



Radionuclide Assessment and Estimation of Radiological Indices in Groundwater Samples from Selected Mining Sites in Osun State, Southwestern Nigeria

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Article Info

Article history:

Received: June 4, 2025

Revised: July 17, 2025

Accepted: July 22, 2025

Keywords:

Contamination,
Groundwater, Mining,
Radiological indices,
Radionuclide
concentration

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ABSTRACT

The radiological contamination of groundwater sources near mining sites has become a significant environmental concern due to the potential risks posed by elevated levels of naturally occurring radioactive materials (NORMs). The study hereby assesses the radioactivity concentrations of 40K, 238U and 232Th in seventy-eight (78) groundwater samples collected from mining sites in the gold belt region of Osun State, Nigeria, using NaI(Tl) based gamma spectrometer and also estimates their associated radiological indices. The results indicated that the mean activity concentrations of all radionuclides exceeded the World Health Organisation's permissible limits of 1.0 Bq/L, raising significant concerns regarding the radiological safety of the water from the area under investigation. The radium equivalent values of 456.75 Bq/L and 272.87 Bq/L were obtained at Olowu and Iganga mining sites, respectively, where contamination levels were highest. The heightened exposure risks underscore the need for urgent interventions to protect public health and the environment in communities affected by mining activities.

INTRODUCTION

Water is among the most vital natural resources on Earth. Hence, all living things need to be sustainable, from the smallest microbes to the largest trees. Water's importance stems from its unique properties and diverse roles in supporting biological processes. Surface water and groundwater are the two major sources of drinking water in the universe (Engelbrecht, 2012), and water from any of these sources is said to be potable provided it is safe to use for domestic purposes without causing damage to human health (WHO, 2011). In addition, it must be aesthetically pleasing

and must be free of harmful concentrations of chemicals, pathogenic microorganisms and radionuclides (WHO, 2011; Akinloye *et al.*, 2018; Isola *et al.*, 2021; Lawal *et al.*, 2023). Radiologically, the distribution of radionuclides in water arises from trace amounts of terrestrial radionuclides comprising the decay series of 238U, 232Th and the singly occurring 40K, most of which are dissolved solids from rocks, soils and mineral deposits (UNSCEAR, 2000). Human activities such as mining, industrial activities and military activities that use or produce man-made radioactive materials

can also contribute to their presence in water (Jibiri *et al.*, 2010; Jibiri *et al.*, 2011; Isola *et al.*, 2019).

The contamination of groundwater sources near mining sites has become a significant environmental concern due to the potential risks posed by elevated levels of naturally occurring radioactive materials (NORMs). Mining activities, especially in the extraction of minerals such as uranium, thorium and radon-emitting ores and sediments, can disturb the natural geochemical balance and lead to the release of radioactive elements into the groundwater system (UNSCEAR, 2000; WHO, 2011; Irunkwor *et al.*, 2022). An activity that does not conform to goal 6, target 6.3 of the United Nations Sustainable Development Goals of minimizing the release of hazardous chemicals and materials into water bodies and improve water quality by reducing pollution globally. Therefore, the occurrence of radionuclides in water may lead to internal exposure in humans, stemming from the decay of these radionuclides that enter the body via ingestion and inhalation (Gorur and Camqoz, 2014). Subsequently, these radionuclides are distributed among the body's sensitive organs based on the metabolism of the corresponding element (Tchokossa *et al.*, 2014, Al-Saadi and Al-Hashimi, 2017; Ononugbo and Ogan, 2017; Arogunjo and Adeniyi, 2020; Isola *et al.*, 2021; Onwuka *et al.*, 2022).

Nigeria engages in considerable mining operations, extracting a range of mineral resources. These activities have made a substantial impact on the nation's economy by generating income, providing employment, and bolstering local industries. However, in view of the fact that mining activities enhance the levels of naturally occurring radionuclides material (NORMs) buried deep beneath the Earth's surface (Isola *et al.*, 2019), it is crucial to assess the extent of contamination and potential environmental hazards at mining sites.

This study, therefore, aims at evaluating the radionuclide concentrations of ⁴⁰K, ²³⁸U and ²³²Th in groundwater samples from mining sites and their radiological implications. The study would help identify whether radiation levels in groundwater at mining sites exceed safe-drinking water standards established by organizations like the World Health Organization (WHO) and national regulatory bodies. This study is essential for safeguarding public health, protecting the environment, and ensuring sustainable mining practices. The results will provide both scientific insight and practical recommendations for managing and mitigating radiation hazards associated with groundwater contamination from mining sites.

METHODOLOGY

Study Area

The study area consists of the gold belt regions in Ilesa and Ijesha region in Osun State, Southwestern Nigeria. Geographically, Ijesha land is situated at approximately latitude 8.92°N and longitude 3.42°E comprising areas such as Ilesa East, Ilesa West, Atakumosa East, and Atakumosa West Local Government Areas, all from which the sample locations were selected for the study. The area is characterized by a forest landscape with significant geological formations, including schist belts that are part of the Nigerian basement complex. These geological features contribute to the region's rich mineral resources, particularly gold, with naturally occurring radioactive materials, which are potential contaminants of air, water and soil.

Study Samples and Preparation

A total of seventy-eight (78) groundwater samples were collected from the ten sampling locations allocated for the study, as shown in Figure 1. These include eight (8) groundwater samples from Igila, Itagunmodi, Iperindo, Oluwo, Aiyetoro, Eyingbin,

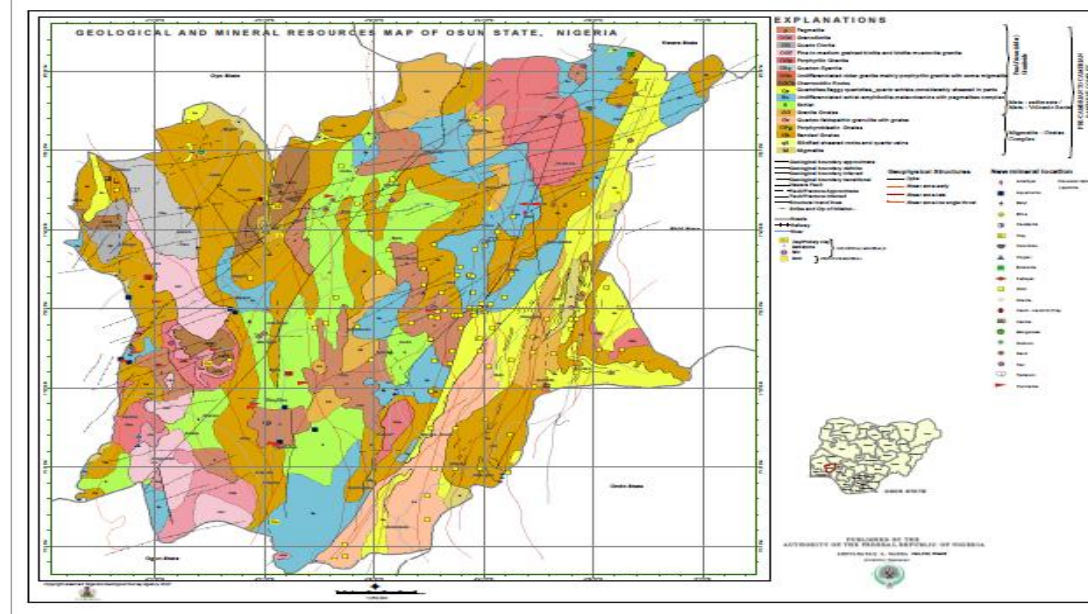


Figure 1: Geological map of the study area

Ajoke and Iganga mining sites, and seven (7) groundwater samples each from Iregun and Okepo mining sites. At each collection, the water samples were then transferred into 2-litre polypropylene containers that had previously been washed with nitric acid (HNO₃) and distilled water. The samples were then acidified with 11M HCl at the rate of 10 ml per litre so as to prevent adsorption of the radionuclides on the container (Akinloye *et al.*, 2018). Thereafter, each sample was packed in an already prepared cylindrical polypropylene container, tightly sealed and kept for a period of 28 days, which was a sufficient time required to attain a state of secular radioactive equilibrium between radium isotopes and their respective daughters before their gamma spectrometry.

Sample Analysis

Potassium-40, uranium-238 and thorium-232 activity concentrations of each water sample were measured using a well-calibrated 3 x 3-inch NaI(Tl) detector coupled to Gamma Spectacular (Model GS-2000 Pro) multi-channel analyser for 18000 s. The analysis of peak areas of gamma spectra and computation was achieved with the aid of

Theremino software. The concentrations of the radionuclides ⁴⁰K, ²³⁸U and ²³²Th were measured through the photo peaks of 1,460 keV for ⁴⁰K, 1,764 keV emitted from the short-lived nuclide ²¹⁴Bi for ²³⁸U, and 2,614 keV emitted from nuclide ²²⁸Ac for ²³²Th, respectively. In addition, an empty container having the same geometry as the sample-filled and the standard-filled container was counted. The background count was subtracted from counts under each corresponding photo peaks of interest (Akinloye *et al.*, 2018; Isola *et al.*, 2021).

Activity Concentration

The activity concentrations of the ⁴⁰K, ²³⁸U and ²³²Th radionuclides in the groundwater samples, according to Ononugbo and Nwaka (2017), were calculated using equation 1.

$$A \text{ (Bq/L)} = \frac{N_C}{\epsilon_\gamma \times I_\gamma \times t \times V} \quad (1)$$

where N_C is the net count at gamma ray energy, ϵ_γ is the emission probability of the radionuclides of interest, P_γ is the efficiency of the detector, t is the total count time in seconds, and V is the sample volume in litres.

Estimation of Radiation Hazard Indices

Radium Equivalent (Raeq)

This index was used to obtain the sum of those activities ^{40}K , ^{238}U and ^{232}Th in (Bq/L) and assess hazards associated with substances that contain ^{40}K , ^{238}U and ^{232}Th in (Bq/L) by using radium equivalent activity and is expressed mathematically by equation 2 (Ikhlas, 2017).

$$Ra_{eq}(\text{Bq/L}) = A_U + 1.43A_{Th} + 0.077A_K \quad (2)$$

where A_U , A_{Th} and A_K are the specific activity in Bq/L of ^{238}U , ^{232}Th and ^{40}K , respectively.

Annual Effective Dose Equivalent

The annual effective dose equivalent (AEDE) resulting from the ingestion of water was estimated using equation 3 based on the assumption that a daily intake of water per person is 2 L d⁻¹ (WHO, 2011) from the following expression (Aladeniyi, *et al.*, 2022; Irunkwo *et al.*, 2022).

$$AEDE (\text{mSvy}^{-1}) = A \times I_W \times DCF \quad (3)$$

where A is the activity concentration of each radionuclide in the groundwater (BqL⁻¹), I_W is the annual intake of groundwater (Ly⁻¹) and DCF is the ingested dose conversion factor for radionuclides (SvBq⁻¹), which varies with both radionuclides and the age of individuals ingesting the radionuclides. The standard dose ingestion conversion factor is equal to 0.28 $\mu\text{Sv Bq}^{-1}$ for ^{228}U , 0.23 $\mu\text{Sv Bq}^{-1}$ for ^{232}Th and 0.0062 $\mu\text{Sv Bq}^{-1}$ for ^{40}K (ICRP, 2012; Ononugbo and Nwaka, 2017).

Lifetime Cancer Fatality Risk (LCFR)

The lifetime cancer fatality risk from the sample to individuals as a result of radiation exposure was estimated using equation 4 (El-Zayat *et al.*, 2022; Mahmoud and El-Zohry, 2020).

$$LCFR = AEDE \times D_L \times C_F \quad (4)$$

where D_L is the average duration of life, estimated to be 70 years for Nigeria, and C_F is the cancer risk factor (Sv⁻¹) i.e., cancer fatality risk per sievert. For

stochastic effects, C_F is 0.05 for members of the public (ICRP, 2012).

RESULTS AND DISCUSSION

Table 1 shows the mean activity concentrations of ^{40}K from the mining sites under consideration in Osun State ranged from 11.18 ± 0.50 Bq/L to 233.29 ± 1.24 Bq/L with a grand mean of 70.96 ± 0.87 Bq/L. The lowest and highest mean activity concentrations of ^{40}K of 6.62 ± 0.21 Bq/L and 422.50 ± 3.02 Bq/L were measured at Iregun and Iganga mining sites. The mean activity concentrations of ^{238}U ranged from 3.16 ± 0.40 Bq/L to 65.47 ± 1.55 Bq/L with a grand mean of 20.48 ± 0.82 Bq/L. The lowest and highest mean activity concentrations of 1.89 ± 0.67 Bq/L and 95.32 ± 1.01 Bq/L at Iregun and Olowu, respectively. The mean activity concentrations of ^{232}Th ranged from 4.04 ± 0.69 Bq/L to 268.92 ± 1.01 Bq/L at Iregun and Olowu mining sites. The lowest and highest values of the mean activity concentrations of 1.09 ± 0.92 Bq/L and 108.12 ± 0.01 Bq/L were obtained from Iregun and Olowu mining sites, respectively. The activity concentration of ^{40}K , ^{238}U and ^{232}Th measured from all mining locations under study in Osun State was above the maximum permissible limit recommended level set by the World Health Organization of 1.0 Bq/L (WHO, 2011). The findings of the study were also compared with both international and local studies on the radioactivity levels of radionuclides in the groundwater from mining sites. For instance, Akuo-ko *et al.*, (2024) reported a comprehensive radiological survey of groundwater resources carried out in artisanal mining communities in the Eastern region of Ghana. In the study, the mean activity concentrations of radionuclides in groundwater samples were 0.1 ± 0.04 Bq/L for ^{226}Ra (^{238}U), 0.36 ± 0.05 Bq/L for ^{228}Ra (^{232}Th), and 1.41 ± 0.10 Bq/L for ^{40}K .

Table 1: Radioactivity concentrations and estimated radiological indices of the investigated groundwater samples

LOCATION	Activity concentration (Bq/L)			Raeq (Bq/L)	Annual effective dose equivalent (mSv/y)			LCFR
	⁴⁰ K	²³⁸ U	²³² Th		Infant	Children	Adult	
IGILA	28.99	19.91	32.09	68.03	3.34E-04	2.92E-05	3.56E-05	1.25E-04
ITAGUNMODI	52.52	14.8	30.83	62.93	5.73E-04	2.24E-05	2.79E-05	9.78E-05
OLOWU	233.29	54.24	268.92	456.75	2.58E-03	9.78E-05	1.34E-04	4.69E-04
IPERINDO	48.48	6.91	52.29	85.42	5.29E-04	1.43E-05	2.07E-05	7.25E-05
IREGUN	11.18	3.59	4.04	10.23	1.21E-04	5.07E-06	6.05E-06	2.12E-05
AYETORO	29.37	9.79	13.41	31.23	3.20E-04	1.41E-05	1.70E-05	5.95E-05
EYINGIBIN	25.13	3.16	4.14	11.02	2.66E-04	4.53E-06	5.46E-06	1.91E-05
AJOKE	26.94	7.51	11.93	26.64	2.92E-04	1.10E-05	1.34E-05	4.69E-05
IGANGA	219.77	65.47	133.2	272.87	2.41E-03	9.86E-05	1.23E-04	4.31E-04
OKEPO	33.88	19.44	35.56	72.9	3.86E-04	2.89E-05	3.56E-05	1.25E-04
MIN	11.18	3.16	4.04	10.23	5.73E-04	5.07E-06	6.05E-06	9.78E-05
MAX	233.29	65.47	268.92	456.75	2.58E-03	9.86E-05	1.34E-04	4.69E-04
MEAN	70.96	20.48	58.64	109.8	7.81E-04	3.26E-04	4.19E-05	1.47E-04

In the study, the mean activity concentrations of radionuclides in groundwater samples were 0.1 ± 0.04 Bq/L for ²²⁶Ra (²³⁸U), 0.36 ± 0.05 Bq/L for ²²⁸Ra (²³²Th), and 1.41 ± 0.10 Bq/L for ⁴⁰K. These values were lower than the values obtained in the current study. Similarly, Oketayo *et al.* (2019) reported that the mean concentrations of ²³⁸U, ²³²Th and ⁴⁰K in water samples from gold mining site at Mokuro-Ile-Ife axis of Osun State are higher than their permissible level in water. In addition, Isinkaye *et al.* (2025) reported that the average concentrations of ⁴⁰K, ²³⁸U, and ²³²Th in water samples from artisanal mining sites within Ife-Ilesha schist-belt, Osun State are far higher than their permissible level in water.

Thus, the high values recorded for the radionuclide activity concentration in the studied mining sites can be attributed to the high concentration of radionuclides in the soil of these mining sites and mining activities. For example, Ademola *et al.* (2014) reported that the radioactivity concentration

of ²³⁸U, ²³²Th and ⁴⁰K was higher in the mining sites than in non-mining areas. In addition, Isola *et al.* (2019) and Amodu *et al.* (2024) both reported that the activity concentrations from the soil samples in the mining site in Iperindo were higher than the world average value. All these can be due to the high-density dust generated from the mining processes, which may raise the possibility of exposure to NORMs from soil, water and air.

Estimated Radiological Hazard Indices

Radium Equivalent

The means of the radium equivalent activity (R_{eq}) ranged from 10.23 Bq/L to 456.75 Bq/L at Iregun and Olowu mining sites, respectively, with a mean value of 109.80 Bq/L. The R_{eq} values of 10.23 Bq/L, 31.23 Bq/L, 11.02 Bq/L, and 26.64 Bq/L were estimated for Iregun, Aiyetoro, Eyingbin, and Ajoke mining sites in Osun State, respectively. These results were comparable to the results obtained from the study on the radiological health risk evaluation of domestic sources of water in selected lead mining

communities of Nasarawa State, North Central, Nigeria, where the Ra_{eq} varied from 3.80 - 4.76, 1.47 - 12.09, 1.47 - 22.31, 2.45 - 13.40 and 9.50 - 10.02 Bq/L for sampled pits, wells, boreholes, streams and ponds water sources respectively. The Ra_{eq} for all the sampled water sources were generally low compared to the recommended permissible limit of 49 Bq/L for domestic water (UNSCEAR, 2010; WHO, 2011; Onwuka *et al.*, 2022). The radium equivalent activity of 68.03 Bq/L, 62.93 Bq/L, 456.75 Bq/L, and 272.87 Bq/L obtained from Igila, Itagunmodi, Olowu and Iganga mining sites were higher than the recommended permissible limit of 49 Bq/L (UNSCEAR, 2000; 2008).

Annual Effective Dose Equivalent

The annual effective dose equivalent, as presented in Figure 2, for infants ranged from 5.73×10^{-4} mSv/y to 2.58×10^{-3} mSv/y at Itagunmodi and Olowu mining sites in Osun State, with a mean of 7.81×10^{-4} mSv/y. The annual effective dose on children ranged from 5.07×10^{-6} mSv/y to 9.86×10^{-5} mSv/y Iregun and Iganga mining sites with a mean of 3.26×10^{-4} mSv/y while the annual effective dose on adults ranged from 6.05×10^{-6} mSv/y to 1.34×10^{-4} mSv/y at Iregun and Olowu with a mean of 4.19×10^{-5} mSv/y.

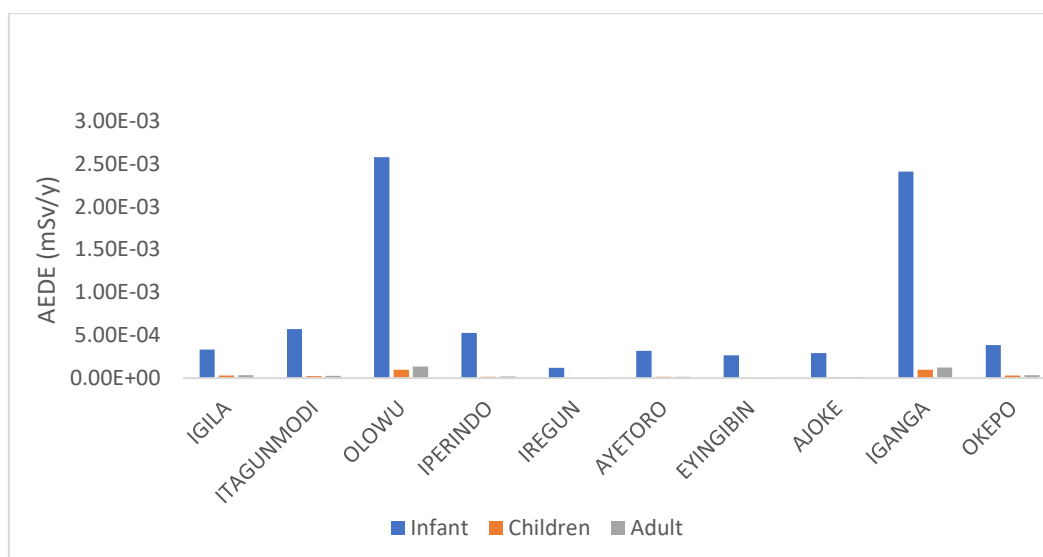


Figure 2: Distribution of annual effective dose equivalent in infant, children and adults

Lifetime Cancer Fatality Risk

The lifetime cancer fatality risk, as shown in Figure 3, on adults ranged from 9.78×10^{-5} to 4.69×10^{-4} with a mean of 1.47×10^{-4} . The lowest and highest lifetime cancer fatality risks were estimated at Itagunmodi and Olowu mining sites, respectively. The results of this study are in agreement with the study carried out on environmental health risk assessment due to radionuclides and metal(loid)s for Igdir province in Anatolia, near the Metsamor nuclear power plant wherein the annual cumulative

biologic effective dose due to radiologic exposure that arises from radioactivity in drinking water in the region was determined as $27.83 \pm 24.81 \mu\text{Sv}$. Accordingly, the average estimated excess cancer risk value was determined as $15.24 \pm 13.6 \text{E-}05$ (Karahan, 2020). It has been reported that from a study that the annual effective dose equivalent (α and β) and excess lifetime cancer risk (α and β) of water samples from a mining site in Jayfi, Paga Tungan Goro of Minna, Niger State, Nigeria were $0.013237/0.645534$ mSv/y and $0.280742/1.77 \times 10^3$ respectively (Abdulkarim *et al.*, 2023). Results from

another study showed that the calculated total annual effective dose due to both ingestion and inhalation for different age groups ranges from 42.40, 57.60 and 65.30 ($\mu\text{Sv/y}$) to 457.10, 622.06 and 704.60 ($\mu\text{Sv/y}$), with an average value of 203.32, 276.70 and 313.51 ($\mu\text{Sv/y}$) for adults,

children, and infants, respectively. The total excess lifetime cancer risk for different age groups varies from 0.00149, 0.00202 and 0.0247 to 0.0160, 0.0220 and 0.0247 with mean values of 0.00703, 0.00956 and 0.0109 for adults, children, and infants. (Hannafi *et al.*, 2024).

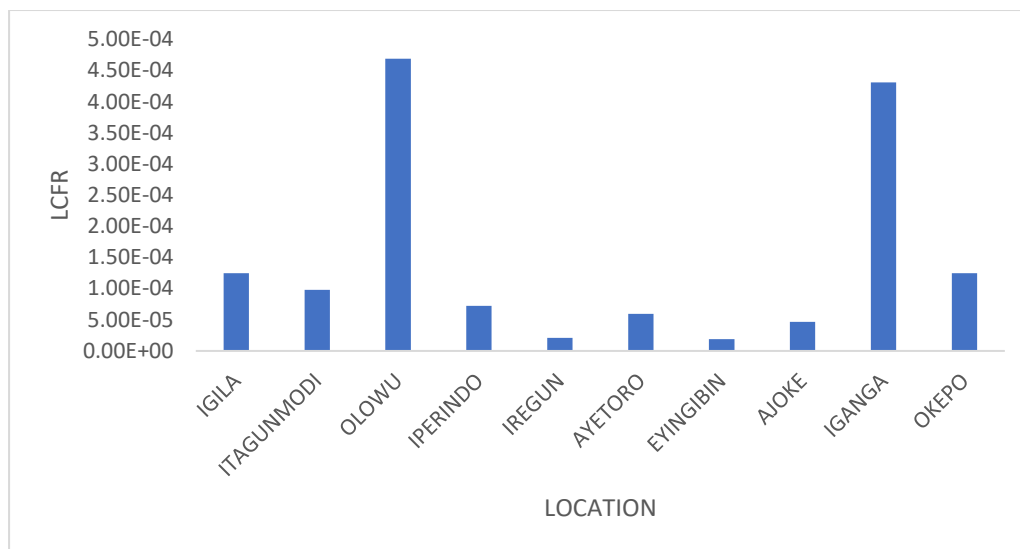


Figure 3: Distribution of lifetime cancer fatality risk for adults across the study locations

CONCLUSIONS

The investigation into the activity concentrations of radionuclides (^{40}K , ^{238}U , and ^{232}Th) in groundwater from mining sites in Osun State revealed concerning levels of contamination that were higher than the safety limits established by the World Health Organization. The findings indicated that these elevated radionuclide concentrations can pose significant risks to human health, including a heightened potential for chronic diseases and various forms of cancer among local populations. Specifically, the calculated radium equivalent activity highlighted alarming radiation exposure levels, particularly in sites like Olowu and Iganga. However, annual effective doses of all the categories of ages investigated and the estimated lifetime cancer fatality of adults were below the recommended safety limits for the public. Based on findings from this study, it is imperative to implement effective monitoring strategies and

regulatory measures to address the potential health risks associated with groundwater contamination from mining activities. Additionally, educating local communities about the dangers of consuming contaminated water is essential for enhancing public health awareness and safety of people in mining-affected regions in order to ensure safe drinking water for affected communities.

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