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# **Estimation of Annual Effective Dose Due to Ingestion and Inhalation of Radon in Groundwater from Kaduna, Nigeria**

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*Authors' contributions* 

*This work was carried out in collaboration between all authors. Author AK designed the study, performed the statistical analysis, wrote the protocol and wrote the first draft of the manuscript. Authors MFH and SG managed the analyses of the study and the literature searches. All authors read and approved the final manuscript.*

## *Article Information*

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## **ABSTRACT**

The variation in the concentration of radon in groundwater sources comprising of boreholes and wells in Kaduna metropolis and environs were determined by 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 boreholes 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) for all samples were observed to increase with radon concentration, age and ACRs, but were significantly lower than the United Nation Scientific Committee on Effect of Atomic

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Radiation (UNSCEAR) and World Health Organization (WHO) recommended limit of 1mSv/yr. The radiation dose rate received by the lung cells due to the inhalation of waterborne radon in the air was considerably 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. INTRODUCTION**

Radon is a noble radioactive gas soluble in water. It exists in three different isotopes but only radon-222  $(^{222}Rn)$  with a half-life of 3.82 days, is of interest as the other isotopes are very shortlived. It originates from the radioactive decay of naturally occurring uranium [1] and radium deposits, which is absorbed by ground water passing through rocks and soil containing such radioactive substances and enters water supplies when this water circulates into a well [2].

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 poses a potential health risk [3,4]. Consequently, exposure to radon and radon progeny is considered to be the dominating source of exposure to ionising radiation in several countries [5]. 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 [6,7]. Additionally, radon, when present at higher concentration is also known to cause lung cancer [8,9].

Literature has shown that though radon is a daughter product of Ra-226, found in the decay series of U-238, its concentration in water is less dependent on the bulk parent radionuclide (e.g. Ra) activity. But it varies from one place to another owing to radon release from solids which primarily depends on the parent radionuclides within host mineral grains (in rocks and soils) relative to the "recoil range"- the maximum distance a daughter product (such as radon) may traverse within a solid and into an adjacent pore due to alpha recoil [10].

Various research on other aspects of radon had culminated into sample measurements of radon concentration in water and estimation of effective doses due to ingestion and inhalation in many parts of United Kingdom and other developed countries [11].

In Nigeria, and most other African Nations, though a few measurements of radon concentration and estimation of effective doses due to ingestion and inhalation have been conducted, the level of awareness on the presence of this carcinogenic radioisotope in groundwater for drinking and for other household use remains; just as the health hazards associated with the inhalation, ingestion and external exposure to radon and its progenies. For instance, radon activities of groundwater samples measured in Nigeria include the following; in the younger granitic rocks of Jos, It ranged between 1.7 and 161.1 Bq/l in open wells; while in the basement complex rocks of Zaria, the value ranged between 0.6 and 5.01 Bq/l for open wells and boreholes; whereas, in the similar locations of predominantly crystalline rock formation and elevated background radiation level in South Western part of Nigeria, the values ranged between 0.95 and 112.00 Bq/l [12-16]. All the measured values are above the 0.1 Bq/l maximum contaminant level set by the Standards Organization of Nigeria (SON). It is also of great concern that despite the low level of awareness of the very high levels of radon measured in some parts of the world, the drilling of deep wells and boreholes as sources of drinking water and for other household use has increased in Nigeria.

Due to the insufficient supply of municipal (treated) water, the practice of drilling deep wells and boreholes as major sources of drinking water in most cities and towns has persisted for about three decades now. The aim of this study, therefore was to measure  $222$ Rn concentration in groundwater in the study area, in an attempt to address the shortcomings as mentioned earlier. In addition, the annual effective doses of radon ingested and inhaled by groundwater consumers in the locations were estimated to ascertain their exposure dosage to the radiation. Data obtained from this study with a liquid scintillation counter particularly has a concise resolving time, so high rates of disintegration could be measured. The radon data obtained in this study may complement other data which may hitherto serve as a baseline for work and policy formulation on <sup>222</sup>Rn concentration in groundwater in Nigeria.

## **2. 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 kilometers [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°25.5ʹ N and 10°36ʹ N, and longitude 7°22ʹ E and 7°30ʹ E covering a total land area of about 131 square kilometers. 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 apart of the extensive but gently undulating peneplain, capped at high elevation by patches of laterised terraces of iron oxides, the concentration of broken-up concretion ironstones and some quartz. The soil type found in this area is classified under the "interior zone of laterite", 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]. At Angwan Dosa and Malali Residential Areas within Kaduna Metropolis, young granite igneous rock formation and quarry activity are known to exist.

## **3. METHODOLOGY**

#### **3.1 Sampling**

Water samples were collected from boreholes and hand-dug wells from Kaduna metropolis and environs during the dry season (i.e., 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 [21]. All samples collected were analyzed within three days to achieve maximum accuracy.

## **3.2 Radon Measurement Using Liquid Scintillation Counting (LSC)**

Measurements were carried out using a liquid scintillation counter. In liquid scintillation analysis, energy from emitted radiation is absorbed by a fluorescent material (scintillation or fluorophore) and re-emitted as light photons.

The light photons are detected by one or more photomultiplier tubes and converted to electrical energy for analysis. The radioactive material is brought into close contact with the scintillator, usually by dissolving both the radioactive material and the scintillator in a suitable solvent; this type of instrument is particularly suitable for the qualitative measurement of radiation with limited penetrating power, such as alpha particles, beta particles and soft x-rays. The instrument also has a concise resolving time, so high rates of disintegration could be measured.

The sample preparation and analyses procedures have been described in details by Packard Instrument Company. The detailed methodology has been reported previously by the authors [22,14].

10 ml of the liquid scintillation toluene based cocktail (trade name is insta-gel) was transferred into a scintillation vial, and then 10 ml of each water sample was drawn into a syringe and injected beneath it. Toluene based or aromatic cocktails are the best solvents for scintillation counting because they provide a target for Binteraction which captures the energy of the incident particle. The energy from these molecules, in turn, allows efficient capture by scintillator molecule (dissolved phosphors). However, they are limited as they are only used for counting directly hydrophobic materials. Whereas other scintillating fluids such as Ultima-AB are safer and less toxic and are used for counting directly hydrophilic materials which include many biological materials.

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 10 ml each and allowed to remain for 14 days before counting for 60 minutes. The <sup>222</sup>Rn activity concentration was then calculated by using the following equation:

$$
A = \frac{(C_s - C_B)(1000mL)}{(CF)(D)(10mL)(1L)}
$$
(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 10 mL aliquot of scintillation mix.
- $D =$  decay correction =

$$
\exp\left(-\frac{0.693(T)}{t_{\frac{1}{2}}}\right),\,
$$

 $T = time$  in days from collection time of sample to midpoint of counting time; and  $t\frac{1}{2}$  = radiological half-life of radon, 3.82 days [23].

## **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<br>on the International Commission on on the International Commission on Radiological Protection (ICRP) age groups in Table 1 was evaluated using the following equation:

$$
D_{E} = A_{R} \times ACR \times DCF
$$
 (2)

**Where** 

 $D_F$  = annual committed effective dose (ACED) received from ingestion of water containing radon  $A_R$  = radon activity concentration

ACR = annual water consumption rate DCF = dose conversion factor

The values for the ACRs and DCFs [24] for ingestion used in Equation 2 are shown in Table 2.

## **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 by using the following equation:

$$
E_{ing} = A_{Rn} \times V \times DCF
$$
 [25,26] (3)

Where  $A_{Rn}$  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 [27]

(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:

$$
E_{inh} = A_{Rn} \times T \times F \times t \times DCF
$$
 [26] (4)

Where  $E_{\text{inh}}$  = effective indoor dose, DCF = 9 x 10<sup>-</sup>  $\mathrm{^{9}S}$ v/Bq/m $\mathrm{^{3}}$  or 9 x 10<sup>-6</sup>mSv/h per Bq/m $\mathrm{^{3}}$  was used to convert radon equilibrium equivalent concentration to population effective dose [28,29] as it lies between the dosimetric and epidemiological dose conversions [30,28,31].

 $A_{Rn}$  = average Rn-222 concentration

- $=$  radon transfer from water to air coefficient  $= 0.1$ Lm<sup>-3</sup>.
- $t = average$  annual indoor occupancy factor in hours = 7,000hr.
- $F =$  indoor radon daughters equilibrium factor = 0.4.



#### **Table 1. ICRP Age Groups and Their ACRs [32]**





#### **4. RESULTS AND DISCUSSION**

#### **4.1 Activity Concentration of Radon 222Rn**

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 estimated volumes of water were presented in Table 3. The radon concentrations was range from 0.35 – 0.85 Bq/L and 0.85 – 2.57 Bq/L with a mean of 0.575 and 1.811 for wells and boreholes respectively. The mean values are higher compared to MCL of 0.1 Bq/L set by the Standards Organization of Nigeria [34] but lower than that of the United States Environmental Protection Agency, (USEPA) reference level of 11.1 Bq/L and the world average of 10 Bq/L prescribed limit of WHO. The spatial variation in radon concentration could be a function of the geological structure of the area, depth of the water source, and also due to differences in the climatic and geo-hydrogeological processes that occur in the area. Hence, spatial variations in radon concentrations are generally related to<br>changes in geology, soil type, and changes in geology, soil type, and structural controls. For example, high radon concentrations in groundwater and soil are observed above structural planes like faults, fractures, folds and lineaments [35]. The concentrations are comparable to other reported works on radon concentration in similar geological formations in North Western Nigeria [36,14,37].

Statistical analyses were performed by using the Microsoft SPSS Software. 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 to those in borehole water (μ = 1.81Bq/L; σ = 0.49 Bq/L), no significant differences in the mean concentration was reported between the two water sources types.

The range of radon concentration and ACED in this study were compared with that determined in several other countries and elsewhere in Nigeria are presented in Table 5. The radon concentration data in this study portray low values in spite of the granitic nature of the area which may be due to the shallowness of most of the wells and boreholes, as well as due to errors associated with the radon measurement method adopted.

These errors are encountered either during sampling, processes in the field in terms of the containers (vials) used in collecting the samples, during transportation and transfer of samples by using a springe and hypodermic needle, into the scintillant. For instance, when collecting samples from a borehole, as a vial is being filled; intense agitation and mixing with the ambient air would affect the result of the radon concentration. Also, temperature increase, sudden pressure drop and high turbulence when radon gas is being released at the faucet valve would affect the radon concentration. During transportation, air bubbles may be observed in the vials which indicate mixing of other gases with radon. While, radon degassing and formation of bubbles may occur during transfer of samples into the scintillant by using a syringe and hypodermic needle in the laboratory, are among other factors that affect the radon concentration [21].

#### **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 a high value 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

 $2.56 - 6.21 \mu\text{Sv/yr}$ , with a mean of  $4.18 \mu\text{Sv/yr}$  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 that 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 in compared to the UNSCEAR and WHO recommended limit of 1mSv/yr for the public.





*Key:\* - hand-dug well; + - borehole*

<b>Sample</b>	Rn-222 concentration	ACED (µSv/year) to individuals in ICRP age groups		ACED (µSv/yr)	ACED (µSv/yr)	<b>Whole body</b>	
No.	(Bq/L)	<b>Infants</b>	Children	<b>Adults</b>	to the stomach	to the lungs	$(\mu Sv/yr)$
		<b>Boreholes</b>					
+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
<b>SD</b>	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
<b>Hand-dug Wells</b>							
*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

**Table 4. Radon concentration of water samples and their respective ACED to internal organs (stomach and lungs)**



*Key: \* - hand-dug well; + - borehole*

### **Table 5. Range of radon concentrations and effective doses in various types of water worldwide: Nigeria, Ghana, India, Poland, Iran, China, and Saudi Arabia**



The foregoing data indicate that the ACED values increase with radon concentration, age and water consumption rates. Hence, the ACED received by adults is higher than that received by children, which is greater in compared to infants. This is in agreement with the findings of a similar kind of study [25].

## **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 into the human body via ingestion (stomach) and through inhalation (lungs). Hence, the ACED to these organs were estimated and are presented in Table 4.

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 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 of 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 clearly shows that the ACED values received by the lungs due to inhalation of radon released in the 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 a 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 hazards when compared to ingestion of water. This is in agreement with a similar kind of work endorsed by Bem et al. [26].

#### **5. CONCLUSION**

The results obtained from this study indicate that majority of the samples of borehole water had  $222$ Rn concentrations higher than those of well water. However, all the samples from two sources had values of  $222$ Rn concentrations much lower than the limit of 11.1 Bq/l set by USEPA, and the world average of 10 Bq/l set by WHO, in spite of the gneissic granitic geology of the study area that may be associated with higher concentration of radon. Additionally, 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. In spite of the low values of the radon concentration in this study, it is still required to carry out extensive work on radon measurements in air and water across the different geological and geo-political zones in Nigeria, to obtain sufficient baseline data for the country and using this data and others before it as a yardstick.

#### **COMPETING INTERESTS**

Authors have declared that no competing interests exist.

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