Determination of concentrations and Annual Effective Dose of Pb, Cr, Rn in Groundwater Sources in Shika and Zaria City, Kaduna State, Nigeria

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ABSTRACT: The levels of some physicochemical parameters, heavy metals and 222Rn in some underground water of Shika and Zaria, Nigeria are reported. The Cr in the hand-dug wells and borehole water was in the range of 0.008 ± 0.006 mg/L to 0.338±0.39 mg/L for the rainy, and 0.09±0.13 mg/L to 0.66 ± 0.06 mg/L for the dry season; while Pb ranged from 0.001 ± 0.00 mg/L to 0.86±0.569 mg/L for rainy season in Shika and Zaria. The 222Rn concentration ranged from 0.1 ± 0.0141 Bq/L to 66.56Bq/L for rainy and 1.5 ± 0.0141Bq/L to 71.53 ± 0.007 Bq/L for dry season. The borehole samples have higher 222Rn than the permissible limit and most physicochemical parameters of Shika water were within the safe limit. The highest annual effective dose was 14.30 μSv/year and 12.0 µSv/year in the rainy and dry seasons respectively. There was significant difference between the parameters across the season. Therefore, caution should be taken to avoid the elevation of these elements in water bodies as a result of anthropogenic activities.

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African countries have a long history of using surface and groundwater sources. Deterioration in the quality and quantity of surface water and public water supply system, in many developing countries have led to dependence on groundwater sources (Okonko et al., 2007). Water shortage or its pollution can cause severe decrease in productivity and deaths of living species (Galadima and Garba, 2008). Water shortage or its pollution can cause severe decrease in productivity and deaths of living species (Galadima and Garba, 2012).

Heavy metals exist in water in colloidal particulate and dissolved phases (Adepoju-Bello et al., 2009). They also exist in water bodies from natural origin such as eroded minerals, leaching of ore deposits and volcanic extruded products (Igwilon et al., 2006).

Rock, soil and water contain 238U, 232Th and their decay products. Since the late 1980s, radon has been identified to be of health concern, causing lung cancer among others. Radon is a radioactive gas, formed when uranium or radium decays. It escapes from the earth's crust through cracks and crevices in the bedrock and either dissolves in ground water or seeps through foundation cracks into the environment/human habitations (Shakir et al., 2011). So, the quality of groundwater is affected by the characteristics of the media through which the water passes on its way to the ground water zone of saturation (Adeyemini et al., 2007). Radiation from natural sources account for the majority of human exposure to radiation, with radon decay product being the largest contributor (Tso and Li, 1987; UNSCEAR, 2000). Adelekan and Abegunde (2011) reported Cr in hand-dug wells near automobile mechanic villages in Ibadan, Nigeria to be lower than the limits set by WHO for drinking water. Also, the levels of Pb in selected underground water have been assessed by various studies (Ekumemogbo et al., 2011). The work of Emad, (2014) showed that groundwater samples in Jordan Valley contained radon-222 in the range of 0.28±1.5 to 44.31±3.2 Bq L⁻¹ by using RAD7 instrument. In addition, well water was reported to have the highest radon content compared to reverse osmosis (drinking water), mineral water and tap water by Abdul Malik (2015). The well water recorded Ra above the maximum contamination limit of 300 pCi/L recommended by United State Environmental Protection Agency. The aim of this work is to determine the radon concentration in groundwater (hand dug well and borehole) in Shika and Zaria city, as well as the lead and chromium concentration in the water samples.

MATERIALS AND METHODS

Site Description: The areas of study are Shika and Zaria city metropolis, in Kaduna State, Nigeria, having scarcity of public water supply; so leading to dependence on groundwater sources for domestic
usage. These areas are bound by Longitude 7° 42' and latitude 11° 3' (Zaria city); and Longitude 7° 33' and Latitude 11° 11' (Shika). The sampling points where the water samples were collected randomly during the rainy and dry season of 2016 are showed in the map below (n = 20).

![Map of Shika and Zaria City Showing Location of Wells and Boreholes. Source: Zaria Quirk Bird Satellite Image 2015.](image)

**Physicochemical Parameters of the Water Sample:**
The methods used to determine the physicochemical parameters of water samples collected in the rainy and dry season of 2016 were those described by APHA (1995). The pH, temperature, total dissolve solids (TDS), colour and conductivity of the water were measured in-situ. In order to maintain quality assurance, triplicate determination of the samples were made and the data presented as means.

**Metal Determination:** To 10.0 cm³ of each water sample collected wet digestion was carried out with 10.0 cm³ concentrated HCl and concentrated HNO₃ acid mixture (3:1) on a hot plate at 90°C (Mendham et al., 2008).

After digestion, each of the digest was filtered using a Whatman number 42 filter paper. The filtrate was analysed for heavy metals using a Plasma Atomic Absorption Spectrophotometer (Varian AA240FS) at the Multi–user Laboratory, Ahmadu Bello University, Zaria – Nigeria. The total Pb was determined at 405.781 nm and Cr at 425.433 nm.

Quality assurance to validate the atomic absorption spectrometry equipment was carried out by spiking experiment. To 50 cm³ of the Multi-element standard solution (MESS) prepared, was drawn with a graduated pipette and used to spike 50 cm³ of pre-digested groundwater sample from Shika, Nigeria in a 100 cm³ beaker. The spiked water sample and 10 cm³ of unspiked water sample was then digested. Then the concentrations of Cr and Pb in the spiked and unspiked samples were used to calculate the percentage recovery (Omoniyi et al., 2016a).

**Analysis of Ground Water for ^{222}\text{Rn} Determination:** The water samples were analyzed as soon as possible (maximum of three days) after collection to achieve maximum accuracy, because the composition of the sample may change if samples are left for long before performing the analysis.

**Sample Preparation and Analysis:** About 10 mL each of the samples was added into a scintillation vial containing 10 mL of the insta-gel scintillation cocktail. The vials were sealed tightly and shaken for more than two minutes to extract ^{222}\text{Rn} in water phase into the organic scintillator.

**Sample Analysis:** The prepared samples were analyzed using a liquid scintillation counter (Tri-Carb-LSA1000) located at the Center for Energy Research and Training (CERT), Ahmadu Bello University, Zaria–Nigeria. The samples were analyzed after they were allowed to stand for three hours after preparation in order to establish radioactive equilibrium between ^{222}\text{Rn} and its daughter progeny. The liquid scintillation counter was calibrated prior to the analysis using IAEA ^{226}\text{Ra} standard solution. For the calibration, the ^{222}\text{Rn} standard samples were counted for 60 minutes. Background count measurements were also made for the same time period (60 min). The ^{222}\text{Rn} activity concentration was calculated using the following equation (Galan Lopez et al., 2004).

\[
\text{Rn} \left( \frac{\text{Bq}}{\text{L}} \right) = \frac{100 \times (\text{SC} - \text{BC}) \times \exp(\lambda T)}{60(KF) \times (D)}
\]

Where, Rn = radon level in Bq/L; SC = sample count rate (count min⁻¹); BC = background count rate (Count min⁻¹); K = calibration value; T = elapsed time from sampling to testing given in minutes.

Also, the annual effective doses due to the intake of radon were calculated from the mean activity concentration using the following equation:

\[
\text{ERn} = \text{DFRn} \times \text{Iw} \times A^{222}\text{Rn}
\]

where DFRn is the effective dose per unit intake of radon in water for adults, taken as 10⁻⁸ Sv/Bq from UNSCEAR 1993 report; and Iw is the water consumption rate (l/a) taken to be 2 l per day.

**RESULTS AND DISCUSSION**

**Physicochemical Parameters of the Water Sample:** The temperature for the water samples as presented in
Table 1 were within the WHO (1993) acceptable limit of 25°C for drinking water, except for sample 6 and 9 from Zaria boreholes that were 27.00 ± 0.00°C and 26.00 ± 0.00°C respectively during the rainy and dry season and sample 11 and 13 from Samaru boreholes, being 24.00 ± 0.00 and 23.00 ± 0.00°C respectively during dry season. The deviation of these temperature might be as a result of absorbed heat from the sun or from bedrock beneath, because temperature is due to flux of heat coming beneath the ground or chemical reaction due to the decomposition of organic matter (Chapman, 1997). The pH of the water samples are in the range of 6.5-9.5 for the wells and boreholes water (Chapman, 1997). The pH of the water samples are in the range of 6.5-9.5 for the wells and boreholes water (Chapman, 1997). The pH of the water samples are in the range of 6.5-9.5 for the wells and boreholes water (Chapman, 1997). The pH of the water samples are in the range of 6.5-9.5 for the wells and boreholes water (Chapman, 1997). The pH of the water samples are in the range of 6.5-9.5 for the wells and boreholes water (Chapman, 1997). The pH of the water samples are in the range of 6.5-9.5 for the wells and boreholes water (Chapman, 1997).

The colour intensity of the water samples across the sampling points ranged from 5.00 to 10.00 Hazen; with all being below the standard limit of 15 Hazen unit for drinking water (WHO, 1997). This indicates that the selected water samples have good aesthetic property. As presented in Table 1, the total dissolved solid contents of the water from all the sampling points were below WHO maximum contaminant levels of 1000 mg/L. Higher total dissolved solid reduce water clarity, which could contribute to decrease in photosynthetic activities and might lead to an increase in water temperature (NRCC, 2011). This might also explain the good water colour reported in this study.

**Heavy Metal Content of the Water Samples:** From the spiking experiment, the mean % recovery in ascertaining the quality assurance ranged from 78.6 ± 0.21 to 89.11 ± 0.32. This signals accuracy of the atomic absorption spectrophotometer. The result of the levels of Pb and Cr in the selected groundwater as presented in Table 2 shows that during the rainy season of 2016, all the water samples contain Cr below the detection limit of the AAS machine, except for samples 6 (ZBS1) having 0.008±0.006 mg/L and 10 (ZBS5) having 0.009±0.006 mg/L; though found to be below the standard permissible limit of Cr in water (0.050 mg/L); the study indicated that sample 7 (ZBS2) and 8 (ZBS3) have mean Cr concentration of 0.338 ± 0.39 mg/L and 0.236 ± 0.169 mg/L which are above the permissible limit of Cr in water (WHO, 1993). This might be as a result of increase in industrial process such as tanning, paint production, and pigment production, as these involve the use of Cr compound and are therefore the most frequent source of hexavalent Cr (Marques, 2000).

The report of this study is similar to that by Saba and Ebrahim (2016) for 100 well water in the plain of...
Determining concentrations and annual results for Cr and Pb in water samples from Shika and Zaria City during the rainy season

Table 2: Concentration (mg/L) of Cr and Pb in Water Samples from Shika and Zaria City during the rainy season

| Site   | Samples | Cr (mg/L) | Pb (mg/L) | Cr (mg/L) | Pb (mg/L) |
|--------|---------|-----------|-----------|-----------|-----------|
| Zaria  | ZWS1    | BDL       | 0.0075 ± 0.005 | BDL | BDL |
|        | ZWS2    | BDL       | 0.001 ± 0.001 | BDL | BDL |
|        | ZWS3    | BDL       | 0.862 ± 0.569 | BDL | BDL |
|        | ZWS4    | BDL       | 0.039 ± 0.028 | BDL | BDL |
|        | ZWS5    | BDL       | 0.0557 ± 0.040 | BDL | BDL |
| Zaria  | ZBS1    | 0.008 ± 0.006 | 0.063 ± 0.044 | BDL | BDL |
|        | ZBS2    | 0.338 ± 0.39 | 0.0475 ± 0.034 | BDL | BDL |
|        | ZBS3    | 0.236 ± 0.167 | 0.301 ± 0.203 | BDL | BDL |
|        | ZBS4    | BDL       | 0.046 ± 0.032 | BDL | BDL |
|        | ZBS5    | 0.009 ± 0.006 | 0.031 ± 0.021 | BDL | BDL |
| Zaria  | ZBS1    | 0.008 ± 0.006 | 0.063 ± 0.044 | BDL | BDL |
|        | ZBS2    | 0.338 ± 0.39 | 0.0475 ± 0.034 | BDL | BDL |
|        | ZBS3    | 0.236 ± 0.167 | 0.301 ± 0.203 | BDL | BDL |
|        | ZBS4    | BDL       | 0.046 ± 0.032 | BDL | BDL |
|        | ZBS5    | 0.009 ± 0.006 | 0.031 ± 0.021 | BDL | BDL |
| Zaria  | SBW1    | BDL       | 0.023 ± 0.016 | 0.12 ± 0.00 | BDL |
|        | SBW2    | BDL       | 0.070 ± 0.049 | 0.11 ± 0.01 | BDL |
|        | SBW3    | BDL       | 0.046 ± 0.033 | 0.12 ± 0.02 | BDL |
|        | SBW4    | BDL       | 0.073 ± 0.052 | 0.11 ± 0.02 | BDL |
|        | SBW5    | BDL       | 0.093 ± 0.066 | 0.11 ± 0.08 | BDL |
| Shika  | SWW1    | BDL       | 0.069 ± 0.049 | 0.10 ± 0.02 | BDL |
|        | SWW2    | BDL       | 0.051 ± 0.036 | 0.66 ± 0.06 | BDL |
|        | SWW3    | BDL       | 0.048 ± 0.034 | 0.09 ± 0.13 | BDL |
|        | SWW4    | BDL       | 0.031 ± 0.022 | BDL | BDL |
|        | SWW5    | BDL       | 0.044 ± 0.030 | BDL | BDL |
| WHO    | 0.05    | 0.01      | 0.05       | 0.01     |       |

BDL - below detection limit

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Radon Concentration in Water: Table 3 shows the $^{222}\text{Rn}$ content in the water samples during rainy and dry season of Shika community and Zaria metropolis, Nigeria. The lowest $^{222}\text{Rn}$ was $0.10 \pm 0.014$ Bq/L (SWW4) and the highest was $66.56 \pm 0.03$ Bq/L (SBS1) in the rainy season. The highest concentration of $^{222}\text{Rn}$ was found during the dry season in ZBS1 (71.53$\pm$0.01 Bq/L). The $^{222}\text{Rn}$ concentration obtained in both the rainy and dry seasons were found to be generally lower than the standard permissible limit for $^{222}\text{Rn}$ intake set by world Health Organization and United State Environmental Protection Agency (11.04 Bq/L). This shows that most groundwater in Shika and Zaria community can be said to be safe for drinking during both season. However, some has higher than the recommended level in water. Also, the borehole water sample from Shika metropolis have $^{222}\text{Rn}$ concentration ranging from 0.80 Bq/L to 11.05Bq/L during the rainy season, and $^{222}\text{Rn}$ concentration between 5.63 Bq/L to 44.08 Bq/L during the dry season of 2016. During the rainy season $^{222}\text{Rn}$ concentration were found to be lower than the World Health Organization (WHO, 1993; UNSCEAR, 1993 maximum permissible value of 10Bq/L and 11.1Bq/L. Therefore, most of the water samples contained tolerable level of $^{222}\text{Rn}$ during the rainy season. Whereas BWS5 have $^{222}\text{Rn}$ concentration of 44.08 Bq/L which is almost four times the permissible limit during dry season, SBW5 has the highest concentration of $^{222}\text{Rn}$ (44.08 Bq/L), this might be as a result of less volume of water in the boreholes compare to the rainy season volume of water. The relatively high $^{222}\text{Rn}$ in the dry season than the rainy season, may be related to the radon source ($^{238}\text{U}$ and $^{226}\text{Ra}$) in the water-rock system present in the areas especially Zaria. This could pose greater health risk when consumed for a long period of time (Lawal, 2008), because wells sunk in areas that are rocky tend to show high content of granites to which radon is associated (Berazina et al., 2005).

Water samples from Zaria metropolis have the concentration of $^{222}\text{Rn}$ in the range of 23.41 to 66.56 Bq/L during rainy season and 17.81 Bq/L to 71.53 Bq/L during dry season. All the water samples have $^{222}\text{Rn}$ concentration higher than the permissible limit stated by WHO (1993) and USEPA (1991). Also having radon concentration above the standard limit during rainy season. This may be as a result of the natural processes, industrial or agricultural activities and increase human activities in the area where the borehole are located (Garba et al., 2008). High concentration of $^{222}\text{Rn}$ was recorded in boreholes in Zaria because most of them are deeper and are therefore closer to the surface sub-soil which are underlined by older granite, also Zaria has high human, agricultural activities, and natural process than Shika (Garba et al., 2008). As presented in Table 4, during the rainy season of 2016, the selected groundwater in Shika and Zaria metropolis, Nigeria has the lowest annual effective dose being 0.90 µSv/year (SWW4) and the highest was 14.30 µSv/year (SBS1). On the other hand, the lowest annual effective dose during the dry season was 1.0 µSv/year in several sites (SWW4) and the highest was 12.0 µSv/year in ZWS1. The borehole samples recorded higher $^{222}\text{Rn}$ concentrations and higher annual effective doses compared to the well water samples.

### Table 3: $^{222}\text{Rn}$ Concentration in Water Sample from Shika and Zaria

| S/No. | Sample ID | $^{222}\text{Rn}$ Concentration | $^{222}\text{Rn}$ Rainy season | $^{222}\text{Rn}$ Dry season | Annual effective dose (µSv/year) -R (µSv/L) -D |
|-------|-----------|-------------------------------|-------------------------------|-------------------------------|--------------------------------------------|
| 1     | ZWS1      | 4.02±0.03                     | 60.07±0.01                   | 12.81-12.0                    |
| 2     | ZWS2      | 0.16±0.00                     | 53.02±0.01                   | 10.63-11.0                    |
| 3     | ZWS3      | 2.50±0.14                     | 28.55±0.00                   | 6.21-6.0                      |
| 4     | ZWS4      | 2.23±0.03                     | 2.26±0.03                    | 0.90-1.0                      |
| 5     | ZWS5      | 25.75±0.01                    | 1.53±0.01                    | 5.31 -5.0                     |
| 6     | ZBS1      | 66.56±0.03                    | 71.53±0.01                   | 14.30 -14.0                   |
| 7     | ZBS2      | 24.04±0.03                    | 26.49±0.01                   | 5.30 -5.0                     |
| 8     | ZBS3      | 54.41±0.01                    | 17.81±0.01                   | 3.60 -3.0                     |
| 9     | ZBS4      | 23.41±0.00                    | 30.70±0.00                   | 6.20 -6.0                     |
| 10    | ZBS5      | 59.60±0.00                    | 31.41±0.01                   | 6.20 -6.0                     |
| 11    | SBW1      | 2.90±0.00                     | 5.85±0.07                    | 1.17 -1.0                     |
| 12    | SBW2      | 11.05±0.07                    | 6.65±0.01                    | 1.32 - 1.0                    |
| 13    | SBW3      | 8.18±0.03                     | 5.71±0.01                    | 1.14 - 1.0                    |
| 14    | SBW4      | 0.80±0.00                     | 11.04±0.01                   | 2.20 -2.0                     |
| 15    | SBW5      | 8.50±0.14                     | 44.08±0.01                   | 1.80 -2.0                     |
| 16    | SWW1      | 5.38±0.00                     | 11.04±0.00                   | 2.20 -2.0                     |
| 17    | SWW2      | 1.79±0.014                    | 9.34±0.03                    | 1.87 -2.0                     |
| 18    | SWW3      | 3.32±0.014                    | 5.29±0.01                    | 1.06 -1.0                     |
| 19    | SWW4      | 0.10±0.014                    | 6.68±0.03                    | 1.34 -1.0                     |
| 20    | SWW5      | 0.81±0.00                     | 6.68±0.00                    | 1.34 -1.0                     |

Table 3: $^{222}\text{Rn}$ Concentration in Water Sample from Shika and Zaria

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Scientists believe that exposure to $^{222}\text{Rn}$ is the second leading cause of cancer. When it decays, it shoots off alpha particles which are a heavy, electrically charged, sub-atomic particles consisting of two protons and two neutrons. If an alpha particle strikes chromosomes in a lung cell, it can alter the way the cell reproduces. And the calculated annual effective dose due to the intake of $^{222}\text{Rn}$ in drinking water (hand-dug well and borehole) at Shika and Zaria was within the permissible limit. The variation in the radon concentration could be a function of the geological structure of the area, depth of the water source and geo-hydrological processes that occur in the area as well as the anthropogenic activities. From the study, there was negative and positive correlation between the amount of $^{222}\text{Rn}$, Cr, Pb and physicochemical parameter of the water sample, also there was significant difference between the parameters across the season.

Generally, the annual effective dose obtained in this study was less than half the total natural radiation exposure of 2400 µSv/year. Also, the effective doses are also within the World Health Organisation (WHO) recommended reference level of 100 µSv per year for intake of radionuclides in water (IAEA, 2001). So the water samples studied are safe for drinking. Notwithstanding, periodical assessment of the levels of heavy metals and radionuclides in the water sources and the environment at large is imperative. In comparison to the work of Nada (2013), this study had higher mean radon concentration. Nada (2013) reported that the radon concentrations in groundwater of Aucashat city (Iraq) was in the range of 8.02 ± 0.14 to 11.7±0.16 Bq/L, with an average of 9.35±1.24 Bq/L, and the annual effective dose from ingestion (stomach) was 840 to 1230 µSv/y; from inhalation (lung) it was 2250 to 3280 µSv/y and for whole body was from 3090 to 4510 µSv/y. Though, the annual effective dose from ingestion of water was higher than radon concentration. Nada (2013) reported for the groundwater of Shika and Zaria, Nigeria. Also the result from this study is lower than the measured radon concentration range of 374.89 ± 37 to 6409.03 ± 130 pCi/L reported by Abdul Malik et al. (2015) for well water of selected areas in Pulau Pinang and Kedah using RAD7 and Rad H$_2$O accessories (1 pci = 0.037 bq). In addition, the well water has radon above the maximum contamination limit (MCL) of 300 pCi/L recommended by United State Environmental Protection Agency (USEPA). In an era of acute water shortage from public water supply system, digging of well and drilling of boreholes is the order of the day, so consideration should be given to installation of water treatment system to remove radon.

Conclusion: From the study most of the physicochemical parameters in the groundwater studied are within the WHO safe limit. Also the levels of Cr and Pb, except Pb in the water collected in the rainy season was below the limit. The $^{222}\text{Rn}$ concentration in the ground water sources were found to be within the maximum permissible limit, except for the samples collected from Zaria metropolis. So, periodic assessment of radon in water and the environment at large is recommended.

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