ESTIMATION OF ANNUAL EFFECTIVE DOSE DUE TO INGESTION AND INHALATION OF RADON IN GROUNDWATER FROM KADUNA, NIGERIA.

Abstract

The variation in the concentration of radon in groundwater sources comprising of boreholes and wells in Kaduna metropolis and environs were determined using Tri-carb LSA 1000 liquid scintillation counter. The radiation dose received by individuals within different age groups categorized under; infants, children and adults, depending on their average annual water consumption rates (ACRs) were also estimated. The mean radon activity in 16 borehole and 18 well water samples were 1.8/Bq/L and 0.57 Bq/L respectively; while the average radon activities ranged from 0.85 to 2.57 Bq/L and 0.35 to 0.85Bq/L respectively with all values far below the United States Environmental Protection Agency MCL of 11.1 Bq/L. All the estimated annual committed effective dose (ACED) values for all samples were observed to increase with increase in radon concentration, age and ACRs, but were significantly lower than the UNSCEAR and WHO recommended limit of 1mSv/yr. The radiation dose rate received by the lung cells due to the inhalation of waterborne radon in air was very much higher when compared to that received by the stomach walls via ingestion.

Keywords: Radon, annual committed effective dose, ICRP age groups, maximum contaminant level.

1.0 Introduction

The necessity of water for human survival underscores the need for water quality assessment, and this entails having regulations on natural radioactivity in drinking water. The occurrence of radionuclides in drinking water gives rise to internal exposure, directly via their decay processes, when they are taken directly into the body through ingestion and inhalation; and indirectly, when they are incorporated as part of food chain [1].

The demand for groundwater in cities, towns and rural areas in Nigeria has risen over the years due to insufficient supply of treated water. Groundwater simply refers to water which accumulates underground as a result of rain, snow, sleet which then seeps into the ground because of gravity. On reaching a soil depth where the pore spaces are already filled or saturated with water, it surfaces to become wells and boreholes [2]. Fractures and joints serve as routes for the flow of groundwater and radon (Rn-222) gas in radon-contaminated soils.

Radon, a noble radioactive gas tends to migrate readily in air or water inspite of the fact that its relatively short half-life (3.82days) restricts the time for which it can migrate. It originates from the radioactive decay of naturally occurring uranium [3] and radium deposits, which is picked up by groundwater passing through rocks and soil containing such radioactive substances and then enters water supplies when this water circulates into a well [4]. All groundwater contain radon from both dissolved radium and from recoil of radon from soil and rocks which diffuse over large distances in water and whose radon concentration varies from place to place.

Public exposure to waterborne Rn-222 and its short-lived decay products (such as Po-218 and Pb-214) may occur by ingestion (drinking water containing Rn-222) and by inhalation (breathing Rn-222 gas in indoor/outdoor air which has been released from household water) and both mechanisms pose a potential health risk [5,6]. Consequently, exposure to radon and radon progeny is considered the dominating source of exposure to ionizing radiation in most countries [7]. A very high level of radon in drinking water can lead to a significant risk of developing internal organ cancers primarily, stomach and gastrointestinal cancer [8,9]. Also, radon, when present at high concentration is also known to cause lung cancer [10,11].

Thus, to protect the public from the consequences of excessive exposure to radiation due to radon in the study area, it is pertinent to evaluate the levels of radon and hence estimate the effective doses to the population due to the consumption of the groundwater which serve as drinking water. In Nigeria, and other parts of the world, some studies have reported relatively high radon concentrations in well and borehole water for public consumption [12,13,14,15]. Specifically, the determination of effective doses due to the ingestion and inhalation of radon has been carried out only in a few locations in western Nigeria [16]. Consequently, in this research work, a liquid scintillation counter was used to study the variation in the level of Rn-222 in groundwater of Kaduna metropolis and environs. Also, the annual effective doses of radon ingested and inhaled by the inhabitants were calculated in order to determine their exposure dosage to the radiation.

2.0 Study Area

Kaduna, the state capital of Kaduna state in north-western Nigeria is situated along the Kaduna river, and is a major transportation hub for the surrounding agricultural areas with its rail and road junction. Its population is estimated to be 1.3 million with a population density of 5,800 person per square kilometer [17]. Kaduna is bordered by Zamfara, Katsina and Kano States to the north; Bauchi and Plateau states to the east; Nasarawa state to the south; and Niger state and Abuja to the west. Kaduna lies approximately, between latitude 10°25.5′N and 10°36′N, and longitude 7°22′E and 7°30′E covering a total land area of about 131 square kilometer. The Kaduna river, a tributary of the Niger river flows roughly east to west through the centre of the state [18]. The state's natural vegetation extends from the Guinea Savannah to the Sudan Savannah woodland in the north.

The study area lies within the North Central Nigeria Basement Complex which is composed mostly of migmatite, granite gneiss, undifferentiated Schists and porphyritic granite [19]. The area is generally part of the extensive but gently undulating peneplain, capped at high elevation by patches of laterised terraces of iron oxides, concentration of broken-up concretion ironstones and some quartz. The soil type found in this area is classified under the "interior zone of laterite" which is made up of sands and clays. They are grey to black clays, poorly drained and seasonally flooded forming the "Fadama". The soil is deeply eroded, generally sticky and impervious to water and has low fertility. Also, for management purposes, Kaduna soils have been grouped under "soils with high base saturation" within the savannah vegetation (grassland). These soils are formed from metamorphic, igneous rocks, volcanic and sedimentary parent materials, and are found in the grain producing areas where water is relatively in adequate supply, and intensive cultivation is possible [20].

3.0 Methodology

3.1 Sampling

Water samples were collected from boreholes and hand-dug wells from Kaduna metropolis and environs during the dry season (that is, between the months of March and May). The water samples from hand-dug wells were fully filled into air-tight clean Polythene bottles to avoid degassing during sample collection. For collection of samples from boreholes the submerged vial method was used. All samples collected were analyzed within three days to achieve maximum accuracy.

3.2 Radon Measurement Using Liquid Scintillation Counting (LSC)

10ml of the liquid scintillation toluene based cocktail (whose trade name is insta-gel) was transferred into a scintillation vial, and then 10ml of each water sample was drawn into a syringe and injected beneath it. The water samples were drawn with the syringe slowly in order to avoid turbulence and collection of air bubbles. After capping, the vials were then shaken for more than two minutes to extract the aliquot of ^{222}Rn which partitioned selectively into a mineral-oil scintillation cocktail, and remained immiscible with the water samples in the process. A standard (^{226}Ra solution) supplied by the IAEA which had been mixed with insta-gel in appropriate proportion and kept for over a month to ensure ^{226}Ra – ^{222}Rn radioactive equilibrium, was used in this work.

The prepared samples were then analyzed for the activities of ^{222}Rn using a liquid scintillation counter (Tri-carb LSA 1000) located at the centre for Energy Research and Training, Ahmadu Bello University, Zaria. Each sample was counted for 1hour only after having been allowed to stay for at least three hours after preparation, to allow for radioactive equilibrium between ^{222}Rn and its daughters to be established.

Calibration of the LSC was done prior to the counting using the IAEA^{226}Ra standard solution; the ^{226}Ra standard samples were counted for 60 minutes. For background count measurement, distilled water sample was mixed with the cocktail of 10ml each and allowed to remain for 14 days before counting for 60 minutes. The ^{222}Rn activity concentration was then calculated using the equation:

$$A = \frac{(C_S - C_B)(1000mL)}{(CF)(D)(10mL)(1L)} \quad \text{Equation 1}$$

where:

A = becquerels of radon per litre (or Activity) of sample

C_S = sample counts per second (CPS)

C_B = background radiation (CPS)

CF = conversion factor calculated as, (CPS measured for calibration standard/disintegrations per second (dps) of Ra-226 contained in 10mL aliquot of scintillation mix.

D = decay correction = $\exp\left(-\frac{0.693(T)}{t_{1/2}}\right)$,

T = time in days from collection time of sample to midpoint of counting time; and $t_{1/2}$ = radiological half-life of radon, 3.82 days.

3.3 Estimation of Annual Effective Dose by Ingestion for ICRP Age Groups

The annual committed effective dose (ACED) received from ingestion of water containing radon to an individual consumer, based on the International Commission on Radiological Protection (ICRP) age groups in table 1 was evaluated using the equation:

Dose (μSv or mSV) = radon activity concentration (Bq/L) x annual water consumption rate, ACR (L) x dose conversion factor (DCF) in Sv/Bq.

Equation 2

The values for the ACRs and conversion coefficient for ingestion used are as follows:-

(i) For adults, age > 18 yrs

$ACR = 730 Lyr^{-1}$, and $DCF = 1 \times 10^{-8} SvBq^{-1}$

(ii) For children ≅ 10 yrs

$ACR = 350 Lyr^{-1}$, and $DCF = 2 \times 10^{-8} SvBq^{-1}$

(iii) For infants between the ages of 1-2yrs

$ACR = 260 Lyr^{-1}$, and $DCF = 2 \times 10^{-8} SvBq^{-1}$

[21,9,22].

3.4 Estimation of Annual Committed Effective Dose (ACED) to Internal Organs

Since radon enters the human body via ingestion as well as inhalation (as radon is released from water to air), the effective doses to internal organs such as the stomach and lungs were evaluated as follows:-

(i) The dose to the stomach which is as a result of ingestion of waterborne radon, was calculated using the equation:

$E_{ing} = A_{Rn} \times V \times DCF$,Equation 3

whereA_{Rn} is the average radon activity in drinking water.

V_A is the estimated annual volume of water consumed in litres (which is 50 litres for adults). The dose conversion factor, DCF equals 3.5nSv/Bq [23]

(ii) The dose to the lungs; which is the dose received by bronchial and pulmonary tissues of human lungs (i.e. inhalation) due to radon gas indoors, was estimated using the equation:

$E_{inh} = A_{Rn} \times T \times F \times t \times DCF$. Equation 4

where E_{inh} = effective indoor dose, DCF = 9×10^{-9}Sv/Bq/m^3 or 9×10^{-6}mSv/h per Bq/m^3 was used to convert radon equilibrium equivalent concentration to population effective dose [24,25] as it lies between the dosimetric and epidemiological dose conversions [26,24,27].

A_{Rn} = average Rn-222 concentration
T = radon transfer from water to air coefficient = 0.1Lm^{-3}.
t = average annual indoor occupancy factor in hours = 7,000hr.
F = indoor radon daughters equilibrium factor = 0.4.

Table 1: ICRP age groups and their ACRs [28].

Age group	Age range (years)	Water consumption (L/day)	Water consumption (L/year)
3 months	0–1	0.55	200
1 year	1–2	0.71	260
3 years	2–7	0.82	300
10 years	7–12	0.96	350
15 years	12–17	1.64	600
Adults	>17	2.00	730

4.0 RESULTS AND DISCUSSION

4.1 Radon Concentration

The Rn-222 concentrations obtained for 18 well water samples and 16 borehole water samples from Kaduna metropolis and environs, along with their age-dependent ACED to individuals belonging to different ICRP age groups, assuming annual consumption of the estimated volumes of water are presented in table 2. The radon concentrations obtained range from 0.35 – 0.85 Bq/L and 0.85 – 2.57 Bq/L with mean of 0.575 and 1.811 for wells and boreholes respectively. The mean values are higher than the MCL of 0.1 Bq/L set by the

Standards Organization of Nigeria [29] but lower than the United States Environmental Protection Agency,(USEPA) reference level of 11.1Bq/L and the world average of 10Bq/L prescribed by the WHO. The variation in radon concentration in the samples from the two sources with boreholes having higher values than wells may be due to the fact that boreholes are more stagnant and there is no loss of radon concentration due to lack of aeration [29]. The concentrations are comparable to other reported works on radon concentration in similar geological formations in North Western Nigeria [31,14,32].

When the mean, μ and standard deviation, σ, of radon concentration in well water (μ = 0.57 Bq/L; σ = 0.16 Bq/L) were compared with those in borehole water (μ = 1.81Bq/L; σ = 0.49 Bq/L), no significant difference in the mean concentration was found between the two water sources types.

The range of the radon concentration data and ACED in this study were compared with that determined in several other countries and elsewhere in Nigeria are presented in table 4. Generally, the radon concentration data portray low values inspite of the granitic nature of the area which may be due to the shallowness of most of the wells and boreholes.

Table 2: Mean radon activity and their respective ACED to ICRP age groups.

Sample No.	Rn-222 Concentration (Bq/L)	ACED (μSv/year) to individuals in ICRP age groups		
		Infants	Children	Adults
Boreholes				
+KDS1	1.288	6.70	9.02	9.40
+KDS2	1.741	9.05	12.19	12.70
+KDS3	2.132	11.09	14.05	15.56
+KDS4	1.788	9.30	12.52	13.05
+KDS5	2.007	10.44	14.05	14.65
+KDS6	1.991	10.35	13.94	14.53
+KDS7	2.132	11.09	14.92	15.56
+KDS8	1.156	6.01	8.09	8.44
+KDS9	1.788	9.30	12.52	13.05
+KDS10	2.335	12.14	16.35	17.05
+KDS11	2.000	10.40	14.00	14.60
+KDS12	2.570	13.36	18.00	18.76
+KDS20	2.210	11.49	15.47	16.13
+KDS21	1.960	10.19	13.72	14.31
+KDS22	1.060	5.51	7.42	7.74
+KDS25	0.850	4.42	5.95	6.21
Mean	1.81	9.43	12.64	13.23

SD	0.49	2.53	3.37	3.55
Minimum	0.85	4.42	5.95	6.21
Maximum	2.57	13.36	18.00	18.76
Hand-dug Wells				
*KDS13	0.510	2.65	3.57	3.72
*KDS14	0.480	2.50	3.36	3.50
*KDS15	0.770	4.00	5.39	5.62
*KDS16	0.650	3.38	4.55	4.75
*KDS17	0.850	4.42	5.95	6.21
*KDS18	0.690	3.59	4.83	5.04
*KDS19	0.350	1.82	2.45	2.56
*KDS23	0.730	3.80	5.11	5.33
*KDS24	0.810	4.21	5.67	5.91
*KDS26	0.750	3.90	5.25	5.48
*KDS27	0.450	2.34	3.15	3.29
*KDS28	0.630	3.28	4.41	4.60
*KDS29	0.530	2.76	3.71	3.87
*KDS30	0.500	2.60	3.50	3.65
*KDS31	0.390	2.03	2.73	2.85
*KDS32	0.380	1.98	2.66	2.77
*KDS33	0.390	2.03	2.73	2.85
*KDS34	0.440	2.29	3.08	3.21
Mean	0.57	2.98	4.01	4.18
SD	0.16	0.85	1.15	1.20
Minimum	0.35	1.82	2.45	2.56
Maximum	0.85	4.42	5.95	6.21

Key:
* - hand-dug well
+ - borehole

4.2 Annual Effective Dose by Ingestion

The estimated annual committed effective dose by ingestion of borehole water for ICRP age groups ranged from 4.42 – 13.36µSv/yr, with a mean of 9.43µSv/yr; 5.95-18µSv/yr, with a mean of 12.64µSv/yr; and 6.21-18.76µSv/yr with a mean of 13.23µSv/yr for infants, children and adults respectively. Since the effective dose depends on the mean radon concentrations, locations with high values of radon concentrations also had high values of annual effective dose. Whereas, the ACED by ingestion of well water for ICRP age groups ranged from 1.82-4.42µSv/yr, with a mean of 2.98µSv/yr; 2.45-5.95µSv/yr, with a mean of 4.01µSv/yr, and 2.56 – 6.21µSv/yr, with a mean of 4.18µSv/yr for infants, children and adults respectively.

Since the ACED by ingestion of borehole for the ICRP age groups has the highest value of 18.76µSv/yr which equals 0.01876mSv/yr for both sources of groundwater, it implies that the overall dose rate received by all ICRP age groups is very low compared to the UNSCEAR and WHO recommended limit of 1mSv/yr for the public.

The foregoing data indicate that the ACED values increase with radon concentration, age and water consumption rates. Hence, the ACED received by adults is greater than that received by children, which is greater than that of infants. This is in agreement with the findings of a similar study [1].

Table 3:Radon concentration of water samples and their respective ACED to internal organs (stomach and lungs)

Sample No.	Rn-222 Concentration (Bq/L)	ACED (µSv/year) to individuals in ICRP age groups			ACED (µSv/yr) to the stomach	ACED (µSv/yr) to the lungs	Whole body (µSv/yr)
		Infants	Children	Adults			
Boreholes							
+KDS1	1.288	6.70	9.02	9.40	0.23	3.2	3.43
+KDS2	1.741	9.05	12.19	12.70	0.30	4.4	4.7
+KDS3	2.132	11.09	14.05	15.56	0.37	5.4	5.77
+KDS4	1.788	9.30	12.52	13.05	0.31	4.5	4.81
+KDS5	2.007	10.44	14.05	14.65	0.35	5.1	5.45
+KDS6	1.991	10.35	13.94	14.53	0.34	5.0	5.34
+KDS7	2.132	11.09	14.92	15.56	0.37	5.4	5.77
+KDS8	1.156	6.01	8.09	8.44	0.20	2.9	3.1
+KDS9	1.788	9.30	12.52	13.05	0.31	4.5	4.81
+KDS10	2.335	12.14	16.35	17.05	0.41	5.9	6.31
+KDS11	2.000	10.40	14.00	14.60	0.35	5.0	5.35
+KDS12	2.570	13.36	18.00	18.76	0.45	6.5	6.95
+KDS20	2.210	11.49	15.47	16.13	0.39	5.6	5.99
+KDS21	1.960	10.19	13.72	14.31	0.34	4.9	5.24
+KDS22	1.060	5.51	7.42	7.74	0.18	2.7	2.88
+KDS25	0.850	4.42	5.95	6.21	0.15	2.1	2.25
Mean	1.81	9.43	12.64	13.23	0.32	4.57	4.88
SD	0.49	2.53	3.37	3.55	0.09	1.24	1.32
Minimum	0.85	4.42	5.95	6.21	0.15	2.10	2.25
Maximum	2.57	13.36	18.00	18.76	0.45	6.50	6.95
Hand-dug Wells							
*KDS13	0.510	2.65	3.57	3.72	0.09	1.3	1.39
*KDS14	0.480	2.50	3.36	3.50	0.08	1.2	1.28
*KDS15	0.770	4.00	5.39	5.62	0.13	1.9	2.03
*KDS16	0.650	3.38	4.55	4.75	0.11	1.6	1.71
*KDS17	0.850	4.42	5.95	6.21	0.15	2.1	2.25
*KDS18	0.690	3.59	4.83	5.04	0.12	1.7	1.82
*KDS19	0.350	1.82	2.45	2.56	0.06	0.9	0.96
*KDS23	0.730	3.80	5.11	5.33	0.13	1.8	1.93
*KDS24	0.810	4.21	5.67	5.91	0.14	2.0	2.14
*KDS26	0.750	3.90	5.25	5.48	0.13	1.9	2.03
*KDS27	0.450	2.34	3.15	3.29	0.08	1.1	1.18
*KDS28	0.630	3.28	4.41	4.60	0.11	1.6	1.71
*KDS29	0.530	2.76	3.71	3.87	0.09	1.3	1.39
*KDS30	0.500	2.60	3.50	3.65	0.09	1.2	1.29
*KDS31	0.390	2.03	2.73	2.85	0.07	1.0	1.07

*KDS32	0.380	1.98	2.66	2.77	0.06	0.9	0.96
*KDS33	0.390	2.03	2.73	2.85	0.07	1.0	1.07
*KDS34	0.440	2.29	3.08	3.21	0.08	1.1	1.18
Mean	0.57	2.98	4.01	4.18	0.10	1.42	1.52
SD	0.16	0.85	1.15	1.20	0.03	0.40	0.43
Minimum	0.35	1.82	2.45	2.56	0.06	0.90	0.96
Maximum	0.85	4.42	5.95	6.21	0.15	2.10	2.25

Key:
* - hand-dug well
+ - borehole

4.3 Annual Effective Dose to Internal Organs

Radon in water is a source of radiation dose to both stomach and lungs as it can enter human body via ingestion (stomach) and through inhalation (lungs). Hence, the ACED to these organs were estimated and are presented in table 3.

The ACED values received by the stomach due to ingestion of waterborne radon in boreholes ranged between 0.15-0.45µSv/yr with a mean of 0.32µSv/yr. While that received by the lungs due to inhalation of radon released by use of borehole water was in the range 2.10-6.50µSv/yr with a mean of 4.57µSv/yr. Whereas, the ACED values received by the stomach due to ingestion of well water was in the range 0.06-0.15µSv/yr with a mean of 0.099µSv/yr. But the ACED values received by the lungs due to inhalation of radon released by use of well water ranged between 0.90-2.10µSv/yr with a mean of 1.42µSv/yr.

These data show clearly that the ACED values received by the lungs due to inhalation of radon released in air when water is used are higher than that received by the stomach due to ingestion of water-borne radon in both boreholes and wells. This suggests that the lung cells are more susceptible to cancer than cells in the stomach walls. However, the maximum ACED values received both by the lungs and stomach due to inhalation and ingestion of radon for both water sources which are 0.00045mSv/yr and 0.0065mSv/yr respectively are far less than the UNSCEAR and WHO recommended limit of 1 mSv/yr for the public.

Though different researchers have used different values for the human annual water intake and the radon water-to-air coefficient, T for any specific conditions; when computing the ACED to the stomach and lungs respectively, the inhalation of the radon escaping from water constitutes the greater radiological hazard when compared to that of ingestion of water. This is in agreement with a similar work by Bem et al. [33].

Table 4: Range of radon concentrations and effective doses in various types of water worldwide: Nigeria. Ghana, Egypt, Kenya, India, China, USA, Saudi Arabia, Brazil, Australia, U.K.

Water type	Country	Range of Radon Conc. (Bq/L)	Range of AED (µSv/yr)	Reference
Borehole	Ghana	5.40-46.74	6.05×10^{-3} – 40.66×10^{-3} (inhalation) 1.71×10^{-5} to 1.32×10^{-4} (ingestion)	[2]
Tap and well water	China	4.63-49	2.75 – 29.40 (ingestion) 28.5 – 301.84 (inhalation)	[34]
Groundwater	Saudi Arabia	0.76-9.15	2.77-33.39	[35]
Ground and surface water	India	11.50-381.20	0.57-71.48 (ingestion) 3.75 – 953.0 (inhalation)	[1]
Ground water	Poland	0.43-10.52	1.15-6.3 (ingestion) 11.8-64.7 (inhalation)	[33]
Ground, surface water and mixture of both.	Iran	0.64-49.09	0.012-8.84 (ingestion) 0.160-122.72 (inhalation)	[36]
Boreholes and wells	Nigeria (Ado Ekiti)	3.09-32.03	2.258×10^{-5} – 2.338×10^{-4} (ingestion)	[13]
Ground water	Nigeria (Ibadan)	2.18-76.75	0.036-1.26 (ingestion) 0.533-18.82 (inhalation)	[16]
Tailings-bearing, domestic and surface water	Nigeria	2.23-3.08	11.11-13.05 (ingestion of surface and domestic water)	[32]
Borehole and well water	Nigeria	0.35-2.57	0.06-0.45 (ingestion) 0.9-6.5 (inhalation)	This work

CONCLUSION

Groundwater samples comprising of borehole and well water were collected from 34 different locations within Kaduna metropolis and environs in Kaduna state, Nigeria, and were analyzed for ^{222}Rn concentration using liquid scintillation counter. In addition, the annual effective dose of the ^{222}Rn ingested by different ICRP age groups and the ACED received via ingestion and inhalation (by the stomach and lungs) were calculated. The study revealed that even though majority of the samples of borehole water had ^{222}Rn concentrations greater than those of well water, all the samples from the two sources had values of ^{222}Rn concentrations much lower than the limit adopted by international regulatory bodies inspite of the gneissic granitic geology of the study area that may be associated with higher concentration of radon in some studies.

However, it was observed that the overall ACED rate due to radon emanating from all the samples in the study area increased with increase in radon concentration, age and ACRs, but were significantly lower than the UNSCEAR and WHO recommended limit of 1mSv/yr.

Also, the ACED received by the lungs cells due to the inhalation of waterborne radon in air was significantly higher than that of stomach walls via direct ingestion of waterborne radon. Therefore, it is recommended that other methods of radon-in-water measurements should be employed for comparison with this work.

However, to mitigate the possible hazard associated with high radon levels, dwellers in the study area are advised to store their drinking water from boreholes and wells in well-ventilated spaces and to also boil their water before drinking.

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