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Evaluation of Radioactive Metals in Surface Soil from an Artisanal Gold Mining Area, Migori, Kenya

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Abstract

Despite the fact that mining for gold causes serious radioactive concerns, there is a scarcity of radiological data in surface soils from artisanal gold mining area in Migori, Kenya. In this study, radioactive metals in 39 surface soil samples collected from gold mine sites were analysed using inductively coupled plasma-mass spectrometry (ICP-MS) in Bureau Veritas Laboratory, Canada. The obtained mean activity concentrations of ^{238}U, ^{232}Th, ^{137}Cs, ^{40}K and ^{226}Ra were 11.34± 3, 25.49 ± 3, 17.27 ± 0.6, 58.25± 4.7, 25± 4Bq/kg, respectively. The estimated average of absorbed dose rate in air (nGy/h), annual effective dose (µSv/y), radium equivalent activity (Bq/kg), external hazard index, internal hazard index, and lifetime cancer risks (1/Sv) were 66.9 ± 1.1, 89.3.3 ±4.6, 129.1 ±5.1, 0.37±0.01, 0.45 ± 0.02, 2.95×10-4±1.4x10-5, respectively. The mean values of the radium equivalent activity, external hazard index, and internal hazard index were all within safe ranges. The Pearson linear coefficient was also used to examine the relationships between radionuclides in surface soil samples. At the 0.05 significance level, ^{238}U was slightly positively associated with ^{232}Th, ^{226}Ra and ^{137}Cs while at t the 0.01 level of significance, there was a substantial positive correlation between ^{232}Th and ^{226}Ra and ^{137}Cs. Both ^{232}Th and ^{226}Ra were negatively and marginally correlated with ^{40}K at 0.05 significance level. In conclusion, the concentrations of radioactive metals in surface soil in the artisanal gold mining region were within the normal range and may present no significant health hazards to the communities in the examined area.

Keywords: Artisanal gold mines, cancer risk, radioactive metals, soil

Introduction

Natural radiation is made up of cosmic radiation and radiation produced by the decay of naturally existing radionuclides. Natural radionuclides comprise the primordial radioactive material in the crust of the earth, their radioactive decay daughters, and radionuclides created through cosmic-

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radiation interactions (Ahmad, et al., 2019). This leads to the natural environment consisting of trace levels of radioactive substances that occur naturally (NORMs), and dating back to earth's existence (Cachada et al., 2018). Their environmental availability is normally at concentrations that are not significantly detrimental to human health (Brusseau & Artiola, 2019). A major concern arises when levels upsurge as a consequence of human activities such as mining or natural catastrophes such as earthquakes (Candeias et al., 2018). Although it is widely assumed that radioactive contamination is mostly caused by manmade activity, investigations have also indicated that naturally occurring radioactive elements account for more than 80 percent of human exposure to ionizing radiation (Zakaly et al., 021). The radioactivity impact of NORM on miners, and other radiation contaminations have all become global issues (Agboola et al., 2020).

Mining is a major source of NORMs in the environment which find their way into the food chains and the atmosphere, posing grave health concerns to humans when consumed or inhaled (Rabuku and Malik, 2020). The most significant NORMs on radiological protection include radionuclides derived from Uranium-238 (^{238}U) and Thorium-232 (^{232}Th) decay family. Potassium-40 (^{40}K), is a non-series radionuclide that makes significant contributions to human exposure in the environments (Habib et al., 2019). According to studies, exposure to high levels of radioactive elements such as potassium (^{40}K), polonium, cobalt, uranium (^{235}U and ^{238}U), cesium, thorium (^{232}Th), radium, radon, plutonium and strontium, pose serious human health threats such as cancer development (Abdel et al., 2021, Ahmed, 2021). As a result, understanding the activity concentrations of ^{238}U, ^{232}Th, and ^{40}K as wells as Cs and Ra is critical in evaluating absorbed doses, which can lead to an assessment of their potential radiological risk to the populations (Botwe et al., 2017). There is no evidence available on the radionuclides' acute (short-term) non-cancer effects on people (Kreuzer, 2018). Animal investigations have shown that inhalation of acute uranium causes inflammatory responses in the nasal passages as well as renal damage (Ma, et al., 2020). Chronic (long-term) inhaling of uranium and radon has been associated with respiratory consequences in humans, including chronic lung illness, whereas radium exposure has been linked to acute leukopenia, anaemia, necrosis of the jaw, and other indicators (Lombardini et al., 2013, Tournieret et al., 2021). Due to a paucity of published environmental data in the study area on radionuclides, the purpose of this investigation was to *(i)* determine the radioactive metals concentration in top surface agricultural soils in the artisanal gold mines in

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Migori, Kenya, *(ii)* estimate the radiation threat for local populations, and *(iii)* analyse the radionuclide relationships.

Study Area

The artisanal gold mines are located in Migori County, which is in the west southern part of Kenya. The study area is about 309 km from the capital Nairobi of Kenya. Migori, Kenya has a latitude of -1.070698 and a longitude of 34.475272 The mean annual temperatures are 25.0^{O}C. The annual precipitation is approximately 1521 mm. Artisanal gold mining and subsistence agricultural practices characterize the study location. The sampling was done in July 2017. Figure 1 show the map of the position of the Migori County in Kenya and the sampling sites.

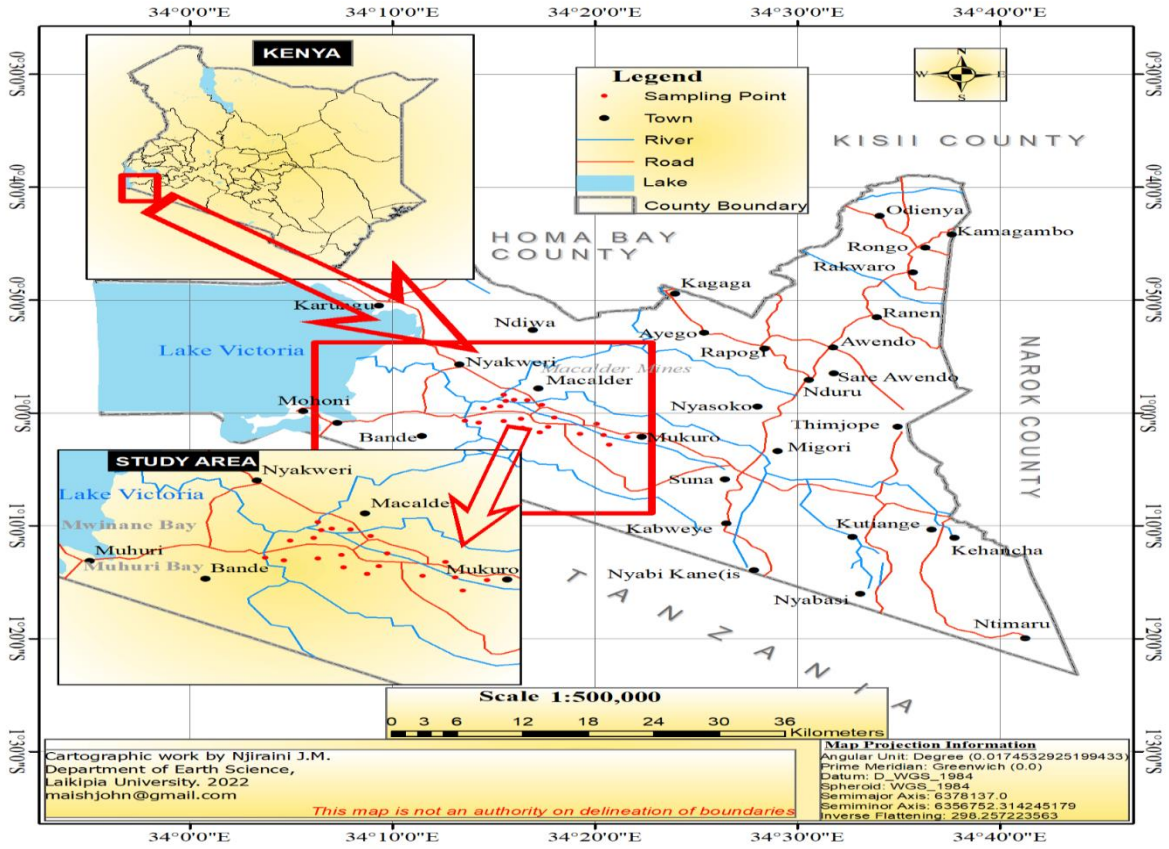

Fig. 1. Map of Kenya showing the location of Migori County and sampling sites

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Sampling and Preparation

Within a 15-kilometre radius in the mining region, 30 agricultural surface soil sample sites were purposively selected for this study (Fig. 1), from whence soil samples were collected. A 1 m^2 square area was mapped out at every sampling point. After eliminating impurities such as stones, gravels, and roots, four samples were obtained from the surface layer (up to 10 cm) of the four corners of the square area (1 m 1 m) using a stainless steel sampler (10 cm H10 cm), combined, and placed in a labelled polythene bag. Each sample was subsequently dried in an oven at 100 C for more than 24 hours to reduce soil water content, homogenized in the laboratory. The samples were filtrated through a 0.25 mm mesh, and a portion of 100.0 g was weighed and preserved in an airtight polythene bag for more than 30 days so that ^{226}Ra and its radioactive elements could attain secular equilibrium before analysis. Soil samples were then lyophilized (72 hours) and acid digested. Each sample was carefully weighed precisely and put in a Teflon digestion vessel with 1.0 mL of concentrated hydrochloric acid (30%), 7.0 mL of concentrated hydrochloric acid (65%), and 1.0 mL of hydrogen peroxide (30%).

To digest the samples, an Ethos D (Type Ethos plus 1) microwave lab station acquired from Milestone Inc. (Monroe, CT, USA) was utilized, using the following microwave program: Step 1: 25-200 degrees Celsius for 10 minutes at 1000 watts; Step 2: 200 degrees Celsius for 10 minutes at 1000 watts. Finally, digests were prepared in acid washed standard flasks with 25.0 mL of ultra-pure de-ionised water. Finally, the diluted solutions were placed in acid-washed plastic bottles. The concentrations of ^{238}U, ^{232}Th, and ^{40}K as wells as Cs and Ra were determined using thermo electron X7 inductively coupled plasma mass spectrometry (ICP-MS), model X series, in Bureau Veritas Laboratory in Vancouver, Canada.

Radiological Parameter Estimations

Calculation of the Radiation Hazard Index

Radiological estimations of naturally occurring radioelements (radionuclides) ^{238}U, ^{232}Th, ^{137}Cs, ^{40}K and ^{226}R from soils collected in gold mining areas of Migori, Kenya were calculated. Because radium and its daughter products account for 98.5% the of the radiological effects of the uranium series, the contribution from ^{238}U has been replaced by the decay product ^{226}Ra (Tufail et al., 2013,

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Ahmad et al., 2015). As a result, the activity levels of U, ^{232}Th, ^{137}Cs, ^{40}K and ^{226}R are typically used to calculate radiation hazard indices.

Radium Equivalent Activity and the External Hazard Index

Various hazard indices are used to quantify radiological risks in environmental contaminants. The Radium Equivalent Activity (Raeq), which represents the actual activity level, is among the most often used radiological parameter. To assess the effect of external radiation on humans, both the radium equivalent activity (Ra_{eq}) and the external hazard index (Hex) were considered.

The radium equivalent activity (Ra_{eq}) index is a useful guideline for managing radiation safety regulations for the general public who live in the region under consideration (Kolo, et al., 2020). The Ra_{eq} index is a weighted total of the natural radionuclide activities and is calculated under the assumption that 1Bq/kg of ^{226}Ra, 0.7 Bq/kg of ^{232}Th, and 13 Bq/kg of ^{40}K generate the same gamma radiation dose rates. The index is given by equation 1:

$$Ra_{eq} = C_{RA} + (1.43C_{Th}) + (0.077C_K) \quad \text{Eq (1)}$$

where C_{Ra}, C_{Th} and C_K are the average activity concentration in the sample in Bq/kg of ^{226}Ra, ^{232}Th, and ^{40}K respectively (Thabayneh, 2012, Dabayneh, et al., 2008).

Equation (2) was used to compute the external hazard index. The external hazard index is a single index that designate the gamma yields from several combinations of radionuclides such as ^{232} Th, ^{226} Ra and ^{40} K in the samples analysed. UNSCEAR (2000), has describes the equation as shown in Eq. 2

The Ra_{eq} threshold value must not be more than 370 Bq/kg, while the Hex value must be less than a unity (Faanu, et al., 2016, Kamunda, et al., 2016). The higher the calculated Bq/Kg value the larger the radiation being emitted from the metal.

$$H_{ex} = A_{Ra}/370 + A_{Th}/259 + A_K/4810 \quad \text{Eq (2)}$$

Internal Hazard Index

The internal hazard index (Hin) was developed to characterize the risk of radon and its short-lived products in built environments, and it is suggested that it should be lower than unity (Korkulu &

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Özkan, 2013; Bekelesi et al., 2017). Equation 3 was used to calculate Internal Hazard Index of the selected radionuclides in this paper.

$$H_{in} = A_{Ra}/185 + A_{Th}/259 + A_K/4810 \quad \text{Eq (3)}$$

Lifetime Cancer Risk (LTCR)

The probability that an individual who is cancer-free will acquire cancer in his or her lifespan is referred to as lifetime cancer risk. The LTCR was determined using the estimated annual effective dose equivalent (AEDE), as described by the equation (4) (Nkpaa, et al., 2018, Elnimr, et al., 2017).

$$LTCR = AED \times DL \times RFSE \quad \text{Eq (4)}$$

where DL is the lifespan expectancy, 70 years, and RFSE is the risk factor for the general populations' stochastic effects given at 0.055/Sv (Sutou, 2018; Luevano-Gurrola et al., 2015).

Annual Effective Dose

The yearly effective dosage is represented by the irradiation dose of the human body from naturally occurring radionuclides in the crust of the earth soil. It is calculated by using the Equation (5) (UNSCEAR 2014, Kamunda, et al., 2016).

$$AED = D \times 8760 \times 0.2 \text{ (or } 0.8) \times 0.7 \times 10^{-3} \quad \text{Eq (5)}$$

wherein AED is the annual effective dose (Sv/y); D is the dose rate (nGy/h); the coefficient 0.7 Sv/Gy is the conversion coefficient from the absorption in air to the optimal dose absorbed by humans; 0.2 is the outdoor occupancy factor; and 8760 hours'/year equals 365 days and 24 hours per year.

Absorbed γ dose Rate in Air

The absorbed dose rate (nGy/h) in air at 1 m above ground for radionuclides (U, ^{232}Th, ^{137}Cs, ^{40}K and ^{226}Ra) evenly scattered on the ground was calculated using Equation (6) (UNSCEAR, 2017).

$$D = 0.462 \times A_{Ra} + 0.604 \times A_{Th} + 0.0417 \times A_K \quad \text{Eq (6)}$$

where A_K, A_{Th} and A_{Ra} are the activity concentrations of ^{40}K ^{232}Th, and 226Ra, (Bq/kg), respectively.

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Parameters Assessed in Soil Metal Exposure in the Migori Gold Mines

Several parameters were considered to determine possible potential radiation exposure from agricultural soils in Migori gold mines in Kenya. These are summarized in table 1.

Table 1: Parameters included in assessment of the potential threat of soil metal exposure in the Migori gold mines, Kenya

Parameters	Symbol	Unit	Value
Soil ingestion rate	IngR	mg/day	100 (adult), 200 (child)
Frequency of exposure	EF	day/year	350
Soil inhalation rate	InhR	m^3/day	20 (adult), 7.6 (child),
Duration of exposure	ED	year	70 [6 (child) for non-cancer effects]
Skin area	SA	cm^2	1530 (adult), 860 (child)
Particle emission factor	PEF	m^3/kg	1.36×10^9
Skin adherence factor	SL	mg·cm^2	0.07 (adult), 0.2 (child)
Body weight	BW	kg	70 (adult), 15 (child)
Average Time	AT	Day	70 × 365 days for carcinogens
			ED × 365 days for non-carcinogens

Radionuclides Concentrations

Radionuclide concentrations in soil samples analysed were measured in ppm and percentage and converted to Bq/kg. To convert ppm to Bq /kg, conversion factors were applied, (^{232}Th; 1 ppm = 4.06 Bq /kg; ^{238}U; 1 ppm = 12.35 Bq /kg, 1 ppm Ra = 11.1 Bq/kg, and Cs; 1ppm =11.1 Bq /kg). Whereas 1% of ^{40}K = 313 Bq/kg (Poschl, and Nollet, 2006, Said, et al., 2010).

Data Analysis

Correlational tests between natural radionuclides in surface soil samples were calculated using the SPSS computer software, Version 19 for Windows. The Pearson correlation's statistical significance was verified using the t- test (Forkapic et al., 2017, Kardan, et al., 2017). If a value was near to zero, there was no relationship between the two elements. For correlation coefficients of 0.2-0.4, 0.4-0.6, and >0.6, the words "weak," "moderate," and "strong" were used (Gulan, 2017). The alpha threshold for testing significance was chosen at 0.01 and 0.05.

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Results and Discussion

Radionuclides Levels

The soil concentrations of radionuclides in 39 surface soil samples in PPM (U,^{232}Th, ^{137}Cs and ^{226}Ra) and % (^{40}K) in Migori gold mining area are presented in Table 2. The mean values of U,^{232}Th, ^{137}Cs, ^{40}K and ^{226}Ra in the surface soil samples from Migori mining sites were; 1.234, 8.65, 1.78, 0.142 and 2.272 respectively, and were lower than the world mean values (UNSCEAR, 2008).

Table 2: Concentration ranges of radionuclides in surface soil samples in Migori gold mining area, Kenya

Radionuclides	^{238}U	^{232}Th	^{137}Cs	^{40}K	^{226}Ra
No. (39 samples)	ppm	ppm	ppm	%	ppm
MDL	0.05	0.1	0.02	0.01	0.01
Max	2.02	13.1	6.07	1.43	2.973
Min	0.33	0.6	0.16	0.01	1.532
Mean	1.234	8.65	1.78	0.142	2.272
World averages UNSCEAR (2008)	(35)	7–50 (45)	-	100–700 (420)	16–116 (33)
MDL=Max detection limit					

Radionuclides Bq/kg

The activity concentration mean values were 11.34± 3, 25.49 ± 3, 17.27 ± 0.6, 58.25± 4.7, 25± 4 for ^{238}U, ^{232}Th, ^{137}Cs, ^{40}K and ^{226}Ra respectively, which were all below the world average values as indicated in table 3 (UNSCEAR, 2008). The activity concentration of ^{40}K was 58.25± 4.7Bq/kg, which recorded the highest calculated value of activity of radionuclides (Bq/kg) in surface soil samples in Migori gold mining area. This could be as a result of the mining activities (Table 3).

Table 3: Activity of radionuclides (Bq/kg) in surface soil samples in Migori gold mining area

Samples/ Radionuclides	^{238}U	^{232}Th	^{137}Cs	^{40}K	^{226}Ra
1	0.62± 2[a]	0.406± 1	0.22± 2.1	72.99± 2.9	24 ± 3
2	0.62± 2	28.01±3	67.38± 2.1	134.59± 2	24 ± 2
3	7.04± 3	30.04±3	63.16± 2.3	134.59± 2	25 ± 2
4	7.29± 2	10.56± 2	7.77± 0.2	12.52± 2.3	26 ± 2
5	7.04± 3	10.56± 2	7.77±0. 2	15.65± 2	29 ± 2

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6	7.41± 2	10.15± 2	7.44± 2.3	12.52± 2	26 ± 2
7	15.81± 2	38.16± 3	19.43± 2.1	62.6± 2	29 ± 2
8	16.18± 2	38.97±3	21.65± 2.1	71.99± 2	26 ± 2
9	20.87±3	40.6± 32	13.43± 2.2	62.6± 2	28 ± 3
10	20.75±3	43.44±3	13.76± 2.3	62.6± 2	25 ± 3
11	20.25±3	30.45±1	14.09± 2.1	65.73± 2	17 ± 2
12	24.95± 3	33.92± 2	13.43± 2	62.6± 2	25 ± 3
13	6.62± 12	6.90± 1.2	5.99± 2	15.65± 2	28 ± 3
14	6.18± 12	7.71± 1.2	5.99± 2	15.65± 2	25 ± 2
15	6.67± 1.2	7.71± 1.2	5.99± 2	15.65± 2	27 ± 2
16	8.03± 1.2	5.28± 2.1	4.33± 2	12.52± 2	26 ± 3
17	7.90± 1.2	5.28± 2.2	4.44± 2	12.52± 2	33 ± 3
18	8.03± 2	5.68± 2.2	4.66± 2	12.52± 2	26 ± 3
19	5.19± 2.2	4.47± 1.2	2.3± 2	9.39± 1.2	22 ± 2
20	5.19± 2.2	4.06± 2	2.3± 2	9.39± 1.2	26 ± 2
21	5.43± 2.2	3.65± 2	2.3± 2	9.39± 1.2	27 ± 3
22	4.32± 1.2	2.84± 2	2.11± 2	6.26± 1.2	25 ± 3
23	4.32± 0.2	2.44± 2	1.89± 2	3.13±2. 2	28 ± 3
24	4.08±0.2	2.84± 2	1.78± 2	3.13± 2.2	23 ± 2
25	12.72± 3	28.83±1	21.87± 2	71.99±2. 2	25 ± 2
26	12.23± 3	26.80±2	19.54±2	68.86± 2	20 ± 2
27	13.09± 3	47.91±3	25.53±2	93.9± 2	27 ± 3
28	14.94± 2	47.09± 3	23.98±2	90.77± 2	28 ± 3
29	17.17± 2	43.85± 3	23.53±2	90.77± 2	21 ± 2
30	17.54± 2	47.50±2	26.64±2	90.77± 2	25 ± 2
31	18.03± 2	51.16±4	31.97±2	100.16± 2	27 ± 2
32	17.91± 2	53.19±4	33.52±2.1	103.29± 2	26 ± 2
33	7.78±1	11.37±2	7.88±0.2	15.65± 2	22 ± 2
34	7.41± 1	9.74± 12	7.22±0.2	15.65± 2	24 ± 2
35	7.53± 1	9.74±1.2	6.77±0.2	15.65± 2	28 ± 3
36	14.45± 2	44.25± 2	15.98±1	59.47± 2	30 ± 3
37	14.20± 2	41.82± 2	15.10±1	59.47± 2	24 ± 2
38	22.85± 3	52.78±3	29.19±2	93.9± 2.2	27 ± 2
39	23.09± 3	51.97± 3	28.19±2	90.77± 2.3	25 ± 2
Mean ± SD [b]	11.34± 3	25.49 ± 3	17.27 ± 0.6	58.25± 4.7	25± 4
WAV	35	45	-	412	32

[a]Activity concentration ± expanded uncertainty, [b] SD represents standard deviation; WAV: world average values.

Annual Effective Dose, Hazard Indices, and Lifetime Cancer Risk

The Absorbed dose rate in air(nGy/h), annual effective dose (μSv/y), Radium Equivalent Activity (Bq/kg), hazard indices (External Hazard and Internal Hazard), and lifetime cancer risk(1/Sv) calculated from radionuclides in soil samples are shown in Table 4. The calculated mean outdoor γ dose rates was 66.9nGy·h^{-1} which was higher than the world average of 60 nGy/h (UNSCEAR, 2008, US EPA, 2002). The mean value of radium equivalent activity was 129.1 ± 5.1Bq/kg, lower

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than the reference value of 370 Bq/kg. The external and internal hazard indices did not exceed unity, which indicates that the γ radiation from the soil was at a safe level. The lifetime cancer risk was $2.95\times10^{-4}\pm1.4x10^{-5}$/Sv, which was also at a very low level indicating that acquiring cancer due to the soil contamination from the mining activities in the region is probably very low.

Table 4: The radiation hazard indices and lifetime cancer risk

	Absorbed Dose Rate in Air (nGy/h)	Annual Effective Dose (μSv/y)	Radium Equivalent Activity (Bq/kg)	External Hazard Index	Internal Hazard Index	Lifetime Cancer Risk (1/Sv)
Mean± SD	66.9 ± 1.1	89.3.3 ±4.6	129.1 ±5.1	0.37±0.01	0.45 ± 0.02	$2.95\times10^{-4}\pm1.4x10^{-5}$
Median	65.7	89.3	128.58	0.33	0.46	2.91×10^{-4}
Min-max	59.6–77.2	71.3–107.3	100.7–157.6	0.31–0.43	0.34–0.569	2.7×10^{-4}–3.2×10^{-4}

Correlation Analysis

Table 5 displays the Pearson correlations for radionuclides studied. At the 0.05 significance level, it was observed that ^{238}U was weakly positively associated with ^{232}Th, ^{226}Ra and ^{137}Cs. At the 0.01 level of significance, there was a moderately positive association between ^{232}Th and ^{226}Ra and ^{137}Cs. Both ^{232}Th and ^{226}Ra are negatively and weakly correlated with ^{40}K at 0.05 significance level.

Table 5: The Pearson correlation matrix for the natural radionuclides

	^{238}U	^{232}Th	^{226}Ra	^{40}K	^{137}Cs
^{238}U	1	0.344 [a]	0.333 [a]	0.032	0.421[a]
^{232}Th		1	0.933 [b]	−0.367 [a]	0.873 [b]
^{226}Ra			1	−0.303 [a]	0.467 [a]
^{40}K				1	0.332[b]
^{137}Cs					1

[a] Correlation is significant at the 0.05 level

[b] Correlation is significant at the 0.01 level.

Conclusions and Recommendations

The activity concentrations of ^{238}U, ^{232}Th, ^{40}K, ^{137}Cs and ^{226}Ra were generally lower than the global average. The results were within acceptable limits. The yearly effective dose and several

radiation hazard indices suggest that the local residents in the gold mining area face negligible radiological threat. Both the non-cancer risk index and the cancer risk were judged to be below the allowed thresholds. A substantial positive association between studied radioactive metals was discovered at the 0.01 significance level, indicating a similar origin. The correlation analysis also revealed negative and weak connections between the radionuclides. As a result, mining activities in this gold mining area of Migori, Kenya do not seem to suggest major radioactive threats to miners or the wider populace. This study produced the baseline information on natural and anthropogenic radioactivity in surface soil from an artisanal gold mining location in Kenya. To the best of my knowledge, this could be one of the first comprehensive studies on radiation levels in a gold mining location in Migori, Kenya. These background data could serve as a significant resource for community environment protection. Given that artisanal goldmining operations are ongoing, there is a greater chance of elevating the concentration levels of ^{238}U, ^{232}Th, ^{137}Cs, ^{40}K and ^{226}Ra in the research region. Continued monitoring of the environmental media, particularly the soils, would go a long way toward detecting any significant health consequences associated with the raised levels of the reported elements in the research region.

Ethical Approval

This study did not include any experiments involving animals or humans, nor did it take place in any private or protected places. For the aforementioned places, no special permits were necessary.

Consent to Participate

The Helsinki 1996 guidelines were followed, and authorization to conduct the study was acquired from Moi University's Research and Ethics Committee (IREC).

Author Contributions

Veronica Ngure contributed to the study conception and design, material preparation, data collection and data analysis as well as the writing of the manuscript.

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Competing Interests

The authors have no relevant financial or non-financial interests to disclose.

Availability of data and materials

Data is available on request. However, the original data sheet printed from the laboratory contains data not reported in this manuscript.

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