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RADIOLOGICAL RISKS OF RADIUM-226 ON GROUNDWATER BASED-DRINKING IN KUBWA AND GOSA AREA OF ABUJA , NORTH CENTRAL NIGERIA

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ABSTRACT: *The radiological risks associated with radium-226 in groundwater samples were analyzed using Inductively Coupled Plasma Mass Spectrometer (ICP-MS). The mean annual effective dose from the natural radionuclide of radium-226 (^{226}R) was estimated to be 2.2 x 10^{-5} mSv. In the groundwater samples, Lifetime average daily dose (LADD) ranged from 1 x 10^{-3} to 2 x 10^{-3} $\mu g\ kg^{-1}\ day^{-1}$. The LADDs and the highest cancer mortality risk was found at Kubwa borehole with a value of 3.1 × 10^{-8} and lower value reported at Gosa borehole with a value of 1.8 x 10^{-8}. The highest cancer morbidity of 9.7 x 10^{-9} was noted at Kubwa whereas lower value of 5.2 x 10^{-9} reported at Gosa borehole. The radiological risks of ^{226}Ra in the water samples were found to be low, typically in magnitude of 10^{-7}. With this low level of radiological implications, it may be the chemical toxicity associated with ^{226}Ra as a heavy metal. The values obtained in the study area may not pose radiological hazards to the residents that rely on groundwater. At the same time, measures of groundwater quality protection and monitoring should be enhanced and new supply source which has lower risk must found.*

Keywords: Abuja**,** Groundwater, Radiological Risks, Chemical Toxicity

1.0 INTRODUCTION

The activity concentrations of ^{238}U, ^{232}Th and ^{40}K in groundwater are connected to the activity concentrations of ^{238}U and ^{232}Th of aquifer bearing formation, and their decay products in subsurface rock formation. This occurs as a result of reactions between groundwater, soil and bedrock which release quantities of dissolved mineral components depending on the mineralogical and geochemical composition of the rock formation [1-2]. It also depends on the chemical composition of the water, degree of weathering of the subsurface rock formation, redox conditions and the residence time of groundwater in subsurface water bearing formation, [3]. ^{238}U and ^{232}Th decay series in soils, bedrocks and groundwater system is controlled by the chemical substances, radioactive decay and surrounding physical factors. As a result of these controlled processes, the radioactive elements either leach into the groundwater or to the surrounding, resulting into decay series of disequilibrium of nuclides [3].

The predominant radium isotopes in groundwater are ^{226}Ra, an alpha emitter with a half-life of 1600 years, and ^{228}Ra, a beta emitter with a half-life of 5.8 years [4-6]. Considering the high radiotoxicity of ^{226}Ra and ^{228}Ra, their presence in water and the associated health risks require particular attention. It is known that even small amounts of a radioactive substance may produce a damaging biological effect and that ingested and inhaled radiation can be a serious health risk [7].

In this study, the main emphasis is to determine the activity concentration and human radiological risk over life time consumption of ^{226}Ra in groundwater-based drinking at Kubwa and Gosa boreholes recommended by [8]. The study area is located within the crystalline basement complex of Nigeria. The rocks of this area are mainly migmatite, Leucocratic granite, quatzmonite and granodiorite. The detailed geology and hydrogeology of the study area was reported elsewhere [8]. The drilling point coordinates at Kubwa lies within lat. 9^o 6'16.7" N and long. 7^o 16'26.0"E whereas that of Gosa area lies within the coordinate of Lat: 8^o 56' 45.6" N and Long: 7^o 13' 26.2" E.

3.0 Materials and Methods:

Four (4) water samples measured two litres each from Kubwa, with a depth of about 60 m, Gosa area with depth of about 50 m, Hand-dug well of shallow depth of 14 m and Water Board (public water supply) in Abuja and suburbs were collected. Water samples were collected in high density polyethylene containers at the site in Nigeria previously washed in a solution of 10 % nitric acid for 15 minutes, followed by repeated rinsing with distilled water and finally rinsing with ultrapure water (resistivity of about 18 MΩ cm^{-1}). The collection containers were kept in sealed polyethylene bags before the collection of samples. The water samples were stabilized with 5 ml of nitric acid in each litre of water in order to prevent it from attacking to the wall of the container. For accurate determination of elemental compositions in each water sample, a solution analytical method was used; a multi-standard calibration method was applied using Elan 9000 instrument that performs analysis at parts-per-trillion and lower. The minimum detectable concentration was 0.01 µg L^{-1}, corresponding to 124 µBq L^{-1} [9]

3.1 Sample Analysis using ELAN 9000 Instrument and Technique

Measurement using ICP-MS was performed at the Universiti Tun Hussein Onn Malaysia Environmental and Soil Science Laboratory. ICP-MS is a relatively new method for determining multi-element analysis and ideal for groundwater, since the vast majority of target compounds can be detected below 0.1 mg L^{-1}. Water Samples were digested according to the previous study [10]. The water samples in Pellets were placed on a sample holder with a small disc of filter paper. A 6 mol L^{-1} NH_4NO_3 solution (50 µL) was added to the filter paper followed by the introduction of a sample holder vessel previously charged with 6 mL of absorbing

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solution (10–100 mmolL^{-1} NH_4OH). All raw data, including methods and parameters used, are stored in an encrypted, checksum-protected data set, in order to guard against data tampering. Audit trails that capture file, system and security-related events provide traceability for most software applications. The powerful quality-control system allows one to set limits, parameters and standards based on U.S. EPA, [11]. Determination methods used in this study for analysing radionuclides and other heavy metals have been accredited according to ISO standard 17025 (European Standard EN ISO/IEC 17025:2000).

4.0 RESULTS AND DISCUSSION

4.1 Accumulation of Radionuclide (^{226}Ra) in Humans and Recommendations for the Maximum Permissible Limit

The activity concentrations of ^{226}Ra that were determined in groundwater samples were used to interpret the human radiological risks of annual effective dose as presented in Table 1 using Equation (1) below.

Table 1: Results of activity concentrations, Annual Effective Dose of ^{226}Ra in Water Samples from the Study area and comparing with various countries and International Standard

Location	Activity Concentration ^{226}Ra (μBq L^{-1})		Annual Effective Dose (mSv y^{-1})	Reference
Dei-Dei	2698		8.9×10^{-5}	Present Study
Kubwa	849		2.8×10^{-5}	Present Study
Gosa	443		1.5×10^{-5}	Present Study
Lugbe	2736		9.0×10^{-5}	Present Study
Water Board	1824		6.0×10^{-5}	Present Study
Hand-dug well	2430		8.0×10^{-5}	Present Study
USE USEPA, CounCouncil Directive 98/83/EY/	19,000	1.0×10^{-1}		[12,13]

The International Commission on Radiological Protection, ICRP, provides recommendations and guidance on all aspects of protection against ionizing radiation, which are published in the commission's own scientific journal, the Annals of the ICRP, was also referred to in this work. The process of exposure starts through ingestion of groundwater that contains radionuclides; after entering the human body, radionuclides are typically accumulated in the skeleton, liver, kidney and soft tissues. Ingested radionuclides are not entirely retained in the human body. Dose coefficients help to determine the effective dose associated with radiation exposure in assessing the health risks to people. The dose coefficient is expressed in Sv Bq^{-1}; the effective dose equivalent per unit water activity concentration of the radionuclide. The annual effective dose is calculated taking into account the activity concentration of the nuclide (Bq L^{-1}), the dose coefficient for ^{226}Ra (Sv Bq^{-1}) is give as 2.8×10^{-7} [14-17] and the annual water consumption is given as 731 L L y^{-1}, [18 - 17]. A reference dose of 0.1 mSv per year corresponds to the activity of 0.5 Bq L^{-1}, from Equation (1).

$$AED\ (\text{mSv y}-1) = AC\ (\text{Bq L}^{-1})\ X\ DC\ (\text{Sv Bq}^{-1})\ X\ AWC\ L\ \text{y}^{-1}\ X\ 1000 \quad (1)$$

where,

AED = Annual effective dose

AC = Activity concentration of ^{226}Ra

DC = Dose coefficient for ^{226}Ra

AWC = Annual water consumption

In the present study, Equation (1) was used to determine the annual effective dose of the water samples for ^{226}Ra radionuclide only in both groundwater based drinking water and Water Board as shown in Table 1. The World Health Organisation (WHO) and Environmental Protection Agency (EPA-USA) used the quantity of 2 litres per day water consumption for adults [18 - 17]. Comparing the two boreholes in Table 1, the annual effective dose reported higher in Kubwa borehole with a value of 2.8×10^{-5} mSv $y^{-1}$and lower value of 1.5×10^{-5} mSv y^{-1} was noted at Gosa borehole. In Comparing the Water Board and hand-dug well with values of 6.0×10^{-5} mSv y^{-1} and 8.0×10^{-5} mSv y^{-1} respectively with the values obtained at Kubwa and Gosa boreholes, 2.8×10^{-5} mSv y^{-1} and 1.5×10^{-5} mSv y^{-1} respectively, they are distinctly higher the values reported at the boreholes. In contrast with the previous report of the international standard [20], 0.1 mSv y^{-1}, the highest value of the borehole water sample obtained in the study area, Kubwa with a value of 2.8×10^{-5} mSv y^{-1} was far below the recommended value. According to ICRP 69 [17], Radium gets to the blood through the soft tissues and excretes in urine. It can be excreted in few months whereas the parents could be retained for years. Radium exchanges with Ca^{2+} accumulate in surface of bone and highly concentrated in the area of growth. The liver accumulates 1.5 to 2% of which most of it removed in a few weeks. Furthermore, the radioactivity of ^{226}Ra has chemical toxicity that predominately affects the kidneys [18, 19]. From the reference work done elsewhere, the Radium concentration is limited mainly by chemical toxicity rather than the effective dose [20]. In 2003, the World Health Organization proposed a provisional guideline of 0.1 mSv per year corresponds to the activity of 0.5 Bq L^{-1}. The result of this present study is below the recommended limit.

4.1 Radiological Risk Assessment of ^{238}U in Groundwater from the Study Area

The radiological risk assessment was to evaluate the life time cancer risk associated with the intake of a given ^{226}Ra R, associated with the intake of a given radionuclide were estimated from the product of the applicable risk coefficient, r and the per capita activity intake, I expressed in Equation (2).

$$R = r\ x\ I \qquad (2)$$

According to WHO, [24], the average life expectancy at birth in Nigeria is 45.5 y and, an annual consumption of water for an individual is about 731 L. This brings the lifetime intake of water to 33,215L. The cancer risk coefficients of ^{226}Ra is 7.17 × 10^{-9} Bq^{-1} for mortality and 1.04 × 10^{-8} Bq^{-1} for morbidity respectively were obtained from the literature [21- 22]. Using Equation 2.0 and the coefficients, the cancer mortality and morbidity risks of ^{226}Ra over lifetime consumption of water were calculated and the results are presented in Table 2.

Table 2: The estimated lifetime cancer mortality and morbidity risk of ^{226}Ra in the water Samples.

Location	Cancer Mortality Risk	Cancer Morbidity Risk	Reference
Dei-Dei	1.01 x 10^{-7}	1.55 x 10^{-7}	Present Study
Kubwa	3.19 x 10^{-8}	4.88 x 10^{-9}	Present Study
Gosa	1.67 x 10^{-8}	2.55 x 10^{-8}	Present Study
Lugbe	1.03 x 10^{-7}	1.57 x 10^{-7}	Present Study
Water Board	6.85 x 10^{-8}	1.05 x 10^{-7}	Present Study
Hand-dug well	9.12 x 10^{-8}	1.40 x 10^{-7}	Present Study
Odeda, Ogun state, Nigeria	2.54 x 10^{-4}	3.39 x 10^{-4}	[23]

In Table 2, the cancer mortality risks ranged from 1.8 × 10^{-8} to 9.5 × 10^{-8} while for morbidity risks, it ranges from 9.7 × 10^{-9} to 2.8 × 10^{-7}. From the two boreholes in Table 3, the highest cancer mortality risk was found at Kubwa borehole with a value of 3.1 × 10^{-8} and lower value reported at Gosa borehole with a value of 1.8 x 10^{-8} . The highest cancer morbidity of 9.7 x 10^{-9} was noted at Kubwa whereas lower value of 5.2 x 10^{-9} reported at Gosa borehole. In contrast with the cancer mortality and morbidity risks from Water Board, 7.2 x 10^{-8} and 2.1 x 10^{-7} , Hand-dug well, 9.5 x 10^{-8} and 2.8 x 10^{-7} respectively were higher than the values obtained from Kubwa and Gosa boreholes. Comparing with a study reported by [23] in Ogun State, Nigeria, both Kubwa borehole, Water Board and Hand-dug well were lower than 2.5 x 10^{-4} and 3.4 x 10^{-4} values obtained for cancer mortality and morbidity risks in Odeda Ogun State, Nigeria. It can be noted that both cancer mortality and morbidity risks reported is higher at Kubwa borehole when compared with Gosa borehole; it may be that the aquiferous zone may have been affected due to higher deformation of fractures which enabled water to trap at the near surface since the subsurface geology permits the rapid downward movement of water sources from the source and enable the escape of radon gas. The cancer risk at 10^{-7} is lower compared to the acceptable level of 10^{-3} for the radiological risk [23].

4.2 Chemical Toxicity Risk of ^{238}U in Groundwater from the Study Area

radionuclide in groundwater. The lifetime cancer risks

The chemical toxicity was to determine the effect of the carcinogenic risks associated with chemical toxicity of ^{226}Ra in the water sample selected for this study. The chemical toxicity risk was evaluated using the lifetime average daily dose of ^{226}Ra through drinking water intake, and compared it with the reference dose (RFD) of 0.6 μg kg^{-1} day^{-1} [24] used as a standard criteria for ^{226}Ra in several foreign organizations and thereby produce the lifetime average daily dose (LADD), Equation (3)

$$\text{Ingestion } LADD \text{ of drinking water} = \frac{EPC\ x\ IR\ x\ EFx\ ED}{AT\ x\ BW} \qquad (3)$$

where, *LADD* is lifetime average daily dose (μg kg^{-1} day^{-1}), *EPC* is the exposure point concentration (μg L^{-1}), *IR* is the water ingestion rate (L day^{-1}); *EF* is the exposure frequency (days $year^{-1}$), *ED* is the total exposure duration (years), AT is the average time (days) and *BW* is the body weight (kg). Using therefore, *IR* = 2 L day^{-1}, *EF* = 350 days, *ED* = 45.5 y, *AT* = 16,607.5 (obtained from 45.5 × 365) and *BW* = 70 kg (for a standard man). The chemical toxicity risk for ^{226}Ra over a lifetime consumption was estimated and presented in Table 3.

Table 3: The estimated lifetime average daily dose (LADD) of uranium in the water samples

Location	LADD (μg $kg^{-1}day^{-1}$)	Reference
Dei-Dei borehole	6 x 10^{-3}	present
Kubwa borehole	2 x 10^{-3}	present
Gosa borehole	1 x 10^{-3}	present
Lugbe borehole	6 x 10^{-3}	present
Water Board	4 x 10^{-3}	Presnt
Hand-dug well	5 x 10^{-3}	present
RFD (Reference Dose)	6 x 10^{-1}	[24]

In Table 3, the Lifetime average daily dose (LADD) ranged from 1 x 10^{-3} to 5 x 10^{-3} μg kg^{-1} day^{-1}. The *LADD*s values were observed to be higher in Kubwa borehole with a value of 2 x 10^{-3} μg kg^{-1} day^{-1} compared to Gosa borehole with a value of 1 x 10^{-3} μg kg^{-1} day^{-1}. This could be due to the interbedding of altrabasic minerals emanated from the deep seated source caused by magmatic and metamorphic processes of granitic intrusions and its interconnectivity with geochemistry and aquifer bearing formation. The lowest value of 1 x 10^{-3} μg kg^{-1} day^{-1} was found in Gosa borehole. Comparing the LADDs from Kubwa and Gosa Boreholes to Water Board and hand-dug well, it can be noted that both boreholes were lower than 4 x 10^{-3} and 5 x 10^{-3} values for Water Board and hand-dug well respectively. Comparing the LADD obtained in this study with the Reference Dose (RFD) (0.6 μg kg^{-1} day^{-1}) that is an acceptable level; the chemical toxicity risk due to ^{226}Ra in the water samples were all below the RFD. This shows that there may not be health risks

associated with ^{226}Ra in the water samples which are mainly due to the chemical toxicity risk of ^{226}Ra and its progeny. However, Hand-dug well reported higher value of LADD of ^{226}Ra than other water samples; it may be due to the formation of soluble complexes in aqueous phase in weathering and alteration caused by metamorphic process. In addition, the Water Board of Abuja which is the public water supply in the region noted higher than Kubwa and Gosa boreholes which may be attributed to solubility and high content of toxic non-carcinogen metals in the source of the surface water of the public water supply.

5.0 CONCLUSION

The mean annual effective dose from the natural radionuclide (^{226}R) for the inhabitants that rely on groundwater was estimated to be 2.2 x 10^{-5} mSv of the annual collective dose. The highest annual effective dose from radionuclide was noted in Kubwa Borehole with a value of 2.8 x 10^{-5} mSv y^{-1}. The lowest value was reported at Gosa borehole which may be due to ultrabasic intrusion of schist belt formation. The magnitude of 10^{-7} obtained for the radiological risks of ^{226}Ra in the water samples may not pose health risk to the consumers within the study area. Adequate measures for groundwater quality is needed for no amount of dose exposure is safe for long accumulation.

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REFERENCES

[1] Internal Commission on Radiological Protection Annual Limits on Intake of Radionuclides by Workers Based on the Recommendations (1990). Annals on the ICRP, ICRP publication 67, Oxf. Press.

[2] Langmuir, D. (1978). Uranium-solution equilibria at low temperatures with applications to sedimentary Ore deposits. Geochi. Cosmochim. Act. ,42 (6): 547-569

[3] Durrance, E. M. (1986). Radioactivity in Geology: Principles and Applications. New York, NY(USA). John Wiley and Sons Inc.

[4] Iyengar, M.A.R (1990). The Natural Distribution of Radium" The Environmental Behavior of Radium, Technical Reports Series No. 310, IAEA, 1: 9-128.

[5] Marovic, G., Sencar, J., Franic Z. and Lokobaner, N. ,(1996). Radium-226 in Thermal and mineral Springs of Croatia and Associated Health Risk", *J. Environ. Radio*. 33: 309-317.

[6] Sidhu, K.S. and Breithart, M.S. (1998). Naturally Occurring Radium-226 and Radium-228 in Water Supplies of Michigan", *Bull. Environ. Contam. Toxicol*., 61, 722-729

[7] Rowland, R.E. (1993). Low-Level Radium Retention by the Human Body. A

Modification of the ICRP Publication 20 Retention Equation", *Health Phys*, 65:

507-513.

[8] Omeje, M., Wagiran, H., Ibrahim, N., Lee, S. K., Soheil, S. (2013b). Measurement of ^{238}U, ^{232}Th, and ^{40}K in boreholes at Gosa and Lugbe, Abuja, North Central Nigeria .*Radia. Protect. Dosim*., 217-277.

[9] Omeje M., Wagiran, H, Ibrahim, N, Lee, S K., Soheil, S. (2013a). Comparison of ^{238}U, ^{232}Th, and ^{40}K in different layers of subsurface structures in Dei-Dei and Kubwa, Abuja, Northcentral Nigeria. *Radiat. Physi. Chem.*, 91, 70-80.

[10] Mesko, M F, Mello, P A, Bizzi, C A, Dressler, V L, Knapp, G, Flores, E. M. M (2010). Iodine determination in food by inductively coupled plasma mass spectrometry after digestion by microwave-induced combustion. *Analytical and Bioanalytical Chemistry*, 398(2): 1125–1131.

[11] Environmental Protection Agency, EPA. (2008). Ground Water and Drinking Water. US Drinking Water Protection Division. Retrieved from http://water.epa.gov/drink/(200.8).

[12] US. Environmental Protection Agency (2000). Methodology for Deriving Ambient Water Quality Criteria for the Protection of Human Health, EPA-822-B-00-004, page 2-3

[13] Council Directive 98/83/EY/ (1996). The quality of water intended for human consumption, Official Journal of the European Communities, L 330, 05/12/1998 s. 0032-0054.

[14] World Health Organization. (2003). Guideline for drinking water quality Health criteria and other supporting Information Edition of Guidelines on Drinking Water Quality, Geneva, 2nd Edit.

[15] Internal Commission on Radiological Protection. (1993). Age-dependent Doses to Members of the Public from Intake of Radionuclides: Part 2 Ingestion Dose Coefficients, Annals on the ICRP, ICRP publication 67, Oxford: Pergamon Press.

[16] Internal Commission on Radiological Protection. (1995). Age-dependent Doses to

Members of the Public from Intake of Radionuclides: Part 3 Ingestion Dose Coefficients, Annals on the ICRP, ICRP publication 69, Oxford: Pergamon Press.

[17] National Research Council. (1999). Risk Assessment of Radon in Drinking Water.

[18] World Health Organization. (2004). 3rd Edition of Guidelines on Drinking Water Quality, Geneva, Washinton D.C. Nation. Academic. Pres.

[19] World Health Organizations WHO (2006). Meeting The MDG Drinking Water and Sanitation Target, The Urban and Rural Challenge of the Decade pp. 1–47). New York.

[20] World Health Organization WHO. (2008). Meeting the MDG drinking water and sanitation target: the urban and rural challenge of the decade. WHO Library Cataloguing-in- Publication Data

[21] U.S. Environmental Protection Agency (EPA) nvironmental Protection Agency (1999). Federal Guidance Report No -13(EPA. 402 R-99-001) on Cancer risk coefficients for Environmental exposure to radionuclides. United State.

[22] United Nations Scientific Committee on the effects of Atomic Radiation, UNSCEAR, (2000). Sources, effect and risks of ionising radiation. Report to the General Assembly with Scientific Annexes. United Nations. New York.

[23] Amakom, C. M., Jibiri, N. N. (2010). Chemical and Radiological Risk Assessment of Uranium in Borehole and Well Waters in the Odeda Area, Ogun State, Nigeria. *International Journal of the Physical Sciences*, 5(7): 1009-1014.

[24] Ye-shin, K., Hoa-sung, P., Jin-yong, K., Sun-ku, P., Byong-wook, C., Ig-hwan, S., Dong Chun, S. (2004). Health risk assessment for uranium in Korean groundwater. *J. Environ. Radioct*, 77-85