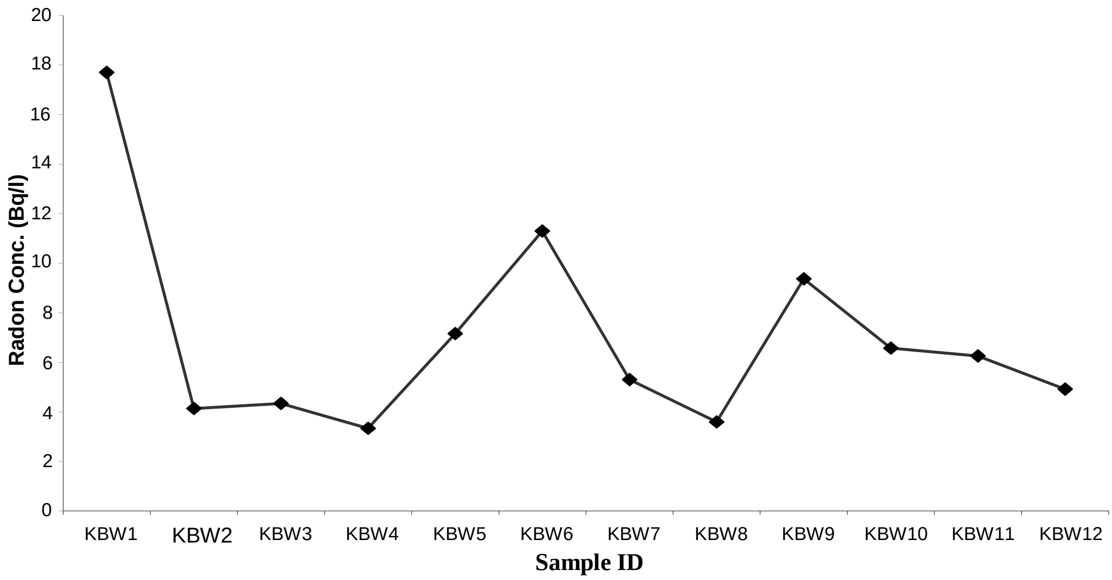


Figure 5. Variability of Rn-222 activity concentration among boreholes in Kaduna

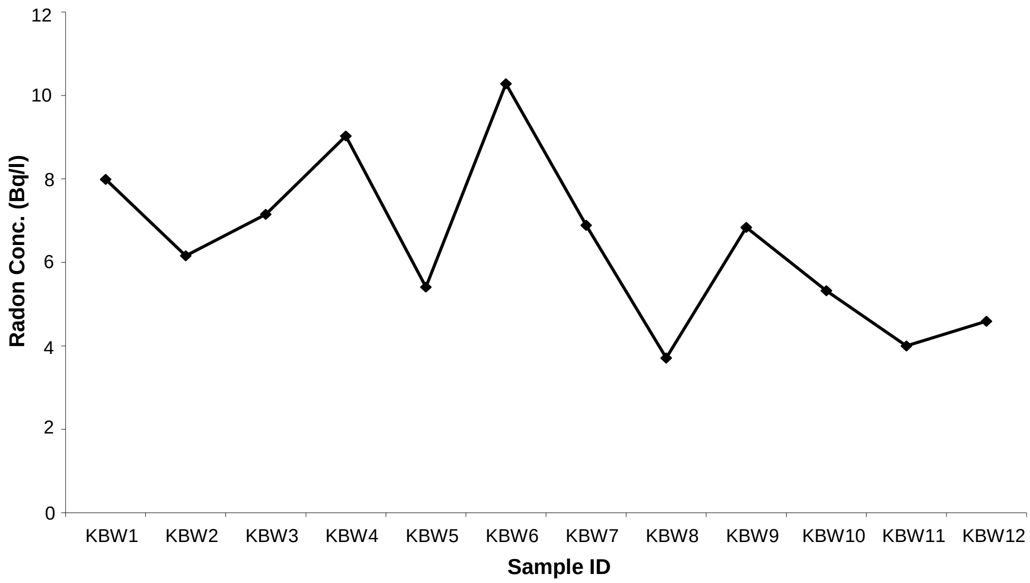


Figure 6. Variability of Rn-222 activity among treated water samples from Kaduna boreholes

Table 7: Sampling data for Borehole and purified water in Kaduna North Metropolis

Sample ID	Effective Dose (mSv/yr) Borehole water	Sample ID	Effective Dose (mSv/yr) Treated water
KBW1	0.06	KDW1	0.03
KBW2	0.02	KDW2	0.02
KBW3	0.02	KDW3	0.03
KBW4	0.01	KDW4	0.03
KBW5	0.03	KDW5	0.02
KBW6	0.04	KDW6	0.04
KBW7	0.02	KDW7	0.03
KBW8	0.01	KDW8	0.01
KBW9	0.03	KDW9	0.02
KBW10	0.02	KDW10	0.02
KBW11	0.02	KDW11	0.01
KBW12	0.02	KD W12	0.02

It should be noted that on the basis of current knowledge of the effects of exposure to radiation, a dose of 1 mSv per annum carries a lifetime risk of one in 20,000 of contracting a fatal cancer. The stomach wall has been identified as the organ most at risk following the ingestion of drinking water containing radon. In the present study, the effective dose calculated is an order of magnitude less than this value and thus ingestion of water from these boreholes may not predispose the consumers to cancer in a short term.

However, it is still advisable to intensify the water purification procedure to reduce the activity concentration of Rn-222 in the borehole water taking into consideration the high variability of radon that may result in much higher concentration of Rn-222 than the average value in some cases and consideration of other radionuclide of the uranium decay series that are always present whenever radon is detected in water.

6. CONCLUSION AND RECOMMENDATIONS

6.1 SUMMARY AND CONCLUSION:

The following provides the summary and conclusions on the present work on monitoring of Rn-222 in boreholes located in Kaduna North metropolis using Liquid Scintillation Counter.

(i) A sampling and analysis methodology for the determination of Rn-222 in drinking water based on liquid scintillation spectrometry was successfully used for analysis of Rn-222 in water samples from boreholes monitored in Kaduna North metropolis.

(ii) The measured activities in boreholes ranged approximately between 3.34±0.02–17.70±1.04 kBq/m^3 with a mean value of 7.002±0.33 kBq/m^3, while the measured activity concentration of radon in the purified water samples ranges between 3.71±0.02 and 10.28±0.27 kBq/m^3 with a mean value of 6.446±0.36 kBq/m^3.

(iii) Rn-222 from the boreholes monitored in Kaduna North metropolis is characterized with high spatial variability implying that the collection of one sample from a particular borehole may be insufficient for radon risk determination.

(iv) The estimated doses due to the consumption of drinking water containing radon vary significantly with the quantity of drinking water consumed and the degree to which the water has been processed prior to consumption. However dose estimates based on measurements made in this study demonstrate that Rn-222 in drinking water may not pose a significant health risk, in the short term, to consumers who depend on drinking water supplies derived from the boreholes monitored.

(v) It should be noted that while significant doses have not been estimated for Rn-222 ingestion in Water samples from the boreholes, it is still advisable to intensify the water purification procedure to reduce the activity concentration of Rn-222 in the borehole water taking into consideration the high variability of radon that may result in much higher concentration of radon than the average value in some cases and consideration of other radionuclide of the uranium decay series that are always present whenever Rn-222 is detected in water.

6.2 RECOMMENDATIONS:

The study hereby recommends that the Ministry of Health (in Nigeria) should establish environmental guidelines on reference limits for reduction of risk to Rn-222 exposure in drinking water.

It is also recommended that Rn-222 reduction methods aimed at reducing activity concentration of Rn-222 in water should be applied on regular basis. These include storing the pumped water in a reservoir longer than 3 days, aerating pump water containing Rn-222 by boiling it before usage etc. These could have significant effect in reduction of Rn-222 levels in drinking water and reduction of cancer cases in the public.

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