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Assessment of Radon-222 in Selected Water Sources at Dutsin-Ma Town, Dutsin-Ma Local Government Area, Katsina State

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Abstract Human exposure to high doses of radon-222 through inhalation as gas or ingestion in water can lead to cancer. In this work, the activity concentrations of Radon-222 (^{222}Rn) were investigated from fifteen (15) water samples collected at different locations within Dutsin-Ma town, Dutsin-Ma Local Government Area, using Liquid Scintillation Counter. The concentrations of ^{222}Rn were found to range from (11.67-152.81$\boldsymbol{Bq/L}$), (0.61-172.25$\boldsymbol{Bq/L}$) and (21.98-47.17$\boldsymbol{Bq/L}$) with mean values of 64.66$\boldsymbol{Bq/L}$, 41.15$\boldsymbol{Bq/L}$ and 34.57$\boldsymbol{Bq/L}$ for Borehole, open well and Earth-Dam respectively. The results were compared with the world average maximum contaminant level (MCL) of 10$\boldsymbol{Bq/L}$ set by World Health Organization and it was noted that 86.67% of the samples exceeded this value with 20% of the samples above the World recommended reference level of 100 $\boldsymbol{Bq/L}$. Also, the Annual Effective Doses due to ingestion of ^{222}Rn in water for the three (3) categories of people were estimated from the measured ^{222}Rn concentrations and their mean values were found to be (0.472, 0.944, 3.304) $\boldsymbol{mSv/y}$ in borehole water, (0.308, 0.616, 2.156) $\boldsymbol{mSv/y}$ in well water and (0.252, 0.504, 1.764) $\boldsymbol{mSv/y}$ in surface water (Earth-Dam) for adults, children and infants respectively. All the mean values of the annual effective doses were above the recommended level of 0.1 $\boldsymbol{mSv/y}$ set by World Health Organization as such it is recommended that the inhabitant of Dutsin-Ma town should always boil their water irrespective of it source before drinking so as to keep the concentration of ^{222}Rn as low as reasonably achievable.

Keywords Radon-222, Nuclear Contamination, Natural Radioactive Sources, Effective Dose, Activity Concentration

Introduction

Water is the major constituent of the Earth's streams, lakes and oceans and the fluid of most living organisms. It covers about 71% of the Earth surface. It is vital for all known forms of life especially man. Man uses water for various reasons such as transportation, power generation, Agriculture and other domestic activities hence its availability and quality as regard radiological, microbiological, chemical and any other form of contamination are delicate and vital issues [1]. Unfortunately, access to potable drinking water in most developing countries such as Nigeria is a major challenge hence most people rely heavily on untreated surface and ground water sources for consumption.

The inhabitants of Dutsin-Ma local Government Area of Katsina State rely solely on untreated groundwater sources (well and borehole) as well as surface water source. This is because there are only few available pipe born water sources and in most places where such sources are available they are not operational. The Zobe Dam located at Dutsin-Ma which is intended to remediate this problem is yet to be completed by the government [2].

Radon-222 is soluble in water and is a naturally occurring radioactive inert gas with a half-life of 3.82 days which is a member of the Uranium decay series [3]. It contributes the largest proportion of the total radiation from natural sources. Studies have shown that Radon-222 (^{222}Rn) and its progeny contribute about 50% of the total effective dose equivalent from natural sources. The concentration of ^{222}Rn in water is due to the decay of

Radium-226 associated with the rock and soil [4]. The radon gas penetrates through soil and rocks and dissolves in water [5]. Normally, drinking water from groundwater sources has higher concentration of radon than surface water [6 – 7].

The exposure of a population to high concentration of radon and its daughters for a long period has significant health effects ranging from respiratory functional changes to cancer of the lungs [8]. Also a very high level of radon in drinking water can lead to stomach and gastrointestinal tract cancer [9].

Access to potable source of water has remained one of the major challenges for most people as well as animals in Dutsin-Ma Local Government Area of Katsina State. As such, majority rely on untreated surface and ground water sources for consumption. The geology of Dutsin-Ma Local Government Area, Katsina State revealed that it is highly enriched in granites and gneisses and studies have shown that high activity concentration of radon-222 is associated with areas rich in granite [10 – 11]. As such, the level of radon in drinking water which in high concentration, can lead to a significant risk of stomach and gastrointestinal tract cancer [12], among others, need to be investigated. This is more so because our knowledge of its level of availability could be of great help in resource planning.

In order to create awareness to the world populace on the risk of drinking water with high concentration of radon and possibly proffer solution to this challenge, this area has been given adequate research backing from all over the world. In a work titled "Estimation of radon concentration in soil and groundwater samples of Northern Rajasthan, India" which measured radon concentrations using RAD7, an electronic Radon detector found the concentration values to range from 0.5 to 22 Bq/L[13].The results were found to be within the recommended level of (4-40) Bq/L set by [14]. In a similar manner, [15] worked on Radon Assessment in ground water sources from Zaria and environs, Nigeria and found that the radon concentrations ranged from 0.77 to 28.37Bq/L, 2.32 to 48.00 Bq/L with geometric means of 11.16 Bq/L and 12.43 Bq/L for open wells and Boreholes respectively which were above the maximum contaminant level of 11.1 Bq/L recommended by USEPA. A total of 64 water samples from Ado-Ekiti were also investigated using calibrated active electronic detector RAD7 connected to RAD H_2O accessory [16] and found that 53% of the results were above 11.1Bq/L MCL suggested by USEPA. Also in 2017, [17] conducted a research work which investigated the concentrations of Radon-222 in 84 water samples found the values to range from 2.18 to 76.75 Bq/L. The corresponding annual effective doses were also found to range from 0.036 to 1.261 mSv/y, 0.071 to 2.521mSv/y and 0.042 to 1.471 mSv/y for adult, children and infants respectively.

However, in Katsina State and Dutsin-Ma in particular there is no established information on Radon activity concentration from the survey of literature despite the fact that its geology revealed that it is highly rich in granite. Thus this present study is intended as a baseline study to ascertain whether Radon from water sources in Dutsin-Ma town possess a health risk to the populace or not.

Figure 1: Map of Katsina State showing Dutsin-Ma Local Government Area [20].

Materials and Method

Study Area

Dutsin-Ma Local Government Area of Katsina State lies between latitude 12^0 17.00N' to 12^0 17.84'N and longitude 007^0 26'E [18]. It is bounded by Kurfi and Charanchi L.G.A to the North, Kankia L.G.A to the East, Safana and Dan-Musa L.G.A to the West, and Matazu L.G.A to the South-East as shown in figure 1. Dutsin-Ma L.G.A has a land size of about 552.323km^2 with a population of about 169,829 as at 2006 national population census with the people being predominantly farmers, cattle rearers and traders [19].

Geology of the Study Area

Katsina State is predominantly underlain by the Basement Complex with 80% of its geology underlined by the Basement Complex of Nigeria which is characterized by Migmatite Gneiss, Metasediments and Old Granite known as Granite Suite. Dutsin-Ma (coined from the Hausa word 'Dutse' meaning rocks) Local Government as a Local Government under Katsina State is not of these features [21].

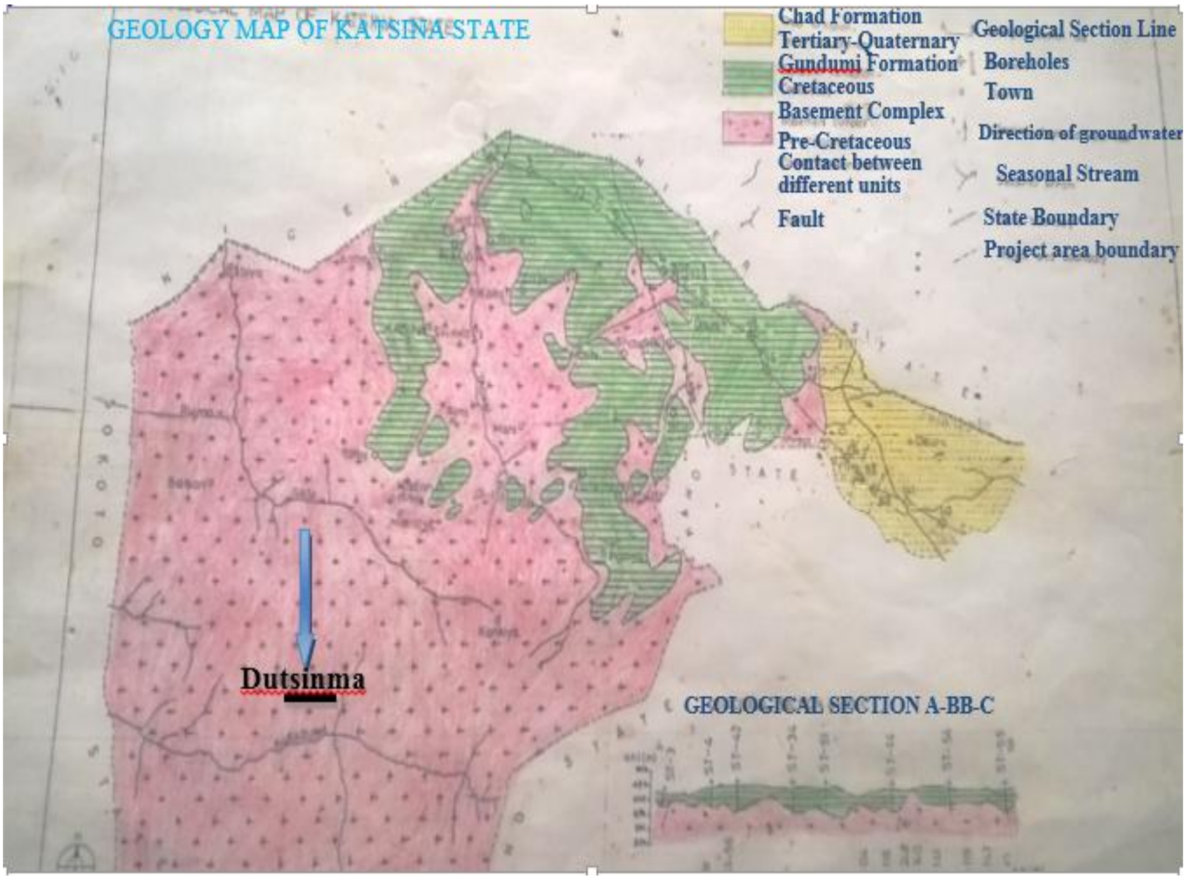


Figure 2: Geologic Map of the Northern Katsina State showing Dutsin-Ma LGA (adapted from GEO INVEST and BOREWELL NIG. LTD Katsina, Modified).

Materials and Reagents

Materials and reagents used in this research had been described by the American Society for Testing and Materials [22].They include; Disposable hypodermic syringe (20ml,10ml capacities), Surgical gloves, Scintillation vials (20ml capacity) with polyethylene inner seal cap liners, Scintillation cocktail, Distilled water, Indelible ink and Masking tape, and Liquid Scintillation Counter (Packard Tri-Card LSA 1000TR) [15]. Also a piece of clean cloth, Session 1 pH meter and Ethanol were used.

Sample Collection

A total of 15 samples were collected for analysis from the selected surface (Earth-dam) and underground (Boreholes and open wells) water sources at different locations in Dutsin-Ma town, Dutsin-Ma Local

Government Area, Katsina State. During sampling, a Global Positioning System (GPS) was used to mark the geographical locations on the earth surface of the sample collection points.

Samples were collected using a plastic vials. When collecting surface water samples, the vials were submerged in the water completely until filled and tightly capped before removing out of the water so as to avoid out gassing of radon-222 gas. Water from wells was first purged before collection of samples. Here samples were first collected with bailers and then transferred into vials. Also, boreholes were pumped and allowed to flow each for at least three (3) minutes before samples were collected in order to ensure that fresh samples were obtained. Each collected sample was properly labeled and the time of sample collection was noted and recorded.

Sample Preparation

10ml of each sample was added into a vial containing 10ml of toluene based cocktail (scintillator) using a hypodermic syringe. The vials were tightly capped and shaken vigorously for three (3) minutes to extract radon-222 in water phase into the organic scintillator. In a similar manner a blank sample for the background was prepared using distilled water that has been kept in a glass bottle for at least 21 days. The prepared samples were allowed to stand undisturbed for at least three (3) hours each in order for ^{222}Rn and its alpha decay products attain equilibrium before counting.

Sample Analysis

The prepared samples and the blank were each analyzed using the Liquid Scintillation Counter (Tri-Card LSA 1000) at the Center for Energy Research and Training (CERT), Ahmadu Bello University Zaria, Kaduna, Nigeria. Radiation emitted from the samples transferred energy to the organic scintillator which in turn emits light photons. This way each emission result is a pulse of light in form of digit.

The activity concentration of Radon-222 was calculated from the samples and background results obtained using the formula below:

$$C_{Rnw}(BqL^{-1}) = \frac{100\times(SC-BC)\times exp\lambda t}{60\times CF\times D} \quad (1)$$

where:

$C_{Rnw}(BqL^{-1})$ = Concentration of Radon − 222 in Becquerel per litre.

SC = Sample Count (Countmin^{-1}), BC Background Count ($Count\ min^{-1}$)

t = Time elapsed between sampling to counting (minutes), λ Decay constant ($1.26\times10^{-4}min^{-1}$).

100 = Conversion factor from per 10ml to per liter, 60 = Conversion factor from minutes to seconds ($second/munite$).

CF = Calibration factor

D = Fraction of ^{222}Rn in the cocktail in a 22 ml total capacity vial for 10ml of sample, 10 ml of cocktail and 2 ml of air.

The corresponding annual effective doses (mSv/y) due to ingestion of Radon-222 in water samples from Dutsin-Ma were also calculated using equation (2) by taking into account the dose coefficient in (Sv/Bq), the annual water consumption (L/Y) and the activity concentration of Radon-222 obtained from equation (1) using equation (2)

$$E = C_{RnW}\times D\times L \quad (2)$$

where:

C_{Rnw} = Concentration of Radon-222, D= Dose coefficient($10^{-8}Sv/Bq, 2\times10^{-8}Sv/Bq, 7\times10^{-8}Sv/Bq$) for adults, children and infants respectively [23].

L = Annual water consumption by an adult of 2 litres per day that is 730L/Y.

According to United Nation Scientific Committee on the Effect of Atomic Radiation [23], doses due to ingestion of radon in water for similar consumption rates could be factor of 2 and 7 higher for children and infants respectively.

Results and Discussion

The results of the analyzed samples are presented in table 1

Table 1: Results of Radon Concentrations in (Bq/L) and their corresponding Annual Effective Doses (AED) in (mSv/y) for Water Samples at Dutsin-Ma town.

S/N	Sample Location	Sample ID	Latitude	Longitude	PH	Elevation (m)	Rn (Bq/L)	AED (mSv/y)		
								Adult	Children	Infant
1	Kadangaru	DB1	12°27'56.6"	07°29' 22.1"	7.81	535	142.1	1.104	2.208	7.728
2	Hayin Gada	DB2	12° 27'25.9"	07°30'17.6"	7.42	527	152.8	1.115	2.23	7.805
3	Yan Dakka	DB3	12°27'5.6"	07°30'4.7"	7.8	529	58.11	0.424	0.848	2.968
4	Bayan Yarima	DB4	12°26'42.6"	07°29'57.5"	7.98	526	21.36	0.156	0.312	1.092
5	Low Coast	DB5	12°26'39.8"	07°29'49.4"	6.68	531	35.28	0.261	0.522	1.827
6	Isa Kaita College	DB6	12°26'20"	07°29'18.4"	6.46	528	27.78	0.203	0.406	1.421
7	Sokoto Rima	DB7	12°26'01.7"	07°28'06.8"	6.71	511	68.11	0.497	0.994	3.479
8	Kandahar	DB8	12°26'34.0"	07°28'48.35"	6.52	527	11.67	0.085	0.17	0.595
9	Kadangaru	DW1	12°27'31.8"	07°29'31.8"	7.64	532	172.3	1.257	2.514	8.799
10	Hospital Road	DW2	12°27'45.6"	07°29'51.2"	6.38	530	13.33	0.097	0.194	0.679
11	Hayin Gada	DW3	12°27'46.7"	07°30'14.2"	6.4	530	11.84	0.086	0.172	0.602
12	Bayan Yarima	DW4	12°27'48.9"	07°29'51.6"	6.68	535	0.61	0.045	0.09	0.315
13	Low Coast	DW5	12°26'33.5"	07°29'37.2"	7.81	534	7.72	0.056	0.112	0.392
14	Earth- Dam	DD1	12°27'47.6"	07°29'52.5"	6.25	533	21.98	0.161	0.322	1.127
15	Earth- Dam	DD2	12°27'48.3"	07°30'03.0"	6.14	530	47.17	0.344	0.688	2.408

Discussion

The mean values of Radon-222 concentrations were found to be 64.66, 41.15 and 34.57 Bq/L for boreholes; wells and surface water (Earth-Dam) respectively (table 1). Also the corresponding Annual Effective Doses due to ingestion of Radon-222 in Borehole water were found to be (0.472, 0.944 and 3.304) mSv/y for adults, children and infants respectively. While that due to the intake of Radon-222 from open wells and surface water (Earth-dam) were found to be (0.308, 0.616, and 2.156) mSv/y and (0.252, 0.504 and 1.764) mSv/y, for adults, children and infants respectively. These results have shown that both the mean values of Radon-222 concentrations and the Annual Effective Doses due to ingestion of borehole, well and surface water samples exceeded the world average recommended values of 10 Bq/L and 0.1 mSv/y set by the World Health Organization, [24 – 25] for Radon concentration and annual effective dose due to intake of radionuclide in water respectively.

Borehole Water

The result of the analysis of radon concentrations for the eight (8) borehole water samples collected at different locations of Dutsin-Ma town, Dutsin-Ma Local Government Area, Katsina State as presented in table 1 revealed that the concentrations of Radon-222 varied from 11.67 Bq/L to 152.81 Bq/L with mean value of 64.66 Bq/L. The maximum concentration was obtained from Hayin Gada while the minimum concentration a was found at Kandahar as shown in figure 3. 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 [24]. The mean concentration of Radon-222 was found to be lower than the recommended guideline level of 100 Bq/L set by WHO [26]. However, 25% of the analyzed borehole water samples which include samples from Kadangaru and Hayin Gada were found to be above 100 Bq/L . These high concentrations of Radon-222 in Kadangaru and Hayin Gada water samples which can pose a threat on the health of the inhabitant 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.

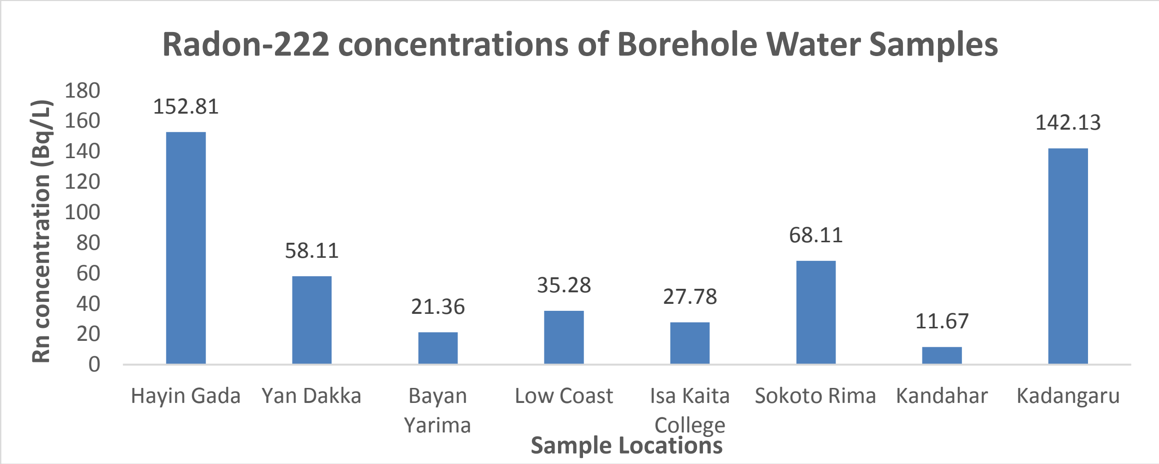


Figure 3: Bar Graph of Radon-222 Concentrations for Borehole Water Samples

Well Water

The concentrations of Radon-222 well in water samples collected from five different wells located at the following areas in Dutsin-Ma town: Kadangaru, Hayin Gada, Bayan Yarima, Hospital road and Low Cost as presented in table 1 were found to range from (0.61 to 172.25 Bq/L) with a mean value of 41.15 Bq/L. Kadangaru recorded the maximum radon concentration of 172.25 Bq/L which exceeded the recommended reference level for action of 100 Bq/L set by WHO [26]. While Bayan Yarima recorded the minimum radon concentration of 0.61 Bq/L as shown in figure 4. Samples from Kadangaru, Hospital Road and Hayin Gada have radon concentrations above the maximum contaminant level (MCL) of 11.1 Bq/L proposed by United States Environmental Protection Agency [27] and world average value of 10 Bq/L set byWHO [24] as such, they pose cancer. However, Radon concentrations of samples obtained from Low Cost and Bayan Yarima were found to be below the Maximum contaminant levels of 10 Bq/L and 11.1 Bq/L World Health Organization and set by United States Environmental Protection Agency [27] respectively.

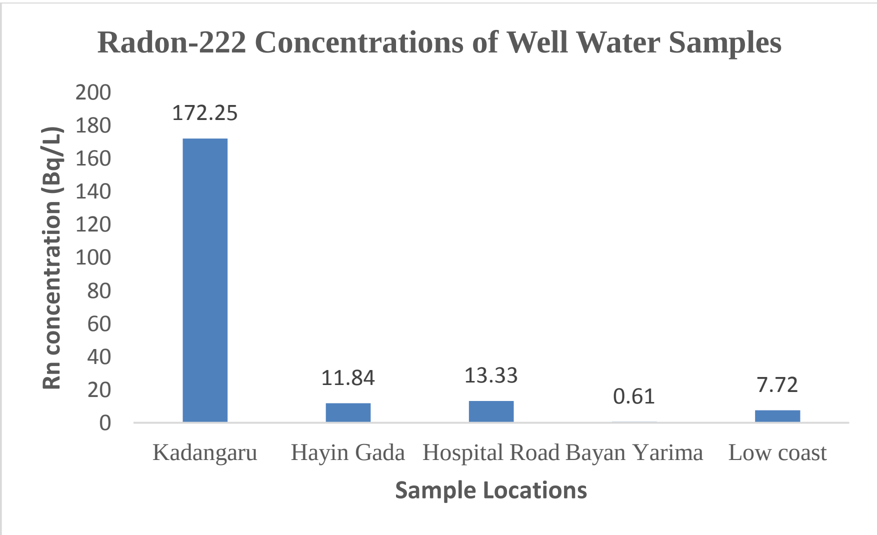


Figure 4: Bar Graph of Radon-222 Concentrations for Well Water Samples

Surface Water

The results obtained from the analysis of the two (2) samples collected at different locations from Dutsin-Ma Earth-dam revealed the maximum concentration of radon in the water as 47.17 Bq/L while the minimum concentration was found to be 21.98 Bq/L with an average value of 34.57 Bq/L as presented in table 1. All the values obtained from the surface water (Dutsin-Ma Earth dam) exceeded the maximum contaminant level of 10 Bq/L set by WHO [24] and 11.1Bq/L set by USEPA [27]. These higher values of radon in surface water could be linked to the granitic rocks used in the construction of the dam bank as well as the soil type. Environmental waste flooded into the dam during heavy rainfall might also be a contributing factor.

Mean Radon Concentration

In order to compare the level of radon in the two categories of water sources: groundwater (wells and boreholes) and surface water (earth-dam) collected from the study area, the mean concentration of each of the water types were calculated and plotted as shown figure 5.

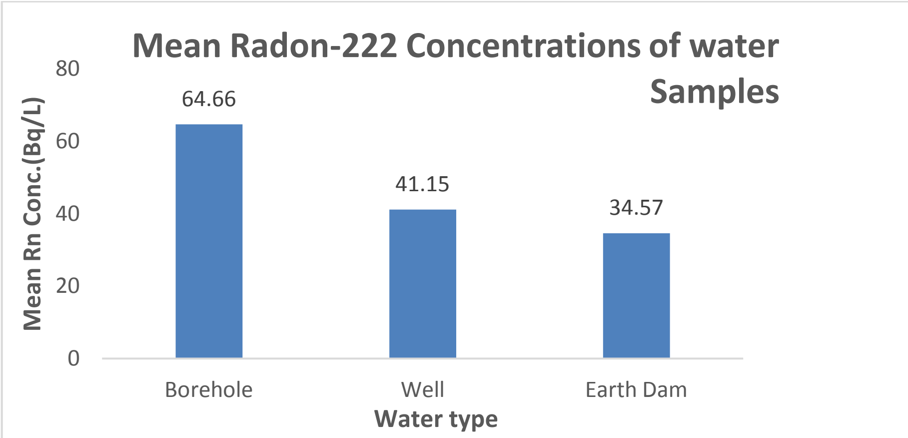
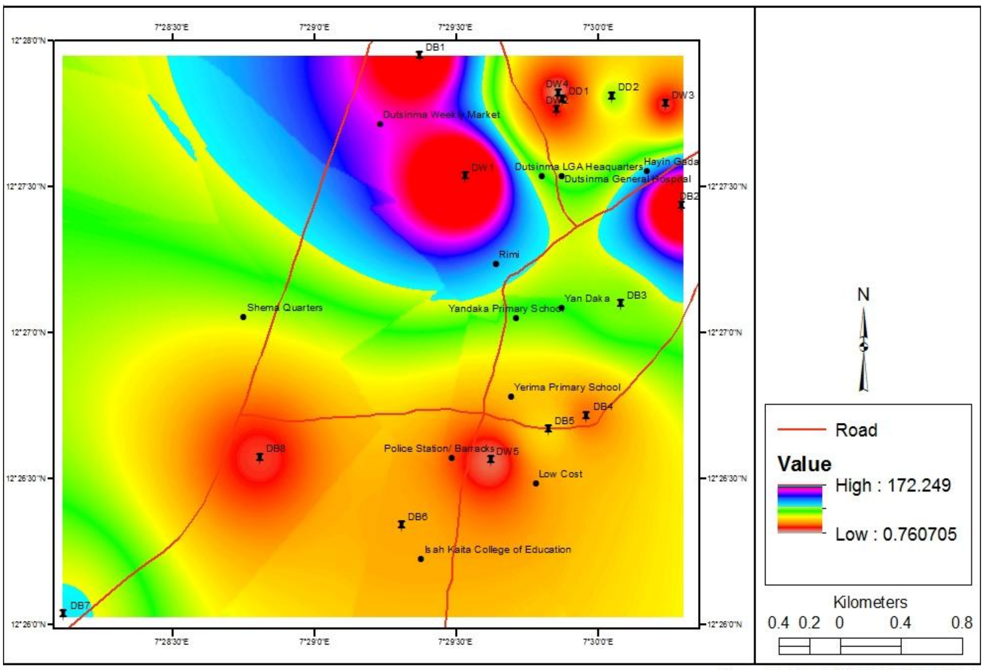


Figure 5: Bar Graph of Mean Radon-222 Concentrations for the three Water Types

Samples from borehole have the highest mean concentration of Radon-222 followed by well water samples while the surface water (earth-dam) has the least mean concentration of radon. All the mean values from the three (3) water types exceeded the maximum contaminant levels set by USEPA [27] and the world average value of 10 Bq/L set byWHO [24]. However, the lower mean value of Radon-222 concentration for surface water as compared to mean radon concentrations of Borehole and well water could be due to the fact that surface water is exposed to the air and since radon is a gas it tends to escape into the air. While the higher mean values recorded for boreholes and wells water may be attributed to the fact that radon readily dissolves in water under pressure which lead to the radon accumulation in groundwater as can be found in [28].



Source: Field work 2017 & FUDMA GIS Lab

Figure 6: Mapping of the Distribution of Radon Concentration in some Selected Settlement ofDutsin-Ma town, Dutsin-Ma Local Government Area, Katsina State.

Distribution of Radon-222 Concentrations

The distribution of Radon-222 concentrations of all the analyzed water samples collected at various locations of Dutsin-Ma town, Dutsin-Ma Local Government Area, Katsina State, as illustrated in figure 6 revealed locations of higher concentrations than normal and those of lower concentrations as shown in table 1 and the various bar charts (figures 3 and 4).

Annual Effective Doses (AED)

The corresponding Annual Effective Doses due to intake of Radon-222 from borehole water samples collected at Dutsin-Ma town, Dutsin-Ma Local Government Area, Katsina State were estimated for the same water consumption rate using equation 2 and found to range from (0.085 to 1.115) mSv/y, (0.17 to 2.23) mSv/y and (0.595 to 7.805) mSv/y as shown in table 1 with corresponding mean values of 0.472, 0.944 and 3.304 mSv/y for adults, children and infants respectively. The estimated annual effective doses due ingestion of Radon-222 from Well water samples were found to range from (0.045 to 1.257) mSv/y, (0.09 to 2.514) mSv/y and (0.315 to 8.797) mSv/y with corresponding mean values of 0.308, 0.616, and 2.156 mSv/y for adults, children and infants respectively. In a similar manner, the annual effective doses due to ingestion of Radon-222 from surface water (Earth-dam) were estimated and found to range from (0.161 to 0.344) mSv/y, (0.322 to 0.688) mSv/y and (1.127 to 2.408) mSv/y with corresponding mean values of 0.252, 0.504 and 1.764 mSv/y for adults, children and infants respectively. 66.67% of the estimated Annual Effective doses were found to be above the recommended reference level of 0.1 mSv/y for intake of radionuclide in water set by WHO [25]as presented in figure 7. These higher values of annual effective doses showed that most of the water sample from the study area could be a threat on the health of the inhabitant of the area if taken directly without proper treatment.

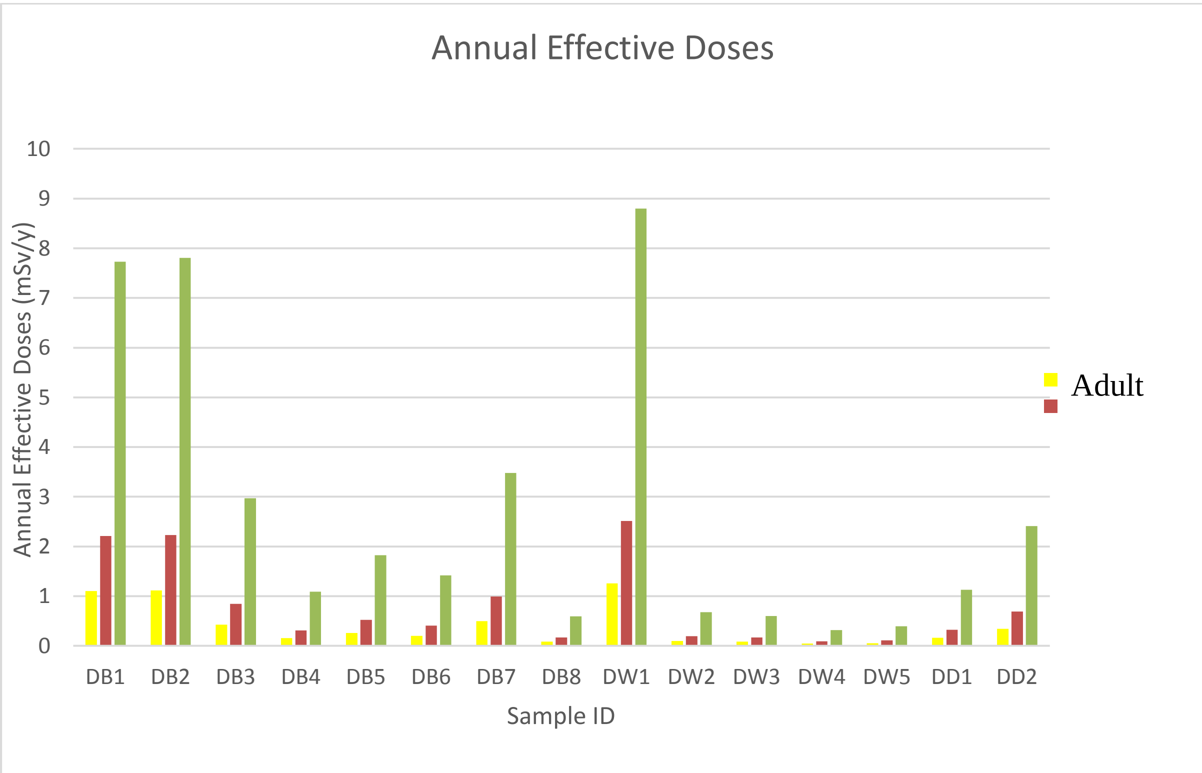


Figure 7: Bar Graph of the Annual Effective Doses for all Water Samples

Mean Annual Effective Dose

The mean Annual Effective Doses due to ingestion of Radon-222 from the three (3) water types were calculated for the three (3) categories of people and found to be 0.472, 0.944 and 3.304 mSv/y in borehole water for adults, children and infant respectively. While mean annual effective doses due ingestion of Radon-222 in both well and surface water samples were found to be 0.308, 0.616, and 2.156 mSv/y and 0.252, 0.504 and 1.764

mSv/y for adults, children and infants respectively. All the mean annual effective doses were found to be above World Health Organization recommended reference level of 0.1 mSv/y for ingestion of radionuclide in water [25]. These results revealed that infants, for the same water consumption rate receive significantly higher doses hence have higher risk to cancer compared to children and adults as illustrated in figure 8 with higher values of the mean annual effective doses for the three (3) categories of people coming from borehole water sources.

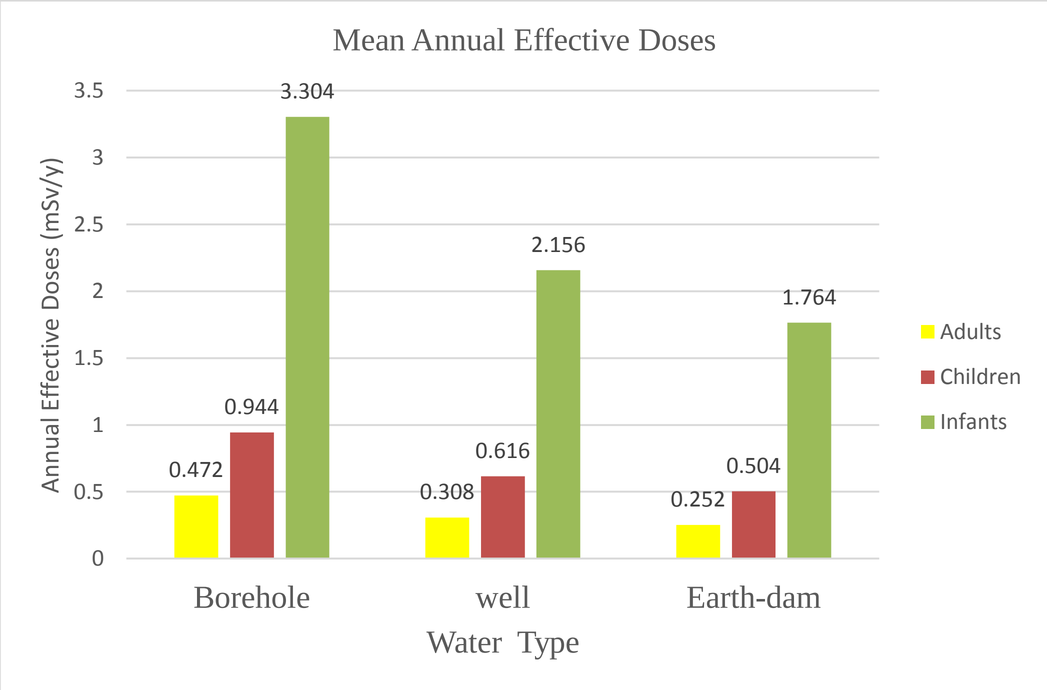


Figure 8: Bar Graph of Mean Annual Effective Doses for the three Water Types

Conclusion

Activity concentrations of radon-222 were calculated from annual effective doses due to ingestion of radon-222 in water samples from Dutsin-Ma town. The annual effective doses were obtained from the analysis of water samples using the Liquid Scintillation Counter (Tri-Card LSA 1000).

Results obtained from the measurement of the activity concentrations of Radon-222 in water samples collected at different locations of Dutsin-Ma town, Dutsin-Ma Local Government Area, Katsina State revealed that 86.67% of the recorded values of radon concentrations in the present study as well as the mean values obtained for the three (3) water types 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 [27]. 20% of the recorded values of radon concentration which include well and borehole wate@&r samples from Kadangaru, and borehole water sample from Hayin gada also exceeded the recommended action level of 100 Bq/L set by the World Health Organization [26]. These significantly high values of radon concentration can be ascribed to the nature of the basement rock and soil type in the study area for groundwater sources while that in the surface water (Earth-dam) could be due to the nature of the granitic rocks used in the construction of the dam bank, the soil type, as well as waste materials flooded into the surface water during raining season. Therefore these water sources pose a threat to the health of the inhabitant of the study area if continually ingested without proper treatment. The likelihood of this threat to health (which could be stomach or lung cancer) is more on infants and children than adults 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 [25] for intake of radionuclide in water.

Considering the above results, it is recommended that, the inhabitants of the study area particularly in locations where concentrations of Radon-222 were found to be higher than normal should boil their water before consumption so as to keep their exposure due to ingestion of Radon-222 as low as reasonably achievable. Epidemiological studies of the general population to determine lung and stomach cancer incidence is strongly recommended.

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