FUDMA Journal of Sciences (FJS)
ISSN online: 2616-1370
ISSN print: 2645 - 2944
Vol. 4 No. 2, June, 2020, pp 615 – 619
DOI: https://doi.org/10.33003/fjs-2020-0402-172

ASSESSMENT OF ANNUAL EFFECTIVE DOSE DUE TO INHALATION AND INGESTION OF RADON IN WATER SAMPLES FROM THE CEMENT INDUSTRIAL AREA OF SOKOTO, NORTH-WESTERN NIGERIA

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ABSTRACT

Water remains the most abundant and critical commodity for guaranteeing the continuity of human lives on earth. Ensuring cleanliness of water for human consumption is of paramount importance. The 222Rn concentration has been assessed in drinking water samples collected from various water resources used by communities around Sokoto Cement Company, with the view of assessing the radiological risk, if any, to human health. The sources of collected water samples were hand pumps and hand dug wells and water seepages. Determination of radon concentration was conducted using liquid scintillation counter (Model: Tri-Carb-LSA1000) following standard procedures. The overall mean value of 222Rn concentration was found to be 34±3.7 Bq/L The resulting mean annual effective doses due to inhalation of radon in the water samples was 41 µSvy-1, while ingestion for adults, children and infants were 248±27 µSvy-1, 372±40 µSvy-1 and 434±47 µSvy-1 respectively. These values are above the recommended benchmarks prescribed by UNSCEAR, WHO, European commission and USEPA guiding the utilization of water for drinking and domestic purposes. This indicates that the water resources around the cement company are not safe for drinking and domestic purposes from the radiological point of view.

Keywords: Radon concentration, Inhalation dose, Ingestion dose, Radiological risk, Sokoto Cement Company

INTRODUCTION

Radiation in the environment originates from a number of naturally occurring and human‐made sources while exposure from it can occur via ingestion, inhalation, injection, or absorption of radioactive materials (Kaur et al., 2017; Malakootian1, 2017; Nejhad, Y. S., 2017). Radiological hazards may be possible due to the presence of large content of radioactive substances in drinking water, Garba et al., (2013). The most common radionuclide of interest in drinking water is radon (^{222}Rn). Radon is a radioisotope of alpha emission from ^{238}U decay chain with a very short half-life of 3.82days. ^{222}Rn have the highest density 9.37 gl^{-1} among other inert gases and solubility of 510cm^{3}l^{-1} among all noble gases. It has been identified that radon is the main source of natural radioactivity in residential areas with short-lived products of ^{214}Po, ^{214}Bi, ^{214}Pb and ^{218}Po, (Bunger and Ruhle, 1994; Bello *et al*., 2020). Radon gas has the largest amount of total annual effective dose to human and this makes it of special attention among all other naturally occurring radioactive minerals in environmental radiological studies, UNSCEAR, (2008). Mining activities such as those from activities of Cement Company in a given area tends to proportionally aggravate this annual effective doses of both surface and underground water resources, UNSCEAR, (2000). The effect of ^{222}Rn in water on human health is majorly through the exposure routes of inhalation and ingestion via breathing and drinking, which leads to lung and gastrointestinal cancers respectively. According to World Health Organization, exposure to ^{222}Rn in water is the second most important cause of lung cancer after smoking and the majority of radon‐induced lung cancers are caused by low and moderate radon concentrations rather than by high indoor radon concentrations WHO, (2009). It has been measured in water in many parts of the world for the last two decades, mostly for the risk assessments due to consumption of drinking water (Ali et. al., 2010; Ben et. al., 2014; Kumar 2016; Kumar 2016). Populace that are exposed to reasonably high levels of ^{222}Rn in drinking water for long periods may develop serious health problems such as stomach and gastrointestinal cancer, lung cancer, anaemia, osteoporosis, cataracts, bone growths, kidney diseases, liver disease and impaired immune Aruwa et al., (2017).

Sokoto cement company, has been in operation since 1967, with communities of Arkilla, Kalambaina, Gidan Gamba and Gidan Belu neighbouring most immediately around the processing plant and quarry site of the company These communities rely predominantly on hand dug wells, boreholes and water seepages from the quarry as their sources of water for domestic and agricultural activities. Radiological characterization of the area is yet to be done despite the perennial anthropogenic activities of the company in the area for over a half century. For this reason, the authors conducted terrestrial gamma radiation survey, an assessment of the activity concentrations of the primordial radionuclides of ^{226}Ra, ^{232}Th, and ^{40}K in soil and associated radiological variables, as well as pollution assessment level in terms of heavy metals using various statistical models of the area. To the best of our knowledge, this research was not conducted in the area for all this while. This may however, be attributed to inadequacies in terms of capacity in terms of techniques, facilities and human resources. This research work is therefore going to be a major contribution to the determination of the level and possible health effects of radioactivity in the area under study and can help in devising efforts by the relevant authorities in ensuring best practice.

MATERIALS AND METHODS

Location of the study area.

Cement Company of Northern Nigeria (CCNN) now known as Sokoto Cement Company was founded by the then Northern Region Government in 1960. It was incorporated in 1962 and commenced production in 1967 with initial installed capacity of 100,000 tons per annum at the Kalambaina plant about 10km westward of Sokoto city, under Sokoto basin. The Basin lies in north-western Nigeria, Yelwa et al., (2015) between latitudes 10°20' and 14°00' N and longitudes 3°30'and 6°58' E (Figure 1). Sokoto Basin is the southern extension of the Iullemmeden Basin, a sedimentary basin which also contains the uranium producing Agades sandstones of the Niger Republic, Obaje, (2009). The area is entirely a cratonic basin created by tectonic epirogenic movements or stretching and rifting of tectonically stabilized crust during the Palaeozoic. These movements become evident from the beginning of Palaeozoic and continued until the Upper Cretaceous when the opening of the Goa Trench was achieved, Yelwa et al. (2015). It falls within a region where rainfall distribution is irregular in time and space and characterized by a prolonged dry season with a short rainy season, Sa'idu et al., (2012).

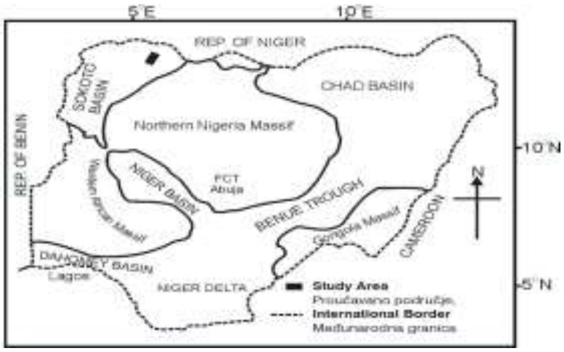

Figure 1: Map of Nigeria showing the study area.

Sample Collection

Water samples were collected from available water sources in the study area. Water samples was collected into the labelled two litres (2 L) plastic bottle. Water samples from seepages were collected with the aid of bailer, the ones from open dug wells were collected after repeated evacuation of the content on the bulk surface to ensure fresh samples while the samples from boreholes were also collected after allowing the remaining water in the pipe to be plug out and then collect the fresh sample. The bottles were first washed clean water and filled to the brim without any head space to prevent CO_2 from being trapped and dissolving in water which might affect the chemical content. Concentrated Trioxonitrate (V) acid HNO_3 was added to the water to ensure radionuclides remain in solution rather than adhering to the walls of the container, Avwiri, (2005). Total of ten (10) water samples was collected based on availability from Arkilla, Kalmbaina, Gidan Belu and Gidan Gamba communities as well as the quarry site. Two (2) samples were collected from each of the locations mentioned. Sampling points are located using Hand held GPS (Model:78s) and presented in Table 1. All samples are transported to the Center for Energy Research and Training, Ahmadu Bello University Zaria (CERT) for preparations and analysis.

Table1: Sample codes and sampling point coordinates.

S/N	Sample Code	Sample Location and Type	Latitude	Longitude
1.	ARB	Arkilla Borehole water	13°02'02"	05°12'0.9"
2.	ARW	Arkilla open-well water	13°01'33"	05°11'52"
3.	GBW	Gidan Belu open-well water	13°03'38"	05°10'30"
4.	GBW	Gidan Belu open-well water	13°03'18"	05°10'34"
5.	GGW	Gidan Gamba open-well water	13°03'17"	05°10'38"
6.	GGW	Gidan Gamba open-well water	13°03'40"	05°10'30"
7.	KLW	Kalambaina open-well water	13°02'36"	05°11'37"
8.	KLB	Kalambaina Borehole water	13°02'19"	05°11'22"
9.	QSW	Quarry seepages water	13°02'42"	05°10'58"
10.	QSW	Quarry seepages water	13°02'44"	05°10'51"

Sample preparation

The sample preparation procedures reported by Bunger and

Ruhle, (1994) were used for the sample preparation. 150mL plastic sample bottles were used for sample collection of both ground from the community areas and surface water from the quarry seepages. The bottles are filled to the brim and hermetically closed before been transported to Centre for energy research and training, ABU Zaria for analysis. The liquid scintillation laboratory at Centre for energy research and training, ABU Zaria had vials that were ready prepared containing 10 ml of liquid scintillation cocktail (toluene). 10mL disposable syringe was carefully used in sucking each sample from its sample bottle and transferred it into the 20mL vial that already held scintillation cocktail from the bottom. The vial containing the content was shaken vigorously for the Radon-222 to be extracted from the water phase to the organic scintillates due to its greater solubility in organic liquids for stand. The content was kept for three hours before counting.

Determination of radon concentrations in water

The counting procedure started by shaking the vials that are already prepared for counting and carefully wiping the outside of each vial with a cloth dampened with ethanol and leave them aside for a minimum of 3 hours to allow for in growth of the short-lived decay products of Radon-222. This is for Radon-222 and its short-lived daughters to attain equilibrium, (Garba, 2011; Garba et al., 2013; ASTM, 1999; Suomela, 1993). The counting vials were placed in the liquid scintillation counter (Model: Tri-Carb-LSA1000) of Center for Energy Research and Training (CERT), Ahmadu Bello University, Zaria, Nigeria. According to the manufacturers Tri-Carb-LSA1000 is equipped with alpha/beta discrimination features and can achieve a detection limit of 0.407 Bql^{-1} or less in 60 min. Each vial was counted for a preset period of time using a calibrated IAEA ^{226}Ra standard solutions (IAEA-423, IAEA-431 and IAEA-427). It was ensured that, when transferring vials from storage to the counter after the 3 hour in growth, they are not shaken as this will greatly disturb die state of equilibrium between radon-222 and its short-lived daughters in the organic scintillate, Bunger and Ruhle, (1994). Background of the counting system was determined by counting a vial with 10 ml of the organic scintillant solution and 10 ml of deionized water. The time and date at which counting commences were noted. The background, calibration and sample solutions were measured over the same spectral range and for the same counting period of 60minutes.The background and sample count rate (counts.min^{-1}) were recorded. ^{222}Rn and its short-lived daughters emit a total of 5 radioactive particles (3 α and 2 β) per every disintegration of ^{222}Rn. Since, secular equilibrium was established between ^{222}Rn and these daughters, all the 5 emissions were used to detect and quantify ^{222}Rn in water. ^{222}Rn activity concentrations was evaluated by considering sample volume, total and background count rates, decay time (time between sample collection and counting), and efficiency of detection. The ^{222}Rn concentration in a sample of water was determined through equation 1, Bello et. al., (2020).

$$C_S = \frac{100 \times \left(R_g - R_o\right) \times \exp(\lambda.\Delta t)}{60 \times 5 \times 0.964} - - - - - - - - - (1)$$

where C_S is ^{222}Rn concentration at the time of sample collection (Bq/L); R_g is the sample total count rate (count min.$^{-1}$); R_o is the background count rate (count min^{-1}); t is the elapsed time between sample collection and counting ((4320 min. (3days)); λ is ^{222}Rn decay factor (1.26 × 10^{-4} min.$^{-1}$); 100 is a conversion factor from per 10 ml to per liter (l^{-1}); 5 is the number of emissions per count; 60 is conversion factor from min. to s. (s. min^{-1}) and 0.964 is the fraction of ^{222}Rn in the cocktail in a vial of 22 ml total capacity, assuming it contains 10 ml cocktail, 10 ml water and 2 ml air.

Estimation of effective dose due to radon in drinking water

The dose due to radon can be divided into two parts, first is dose from ingestion (drinking water containing radon) and second is the dose from inhalation. The annual mean effective dose for ingestion and inhalation were calculated according to UNSCEAR report (UNSCEAR 2008)
equations 2 and 3 respectively.

$$E_{ing}(\mu Sv\,y^{-1}) = Rn\,^{222}conc.\,(Bq\,l^{-1}) \times W_{in} \times DCF_{ing} \quad - - - - (2)$$

$$E_{inh}(\mu Sv\,y^{-1}) = Rn\,^{222}conc.\,(Bq\,l^{-1}) \times R_{aw} \times F \times O \times DCF_{inh} - (3)$$

where, E$_{ing}$ ($\mu Sv\,y^{-1}$) is the effective dose from ingestion, W_{in} water ingestion rate (730, 547.5 and 182.5 $l\,yr^{-1}$ for adults, children and infants respectively), DCF_{ing} is the ingestion dose conversion factor (3.5 × $10^{-3}\mu Sv\,Bq^{-1}$), $E_{ing}(\mu Sv\,y^{-1})$ is the effective dose from inhalation of radon released from water into air, $Rn\,^{222}conc.\,(Bq\,l^{-1})$ is the radon concentration in water, R_{aw} is the ratio of radon released to air when water is used to radon in water (10^{-4}), O is the average indoor occupancy time per (7000 hy^{-1}), F is the equilibrium factor between radon and its products (0.4), DCF_{inh} is the conversion dose factor ($9n\,Sv\,h^{-1}(Bq\,m^{-3})^{-1}$).

RESULTS AND DISCUSSION

Table 2 presents the radon specific activity (Bq/L) and resulting annual effective doses of the water samples used for drinking and domestic purposes in the study area. The overall mean radon activity concentration was found to be 34±3.7 Bq/L value three times the maximum permissible limit by European Commission and United States Environmental Protection Agency (E. C., 1998; USEPA, 2003) of 11.1 Bql^{-1} and 10 Bql^{-1}. The radon activity concentrations from all the samples collected were evaluated through equation 1 with overall range of 28.6-39.3 Bql^{-1}. The lowest and the highest ranges from the ground (open-well) water of Kalmbaina and Gidan Belu respectively. The values of radon concentration in water increases as one move from Gidan Belu, a town closest to the cement company and to Kalambaina the distance town to the cement company. Regardless of the source, all of the 10 water samples analyzed including the control samples had radon activity concentrations above 11.1Bql^{-1} maximum permissible limit by (USEPA, 2003). The lower values recorded but yet above permissible limit include water of open hand dug wells of Kalambaina, and Gidan Gamba as well as Quarry seepages.

This observation may be attributed to continuous aeration and diffusional loses to the atmosphere (USEPA, 2003). Generally, water from ground sources in the study area that recorded higher values are consistent with the reported results in Nigeria (Bello *et al*., 2020; Aruwa *et al.,* 2017) and beyond (Oni *et al*., 2014: Binesh *et al.,* 2010). However, values recorded for Quarry seepages, 35.8-33.3 Bql^{-1} from this study are variance with similar reported researches that indicated a very low radon concentration of Rn-222 because of the diffusional losses and perennial aeration to the atmosphere (Bello *et al*., 2020; Oni *et al*., 2014: Binesh *et al.,* 2012). This unusual observation may not be unrelated to continuous anthropogenic activities of mining limestone from the quarry for cement production from where seepages are formed (Olise *et al*., 2016).

The overall mean annual effective dose due to inhalation of radon in water has been 85.7±9.3 μSvy^{1} with the range of 71.9-99.1μSvy^{-1} having lowest and highest from Kalambaina and Gidan Belu respectively. Also, the overall mean annual effective doses due to ingestion of radon in water for adults, children and infants were 248.2±27 μSvy^{-1}, 372.4±40 and 434.4±47 respectively.

Moreover, the mean annual effective doses of all locations due to ingestion considered in this study including the control were found doubled the recommended limit of 100 μSvy^{-1} and 200 μSvy^{-1} for adult and children respectively. (WHO 2003: E. C., 1998). These high values are in agreement with results from similar researches (Bello *et al*., 2020; Olise *et al*., 2016; Binesh *et al.,* 2010) but still below other reported results (Przylibski *et. al.,* 2014; Pareira *et. al.,* 2015; Skeppstram *et. al.,* 2006). Meanwhile, results recorded from this study indicated serious radiological hazard due to Rn-222 concentration in water uses for drinking and other domestic purposes in the area.

Table 4: Radon concentration in water samples and their annual effective doses for studied area.

		Radon activity concentration (Bql^{-1})	Annual effective dose effective from Inhalation and Ingestion (μSvy^{-1})			
Sampling locations	Statistical factors	Rn-222 Conc.	E_{inh}	E_{ing} (Adults)	E_{ing} (Children)	E_{ing} (Infants)
Arkilla	Range	35-37	89-94	256-273	387-410	451-478
	Mean±S.D	36 ±1.5	92±3.7	265 ±11	398±16	465±19
Gidan Belu	Range	37-39	94-99	272-287	409-431	477-502
	Mean±S.D	38±1.4	97±3.6	280±10	420±16	490±18
Gidan Gamba	Range	33-29	84-74	216-243	325-366	379-426
	Mean ±SD	32±2.6	79 ±6.7	230±19	345±29	403±34
Kalambaina	Range	30-28	71-75	208-218	313-328	365-382
	Mean ±SD	29±1	74±2.4	213±07	320±11	373±12
Quarry seepages	Range	33-36	84-90	243-261	365-392	426-457
	Mean ±SD	35±1.7	87±4.3	252±13	379±19	442±22
	Overall Mean	34±3.7	41	248.2±27	372±40	434±47

CONCLUSION

This research determines the radon activity concentration and its corresponding annual effective doses in the samples from the available water resources used for drinking and domestic purposes by the communities living around cement processing facility of Sokoto. This is with the view to establish the radiological safety level, in terms of radon, of utilizing the water for the said purposes. The values of overall mean of radon levels in and the subsequent ingestion annual effective doses in adult, children and infants in all the water samples, were above the limit recommended by UNSCEAR, WHO and European Commission. Hence the available drinking water resources of the studied area are not safe in relation to the possibility of acquiring hazard due to radon.

ACKNOWLEDGMENTS

The authors thank the management and entire staff of Center for Energy Research and Training, Ahmadu Bello University, Zaria for making the laboratory facilities available for undertaking this research. The corresponding, a current PhD student, acknowledge the erudite effort of his supervisory team throughout this research. I will also like to appreciate the management of my employer, Katsina State College of Nursing and Midwifery for their support and understanding while I'm undertaking this study.

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