



Spectroscopic Analysis of Radionuclide Concentrations and their Health Implications in Water from Artisanal Mining Sites, Ile-Ife, Nigeria

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Abstract

Water is a vital resource that plays a crucial role in human life. Anthropogenic activities, particularly mining, contribute significantly to the increased concentrations of naturally occurring radioactive materials in water sources, thereby posing potential health risks. Compared with other regions where risk assessment studies have been conducted in Nigeria, the gold mining area in Ile-Ife requires a comprehensive evaluation of radionuclide risks. This study's objective is to evaluate the concentration level of radionuclides and the related health risks in water from gold mining locations in Ile-Ife. In total, 30 water samples were analyzed to determine radionuclide activity concentrations and assess radiological safety indices. The mean radioactivity concentrations of radionuclides in Bq/L were found to be 135.78 ± 2.52 , 11.17 ± 0.64 , and 9.69 ± 0.24 for ^{40}K , ^{232}Th , and ^{238}U , respectively, which were notably elevated compared to the WHO tolerable exposure levels for drinking water. The yearly radiation dose absorbed by the population is 0.04 mSv/y, posing no significant health concerns as it remained below the 1 mSv/y international safety limit. On average, annual effective dose equivalent ingested for adults (2.81 mSv/y) exceeded the UNSCEAR limit, while it remained below for children (1.35 mSv/y). Excess lifetime cancer risk values were 11.27×10^{-3} for adults and 5.41×10^{-3} for children, indicating a substantial overall cancer risk due to the consumption of water from these mining sites. Statistical analysis of the data analyzed in this study revealed a significant positive correlation ($p < 0.05$), suggesting a strong correlation between primordial radionuclides present in water samples and the assessed radiological safety indices.

Keywords Artisanal mining, water contamination, radionuclide concentrations, environmental radioactivity, health risks

1. Introduction

Radiation is the breakdown of unstable atomic nuclei, characterized by the release of particles or energy [1,2], and is extensively used in diverse fields, such as medicine, industry, and research. Its diverse uses include cancer treatment, medical equipment sterilization, carbon dating of archaeological artifacts, and smoke detection [3]. While these applications offer significant benefits, the environmental consequences of radiation are complex issues that affect human health, wildlife, aquatic ecosystems, and the overall ecological balance [4]. The presence of radioactive contaminants, both natural and anthropogenic, has emerged as a substantial environmental concern with profound implications for ecosystems [5–7]. Assessing radionuclides is crucial for estimating the threats humans face, particularly from anthropogenic activities. Sectors, such as mining, mineral exploration, and milling, contribute to increased background radioactivity levels in our surroundings, highlighting the need for comprehensive radionuclide assessments.

Artisanal mining activities have drawn global attention owing to their contribution to environmental radioactivity. Artisanal miners, often characterized by their utilization of rudimentary tools such as pans, chisels, hammers, and shovels, engage in small-scale mining activities to extract valuable minerals, such as gold. Despite the documented global threat posed by environmental degradation resulting from the unskilled practices of these miners [8–12],

their activities persist unabated. Extensive research has also focused on understanding the characteristics of long-lived radionuclides (such as ^{40}K , ^{232}Th , ^{238}U) in water, soils, and plants [13–20]. These radionuclides are transferred from soils to crops on land, and from water to aquatic organisms in rivers, lakes, and seas through osmotic and diffusion processes. The interaction between humans and the environment facilitates the entry of these radionuclides into the human digestive tract via ingestion, skin contact, and other pathways [21–23]. Over an extended period, these radionuclides tend to bioaccumulate in humans, posing potential risks, such as cancer, liver damage, impairment, and genetic mutations, as evidenced by incidents such as Fukushima and Chornobyl [24–28].

Water, constituting the vast majority of the Earth's surface, reaching over 70%, is a vital and indispensable natural resource, fostering a conducive environment and providing sustenance for all life forms on Earth. Surface and groundwater movement is crucial for transporting NORMs across different environmental layers. Unfortunately, human activities, particularly mining, have contaminated these water sources over time, contributing to elevated levels of NORMs and further increasing the overall radioactivity in these water sources [29–31]. Therefore, the evaluation of environmental and public health necessitates the assessment of radioactivity levels in water. ^{238}U isotopes exhibit greater water solubility than ^{232}Th , resulting in increased mobility within groundwater [32,33]. In mining areas, contaminated materials can be mobilized by water, which percolates through rock formations and soil layers, leading to the degradation of water sources. In sub-Saharan African countries such as Nigeria, rural populations face challenges in accessing potable drinking water, as many communities depend on natural sources such as rivers, lakes, and streams for their water supply [34–37].

Unfortunately, the exploration of valuable minerals in various parts of the globe has led to pollution of water sources, rendering them unsuitable for consumption. Clean water sources that were formerly accessible have become increasingly challenging because of the continuous activities of artisanal miners. This challenge has prompted extensive global research on the impact of artisanal mining on water sources [9,12,16,19,36,38–43]. Liu et al. [44] investigated radionuclide concentrations in water sources near a uranium tailing reservoir in China and found that it exceeded the reservoir's concentrations and surpassed national and global average values. The study also observed that radionuclides in soil tailings diffused into the surrounding water, posing a considerable potential danger to the health of residents. A similar study in South Africa by Kamunda [16] on water samples collected around gold mining sites in Gauteng province revealed elevated radionuclide concentrations in surface water samples, followed by borehole water. Although the radiological hazard index results are below the global permissible limits, this outcome is anticipated because of South Africa's advanced water treatment system, reflecting its urban and industrial significance.

Several investigations have been undertaken in Nigeria to evaluate the radioactivity emanating from artisanal mining areas and its radiological health impact [9,10,21,30,34,35,45–50]. Akpanowo et al. [49] assessed radionuclide concentration in water sources from artisanal mines in Anka, Northwestern Nigeria. The findings revealed activity concentration values below the World Health Organization (WHO) thresholds, indicating an insignificant lifetime cancer risk for adults and children. Muhammad et al. [30] observed traces of radionuclides in drinking water collected from tin mining sites in northwestern Nigeria, with concentrations in both surface and groundwater exceeding the International Atomic Energy Agency (IAEA) drinking water standard [51], but within the recommended safety guidelines established by the WHO. These findings underscore hidden health risks to communities residing in the proximity of these mining areas. It emphasizes the importance of assessing water sources around mining areas to illuminate potential threats to public health, the community, particularly adults and children, and mine workers who rely on these water sources, especially during dry seasons when water scarcity is prevalent.

A careful analysis of the existing literature reveals a notable gap in the data concerning the potential adverse health impacts of naturally occurring radioactive components present in the surface and groundwater at the Ile-Ife gold mining site. This study seeks to fill this gap by estimating the radionuclide concentrations in water sources within gold mining sites and assessing the potential health risks for adults and children in the Ile-Ife mining area. Given that the waters surrounding these mining sites are ingested by both mine workers and local inhabitants, who rely on streams and rivers flowing through these sites for various activities, this study aims to present comprehensive data on radionuclide concentrations and the associated health implications in water from the mining site in Ile-Ife, Osun, Nigeria.

2. Wastewater

2.1 Study location

The study location is Alaagba village, in Ile-Ife, Osun, Southwestern Nigeria, with geographical coordinates of longitude (4.579°E) and latitude (7.384°N). Ile-Ife lies within the Ife-Ilesha schist belt, a geological formation within the Archean-Proterozoic crystalline basement in Southwestern Nigeria [52–54]. The area encompasses a gneissic migmatite assemblage and metavolcanic-sedimentary rocks, primarily characterized by gray and granite gneiss, which form a significant part of the landscape. Notably, the gneissic rock with granite composition appears as an elevated peak intruding into the gray gneiss. The metavolcanic-sedimentary rocks in the tectonic boundary with the Ile-Ife gneissic migmatite assemblage consist mainly of micaceous schists and are situated within the western part of the Ife-Ilesha schist belt in Southwestern Nigeria [55,56].

Micaceous schists comprise minerals such as crystalline silica, potassium mica, ferrous mica, cross-shaped crystals, deep-red minerals, and fibrous aluminosilicate [57]. Geological features such as flexures, fractures, displacements, and localized stretching are observable in various rock formations, signifying multiple deformation episodes, likely relicts of the Pan-African Orogenic event. The lithological characteristics, tectonic features, and presence of gold-bearing eluvial deposits contribute significantly to small-scale gold mining practices in the area. Alaagba Village, primarily a residential community with agriculture as the main occupation, is characterized by a low-income population. However, artisanal mining activities for gold have led to noticeable environmental consequences, including vegetation cover loss, soil quality decline, and the degradation of water and soil resources, disrupting Earth's terrestrial systems.

2.2 Sample acquisition and preparation

A random sampling procedure was employed to collect 30 water samples for three consecutive months from water bodies within a 2 km radius of the gold mines at Ile-Ife, Osun State, Nigeria. Each month, ten water samples were obtained from the surface water bodies and discharged at the bottom of the tailings. The samples were carefully poured into 750 mL high-density polyethylene (HDPE) bottles and labeled appropriately for easy identification during analysis. The GPS coordinates for each water sample collection point were identified using a GPS locator, as depicted in Fig. 1. To minimize the sequestration of radionuclides of interest within the container lining and inhibit microbial growth, 0.1 M HCl was added to the HDPE water sample bottles. Subsequently, 500 mL aliquots of each water sample were transferred into beakers securely sealed with cellulose wrap and left undisturbed for 30 days to establish a steady-state distribution between the short-lived radionuclide derivatives, following the method described by [44,49,58,59].

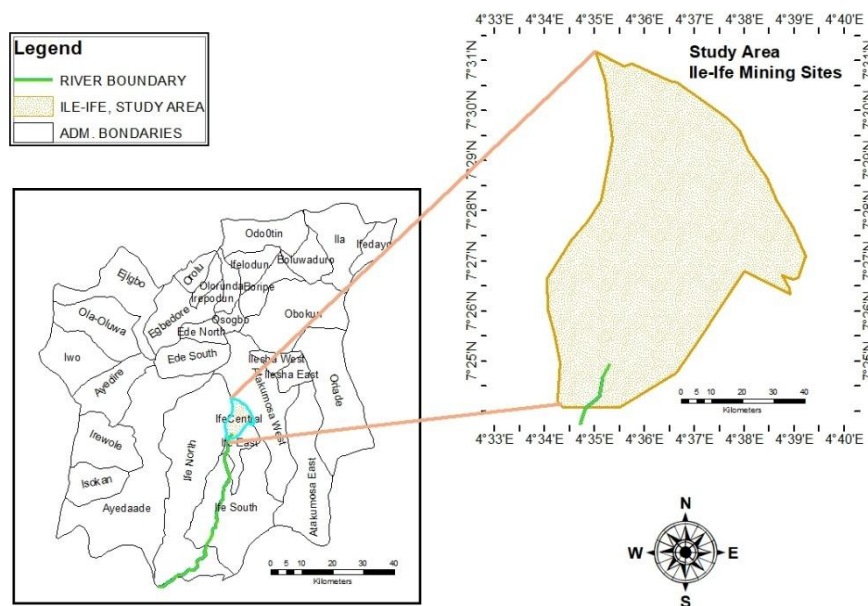
2.3 Radionuclide activity concentrations

Radionuclide activity concentrations of water samples from Ile-Ife gold mining sites was measured using gamma (γ) spectrometry. The gamma-ray detection system integrates a 3-inch by 3-inch sodium-iodide (NaI)(TI) sensor manufactured by Princeton Gamma Technology, USA. The sensor is shielded by a tubular lead enclosure to mitigate the influence of ambient radiation and connected to a Gamma multichannel analyzer (model GS-2000 Pro), which, in turn, is interfaced with a computer for visual monitoring. The Theremino software was used to acquire data and analyze the gamma-ray spectra. The spectrometry system was calibrated to establish a direct correlation between the spectrum peak intensity and gamma-ray energy. Energy calibration of the sensor was achieved through the use of the RSS8 gamma source kit, which is a primary reference material from Spectrum Techniques LLC, USA. This calibration process encompassed the acquisition of gamma-ray spectra from point sources with well-defined energies (^{137}Cs and ^{60}Co).

Additionally, to determine the detector's efficiency, a reference source emitting gamma rays from nuclides with known activities was used. These standard sources, designed to determine natural radionuclides in environmental matrices, were counted for 36,000 s. The total energy peak efficiency, denoted by ε , was utilized, correlating the spectrum peak point to the radioactivity level detected. By determining the net peak count (N_p) and the activity concentration (A_c) for each radioactive isotope within the source, along with considering absolute gamma-ray emission probability (P_γ) of specific radionuclides of interest, the volume of the sample (V), and the acquisition time (T), we estimated the efficiency of the total peak energy using [60,61]:

$$\varepsilon = \frac{N_p}{A \times P_\gamma \times V \times T} \quad (1)$$

Fig. 1. Map of Osun, Southwestern Nigeria highlighting the study area.



Before measuring the water samples, an empty container was subjected to a 36,000-second count to establish the count of background gamma-ray distribution. After reaching secular equilibrium, each sealed water sample was individually positioned on the detector for analysis. The limit of detection (LOD) of the detector was estimated from the background count (B_c) using Eq. (2). Each water sample obtained from the Ile-Ife mining site underwent counting for the same duration as the vacant container. The essential features of the radionuclides employed to identify the most notable ones within the samples are as follows: 1460 keV for ^{40}K , 1765 keV for ^{214}Bi (^{238}U), and 2615 keV for ^{208}Tl (^{232}Th). The radioactivity level in Bq/L of each detected radioactive isotope in the water sample was determined using Eq. (3) [62].

$$LOD = 2.71 + 4.66\sqrt{B_c} \quad (2)$$

$$A = \frac{N_p}{P_y \times V \times \varepsilon \times T} \quad (3)$$

2.4 Radiological safety indices

The radiological safety indices estimated in this study include the absorbed dose rate (D), annual effective dose rate (AEDR), radium equivalent (Ra_{eq}), external hazard index (H_{ex}), internal hazard index (H_{in}), total annual effective dose equivalent from ingestion of water sample (AEDE_{ing}), and excess lifetime cancer risk (ELCR).

The UNSCEAR guidelines [63] can be used to estimate the D in air from gamma radiation at an elevation of one meter above ground level, considering a homogeneous dispersion of primordial radionuclides (^{40}K , ^{232}Th , and ^{238}U). By assuming contributions from other primordial radionuclides to be negligible, D is estimated using [4,63,64]:

$$D \text{ (nGyh}^{-1}\text{)} = 0.042A_K + 0.604A_{Th} + 0.462A_U \quad (4)$$

AEDR provides an efficient method to evaluate the radiological risk to which the population of the mining area are exposed. The AEDR can be computed using 0.2 as the outdoor occupancy factor (OF), 0.7 SvGy⁻¹ as dose conversion factor (DCF), 8760 hours as time (T), and D . Therefore, AEDR for outdoor terrestrial gamma radiation is evaluated using [63,65,66]:

$$AEDR \text{ (}\mu\text{Svy}^{-1}\text{)} = OF \times DCF \times D \text{ (nGyh}^{-1}\text{)} \times T \times 10^{-3} \quad (5)$$

The Ra_{eq} value represents the gamma radiation dose absorbed from a mixture of naturally occurring radioactive elements. It is the weighted sum of the radioactivity concentrations for ^{40}K (A_K), ^{232}Th (A_{Th}), and ^{238}U (A_U) at 4810, 259, and 370 Bq/L, respectively given that these radionuclides generate equal dose rate of gamma rays [16], thus, Ra_{eq} is estimated as [64,67]:

$$Ra_{eq} = A_U + 1.43A_{Th} + 0.077A_K \quad (6)$$

The H_{ex} functions as a tool for assessing the potential health risks resulting from radionuclides emission of gamma rays. It is crucial to ensure that the value of H_{ex} does not surpass unity, as this criterion was introduced as a model to restrict radiation exposure from primordial radionuclides in water sources to a single dose equivalent threshold. In this study, the H_{ex} is estimated using [68,69]:

$$H_{ex} = (27.03 \times 10^{-4} A_U) + (38.61 \times 10^{-4} A_{Th}) + (2.08 \times 10^{-4} A_K) \leq 1 \quad (7)$$

The H_{in} signifies the potential risks of radon decay products regarding intrinsic exposures within cellular materials. For the radiation threat to be deemed insignificant, the H_{in} value must be strictly less than unity. In this study, the H_{in} is estimated using [70,71]:

$$H_{in} = (54.05 \times 10^{-4} A_U) + (38.61 \times 10^{-4} A_{Th}) + (2.08 \times 10^{-4} A_K) \leq 1 \quad (8)$$

The annual AED_{ing} of ^{40}K , ^{232}Th , and ^{238}U through water samples is calculated for children and adults within the mining population area. This estimation is based on the quantities of radioactivity of each radionuclide present in the sample. The AED_{ing} for the water samples obtained in this study was estimated using the yearly water consumption rates of 730 L and 350 L, respectively for adults and children [72]. By applying the average water consumption rates per year (IRW_{ing}) for adults and children, along with the activity concentration (A) and effective dose coefficient (DCF_{ing}) values in Sv/Bq of 6.20×10^{-9} for ^{40}K , 2.30×10^{-7} for ^{232}Th , and 4.50×10^{-8} for ^{238}U in accordance with the International Commission on Radiological Protection (ICRP) [73], AED_{ing} is estimated using [47,74]:

$$AED_{ing} = A \times IRW_{ing} \times DCF_{ing} \quad (9)$$

Gamma rays generated in water can potentially lead to carcinogenic effects through routine interactions and consumption of NORM. The ELCR was calculated by utilizing the outdoor annual effective dose (AED) over a lifetime duration (DL) of 55 years for Nigeria [30,61]. The cancer risk factor (RF) employed was 0.05 S/v, per the ICRP guidelines [75]. It is essential to note that this particular value in the context of low background radiation is recognized as causing stochastic effects in the case of public exposure [49]. In this study, the ELCR is estimated using [4,64,76,77]:

$$ELCR = AED \times DL \times RF \times 10^3 \quad (10)$$

3. Results and Discussions

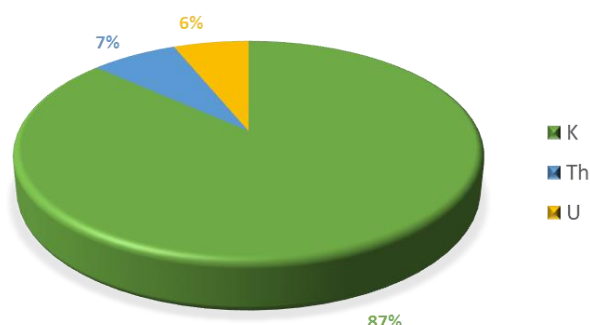
3.1 Radionuclide Activity concentration

The results of γ -spectroscopy measurements, conducted to assess radioactivity concentration of ^{238}U , ^{232}Th , and ^{40}K in water samples from Ile-Ife mining sites, are presented in Table 1. For three months of evaluation, activity concentrations for ^{40}K varied from 29.95 ± 1.21 to 304.08 ± 3.91 Bq/L, with 135.78 ± 2.52 Bq/L as the mean, for ^{232}Th , it ranged from 2.95 ± 0.33 to 23.09 ± 1.08 Bq/L, with 11.17 ± 0.64 Bq/L as the mean, and for ^{238}U , it varied from 1.86 ± 0.10 to 21.17 ± 0.40 Bq/L, with a mean of 9.69 ± 0.24 Bq/L. The peak activity concentrations for all radionuclides were observed in December (month three), with mean Bq/L values of 158.74 ± 2.79 , 12.04 ± 0.66 , and 10.63 ± 0.26 , respectively, for ^{40}K , ^{232}Th , and ^{238}U . November recorded the lowest radionuclide activity concentrations, with mean values of 158.74 ± 2.79 Bq/L, 12.04 ± 0.66 Bq/L, and 10.63 ± 0.26 Bq/L for ^{40}K , ^{232}Th , and ^{238}U , respectively.

The observed unusual climate pattern includes a late cessation of rainfall in October 2022, followed by a resurgence of rainfall primarily in November 2022, and the onset of the dry season in December 2022. This pattern highlights the significant impact of climate change, which makes it increasingly challenging to predict Nigeria's dry and rainy seasons. The elevated radionuclide concentrations observed in December can be attributed to decreased water levels, mainly due to rapid evaporation under intense sunlight. This observation has implications for public health during the dry season, when the availability of drinking water sources diminishes. The local community relies more on water from flowing streams and rivers, which cuts through mining channels for their daily activities. The decrease in radionuclide concentrations in November was primarily caused by increased rainfall. Rainfall has a dilution effect on the radionuclide concentrations in water, which can result in a temporary decrease in its concentration. Otherwise, the trend would have shown a consistent increase in radionuclide concentrations as the effects of the dry season intensified. Fig. 2 is a chart showing the percentage distribution of primordial radionuclides found in water samples collected from artisanal mine sites in Ile-Ife.

Table 1. Radionuclides activity concentration in water samples from Ile-Ife mine locations.

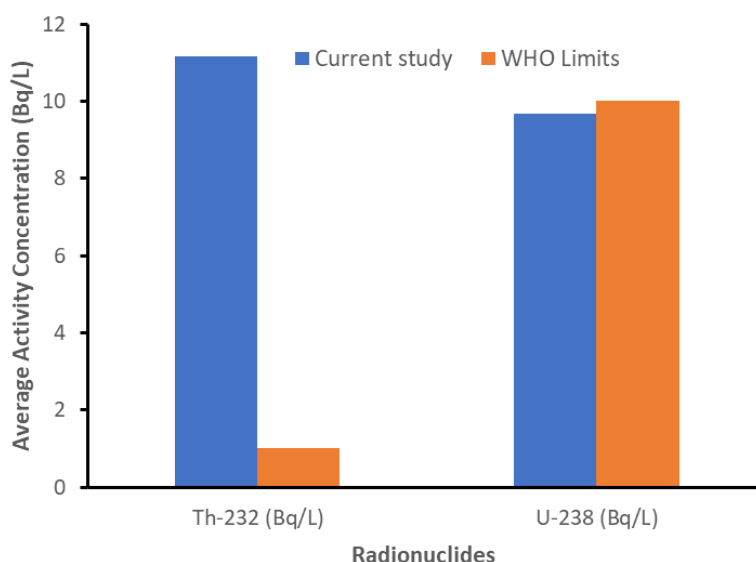
| Sample no | Sample Size | ⁴⁰ K (Bq/L) | | ²³² Th (Bq/L) | | ²³⁸ U (Bq/L) | |
|--------------------|-------------|------------------------|----------------|--------------------------|--------------|-------------------------|--------------|
| | | Mean | Range | Mean | Range | Mean | Range |
| SM-1 | 10 | 133.35 ± 2.45 | (29.95-304.08) | 11.42 ± 0.65 | (4.17-21.26) | 10.08 ± 0.24 | (2.95-18.94) |
| SM-2 | 10 | 115.26 ± 2.32 | (41.85-241.35) | 10.04 ± 0.61 | (2.95-20.06) | 8.36 ± 0.22 | (1.86-17.92) |
| SM-3 | 10 | 158.74 ± 2.79 | (53.05-284.43) | 12.04 ± 0.66 | (3.04-23.09) | 10.63 ± 0.26 | (1.94-21.17) |
| Mean | | 135.78 ± 2.52 | | 11.17 ± 0.64 | | 9.69 ± 0.24 | |
| Min | | 29.95 ± 1.21 | | 2.95 ± 0.33 | | 1.86 ± 0.10 | |
| Max | | 304.08 ± 3.91 | | 23.09 ± 1.08 | | 21.17 ± 0.40 | |
| Standard deviation | | 86.68 | | 6.71 | | 6.31 | |
| Kurtosis | | -1.15 | | -1.41 | | -1.38 | |
| Skewness | | 0.46 | | 0.35 | | 0.32 | |

Fig. 2. Percentage variation of primordial radionuclides in water samples from Ile-Ife mining sites.**Table 2.** Evaluating the average radioactive content of different radionuclides in water samples of Ile-Ife gold mining sites and those around the globe.

| Location | Activity Concentrations (Bq/L) | | | | Ref. |
|--|--------------------------------|-------------------|------------------|-------------------|------------|
| | ⁴⁰ K | ²³² Th | ²³⁸ U | ²²⁶ Ra | |
| Tin mining areas, Northwest Nigeria | 6.15 | 4.35 | - | 5.88 | [78] |
| Asikam-gold mining areas, Ghana | 2.44 | 0.30 | - | 0.49 | [42] |
| Uranium tailings reservoir, China | 2.17 | 0.44 | 4.73 | 3.92 | [44] |
| Gold mine, Gauteng Province, South Africa | 7.36 | 0.56 | 0.66 | - | [16] |
| Biseni flood plains, Niger-Delta, Nigeria | 97 | 12 | - | 12 | [79] |
| WHO | N/A | 1 | 10 | 1 | [80] |
| Gold mining areas, Ile-Ife, Southwest, Nigeria | 135.78 | 11.17 | 9.69 | - | This study |

Similar studies around mining areas like Anka in Northwest Nigeria reported an average activity concentration of 15.74 ± 3.4 and 28 ± 7.2 mBq/L, respectively, for α and β gross values for stream water samples [49], which are far less than what was obtained in our study. Gross α activity is linked mainly to dissolved uranium isotopes (^{238}U , ^{235}U , and ^{234}U) and ^{226}Ra , while the primary source of gross β activity is linked to ^{40}K and short-lived progenies of ^{234}Th and ^{238}U [33]. The result of this study is lower when compared to Atule et al. [42], Nasiru et al. [78] and Liu et al. [44] for studies conducted in Asikam-gold mining community in Ghana, tin mining areas in Northwest Nigeria, and uranium tailings reservoir in China. However, the results were similar to that reported by Algalagba and Onoja [79] for the Niger Delta flood plains in Nigeria. This comparative study suggests that the radionuclides concentrations are mostly influenced by the geological distribution of the region being analyzed.

Fig. 3. Radioactivity concentration of ^{232}Th and ^{238}U compared with WHO limits.



Mean activity concentration of ^{40}K in our study at a significant value of 135.78 ± 2.52 Bq/L is consistent with those reported in related studies, as indicated in Table 2. However, ^{40}K is a natural radionuclide that is challenging to control, and is typically not considered when establishing regulatory standards [42]. Consequently, our research emphasizes ^{232}Th and ^{238}U as elements with distinctive characteristics and potential health implications. The WHO recommended protection limits for radionuclides (^{232}Th and ^{238}U) in ingested water are 1 Bq/L and 10 Bq/L, respectively [80]. From Fig. 3, it is evident that the radioactivity concentration of ^{232}Th obtained is significantly greater than the global permitted threshold of 11.17 Bq/L. Although the activity concentration of ^{238}U is approximately 3.1% lower than the WHO-established limits, this value illuminates a hidden threat in the radionuclide concentration found in water sources around the artisanal mining sites in Ile-Ife. This concern is particularly pronounced with ^{232}Th and ^{238}U , which are able to leach into groundwater, potentially contaminating drinking water sources. This issue becomes acute during the dry season when water scarcity is prevalent in these communities.

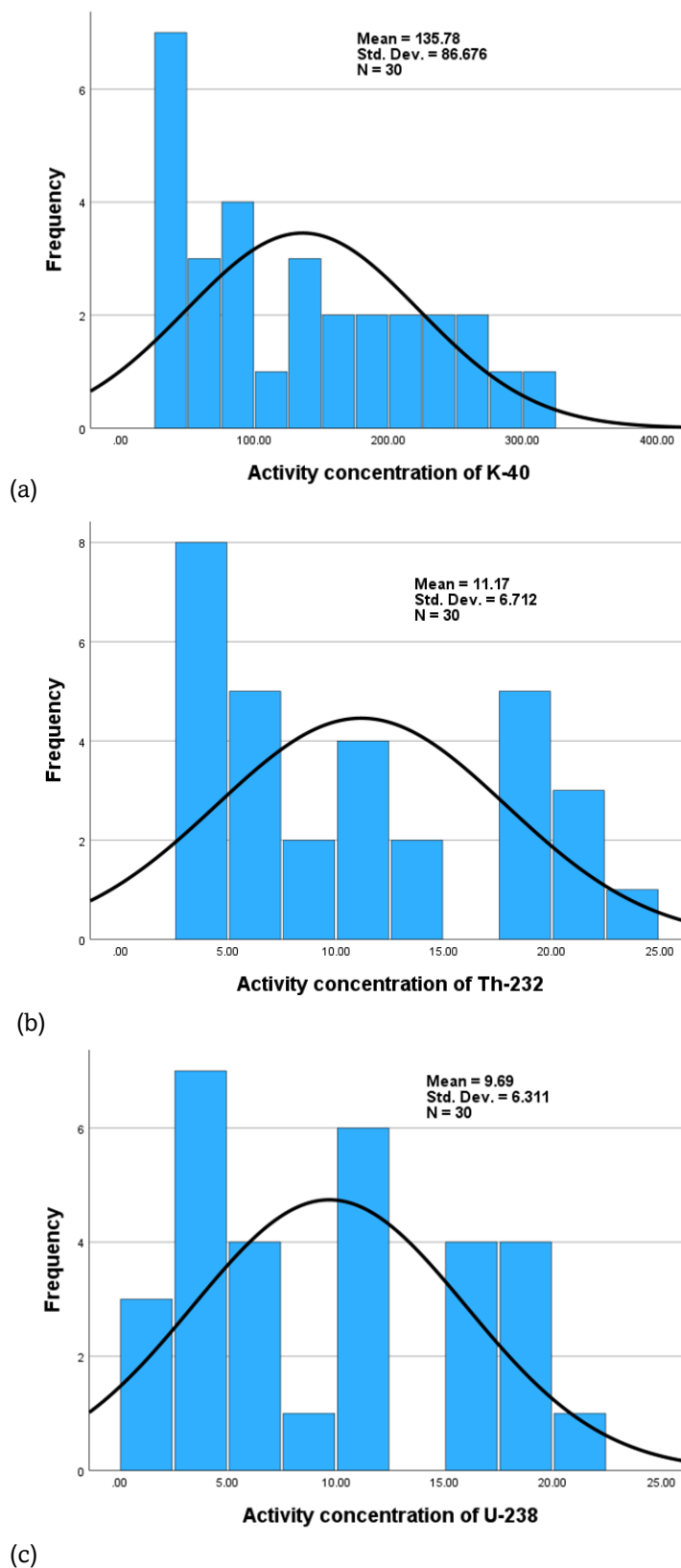
The primary statistical parameters, mean, range, standard deviation (SD), kurtosis, and skewness coefficients, are presented in Table 1. A high SD value for ^{40}K indicates a significant degree of variability in the radionuclide concentration of the water samples, while lower SD values for ^{232}Th and ^{238}U suggest less variability. However, the SD values for all three natural radionuclides are smaller than the mean, implying a substantial degree of consistency in the estimated radionuclide levels within the artisanal mining water samples of Ile-Ife. As observed in this study, negative kurtosis values indicate that the radionuclides' distributions are platykurtic, signifying relatively flat distributions with lighter tails compared to a normal distribution. The skewness values obtained in this study suggest that all three radionuclides exhibit positive skewness, indicating an asymmetric distribution. Fig. 4 shows the frequency distribution curves of ^{40}K , ^{238}U , ^{232}Th within the water samples taken from Ile-Ife artisanal mine sites.

3.2 Radiological safety indices

The radiological safety indices of water samples from Ile-Ife artisanal mining sites accessed in this study are presented in Table 3. The average D from the gold mining sites due to ^{40}K , ^{232}Th , and ^{238}U obtained varied from 4.39 to 35.59 nGy/h. The average value of 16.88 nGy/h is significantly larger than that reported by [16], but lower than the 59 nGy/h global threshold. Despite the elevated D value observed in the water samples, it is essential to note that the calculated values remain within permissible limits, ensuring that the associated health risks for both miners and the community are minimal.

The AEDE water sample value obtained from the mine locations ranged from 0.01 to 0.04 mSv/y. The average AEDE of 0.02 mSv/y obtained is less than the 0.48 mSv/y global threshold value [63]. The value, therefore, suggests that the mining population is not subject to excessive radiation exposure through water consumption. This result provides reassurance regarding the safety of water sources in the gold mining sites and suggests a low health risk for miners and the surrounding community.

Fig. 4. Frequency distribution curves of radionuclides from Ile-Ife water samples.



The Ra_{eq} in Bq/L obtained for water from the mining sites varied from 9.31 to 76.09, with an estimated average of 36.12 Bq/L. This Ra_{eq} value is lower than the 370 Bq/L the IAEA threshold [81]. The average external and internal hazard indices estimated for water samples obtained at the Ile-Ife gold mining areas were 0.10 and 0.12,

respectively. The R_{eq} values are consistent with [16], but less than the unity threshold [63], implying that the water bodies around the mining areas are still safe for drinking by the community and artisanal miners.

The calculated total AED_{ing} due to external terrestrial radiation and ingestion of water at the mine sites in Ile-Ife averages 2.81 mSv/y and 1.35 mSv/y for adults and children, respectively. Average total AED_{ing} obtained for adults exceeds the UNSCEAR standard of 2.4 mSv/y [63]. In contrast, the AED_{ing} for children is below the UNSCEAR limit. Nevertheless, both adults and children in this study have average AED_{ing} values surpassing the standards set by ICRP (1 mSv/y) for the public and WHO (0.10 mSv/y) as the upper limit in drinking water [80,82]. These values raise concerns about potential health implications for both miners and the broader community in the long term. The elevated AED_{ing} levels, particularly in adults, suggest increased radiation exposure-related health effects. While AED_{ing} for children falls below certain established limits, it is essential to remain vigilant as any exposure, especially at a young age, will result in long-term accumulation, thereby posing serious health risks with acute effects ranging from hair loss, skin redness, and cancer induction [83].

The estimated excess lifetime cancer risk associated with water samples from the Ile-Ife mining site raises notable health concerns for both miners and the broader community. In adults, the ELCR ranges from $(3.00 \text{ to } 23.53) \times 10^{-3}$, indicating a potentially elevated risk of cancer occurrence due to exposure. Similarly, in children, the ELCR varies from $(1.44 \text{ to } 11.28) \times 10^{-3}$, suggesting a concerning level of risk for this demographic. The average ELCR respective values of 11.27×10^{-3} and 5.41×10^{-3} for adults and children highlight a substantial overall cancer risk associated with the consumption of water from these mining sites. Miners, who are likely to have prolonged exposure, face an increased risk of developing cancer, underscoring the significance of occupational health and safety measures. Additionally, the broader community, including children, may experience health implications from prolonged accumulation through exposure to water sources with elevated cancer risk.

3.3 Statistical analysis

The Pearson Correlation matrix, generated using IBM Statistics, serves as a tool to assess the correlations among the radionuclide's activity concentrations, D, AEDR, and other radiological safety indices presented in Table 4. Positive significant correlation ($p < 0.05$) was observed among the radiological parameters in Table 4.

^{40}K exhibits strong positive correlations with ^{232}Th , ^{238}U , D, AEDR, and other radiological hazard estimates, suggesting that potassium concentration in the water samples is relatively linked to the levels of these variables. Thorium and uranium show high positive correlations of 0.951 and 0.949, respectively, indicating a strong association between thorium and uranium concentrations in the investigated water samples.

AEDR show strong positive correlations with ^{40}K , ^{232}Th , ^{238}U , D, R_{eq} , H_{ex} , H_{in} , AED_{ing} (both Adult and Children), and ELCR (both Adult and Children), thus suggesting that the concentrations of these radionuclides and radiological hazard indices significantly influence the AEDR. Likewise, the strong positive correlations observed by R_{eq} when compared with other variables is an indication of its dependence on the concentrations of ^{40}K , ^{232}Th , ^{238}U and other radiological indices.

External and internal hazard indices show strong positive correlations with other variables, indicating a coherent relationship between these hazard indices and the other parameters evaluated in this study. The total AED_{ing} ingested and the ELCR observed in adults and children are also highly positively correlated with other variables. This result underscores the significance of the radioactivity concentrations and radiological hazard indices influencing the AED_{ing} and ELCR.

Generally, exceptionally high positive correlation values observed in Table 4 indicate very strong relationship between primordial radionuclides and the radiological parameters evaluated from Ile-Ife mine sites. The robust correlation may suggest that the radionuclides (^{40}K , ^{232}Th , ^{238}U) significantly generate gamma radiation in water samples at the artisanal mining sites in Ile-Ife, Southwestern Nigeria.

4. Conclusion

Water samples from artisanal mines in Ile-Ife, Southwestern Nigeria, have been assessed in this study using a thoroughly calibrated NaI (TI) gamma-ray spectrometer. This study provides data on the levels of radionuclide activity in water samples collected from Ile-Ife gold mining areas. Average radioactivity concentration values of 135.78, 11.17, and 9.69 Bq/L, respectively, were obtained for ^{40}K , ^{232}Th , ^{238}U . The thorium concentration was 10.17 Bq/L above the WHO permissible limit, while the uranium concentration was 0.31 Bq/L lower than the world permissible limit. Statistical analysis of results obtained for radionuclide activity and radiological hazard indices shows a very high positive correlation, which emphasizes the interconnectedness of the variables studied.

Table 3. Results of estimated monthly average radiological hazard indices of water samples from Ile-Ife mining sites.

| Sample Code | D (nGy/h) | AEDE (mSv/y) | Ra _{eq} (Bq/L) | H _{ex} | H _{in} | AED _{ing} (mSv/y) | | ELCR (10 ⁻³) | |
|-------------|--------------|--------------|-------------------------|-----------------|-----------------|----------------------------|-------------|--------------------------|-------------|
| | | | | | | Adult | Children | Adult | Children |
| SM-1 | 17.12 | 0.02 | 36.68 | 0.10 | 0.13 | 2.85 | 1.37 | 11.45 | 5.49 |
| SM-2 | 14.73 | 0.02 | 31.59 | 0.09 | 0.11 | 2.48 | 1.19 | 9.97 | 4.78 |
| SM-3 | 18.80 | 0.02 | 40.07 | 0.11 | 0.14 | 3.09 | 1.48 | 12.40 | 5.95 |
| Mean | 16.88 | 0.02 | 36.12 | 0.10 | 0.12 | 2.81 | 1.35 | 11.27 | 5.41 |
| Min | 4.39 | 0.01 | 9.31 | 0.03 | 0.03 | 0.75 | 0.36 | 3.00 | 1.44 |
| Max | 35.59 | 0.04 | 76.09 | 0.21 | 0.26 | 5.86 | 2.81 | 23.53 | 11.28 |

Table 4. Pearson correlation of ⁴⁰K, ²³²Th, ²³⁸U, and radiological safety indices in water from Ile-Ife mining sites.

| | ⁴⁰ K | ²³² Th | ²³⁸ U | D | AEDR | R _{eq} | H _{ex} | H _{in} | AED _{ing} | | ELCR | |
|--------------------------|-----------------|-------------------|------------------|-------|-------|-----------------|-----------------|-----------------|--------------------|-------|-------|-------|
| | | | | | | | | | Adult | Child | Adult | Child |
| ⁴⁰ K | 1 | | | | | | | | | | | |
| ²³² Th | 0.951 | 1 | | | | | | | | | | |
| ²³⁸ U | 0.949 | 0.999 | 1 | | | | | | | | | |
| D | 0.978 | 0.994 | 0.993 | 1 | | | | | | | | |
| AEDR | 0.964 | 0.964 | 0.961 | 0.974 | 1 | | | | | | | |
| R _{eq} | 0.975 | 0.996 | 0.995 | 1.000 | 0.973 | 1 | | | | | | |
| H _{ex} | 0.976 | 0.994 | 0.993 | 0.999 | 0.973 | 0.999 | 1 | | | | | |
| H _{in} | 0.969 | 0.996 | 0.996 | 0.998 | 0.967 | 0.999 | 0.998 | 1 | | | | |
| AED _{ing} Adult | 0.970 | 0.997 | 0.996 | 0.999 | 0.972 | 1.000 | 0.998 | 0.999 | 1 | | | |
| AED _{ing} Child | 0.970 | 0.997 | 0.996 | 0.999 | 0.972 | 1.000 | 0.998 | 0.999 | 1.000 | 1 | | |
| ELCR_Adult | 0.970 | 0.997 | 0.996 | 0.999 | 0.972 | 1.000 | 0.998 | 0.999 | 1.000 | 1.000 | 1 | |
| ELCR_Child | 0.970 | 0.997 | 0.996 | 0.999 | 0.972 | 1.000 | 0.998 | 0.999 | 1.000 | 1.000 | 1.000 | 1 |

The silent health implications uncovered in this study show that residents will likely have an increased risk of radionuclide-related illnesses during the dry season, primarily due to the long-term accumulation through the ingestion of water from the mining sites and their associated tailings. During the dry season, elevated levels of radionuclides are typically observed in water sources that are experiencing scarcity. This is demonstrated by the exceptional AEDing values that have been recorded cumulatively for both adults and children, as well as the ELCR values that have been identified, which far exceed the global acceptable limits.

Consuming water from rivers or streams that flow across the Ile-Ife gold mining sites and their tailings poses a latent danger. Despite the radiological hazard indices (except for AEDing and ELCR) falling below the global permissible limit, it is crucial to recognize that this does not negate the importance of residents being vigilant about their water sources, especially during dry seasons. Proximity to mining sites amplifies the need for residents to be conscious of their water supply and to take precautions to mitigate potential health risks linked with radionuclide exposure and accumulation.

Declarations

Data availability Data will be made available upon reasonable request.

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Competing interests Authors declare no known competing or financial interests.

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