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Radiotoxicity and groundwater quality for drinking in a multi-layered saline sedimentary terrain of Keana, Central Benue trough, Nigeria

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Abstract

The evaluation of uranium concentrations in the groundwater is pertinent to advise on the known connection between the chemical and radiological toxicity of the element with certain chronic lung cancer and kidney diseases. Ten (10) groundwater samples were collected from boreholes and wells around the research area to determine the ^{238}U level and related risk. Water samples were kept in a 50ml High-Density Polyethylene (HDPE) plastic container and acidified with strong nitric acid to prevent metal leaching. Temperature and conductivity were recorded on-site with a portable WTWLF 91 model pH/conductivity meter. The activity concentrations in all water samples vary from 372 to 19964 μgL^{-1} with the highest value in the AG borehole samples and the lowest in both the AB and AC samples, respectively. The estimated Annual Effective Dose (AED) for all age groups that rely on groundwater-based drinking varies from 0.012 to 0.991 mSvy^{-1} with the highest value of 0.991 mSvy^{-1} reported in sample AG for children in the age range from 7-12 years. The Lifetime Average Daily Dose (LADD) of ^{238}U varies from 8.58×10^{-4} to 4.61×10^{-2} with the highest value of 4.61×10^{-2} μgkg^{-1}day^{-1} noted in sample AG. The LADD for all the samples is lower than the acceptable value of 0.6×10^{-1} μgkg^{-1}day^{-1}. Although the risk at some locations is close to the threshold value, the present concentration of ^{238}U in the groundwater may pose a serious threat to the users unless adequate treatments are taken before consumption.

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1. Introduction

A radionuclide or radioisotope has the same atomic number but differs in atomic mass, i.e., the same number of protons and electrons, but with a different number of neutrons, such as ^{12}C6, ^{13}C6, and ^{14}C6. The excess nuclear energy they carry makes them unstable in the environment. Excess nuclear energy can be utilized in several ways, including nuclear radiation emitted directly from the nucleus in the form of gamma (γ) radiation. Then, it transfers an electron to one of its atoms, which is released as a conversion electron, or from the nucleus, new particles like alpha (α) or beta (β) can be released. Radioactive decay is the name given to this process of energy conversion. There are two types of radionuclides: primordial radionuclides, which the Earth has been containing

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radionuclides since its formation, as well as cosmogenic radionuclides formed in the upper layer of the crust and atmosphere. These ionizing radiation sources in the environment are both natural and man-made. Uranium (^{234}U, ^{235}U, ^{238}U), Potassium 40, Thorium, and other naturally occurring radionuclides. The radioactive elements can be used to generate electricity in nuclear power plants and as a high-density penetrator in the military. However, it is also used to test nuclear weapons. Natural radioactive elements such as Uranium and Thorium are commonly used to introduce radioactive materials into drinking water. Mineral processing, mining, and phosphate fertilizer production, etc., are examples of technological processes. Through the nuclear fuel cycle, radionuclides are also released into the environment, improper use of radionuclides produced in an unsealed form, inappropriate methods for use in the medical and industrial fields, unsuitable methods of dumping radioactive substances, and Radionuclides have been released into the environment in the past, which ultimately contaminated the water sources. Uranium has been described as an element that occurs naturally as a trace constituent in the subsurface [1].

It is also radioactive and chemically toxic, and the consequences of radiation versus chemical effects pose grave concerns. Moreover, uranium in groundwater occurs due to the weathering of rocks and soil. However, in groundwater, the activities of nuclides are governed by various methods such as adsorption desorption and dissolution processes [2]. The information on uranium concentration in surface and groundwater is crucial in investigating radiological assessment and the impact of various anthropogenic activities to secure the increased standard of living in present-day societies. The current drinking water guidelines are based on a Reference Dose Level (RDL) of 0.1 mSv from possible radioisotope consumption through drinking water per year, as recommended by the International Commission on Radiological Protection (ICRP) [3] in 1991 and the IAEA International Basic Safety Standard (IBSS) [4] in 1996 and accepted by the World Health Organization (WHO). Over the decades, interest in the environmental toxicity risk of uranium has been placed on nuclear power generation and depletion of uranium from military ammunition use, while that of groundwater is neglected [5]. Recent studies have shown that water in permeable rock across various parts of the globe is becoming polluted with uranium from both natural and man-made sources, European Food Safety Authority [6], the Agency for Toxic Substances and Disease Registry [7]. Drinking water exposure to uranium is increasing rapidly as more people rely on groundwater for their basic needs around the globe [5]; the high concentration of uranium poses a potential threat in terms of health effects on society. The long-term health consequences of uranium in drinking water include kidney damage and bone toxicity caused by alpha radiation [8, 9]. Human radiation exposure is influenced by factors such as elevation above mean sea level, radionuclide composition in the air, food, and drinking water, radionuclide types and amounts in soil, and others. The quantity of radionuclide absorbed into the body via food and ingestion has a significant impact on health. Such ionizing radiations are more powerful and harmful to human health and the environment. Such radiations cause hereditary effects, genetic deformity, non-fatal cancer, fatal cancer, and a drop in blood cell count. Higher dose exposure can sometimes result in death.

The standard value of 30 μg/L for uranium has been set for drinking water by the World Health Organization [10], the U.S. Environmental Protection Agency [11, 12], which is above the threshold value of 10 μg/L adopted in Germany [13]. Uranium at higher concentration levels in water has been reported globally; for example, in Switzerland [14], Finland [8] , Bangladesh [15], U.K. [16], Vietnam and Cambodia [17], Canada [18], India [19], Korea [20], Sweden [21], China [12], U.S. [22, 23], and Nigeria [24]. Aeration, air stripping, and activated carbon absorption has efficiencies of 67-99 %, >99 %, and 70-100 %, respectively, to reduce heavy radionuclide concentrations in water. In this present study, the concentration of uranium in groundwater for drinking at Awe Area Central Nigeria was examined for potential risk exposure to water users.

1.1. Location, climate and vegetation

The Awe Brine fields are part of the Benue Brine fields in north-central Nigeria (Figure 1). It is situated in Nigeria's Central Benue Trough, between longitudes 9° 07'E - 9° 09'E and latitudes 8° 06'N - 8° 08'N. The landscape has relatively low relief of 140 meters on average. It is drained by one of the major tributaries of the River Benue, the River Tunga, as well as several smaller rivers and rivulets. The research area, which is made up of Guinean savannah vegetation, has two distinct seasons: wet and dry. The rainy season runs from March to October, whereas the dry season is from November to February. It is unusual for rain to fall between January and February. The annual average rainfall and mean relative humidity range from 1000 to 1500 mm and 60 to 80 %, respectively [25], while the yearly average temperature is 28.50°C, with temperatures ranging from 33 to 360°C during the dry season [26].

1.2. Geology of the study area

The Awe Formation, which was formed as passage (transitional) beds during the Late Albian Early Cenomanian regression, underlies the locality. Its most common parts are found near the town of Awe, where Ref. [27] estimated the thickness to be about 100 meters. It is made up of flaggy, white, medium- to coarse-grained calcareous sandstone, carbonaceous shale, and clays [28]. The Cenomanian regression produced fluviodeltaic sediments, giving rise to the Keana Formation. The Keana Formation, which consists of cross-bedded, coarse-grained feldspathic sandstones, occasional conglomerates, and areas of shale and limestone from which brine emerges, is directly overlain by the Awe Formation. Massive outcrops may be seen in Keana, Noku, Jangerigeri, Azara, and Daudu [27, 29, 30]. Underneath the Awe Formation is the Asu River Group. According to Offodile [30] and Obaje [29], this is the oldest sedimentary rock unit in the Trough, deposited during the Mid-Albian marine transgression of the South Atlantic-Gulf of Guinea. Limestone, shale, calcareous shale, micaceous siltstones, mudstone, and clay make up the lithology of the group.

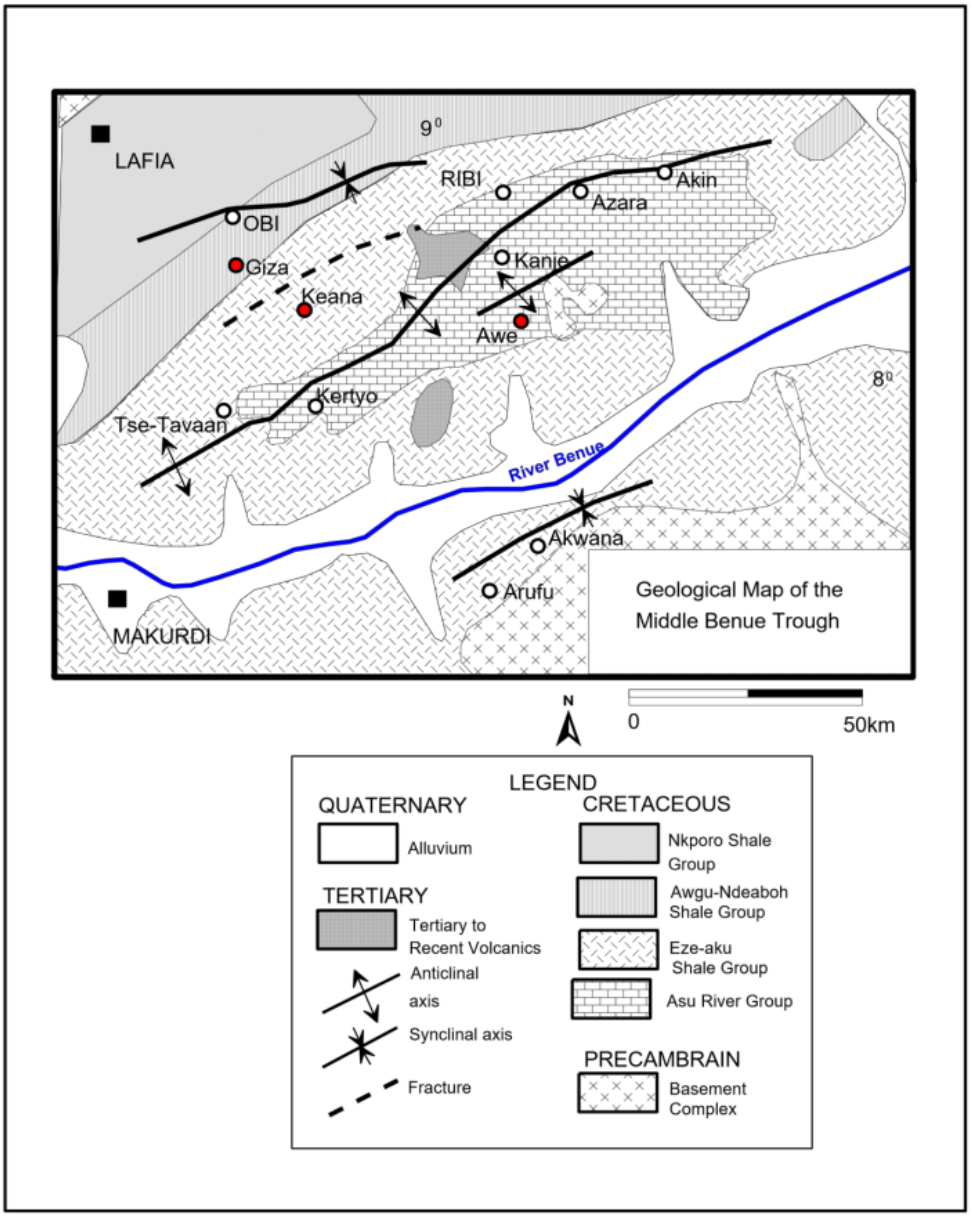

Figure 1. Geological map of the study areas (modified from Ref. [27]).

1.3. Hydrogeology of the study area

The primary aquifer's units found within the study area are the Awe, Keana, and Ezeaku Formations, with alternating shale and sandstone strata creating some unusual and complex hydrogeological settings. The aquifer's sandstone is strongly hardened, lightly developed with alternating shale and sandstone strata, confined to the point where only secondary pores are caused by tectonic activities, which result in rock fracturing of hydrogeological interest [31].

2. Materials and methods

To assess the level of ^{238}U and the associated risk in the groundwater, ten (10) groundwater samples were obtained from boreholes and wells throughout the research area. The water samples were stored in a 50ml High-Density Polyethylene (HDPE) plastic container and acidified with concentrated nitric acid to prevent the leaching of metals. A portable WTWLF 91 model pH/conductivity meter was used to measure temperature and conductivity in situ. The concentration of radionuclide levels was determined using enhanced ICP-ES/ICP-MS at Acme Analytical Laboratories (Vancouver Ltd), Canada. According to the previous investigation, all the samples were digested. Pellets of water samples were placed in a sample container with a small filter paper disc. After adding a solution of six (6) $molL^{-1}$ NH_4NO_3 (50L) to the filter paper, a sample holding vessel previously filled with 6mL of absorbing solution

Table 1. Concentration and activity concentrations of ^{238}U in water samples for the age group in the study area.

Sample	Concentration of ^{238}U (mg/L)	Concentration of ^{238}U (μg/L)	Activity concentration of ^{238}U (μBeq/L)
AB	0.00003	0.03	372
AC	0.00003	0.03	372
AD	0.00037	0.37	4588
AE	0.00026	0.26	3224
AF	0.00033	0.33	4092
AG	0.00161	1.61	19964
AH	0.00021	0.21	2604
AI	0.00021	0.21	2604
AJ	0.00062	0.62	7688
AK	0.00136	1.36	16864
WHO (2008)	0.030	30	37200

(10–100 mmolL^{-1} NH_4OH) was introduced. After closing and capping the rotor, the vessels were pressurized with twenty (20) bars of oxygen, and the rotor was inserted into the microwave cavity, where the heating stage began at 1400W for 5 minutes and the cooling stage began at 0W for 20 minutes. To determine elements by ICP-MS using this technique, the digests were transferred into a 25 mL volumetric flask and diluted with water after releasing the pressure in each vessel [32]. Microwave-heated concentrated HNO^3 was used to clean all vessels for 10 minutes at 1000W and then cooled at 0W for 20 minutes. Before use, glass and quartz constituents were immersed in 1.4 mol L^{-1} HNO^3 for 24 hours and then rinsed with water. Therefore, the results obtained from the analysis were used to assess the radioactive and radiotoxic risks posed by 238U in the groundwater of the Central Benue Trough, considering the characteristics of the multi-layered saline sedimentary terrain.

3. Results and discussion

3.1. Concentrations of ^{238}U in groundwater samples from the study area

Table 1 shows the different concentrations of ^{238}U in all groundwater samples. The ^{238}U content values vary from 0.03 to 1.61$\mu g l^{-1}$, with sample AG having the highest value of 1.61$\mu g l^{-1}$. The concentration levels in all the water tests are below the standard value of 1.9$\mu g l^{-1}$ [3]. The present calculated values for uranium concentration in water samples are on the lower side than the maximum acceptable uranium concentration value of 15$\mu g l^{-1}$ [33] and the recommended safe value of 30$\mu g l^{-1}$ [34]. The measured uranium concentration values for water samples are well below the range of 15-30 ppb proposed by Refs. [10, 34, 35].

3.2. Activity concentrations of ^{238}U in water samples found in the study area

The amounts of ^{238}U, and activity concentrations discovered in water samples taken from the research area as shown in Table 1. It varies from 372 to 19964 $\mu BeqL^{-1}$ with the highest value of 19964 $\mu BeqL^{-1}$ obtained from sample AG. The lowest value of 372 $\mu BeqL^{-1}$ is reported in both sample's AB and AC. This could be attributed to geologic features that control the subsurface formation. The high concentration found in water sample AG could be because of an intrusion of volcanic rocks within the study area. Comparing the highest value of the activity concentration of 19964 $\mu BeqL^{-1}$ with the International Reference Standard of 37200 $\mu BeqL^{-1}$ (30μgL^{-1}), The current study is low by a factor of 1.9. Approximately 99 % of uranium absorbed in food or drink is excreted by the human body, with the remaining entering the circulation [34]. Within a few days, the kidneys will eliminate most of this uranium, which will be expelled in the urine. A little quantity of uranium in the circulation will be deposited in the bones, where it will most likely stay for years. The danger of cancer presented by uranium and all other radionuclides is one of its harmful consequences; uranium also has non-cancer impacts. The kidney is the "target organ" of uranium's chemical toxicity. Uranium prevents proteins from being absorbed. Miners and millers who were exposed to large levels of uranium experienced modest and transitory side effects. Ref. [34] set a Maximum Contaminant Level for uranium of thirty (30) micrograms per liter (0.031 milligram per liter) based on the chemical toxicity of uranium. The highest concentration value of 1.61μgL^{-1} acquired in this current research is lower than the thirty (30) μgL^{-1} value recommended by the Environmental Protection Agency (EPA).

3.3. Ingestion of ^{238}U in humans and dose coefficients for radiological protection

The International Commission on Radiological Protection (ICRP) makes recommendations and gives guidance on all areas of ionizing radiation protection, which are published in the ICRP's own scientific publication, the Annals of the ICRP. The process of contact begins with the consumption of radionuclide-containing groundwater. Radionuclides often accumulate in the bones, liver, kidney, and soft tissues after entering the human body. Radionuclides consumed are not completely absorbed in the human body.

Table 2. Results of annual effective dose of ^{238}U for all the age groups in water sample from the study area and comparing with various countries and international standard.

Sample	AED for Adults ^{238}U (μSvy^{-1})	AED for Adults ^{238}U (12-17 years) (μSvy^{-1})	AED for Adults ^{238}U (7 - 12 years) (μSvy^{-1})
AB	12.22	18.89	18.47
AC	12.22	18.89	18.47
AD	150.72	224.39	227.75
AE	105.91	157.69	160.04
AF	134.42	200.14	203.13
AG	655.82	976.44	991.01
AH	85.54	127.36	129.26
AI	85.54	127.36	129.26
AJ	252.55	376.02	381.63
AK	553.98	824.82	837.13
Council directive [10, 20, 36]	100	100	100

Dose coefficients aid in determining the effective dose related to radiation exposure when evaluating the dangers to people's health. The dose coefficient of ^{238}U is 0.28 μSvBq^{-1}, which corresponds to a Reference Dose Level (RDL) of 0.1 mSvy^{-1} [34]. The annual effective dosage is calculated utilising nuclide's activity concentration (Bq L^{-1}), the dose coefficient of ^{226}Ra (Sv Bq^{-1}) is given as 4.5×10^{8}, 6.7×10^{8}, and 6.8×10^{8}, for an adult, children (12-17 years), and children (7-12 years) respectively, and the annual water consumption is 731 Ly^{-1} [35]. A reference dose of 0.1 mSv per year corresponds to the activity of 0.5 Bq L^{-1}, was calculated using Equation (1).

$$AED(mSvy^{-1}) = AC(BqL^{-1}) \times DC(SvBq^{-1}) \times AWC\ (Ly^{-1}) \times 1000, \quad (1)$$

where AED is annual effective dose, AC is activity concentration of ^{236}Ra, DC is dose coefficient for ^{236}Ra, and AWC is annual water consumption.

In this current study, Equation (1) has been used to determine the annual effective dose of groundwater samples for the ^{238}U radionuclide only in groundwater for drinking, in microSievert per year (μSv y^{-1}) as shown in Table 1. The World Health Organization (WHO) and the Environmental Protection Agency (EPA-USA) used a quantity of 2 and 1 liters per day of water consumption for adults and children, respectively [8, 10].

The values obtained in Table 2 were converted to milliSievert per year (mSv y^{-1}) for further discussions on the level of risks in age groups, as shown in Table 3. In Table 3, the annual effective dose for adults varies from 0.012 to 0.656 mSvy^{-1}, with the highest value of dose exposure of 0.656 mSvy^{-1} obtained in sample AG, and the lowest value of 0.012 mSv y^{-1} noted in samples AB and AC, respectively. For children between 12-17 years who rely on groundwater for daily consumption, the annual effective dose (AED) varies from 0.018 to 0.976 mSv y^{-1}, with the highest value of 0.976 mSv y^{-1} noted in sample AG. The lowest value of 0.018 mSv y^{-1} was reported in both samples AB and AC. At the same time, the exposure annual effective dose for children within the age limit of 7-12 years varies from 0.019 to 0.991 mSv y^{-1}. It can be observed that the highest value of 0.991 mSv y^{-1} was reported in the AG borehole water sample, whereas AB and AC respectively reported the lowest value of the same AED of 0.019 mSv y^{1}. In all the water samples, AG borehole water reported the highest AED in all the age groups that rely on groundwater use. Comparing this value with the International Reference Dose of 1.0 mSv y^{-1}, all the water samples are distinctly closer to the value for all the age groups, except for samples collected from boreholes AB and AC, which are lower. This is because of granitic intrusion and high deformation and permeability that form the secondary aquifer in fractured zones where the groundwater is tapped. According to USEPA [37], the liver stores 1.5 to 2% of ^{238}U, most of which is excreted in a few weeks. In addition, ^{238}U's radioactivity causes chemical toxicity in the kidneys [24]. It has also been found that ^{238}U concentration is restricted by chemical toxicity instead of the effective dose [4]. The World Health Organization (WHO) set a tentative recommendation of 1.0 mSv per year as the recommended threshold in 2003, which equates to 0.5 Bq L^{-1} activity. Except for AB and AC, the findings of this study are greater than the suggested limit for all age categories in all samples.

3.4. Radiological risk assessment of ^{238}U in groundwater from the study area

The cancer risks were calculated using the ^{238}U concentration dispersion in the underground water of each studied location. A cancer mortality rate is the number of deaths in a given population due to cancer as the major cause of death over the course of a year. Cancer mortality is often stated as the number of cancer-related deaths per 100,000 people. That is, the mortality rate is calculated as follows: (Cancer Deaths / Population) / 100,000.

At the same time, morbidity risk is defined as illness and is the term used to refer to measures of illness. Also, it may be defined as the frequency with which a disease appears in a population. The lifetime cancer risks, R, related to consumption of a given

Table 3. Results of annual effective dose of ^{238}U for all the age groups in water sample from the study area and comparing with various countries and international standard.

Sample	AED for Adults ^{238}U (μSvy^{-1})	AED for Adults ^{238}U (12-17 years) (μSvy^{-1})	AED for Adults ^{238}U (7 - 12 years) (μSvy^{-1})
AB	0.012	0.018	0.018
AC	0.012	0.018	0.018
AD	0.151	0.224	0.228
AE	0.106	0.158	0.160
AF	0.134	0.200	0.203
AG	0.656	0.976	0.991
AH	0.086	0.127	0.129
AI	0.086	0.127	0.129
AJ	0.252	0.376	0.382
AK	0.553	0.825	0.837
Council directive [10, 20, 36]	1.0	1.0	1.0

Table 4. The estimated Lifetime cancer mortality and morbidity risk of ^{238}U in water samples.

Sample	Activity concentration	Cancer mortality risk	Cancer morbidity risk
AB	372	1.396×10^{-2}	2.138×10^{-2}
AC	372	1.396×10^{-2}	2.138×10^{-2}
AD	4588	1.722×10^{-1}	2.636×10^{-1}
AE	3224	1.210×10^{-1}	1.853×10^{-1}
AF	4092	1.536×10^{-1}	2.351×10^{-1}
AG	19964	5.616×10^{-1}	8.599×10^{-1}
AH	2604	9.744×10^{-2}	1.496×10^{-1}
AI	2604	9.744×10^{-2}	1.496×10^{-1}
AJ	7688	2.886×10^{-1}	4.442×10^{-1}
AK	16864	4.330×10^{-2}	6.332×10^{-1}
Reference dose level		1×10^{-3}	1×10^{-3}

radionuclide were evaluated by using equation (2) given as [34]:

$$R = r \times l, \tag{2}$$

where r is risk coefficient, l is per-capita activity intake. For radiological risk assessment, the international reference values of cancer risk coefficients of ^{238}U were used for comparison purposes. Note that this type of radiological risk assessment has not been estimated based on Nigeria's population data. According to WHO [12], the average life expectancy at birth in Nigeria is 45.5 years, with an average yearly water usage of 731 liters per person. This equates to 33,215L of water consumed throughout a lifetime. The cancer risk coefficients for ^{238}U are 1.13×10^{-9} Bq^{-1} for mortality and 1.73×10^{-9} Bq^{-1} for morbidity [38, 39]. The cancer mortality and morbidity risks associated with ^{238}U over a lifetime consumption of water in the study location were evaluated using equation (2) and the coefficients, and the results are shown in Table 4. The cancer mortality risk from the intake of water samples varies from 1.40×10^{-2} to 5.6×10^{-1}, with the highest value of 5.6×10^{-1} Bq^{-1} noted in sample AG, whereas the lowest value of 1.40×10^{-2} was reported in samples AB and AC, respectively. Comparing the present study with the International Reference Level of 10^{-3}, the entire water sample is distinctly higher for radiological risk, according to Kim *et al.* [4]. For cancer morbidity risk, it varies from $2.14x10^{-2}$ to 8.60×10^{-1}, with the highest value of 8.60×10^{-1} reported in sample AG; the lowest value of 2.14×10^{-2} was obtained from samples AB and AC, respectively. Comparing the present study with the International Reference Level of 10^{-3}, the entire water sample is distinctly higher for radiological hazard [4]. Such a higher value could be attributed to the weathered nature of the water-bearing formation, which might have affected the geochemical activities of the aquifer.

3.5. Chemical toxicity risk of 238U in groundwater at Awe area

The chemical toxicity risk of groundwater in the Awe area was calculated to evaluate the impact of the carcinogenic associated risk of ^{238}U as a chemical in the water. This was calculated using equation (3), the lifetime average daily dose of ^{238}U obtained via water intake, and the result was equated to the Reference Dose (RFD) of 0.6 gkg^{-1} day^{-1} [31], which is used as a standard criterion for ^{238}U by several foreign organizations, yielding the Lifetime Average Daily Dose (LADD), see equation (3).

$$\text{Ingestion LADD of drinking water} = \frac{EPC \times IR \times ED}{AT \times BW}, \tag{3}$$

Table 5. The estimated lifetime average daily dose (LADD) of ^{238}U in the water samples.

Sample ID	LADD of ^{238}U (μkg^{-1}day^{-1})	Reference dose (μkg^{-1}day^{-1})
AB	8.58×10^{-4}	Present
AC	8.58×10^{-4}	Present
AD	1.06×10^{-2}	Present
AE	7.44×10^{-3}	Present
AF	9.44×10^{-3}	Present
AG	4.61×10^{-2}	Present
AH	6.01×10^{-3}	Present
AI	6.01×10^{-3}	Present
AJ	1.77×10^{-2}	Present
AK	3.75×10^{-2}	Present
U.S. Department of Health and Human Service	6.0×10^{-1}	ATSDR, 2003

where LADD denotes Lifetime Average Daily Dose (μg kg^{-1} day^{-1}), EPC is Exposure Point Concentration (μg L^{-1}), IR is Water Ingestion Rate (L day^{-1}), EF is Exposure Frequency (Day's year ^{1}), ED is Total Exposure Duration (years), AT is Average Time (Days), and BW is Body Weight (Kg).

Therefore using, IR=2 LDay^{-1}, EF= 365days, ED= 45.5y, AT= $16,607.5$ (obtained from 45.5×365) and BW = 70 kg (for a standard man). The chemical toxicity risk for ^{238}U in the study area over a lifetime consumption was estimated and presented in Table 5.

4. Conclusion

From the analysis of activity concentrations, annual effective dose, and radiological risks of ^{238}U in all the studied samples, the Annual Effective Dose (AED) and Lifetime Average Annual Dose (LADD) at a few locations are close to threshold values. In addition, the cancer mortality and morbidity risks are higher than the recommended values suggested by Refs. [3, 10, 39], respectively. Importantly, the cancer mortality and morbidity risks of 5.616×10^{-1} and 8.599×10^{-1}, respectively, when compared to the international reference dose magnitude of 10^{-3}, pose a health risk to the residents who rely on underground water for drinking. Although the average exposure level of ^{238}U in water samples is high in the study area, it is strongly recommended to continuously monitor and treat the water before consumption to reduce the risk of cancer-related diseases. The entire area needs to be thoroughly investigated soon, and stringent remedial measures should be implemented in high-risk locations. This study can be used to investigate any cancer-related diseases in the population and further research on heavy metals and their toxic levels in the region is recommended.

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