



The radioactive contamination of ground and surface water near a uranium mine in Malawi

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Abstract: Ground and surface water in the uranium mining area of Kayelekera in Malawi was assessed for concentration levels of radioactive metals. Potential health risks associated with the intake of these metals in drinking water from various sources were also estimated. Surface, groundwater and mine discharge water samples were collected and analysed for radio elemental concentration using inductively coupled plasma mass spectrometry analytical technique. The results indicated a high concentration of ²³⁸U in water samples from lower Sere river. The activity concentrations of ²³⁸U, ²³²Th and ⁴⁰K were however below WHO recommended limit. Health risk assessment using average committed effective dose were below the global average. Excess lifetime cancer risk values with an average of 2.92×10^{-6} for borehole water was calculated and found to be below the global average. Radiologically, the water quality of Kayelekera area post uranium mining activities has not been compromised, however close monitoring and treating of drinking water is recommended.

Keywords: Kayelekera, Uranium mine, Radioactive element, ICP-MS.



La contaminación radiactiva de las aguas subterráneas y superficiales cerca de una mina de uranio en Malawi

Resumen: Se evaluaron los niveles de concentración de metales radiactivos en las aguas subterráneas y superficiales de la zona minera de uranio de Kayelekera, en Malawi. También se estimaron los riesgos potenciales para la salud asociados a la ingesta de estos metales en el agua potable procedente de diversas fuentes. Se recogieron muestras de aguas superficiales, subterráneas y de descarga de minas y se analizaron para determinar la concentración de radioelementos mediante la técnica analítica de espectroscopia de masas con plasma acoplado inductivamente. Los resultados indicaron una elevada concentración de ^{238}U en las muestras de agua del curso inferior del río Sere. Sin embargo, las concentraciones de actividad de ^{238}U , ^{232}Th y ^{40}K estaban por debajo del límite recomendado por la OMS. La evaluación del riesgo para la salud utilizando la dosis media efectiva comprometida se situó por debajo de la media mundial. Se calcularon los valores de riesgo excesivo de cáncer a lo largo de la vida con una media de 2.92×10^{-6} para el agua de pozo y se comprobó que estaban por debajo de la media mundial. Desde el punto de vista radiológico, la calidad del agua de la zona de Kayelekera tras las actividades de extracción de uranio no se ha visto comprometida, aunque se recomienda una estrecha vigilancia y el tratamiento del agua potable.

Palabras clave: Kayelekera, Mina de uranio, Elemento radiactivo, por inducción de plasma espectrometría de masas.

1. INTRODUCTION

Radioactive metals are known for their widespread availability owing to their difficulty to degrade. They are found in the environment, and they have attracted a significant amount of attention all over the world [1-3]. This is due to their persistency and the risks they pose to people, animals and the environment. The term radio-element along heavy metals are often researched in geochemistry and environmental pollution. [4-5]. Radioisotopes like heavy metals are also persistent in aquatic environments thus affecting the water quality as a result of pollution. In regard to the fact that water is a significant natural resource for varied life, it is thereof impossible for people, crops, and other living things to survive in an environment where the water quality is compromised.

As a consequence of the expansion of industrial and agricultural production as well as the extensive human activities that have taken place, large quantities of potentially toxic metals have been discharged into rivers and other bodies of water all over the world. Potentially dangerous metal residues may accumulate in the bodies of water that have been contaminated. Due to changes in sedimentary environments making it easy for these metals to cause "secondary pollution," and because they pose a significant threat to the environment, human and animal health via the food chain, and other migration routes [6], it is important to monitor them. Radioactive metals are an important category of environmental pollutants, and they can enter water bodies through a variety of natural or anthropogenic pathways [7]. They can also be found in water phases, sediments, and organisms, and they can exhibit a variety of environmental geochemical behaviours and toxic biological effects [8-10].

There are a number of factors, including geological location, the quality of uranium ore deposits, and the hydro-geometry of aquifers, that have the potential to affect the

concentration and distribution of toxic heavy metals [11]. Whenever water flows through waste rocks and soil in an area that was formerly or currently mined for uranium, the water eventually becomes tainted with radioactive particles. Surface water and groundwater are impacted by a decline in water quality as a result of an increase in the concentration levels of these pollutants [11-13]. This is because the deterioration affects both types of water.

Exposure to radioactive elements such as uranium can lead to a number of unfavourable outcomes during pregnancy, including birth defects and stillbirths [14-15]. These outcomes are in addition to the neurotoxicity, nephrotoxicity, and hepatotoxicity that they cause, as well as the impaired embryogenic development. Some research suggests that consuming water with a uranium content of more than $30 \mu\text{g}/\text{l}$ can have long-term negative effects on one's health, including kidney damage and the possibility of toxicity to the bones caused by alpha radiation [16-17].

Uranium mining pollutes the air, water and the soil and exposure to high concentration of uranium is harmful to both public health and the environment. However, mining laws have become more stringent particularly in developed countries, as a result of increased uranium mining and exploration in African countries [18-20]. Drinking water in Nigeria, South Africa, and Namibia has been found to have uranium levels that are higher than the limits set by the World Health Organization (WHO) [14,21-22]. On the other hand, there is a dearth of research that focuses on the concentrations and distributions of potentially toxic metals in surface water and sediments, particularly in the waters that are in the vicinity of uranium mines.

Investigations into the radioactive elements and toxic heavy metal concentrations in Kayelekera water were carried out as part of the preliminary environmental impact assessment that preceded the uranium mining. This was a requirement of the mining license regulator. There has been no follow-up research conducted on the impact of uranium mining on the levels of radioactive elements found in the water in the area. The purpose of this study

was to ascertain the levels of concentration of radioactive elements in the water sources of Kayelekera after uranium mining. The health risks associated with the water were also estimated in the study. The levels of radioactive element concentrations that are permissible according to the WHO and the United States Environmental Protection Agency (USEPA) were compared with the concentrations that were found in the water sources for this study in order to determine the area's water quality.

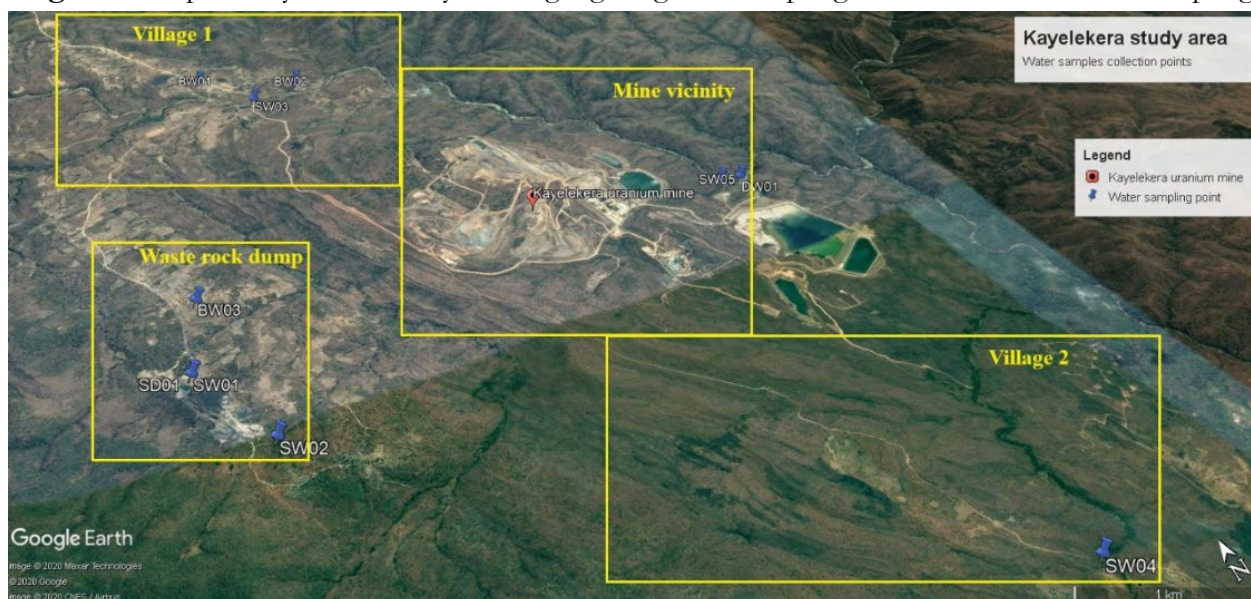
2. MATERIALS AND METHODS

2.1. Study Area

Malawi is in southeast Africa's tropical region. Kayelekera is 52 km west of Karonga, Malawi. Kayekelera is 33°41' east of the equator and 9°59' south. The faulted rift escarpment, topography, and heavily wooded landscape influence the microclimate of the area [23]. Only three main village settlements are within the mining area's borders. Locals in the study area mine and process uranium. Residents also grow local food crops. Around small village settlements and along streams and riverbanks, cultivation occurs.

The study area was divided into four sampling zones as illustrated in Figure 1. Two ground water samples from boreholes and one stream water sample were collected from village 1 sampling zone situated to the north of the Kayelekera uranium mine. One stream water sample was collected from village 2 located to the south of the Kayelekera uranium mine. One ground water sample from a borehole and two stream water samples were collected from a waste rock dump to the west of the Kayelekera uranium mine. One stream water sample and a sample of discharge water from the mine were collected from the mine vicinity sampling zone. The selection of sampling locations was based on accessibility, proximity to the mine and areas that are most frequented by the local inhabitants.

Figure 1: Map of Kayelekera study area highlighting four sampling zones and locations of sampling



2.2. Sample collection and preparation

A total of ten water samples were collected from the various locations within the four sampling zones as shown in Figure 1. Each sampling location was marked using a global positioning system (GPS), the geographical coordinates were recorded against corresponding sample codes for easy identification of each sample. The water samples were transported from Malawi to Centre for Applied Radiation Science and Technology (CARST) laboratory facilities at North-West University, Mafikeng campus in South Africa for subsequent treatment and analysis. At CARST laboratory facilities, the water samples were filtered with 4.5 μm pore size filter paper to remove all solid particles in the water. The filtered water samples were transferred into 20 ml vials.

2.3. Sample analytical method

Elemental concentrations of radioactive elements of uranium, thorium and potassium were analysed in the water samples using inductively coupled plasma mass spectrometry (ICP-MS). The spectrometer, a Perkin Elmer NexION 2000C, was operated under the total

quant method. The accuracy of the analytical method was validated using a 10 ml multielement calibration standard 3 (Perkin Elmer) with elements. ICP-MS calibration is done by measuring the instrumental response to a reference standard solution (a 10 µg/l multielement calibration standards Ag, Al, As, Ba, Be, Bi, Ca, Cd, Co, Cr, Cs, Cu, Fe, Ga, In, K, Li, Mg, Mn, Ne, Ni, Pb, Rb, Se, Sr, Tl, U, V and Zn) with Table 1 showing the calibration summary report and the calibration curves were obtained. Reliability and precision of the data was ensured by duplicating the analysis three times and blank samples were run in between analysis and reference standards as part of the quality management of the process.

The elemental concentration of radioactive elements in µg/l was estimated using the expression in equation 1 [24].

$$Concentration \left(\frac{\mu g}{l} \right) = C_i \times \frac{V_f}{W \times S} \times \frac{DF}{1000} \tag{1}$$

where C_i = instrument value in 11 g/l (the average of all replicate integrations), V_f = final digestion volume (ml), W = initial aliquot amount (g), S = % solids/100 and DF = the dilution factor.

Table 1: Perkin Elmer NexION 2000C ICP-MS instrumental working parameters

PARAMETERS	TYPE/ VALUES
Nebulizer	Meinhard® glass micro concentric
Cones	Nickel
Spray chamber	Glass cyclonic
Plasma gas flow	15.0 L/min
Nebulizer gas flow	1.00 – 1.05 L/min
RF power	1600 W
Mode of Operation	Standard mode (using argon gas)
Sweeps/readings	9
Replicates	3
Time per sample	4.5 minutes

2.4. Elemental concentration

The radiological hazard indices were assessed based on uranium, thorium and potassium activity concentrations in the water samples. Uranium has been identified as a nephrotoxin by the WHO and USEPA, as such, its effects on human health need to be investigated [25]. Activity concentrations of ^{238}U , ^{232}Th and ^{40}K were calculated from elemental concentrations of U and Th and the one percent of K using conversion factors as recommended by the IAEA technical document 1363 [26]; as shown in equations 2-4.

$$1\% K = 313 \text{ Bq/kg of } ^{40}\text{K} \quad (2)$$

$$1 \text{ ppm U} = 12.35 \text{ Bq/kg of } ^{238}\text{U} \quad (3)$$

$$1 \text{ ppm Th} = 4.06 \text{ Bq/kg of } ^{232}\text{Th} \quad (4)$$

2.5. Uranium source

The source of the uranium measured in groundwater was determined using the isotopic ratio of ^{234}U to ^{238}U and was based on the concentration of uranium radionuclides in the water, that is, 0.00548% of ^{234}U and 99.3% of ^{238}U [27].

2.6. Radiation hazard indices

In order to quantify the radiation hazard introduced by the radionuclides in water to the local inhabitants, Equation 5 was used to calculate the ingestion dosage E_{ing} (mSv/y) from the consumption of ^{238}U , ^{232}Th , and ^{40}K in the study area [28]:

$$E_{ing}(\text{mSv/y}) = CI_{ing} \sum_{j=1} DCF_{ing} \quad (5)$$

where C is the activity concentration of the radionuclides in a sample, I_{ing} is the consumption rate per year, DCF_{ing} is the effective dose coefficient in Sv/Bq for the ingestion

of natural radionuclides. Effective dose coefficients of 4.50×10^{-8} Sv/Bq, 2.30×10^{-7} Sv/Bq, 6.20×10^{-9} Sv/Bq were used for ^{238}U , ^{232}Th and ^{40}K , respectively. The average consumption rate of 600 l/y was used to estimate annual effective doses in the sampled water [29-30].

Excess lifetime cancer risk (ELCR) provides an estimate of the number of extra cancers expected to develop in a population as a result of exposure to gamma radiation from NORMs [31]. It therefore, gives the probability that a certain stochastic effect will occur in an individual or population exposed to low doses of ionizing radiation over a given period of time [28,31]. ELCR for the population of the Kayelekera study area was estimated using equation 6 [32]

$$ELCR = E_{ing} \times DL \times RF \quad (6)$$

where E_{ing} is the ingestion dose in equation 5, DL is duration of life (estimated to be 64 years) [33] and RF is the risk factor (Sv^{-1}). For risk assessment, the nominal probability coefficient recommended by the International Commission on Radiological Protection (ICRP) for radiation-induced stochastic health effects, which include fatal cancer and severe hereditary effects for the whole population, of $7.3 \times 10^{-2} \text{Sv}^{-1}$ is used [32].

3. RESULTS AND DISCUSSIONS

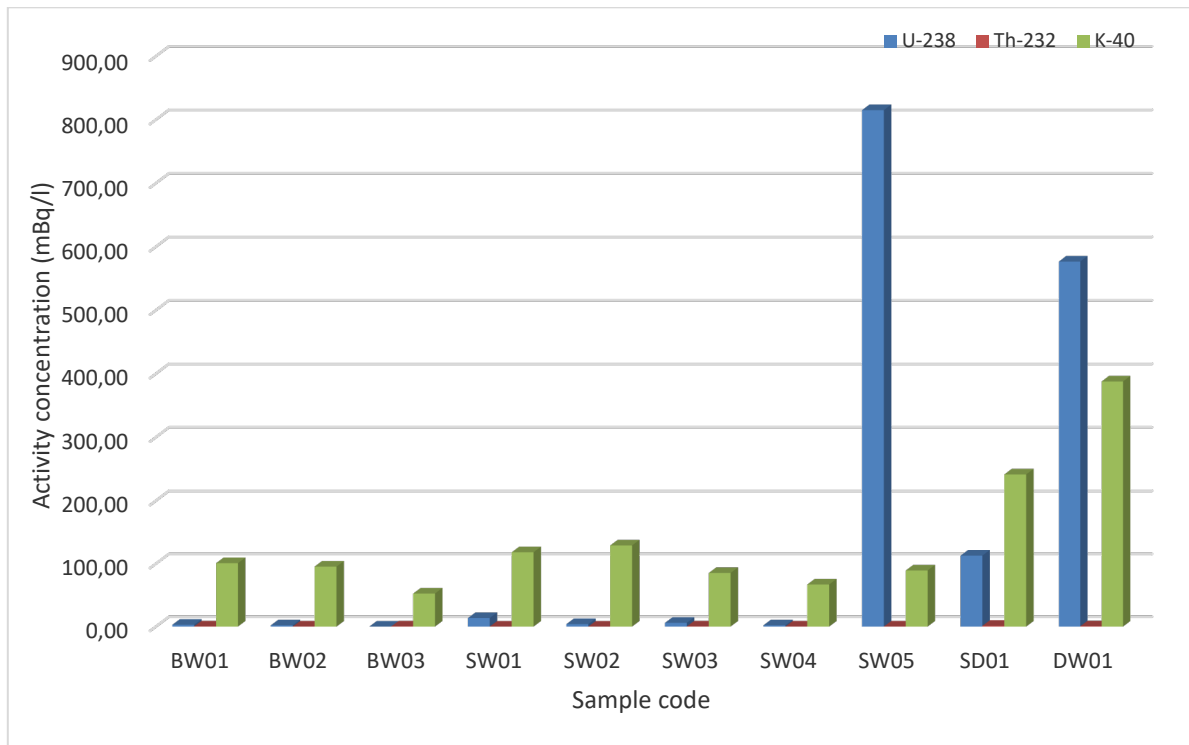
The dissolved radionuclide content in water samples from aquatic streams, boreholes and effluent from mine are presented in Table 2. The lower Sere stream which is the primary recipient of the effluent in the environment showed high concentrations of uranium at SW05 and DW01, respectively, as can be seen from Figure 2. Water samples sourced from boreholes around the study area showed insignificant concentrations of dissolved uranium in the village and waste rock dump but the high concentrations in the streams and the discharge can be attributed to the effluent from the mine tailings.

Table 2: The activity concentration of water samples collected from the study area

LOCATION	SOURCE OF WATER	SAMPLE CODE	ACTIVITY CONCENTRATIONS (mBq/l)		
			U-238	Th-232	K-40
Village 1	Borehole	BW01	2.84	0.27	100.94
Village 1	Borehole	BW02	1.99	0.04	95.51
Waste rock dump	Borehole	BW03	0.02	0.19	52.59
Waste rock dump	Stream	SW01	13.58	0.02	118.26
Coal mine	Stream	SW02	3.87	ND	129.16
Upper Sere river	Stream	SW03	5.91	0.26	85.32
Muswanga river	Stream	SW04	2.06	0.04	66.96
Lower Sere river	Stream	SW05	815.07	0.03	89.38
Waste rock dump	Stream	SD01	112.98	1.23	241.4
Lower Sere river	Mine discharge	DW01	576.61	0.41	387.81
		Min	0.02	ND	52.59
		Max	815.07	1.23	387.81
		Average	106.48	0.26	108.84
WHO 2011			10000	1000	-

The activity concentration of ^{40}K ranged from 52.59 to 387.81 mBq/l and an average of 108.84 mBq/l, while the highest activity concentration of ^{232}Th of 1.23 mBq/l was found in SD01. The mean activity concentrations of ^{232}Th in both surface and groundwater from the studied samples show no significant variations, this could be attributed to the same geological [compositions](#) of the studied areas, which are believed to be the major factor of radionuclide content in water [34]. The activity concentration in this study was compared with similar investigations from Namibia [35].

Figure 2: Activity concentration of radionuclides in water sample location near Kayelekera uranium mine



The activity concentration of the natural radionuclides from the boreholes, streams and mine discharge in Figure 3, has a value below the WHO guidance level of 10 Bq/l and 1 Bq/l for ^{238}U and ^{232}Th respectively [36]. This guidance level of radionuclide concentration represents the concentration that if present in drinking water consumed through the year would result in an individual dose of 0.1mSv and does not exceed the acceptable nominal risk to the health of the population over a lifetime of consumption [36-38]. ^{40}K does not accumulate inside the body and remains at a stable level independent of its intake [39]. This shows that there is little natural radioactivity present in the water sources in the research area that are intended for drinking and domestic use. It is vital to keep in mind though, that some of the surface water bodies in the mines' restricted access regions are inaccessible to the general public for residential use. Nevertheless, some residents of the communities still use nearby surface water bodies for domestic reasons, despite the fact that the boreholes provide

drinking water for the communities. The primary source of the mines' subterranean water supply is used for both household purposes and processing facility.

Figure 3: The activity concentration of the natural radionuclides from boreholes, streams and mine discharge

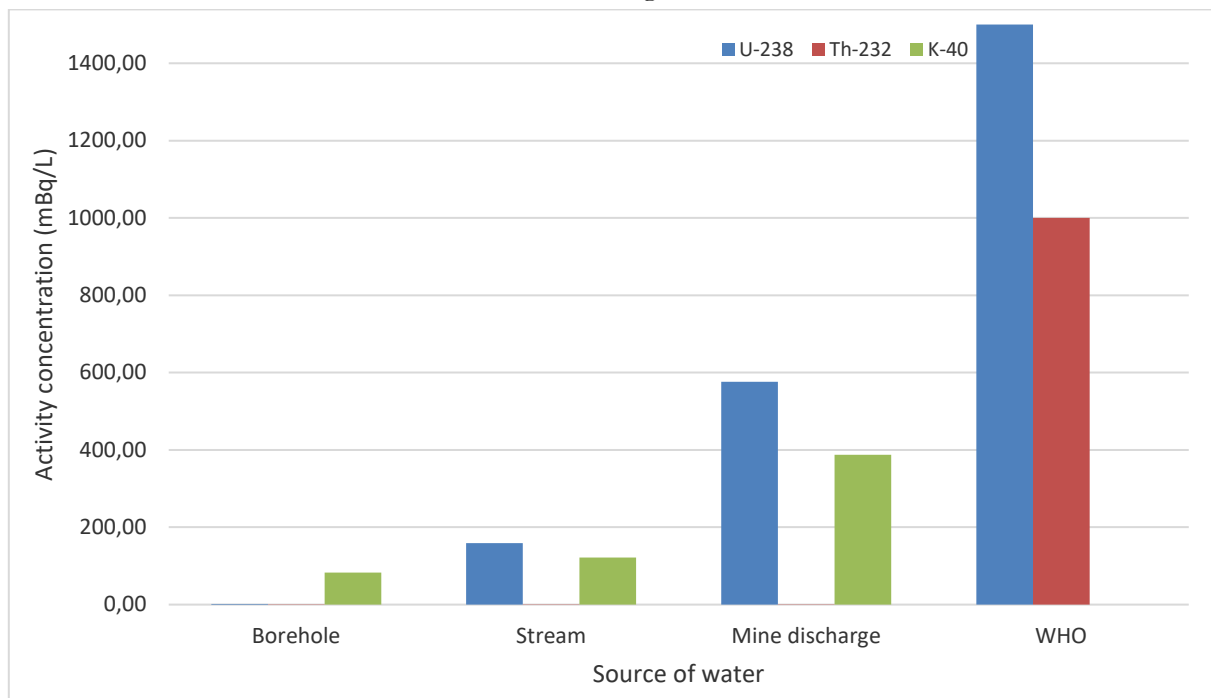
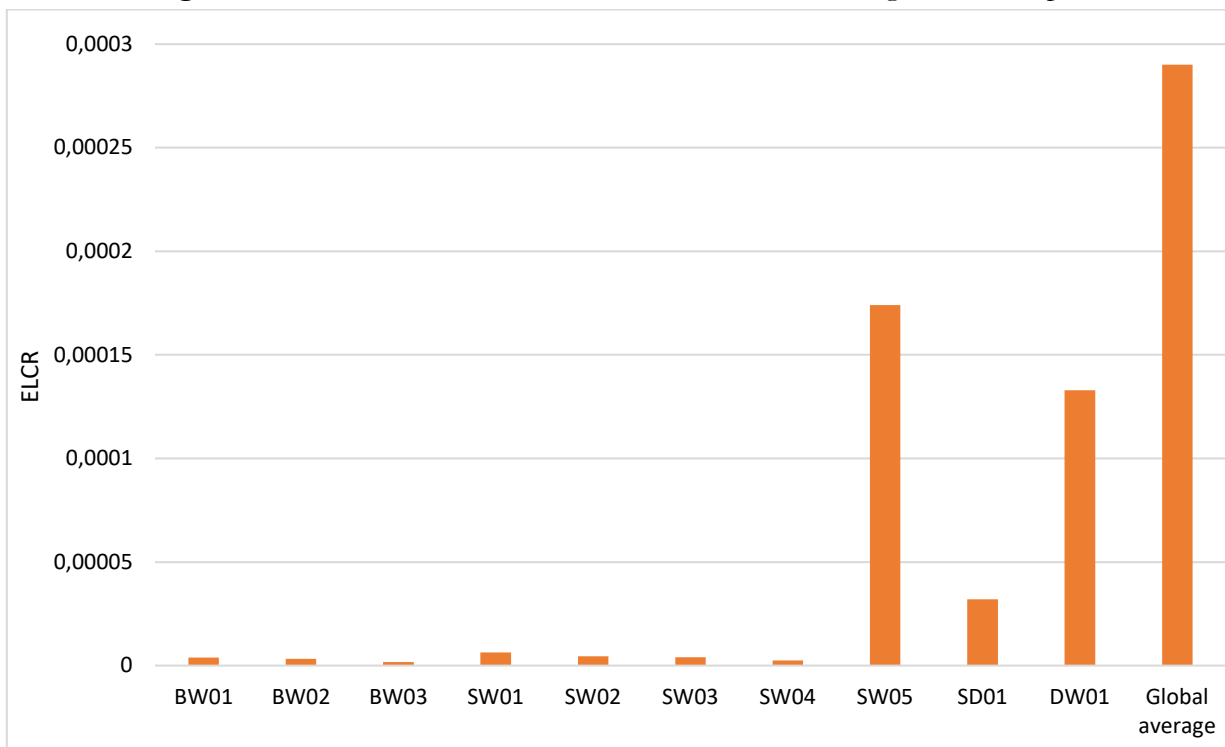


Table 3: Ingestion dose of NORMS in water around Kayelekera uranium mine

SAMPLE SOURCE	SAMPLE CODE	INGESTION DOSE, E_{ing} (mSv/y)				ELCR
		U-238	Th-232	K-40	TOTAL	
Borehole	BW01	1.28E-07	6.21E-08	6.26E-07	8.16E-07	3.81E-06
	BW02	8.96E-08	9.20E-09	5.92E-07	6.91E-07	3.23E-06
	BW03	9.00E-10	4.37E-08	3.26E-07	3.71E-07	1.73E-06
Stream	SW01	6.11E-07	4.60E-09	7.33E-07	1.35E-06	6.30E-06
	SW02	1.74E-07	0.00E+00	8.01E-07	9.75E-07	4.55E-06
	SW03	2.66E-07	5.98E-08	5.29E-07	8.55E-07	3.99E-06
	SW04	9.27E-08	9.20E-09	4.15E-07	5.17E-07	2.42E-06
	SW05	3.67E-05	6.90E-09	5.54E-07	3.72E-05	1.74E-04
Mine discharge	SD01	5.08E-06	2.83E-07	1.50E-06	6.86E-06	3.21E-05
	DW01	2.59E-05	9.43E-08	2.40E-06	2.84E-05	1.33E-04
	Min	9.00E-10	0.00E+00	3.26E-07	3.27E-07	1.73E-06
	Max	3.67E-05	2.83E-07	2.40E-06	3.94E-05	1.74E-04
	Average	6.91E-06	5.73E-08	8.48E-07	7.81E-06	3.65E-05

Table 3 shows the ingestion dosage and ELCR associated with water intake. The average committed effective dose due to ^{238}U , ^{232}Th , and ^{40}K intake in water is $6.91\text{E-}06$, $5.73\text{E-}08$, and $8.48\text{E-}07$ mSv/y, respectively. They are lower than the global average of 2.4 mSv/y [28,40]. The ELCR values were below the global average published by the United Nations Scientific Committee on the Effects of Atomic Radiation of $0.29\text{E-}03$, as can be seen in Figure 4. This shows a negligible radioactive risk for the tested radionuclides (^{238}U , ^{232}Th , and ^{40}K), indicating that the water in the studied region is safe for residential and other uses.

Figure 4: ELCR for the borehole, stream and mine discharge water sample



Statistical description

Descriptive statistical data, which include the minimum, maximum, mean, standard error and standard deviation of the measured radionuclides are presented in Table 4. The standard deviations of the measured primordial radionuclides, ^{238}U , and ^{232}Th , were higher than their respective mean values, which suggests a significant level of non-uniformity in

their distribution as against the symmetry distribution [41], while potassium is evenly distributed which can be attributed to its abundance in the earth’s crust.

Table 4: Descriptive statistical data of the measured radionuclides in water around Kayelekera area

	N STATISTIC	MINIMUM STATISTIC	MAXIMUM STATISTIC	MEAN STATISTIC	STD. DEV STATISTIC	SKEWNESS		KURTOSIS	
						STATISTIC	STD. ERROR	STATISTIC	STD. ERROR
U-238	10	0.02	815.07	153.49	293.30	1.88	0.69	2.36	1.33
Th-232	9	0.02	1.23	0.28	0.38	2.32	0.72	5.90	1.40
K-40	10	52.59	387.81	136.73	102.31	2.03	0.69	4.00	1.33
Valid N (listwise)	9								

Data variability of the measured radionuclides sequence as reflected by the frequency distribution histograms in Figure 5-6, was tilted to a region of the study area through the skewness level for the investigated uranium and thorium isotope.

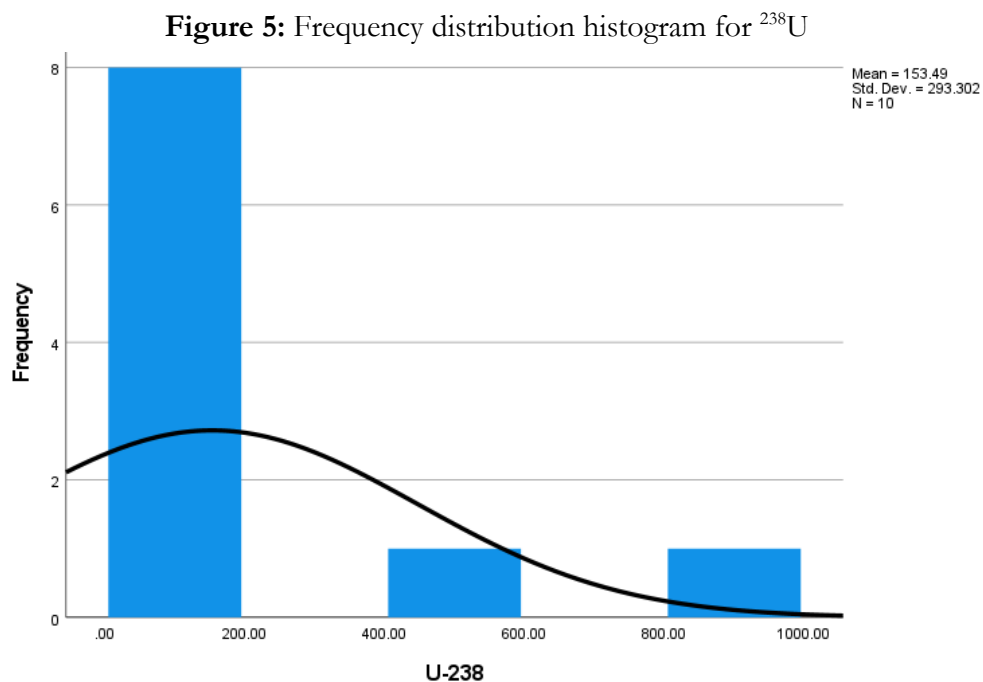
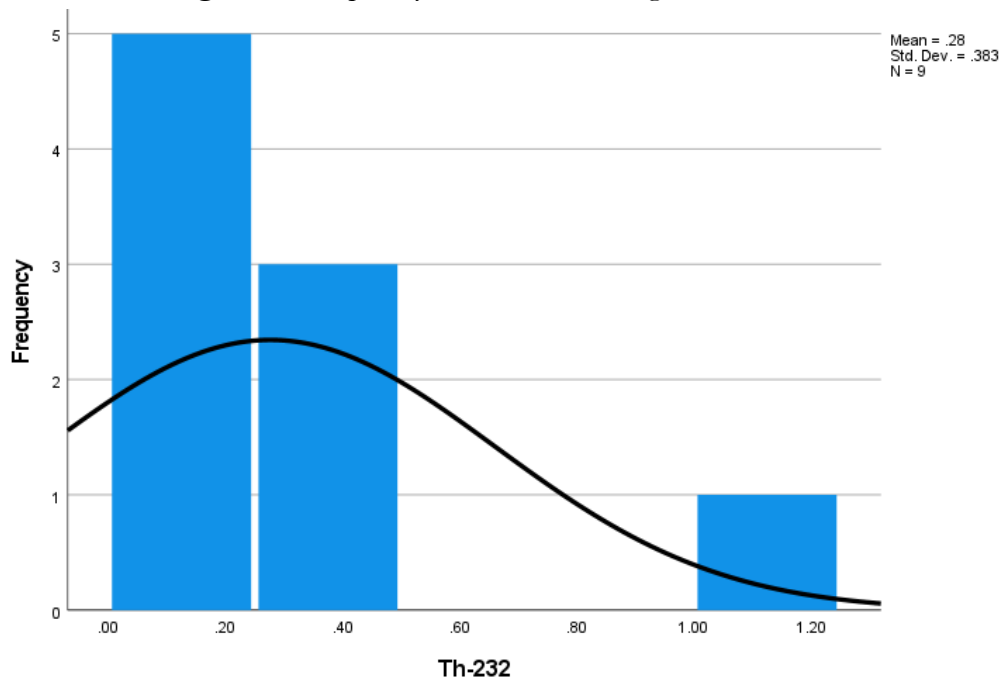


Figure 6: Frequency distribution histogram for ^{232}Th



4. CONCLUSIONS

The levels of ^{238}U , ^{232}Th and ^{40}K concentrations in water sources around the Kayelekera study area were measured utilizing the inductively coupled plasma mass spectrometry technique in order to assess the water quality in the area. High concentration of ^{238}U were recorded at SW05 and DW01 along the lower Sere river. The measured activity concentrations of ^{238}U , ^{232}Th and ^{40}K were however below the recommended limits set by the WHO [37]. The results indicate that the water sources of the Kayelekera study area generally have a low radio-elemental content for these species.

Potential health risks associated with ingestion of these metals evaluated through average committed effective dose were found to be lower than the global average. This indicates that the water from these sources have no potential of causing health issues radiologically if ingestion occurs for prolonged periods of time. The low values in this study suggest that the population of the Kayelekera study area is not at any imminent health risk

due to radioactive metals in water, however long-term exposure to ingestion of the water may be hazardous to the population overtime.

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CONFLICT OF INTEREST

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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