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Determination of Natural Radioactivity Levels and Radiation Hazards from the Soil of Tin Mining in Kyerwa District, Tanzania

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Authors' contributions

This work was carried out in collaboration among all authors. All authors read and approved the final manuscript.

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ABSTRACT

Gamma spectrometry was used to determine the activity concentrations of the natural radionuclides 226Ra, 232Th, and 40K in soil samples from the Tin mining in Kyerwa District, Tanzania. The study findings revealed that the mean activity concentrations of 226Ra, 232Th, and 40K were 90.68, 65.45, and 630.95 Bq/kg, respectively. These values were all greater than the average global activity concentrations for 226Ra, 232Th, and 40K which are 32 Bq/kg, 45 Bq/kg, and 420 Bq/kg, respectively. Assessments of the radiological risks associated with these naturally occurring radionuclides were done. In this case, the radium equivalent activity (Ra_{eq}), external hazard index (H_{ex}), internal hazard index (H_{in}), and radioactivity level index (Iy) were calculated. The results show

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that the mean values of R_{aeq} , H_{ex} , H_{in} , and $I\gamma$ were 230.6 Bq/kg, 0.628, 0.861, and 0.831, respectively. These results were below the global criteria levels for R_{aeq} , H_{ex} , H_{in} , and $I\gamma$, which are 370 Bq/kg, 1, 1, and 2, respectively. Estimates for the absorbed dose rate in the air (DR), the annual effective dose equivalent (AEDE), and the annual gonadal equivalent dose (AGED) were also made. The findings revealed that the corresponding mean values for DR, AEDE, and AGED were 106.8 nGy/h, 1.07 mSv/y, and 744.4 μ Svy-1. The mean values exceeded the global average value of 57 nGy/h for DR, 0.07 mSv/y for AEDE, and 2984 Sv/y for AGED. Therefore, Tin mining activities in the Kyerwa District could endanger the mining community from natural radioactive sources. In this study, therefore, we recommend performing a periodic inspection of the mining areas, monitoring the mining workers, and determining the level of activity and concentrations of different radionuclides in the mining area.

Keywords: Natural radioactivity levels; activity concentration; radiation hazards; radionuclides.

1. INTRODUCTION

Humans are exposed to varied levels of radioactivity depending on the natural radioactive elements found in each place. To undertake background tests and identify environmental radioactivity levels, researchers investigated natural environmental radiation and radioactivity in soil [1]. Radioactivity levels can be used to evaluate radioactive pollution and population exposure rates, as well as to forecast changes in environmental radioactivity that might be caused by nuclear accidents, industrial processes, and other human activities [2]. The majority of radiation doses that people receive are caused by the natural elements potassium-40, uranium-238, thorium-232, and their decay products. There have been detected about 60 radionuclides that are widely dispersed. Radionuclides can guickly build up in the food chain and can be found in geological formations like soil or rocks, and water bodies such as rivers, lakes, and seas [1,3]. One of the potential causes of exposure to naturally occurring radioactive materials (NORM) is Tin mining. NORM makes up a sizable portion of radiation doses from internal or external radiation sources; therefore, tin mining and processing may result in higher concentrations.

Depending on the type of rock from which the soil is formed, different dose levels apply. Igneous rocks like granite have higher radioactivity levels, while sedimentary rocks have lower levels [4]. Higher activity concentrations of 226Ra, 232Th, and 40K have been reported by the studies on uranium and gold mining in Tanzania that assessed the activity concentration of natural radionuclides in soil [5,6]. The reported concentrations were higher than the global average concentrations of these radionuclides described in UNSCEAR (2000). Additionally, research has demonstrated that human activities like mining and oil and gas production might increase radioactivity levels from naturally occurring radioactive materials that might become high enough to require regulatory control [2,7].

Radium, thorium, and potassium concentrations in mining operations range from an order of magnitude greater than the typical background value [8]. When heavy minerals like tin are mined and processed in any sizeable quantity, the requirements for suitable radiation safety measures become apparent in such radioactive element concentrations. This article aims to assess the natural radioactivity levels and radiation hazards in the soil of Tin Mining in the Kyerwa District to compare the radionuclide concentration levels with the recommended set by National Authorities values and International Organizations.

2. MATERIALS AND METHODS

2.1 Description of the Study Area

One of the eight districts that make up the Kagera region is Kyerwa District. It is located between 31° and 32° east of the standard meridian and 0° to 5° south of the equator (Fig. 1). It covers a 3,086 km² area within the Kagera region. Both Rwanda and Uganda share a border with this area. The Kagera River demarcates the border between Kyerwa, Rwanda, and Uganda. The District is made up of mountain ranges that are divided by wetlands and marshy valley bottoms. While valley bottoms and moist areas are 1,150 to 1,450 meters above sea level, the altitudes range from 1,500 to 1,800 meters above sea level. The climate in the District is tropical highland with 26°C mean temperature annually. The bimodal rainfall distribution peaks in March and May and from September through December. The District experiences yearly precipitation of 800 to 1000 mm. Rainfall typically declines from east to west and exceeds 1000mm/yr on high hills.

The parent material, age, prehistoric terrain, climatic circumstances, and biological history of Kyerwa soils all vary substantially from one location to the next. Approximately 60% of the district is coated with minerals, with tin accounting for the majority of this coverage. Tin is mined by small-scale miners, and various firms have submitted plans to the executive director of the Kyerwa district for larger-scale mining [9].

2.2 Sampling Methodology

The sampling area from the mine was divided into three zones: the northern (Kabingo, Kabagambe mine), the central (Kabushoke mine), and the southern (Chabitembe mine). Sampling locations are depicted in Fig. 1. Six samples of soil were collected randomly from each sampling zone to make a total of 18 samples of soil. The samples were taken at a depth of 0 to 5 cm, as previously described by Ajayi [4,10]. The collected samples were then sent to the TAEC laboratory for processing and analysis in labeled polythene bags.

2.3 Sample Preparations

To eliminate larger items, soil samples were sieved in the lab using a 2 mm sieve, and then

they were ground into a fine powder using a mortar and pestle to match the reference sample's matrix. The samples were then placed in a desiccator to prevent moisture absorption after being dried for several hours in an oven at about 45 to 50°C until the weight remained constant [10]. Each sample weighed around 500 g, and the Marinelli beaker's 500 cm3 volume was sealed with silicone and plastic tapes to ensure airtightness. To achieve a radioactive secular equilibrium between radon gas 222Rn and its decay products (214Pb, 214Bi, and 226Ra) from the 238U decay series, the samples were stored for a month.

2.4 Gamma-Ray Spectrometry

HPGe detector (Hiah Α coaxial Purity Germanium Detector) serial number 57-P51572A with relative efficiency of 51% and resolution sufficient to resolve the relevant gamma-ray energies at Full Width Half Maximum (FWHM) of roughly 7.2% at any energy of 0.662 MeV. Three layers of copper, cadmium, and lead, each measuring 30 mm, 3 mm, and 100 mm thick, respectively were used to shield the detector chamber. Daily calibrations of the gamma-ray energy were carried out using standard radiation sources such as 133Ba, 109Cd, 54Mn, 57Co, 60Co. 65Zn. 22Na. and 137Cs. Using the Genie 2000 software's In Situ Object Counting-system (ISOCS), efficiency calibration was established.



Fig. 1. Map of the study area in Kyerwa Mining

The corresponding gamma-ray lines emitted by the decay products were used to calculate the activity concentrations of the radionuclides in the samples. The gamma lines with weighted mean activity levels of 911.1 keV (228Ac), 2614.5 keV of 208TI, and 583.1 keV (212Pb) were used to calculate 232Th. To calculate 226Ra, gamma lines at 609.3 keV (214Bi), 1764 keV of 214Bi, 295.2 keV (214Pb), and 186.1 keV (226Ra) were employed. Its gamma line energy of 1460.8 keV was used to calculate the 40K [11].

International Atomic Energy Agency (IAEA) soil 375 was utilized as a standard reference material to evaluate the findings' precision and accuracy. The certified value was compared to the standard's activity at various energies following decay modification, using the datasheet of December 31, 1991, as a guide. The standard was counted for 10 hours, the same as that used for the samples. The experimental concentration values matched the designed values within 10% as shown in Table 1.

The following analytical expression, which is stated in equation (1) below, was used to compute the activity concentration (Bqkg⁻¹) of 226Ra, 232Th, and 40K in the samples [10,11].

$$XEi = \frac{BEi}{\operatorname{yd} x \operatorname{EE} x \operatorname{ms} xT}$$
(1)

Where BEi is the net peak area at energy E, $\mathcal{E}E$ is the detection efficiency at the energy E, T is the counting live time in seconds, $\mathbf{y}d$ is the gamma-ray yield per disintegration of the specific radionuclide for a transition at energy E, and ms is the mass of the dry weight in kg of the measured sample.

2.5 Radiological Hazard Assessment

2.5.1 Assessment of radium equivalent (*Ra_{eq}*)

Calculating radium equivalent activity allows one to evaluate the risks of gamma radiation from people who are engaged with the soil from the mining locations listed in Table 5. This yields a single index that characterizes the gamma output from various mixtures of 226Ra, 232Th, and 40K in the samples [12]. The mathematical formula for radium equivalent activity (Raeq) is expressed in equation (2) below [2].

$$Ra_{eq}$$
 (Bqkg-1) = CRa+1.43CTh+0.077CK (2)

Where *C*Ra, *C*Th, and *C*K are the activity concentrations of 226Ra, 232Th, and 40K (in Bqkg-1), respectively.

2.5.2 The absorbed dose rate in air

226Ra. 232Th. 40K When and are present in a certain activity concentration in the air at a height of 1 meter above the ground, the absorbed dose rate (DR) Bq/kg in indicates the effects of gamma radiation [13]. The value of DR estimated in Table 6 is defined by Equation (3).

DR (nGy/h)=0.462CRa+0.604CTh+0.042CK (3)

Where DR is the dose rate in nGyh-1 and CRa, CTh, and CK are the activity concentrations (Bqkg-1) of radium (226Ra), thorium (232Th), and potassium (40K), respectively.

2.5.3 The annual effective dose equivalent (AEDE)

The Annual Effective Dose Equivalent (AEDE), which was determined in Table 6 as part of the radiological hazard assessment for miners and surrounding communities in Kverwa. the described the dose rate that will be responsible for biological damage to human tissues [14]. Equation (4) was used to calculate the annual effective dose equivalent (AEDE) for miners or the general population in (mSv/y) [2].

2.5.4 External hazard index (Hex)

The external hazard index for samples under investigation was calculated using the equation (5) defined by [15]. The estimated results of Hex are depicted in Table 5.

$$H_{\text{ex}}$$
=CRa/370+CTh/259+CK/4810 (5)

Where CRa, CTh, and CK are the activity concentrations of 226Ra, 232Th, and 40K in (Bqkg-1), respectively.

2.5.5 Internal hazard index (H_{in})

The underground miners' respiratory systems are also at risk from radon and its transient products. Hence, the internal hazard index, as given in Table 5, is used to quantify internal exposure to radon and its transient derivatives. Equation 6 in [14,15] expresses this mathematically.

$$H_{in}=CRa/185+CTh/259+CK/4810 \le 1$$
 (6)

Radionuclides	Energy (keV)	Certified reference value	Experimental activity concentration	Accuracy (%)
226Ra	186.2	20.0 ± 0.9	20.54 ± 09	3.0
214Bi	1764	19.96 ± 0.4	19.97 ± 0.9	4.5
	2477.4	19.96 ± 0.9	20.46 ± 12	6.4
228Ac	338.5	20.46 ± 0.3	20.58 ± 1.5	7.8
	910.8	20.46 ± 0.5	20.29 ± 0.7	3.7
	968.5	20.46 ± 0.5	23.69 ± 0.9	4.8
40K	1460.7	423.40 ± 0.2	499.91 ± 2.2	0.4
208TI	860.4	20.46 ± 0.3	21.20 ± 1.1	4.7
	2614.5	20.46 ± 0.6	22.44 ± 1.7	8.0
214Pb	295.2	19.96 ± 0.8	19.46 ± 0.8	3.6
	351.92	19.96 ± 0.5	20.61 ± 1.1	5.7

Table 1. The experimental activity values ($Bq/kg \pm SD$) and the standard reference values

2.5.6 The radioactivity level index

The radioactivity level index ($I\gamma$) is an additional radiological factor that can be calculated. The radioactivity level index ($I\gamma$) given in Table 5 is used to estimate the rate of γ -radiation hazards linked with natural radionuclide in investigated samples [15,16]. The $I\gamma$ is defined by Equation 7.

 $I\gamma = CRa/300 + CTh/200 + CK/3000$ (7)

2.5.7 Annual gonadal equivalent dose

Gonads, active bone marrow, and bone surface cells are regarded as the organs of interest by [12]. Hence, Equation (8) was used to estimate the Annual Gonadal Equivalent Dose (AGED mSv y-1) for the miners and the public around the mining areas due to the specific activity of 226Ra, 232Th, and 40K given in Table 6 [16,17].

AGED (μ Svy-1) = 3.09CRa+4.18Th+0.314CK (8)

3. RESULTS

3.1 Radioactivity Concentrations in Soil

Tables 2, 3, and 4 provide the activity concentrations of 226Ra, 232Th, and 40K in soil samples taken from various places in the three zones of the Kyerwa district as well as the average values for the zones. Compared to the Northern zone (NS) and Southern zone, the Central zone (CZ) has the highest average activity concentrations of 226Ra and 40K. (SZ). In comparison to the Central Zone and the Southern Zone, the Northern Zone (NS) has the greatest average activity concentrations of 232Th.

The Northern (NZ) includes the area around the Kabingo and Kabagambe mines, the Central Zones (CZ) includes the area around the

Kabushoke mine, and the Southern Zones (SZ) includes the area around the Chabitembe mine. Table 4 compares the average values of the three radionuclides from the three Kyerwa District zones with those from other locations in Tanzania and around the world. The measured activity concentration of 226Ra in Tin mining soil samples ranged from 24.10±2.3 Bqkg-1to to 207.9±1.7 Bqkg-1, with a mean value of 90.68±2.6 Bqkg-1. The concentration of 232Th ranged from 40.16±2.0 Bgkg-1 to 88.20±3.3 Bqkg-1, with an average value of 68.12± 5.5 Bq kg-1. The activity concentration of 40K ranged from 187.80±7.7 Bqkg-1 to 937±17.1 Bqkg-1, with an average value of 630.95 ± 15.9 Bgkg-1. The world average value of soil for 226Ra, 232Th, and 40K are 35, 30, and 400 Bgkg-1, respectively, as registered in the world average concentrations published by UNSCEAR (2000). The average concentrations of 226Ra and 232Th obtained in the current study are higher than the recommended reference levels.

The range and average of the natural radioactivity concentration levels reported in different locations in Tanzania were compared to the mean radioactivity concentrations of the soil samples from this study (Table 4). The results show a greater mean concentration of 226Ra, 232Th, and 40K than those found in Bahi and Likuyu village, except 40K, which is slightly higher in Bahi. The current study's mean activity concentrations are lower than those found in Mkuju, Tanzania.

These results were also compared with those reported in studies conducted in other countries (Table 4). The mean activity concentrations of natural radioactivity of 226Ra, 232Th, and 40K in this study are higher than those reported in Egypt, Iraq, Ghana, Nigeria, and Namibia, although the concentration of 232Th is slightly higher in Nigeria.

4. DISCUSSION

Gamma spectrometry was used to measure the radioactivity concentration of 18 soil samples collected from Tin Mining in Kyerwa

District, located in North West of Tanzania. Results showed that the mean activity concentrations of 226Ra, 232Th, and 40K for the mining area are 90.68 \pm 2.5, 65.45 \pm 2.7, and 630.95 \pm 15.9Bqkg⁻¹, respectively. The measured values are higher than the world-recommended values.

Table 2. Activity concentration with their total uncertainties in the soil samples from Tin mine in the Northern Zone (NZ) together with the average activity concentration of the zone

Sample Code	Activity concentrations (Bqkg ⁻¹)			
-	²²⁶ Ra	²³² Th	⁴⁰ K	
KS1	56.4 ±3.4	91.00±2.4	1397.9±3.3	
KS2	51.9±1.7	92.6±3.7	1616.6±26.2	
KS3	45±2.30	64.00±2	466.0±23	
KS4	948.4±1.1	84.3±1.5	23.00±8.1	
KS5	82.60±20	42.2±1.4	667.9±19.4	
KS6	56.4 ± 3.4	84.9±3.3	1456±26.6	
Mean	207.9±1.7	76.00±2.4	937±17.1	
Range	(45-948)	(42-92)	(23-1616)	

 Table 3. Activity concentration with their total uncertainties in the soil samples from Tin Mine in the Central Zone (CZ) together with the average activity concentration of the zone

Sample Code	Activity concentrations (Bqkg ⁻¹)			
-	²²⁶ Ra	²³² Th	⁴⁰ K	
CB1	23.8 ±2.2	41.80±1.6	615.0±22	
CB2	18.20±1.2	36.40±1.0	84.0 ±7.4	
CB3	20.00±2.0	37.00±2.0	102.0±8.2	
CB4	19.01±0.9	42.01±0.1	106.0±1.2	
CB5	23.00±0.8	38.80±4.0	122.0±0.2	
CB6	40.60 ±9.7	45.00±0.1	98.40±7.2	
Mean	24.10±2.3	40.16±2.0	187.80±7.7	
Range	(18-40)	(41-88)	(84-1543)	
Mean(mining area)	90.68 ±2.5	65.45±2.7	630.95±15.9	

Table 4. Comparison of ²²⁶Ra, ²³²Th and ⁴⁰K activity concentrations in the Soil samples under this study with those from other places

Location	Activity concentrations (Bqkg ⁻¹)			References
	²²⁶ Ra	²³² Th	⁴⁰ K	
	Mean (range)	Mean (range)	Mean (range)	
NZ	40	88	768	Present study
	(24-75)	(69-130)	(220-1543)	-
CZ	207	76	937	Present study
	(45-948)	(42-92)	(23-1616)	
SZ	24	40	188	Present study
	(18-40)	(41-88)	(84-1543)	-
Bahi, Tanzania	37	51	875	[18]
	(9–60)	(28–107)	(542–1385)	
Likuyu-Tanzania	52	36	564	[5]
Mkuju,-Tanzania	245	80	1407	[19]
	(132 – 316)	(39 – 114)	(1169 – 1651)	
Indonesia	1042	1756	232	[15]
	(320-2195)	(993- 2683)	(116-438)	

Location	Activity concentrations (Bqkg ⁻¹)			References
	²²⁶ Ra	²³² Th	⁴⁰ K	
	Mean (range)	Mean (range)	Mean (range)	
Nile Delta-Egypt	23.59	35.53	266.41	[20]
Iraq	26	10	360	[16]
	(17-38)	(2-22)	(278-395)	
Ghana	14	24	162	[21]
	(2–31)	(6-94)	(40–552)	
Nigeria	55	91	287	[22]
	(9-198)	(5-502)	(35-1,359)	
Namibia	25	35	518	[8]
	(18-34)	(25-48)	(382-622)	
China	27	9	454	[23]
	(33-20)	(10 – 7)	(91 -51)	
World wide	32	45	420	[2]
	(16-110)	(11-64)	(140-850)	

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Table 5. Radium Equivalent (Ra_{eq}), External Hazard Index (H_{ex}). Internal hazard index (H_{in}) and Radioactivity Level Index estimated for the mining areas and the normal living areas in Kyerwa mining

Location	Ra _{eq} (Bqkg⁻¹)	H _{ex}	H _{in}	lγ
KB1	301.4	0.814	1.016	1.121
KB2	348.3	0.940	1.057	1.310
KB3	207.7	0.561	0.627	0.759
KG1	192.1	0.518	0.607	0.687
KG2	148.5	0.401	0.468	0.529
KG3	161.2	0.434	0.545	0.574
KS1	294.1	0.793	0.946	1.108
KS2	308.7	0.833	0.974	1.174
KS3	172.4	0.465	0.587	0.625
KS4	1070	2.893	5.456	3.590
KS5	194.4	0.525	0.748	0.708
KS6	289.9	0.781	0.935	1.097
CB1	130.9	0.353	0.417	0.493
CB2	76.72	0.317	0.256	0.270
CB3	80.76	0.214	0.272	0.285
CB4	87.25	0.235	0.286	0.308
CB5	87.87	0.237	0.299	0.311
CB6	112.5	0.303	0.413	0.393
Mean (mining area)	230.6	0.628	0.861	0.831

Table 6. Absorbed dose rate in air (DR), Annual effective dose equivalent (AEDE) and Annual gonadal equivalent dose (AGED) estimated for the mining areas and the normal living areas in Kyerwa mining

Location	D _R (nGy/h)	AEDE (mSv/y)	AGED (µSvy-1)
KB1	142.0	1.45	1004.2
KB2	163.5	1.72	1162.8
KB3	98.0	1.03	699.1
KG1	85.6	0.91	596.4
KG2	65.8	0.71	457.7
KG3	72.3	0.76	502.6
KS1	139.7	1.44	993.5
KS2	147.8	1.52	1055
KS3	79.01	0.82	552.8

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Location	D _R (nGy/h)	AEDE (mSv/y)	AGED (µSvy-1)
KS4	490.0	4.41	3290
KS5	91.70	0.91	641.3
KS6	138.4	1.42	986.3
CB1	62.07	0.64	441.3
CB2	32.92	0.36	234.7
CB3	35.87	0.37	242.8
CB4	38.60	0.41	267.6
CB5	39.18	0.41	271.5
CB6	50.07	0.51	344.5
Mean (mining area)	106.8	1.07	744.4

On the other hand, the activity concentrations of each radionuclide were compared from each zone (Fig. 2). In that case, the activity concentration of 226Ra at the central zone (CZ) was higher than the southern zone (SZ) by 88% and 81% from the north zone (NZ). The activity concentration of 232Th at the northern zone (NZ) was higher than the central zone (CZ) by 14% and 55% from the southern zone (SZ). Similarly, the activity concentration of 40K at the central zone (CZ) was higher than the northern zone (NZ) by 18%and 80% from the southern zone (SZ).

Radiological hazard assessments due to these natural radionuclides were also performed. Radium equivalent (Ra_{eq}) which is a single term that represents the combined specific activities of 226Ra, 232Th, and 40K and is used to indicate the external dose to the public was estimated. Other radiological hazard indexes such as external hazard index (Hex), internal hazard index (H_{in}), and radioactivity level Index ($I\gamma$) were also calculated. The results revealed that the

mean values of Ra_{eq} , H_{ex} , H_{in} , and $I\gamma$ were 230.6, 0.628, 0.861, and 0.831Bq/kg, respectively.

These values were lower than the worldrecommended values, which are 370 for $Ra_{eq} \leq$ 1 for H_{ex} , \leq 1 for H_{in} , and \leq 1 for Iv respectively [24]. Absorbed Dose Rate in Air (DR), Annual Effective Dose Equivalent (AEDE), and Annual gonadal equivalent dose (AGED) were also calculated. The results indicate that the mean values for DR, AEDE, and AGED were 106.8nGy/h. 1.07mSv/y, and 744.4µSvy-1 respectively. The estimated mean values were higher than those of the worldwide average which are 57 nGyh-1 for DR, 0.07mSv/y for AEDE, and 2984µSv/y for AGED [25]. Due to the elevated mean calculated values of the radiological hazard indexes, therefore, the mining activities in Kyerwa District can pose a radiological hazard to the mining community. Regular inspection of Tin Mining and monitoring of mining workers is recommended for further assessment of the level of activity concentrations of different radionuclides in the mining area.



Fig. 2. Comparison of activity concentration of ²²⁶Ra, ²³²Th, and ⁴⁰K in the study locations

5. CONCLUSION

Mining activities are among the major sources of radiation exposure [26]. The activity concentrations of 226Ra, 232Th, and 40K in soil samples from the Kyerwa Tin Mining area have been studied using Nal (TI) gamma-ray spectrometry. The mean activity concentrations of 226Ra, 232Th, and 40K in the soil samples from the mining sites were estimated to be 90.68, 65.45, and 630.95Bq/kg, respectively. The results of the mean activity concentrations of 226Ra, 232Th, and 40K in the mining sites are higher than the world average. The mean effective dose in this study is about 100% higher when compared to the world average. The results in the study area show that there are higher levels of natural radionuclides in the mining areas than the worldwide average. Therefore, mining activities in Kyerwa might pose a radiological hazard to the mining communities.

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COMPETING INTERESTS

Authors have declared that no competing interests exist.

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