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Evaluation of Radiological Risk Hazards in Sediments and Selected Streams Around Kalulushi and Kitwe Towns

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Abstract

Mining operations in the uraniferous Copperbelt Province of the Katanga Basin have raised significant environmental and health concerns, primarily due to the generation of waste during copper and cobalt extraction processes. Kitwe town heavily relies on the Kafue River for its domestic water supply. However, studies measuring the levels of natural radionuclides in several tributaries of the Kafue River, namely, the Fikondo Stream, Mindolo Stream, Kitwe Stream, and Mwambashi River, are lacking. This study utilized gamma-ray spectrometry to assess the presence of radionuclides, specifically radium-226, thorium-232, and potassium-40, in sediments and water near Kitwe and Kalulushi. The radionuclide concentrations varied, with sediments showing 51.8 to 104.6 Bq/kg and 43.1 to 72.2 Bq/kg and water showing 1.7 to 28.5 Bq/l and 0 to 1.3 Bq/l. These concentrations were compared to permissible limits set by the UNSCEAR and WHO. Sediment samples had higher radionuclide activity than water with ^{226}Ra often exceeding the WHO limits, while ^{232}Th and ^{40}K were generally greater in all stream samples. To assess the radiological hazard, parameters, such as the radius equivalent, absorbed dose rate, absorbed equivalent dose, internal hazard index, external hazard index, and excess lifetime cancer risk, were calculated.

Keywords: Radiological hazards, Natural radioactivity, Absorbed dose rate, Mining activities, Excess lifetime cancer risk, Environmental health, Health concerns

Introduction

Large mining activities in the uraniferous Copperbelt Province of the Katanga Basin have raised many concerns about environmental and health implications due to the waste generated during copper and cobalt mining and processing [1]. Since the levels of uranium and other radioactive nuclides emanating from these mining activities are thus far poorly characterized, regulatory authorities such as the Radiation Protection Authority (RPA), the Zambia Environmental Management Agency (ZEMA) and the Mine Safety Department (MSD) hitherto lack radiometric environmental data for mining industry wastewater receiving streams around Kitwe and Kalulushi, as well as baseline data or data showing evidence of pollution. Establishing significant levels of radionuclides in freshwater streams, such as the Fikondo, Mindolo, Kitwe and Mwambashi Rivers, could be a potential environmental health problem since industrial wastewater from mining and metallurgical operations is discharged in these streams, which are tributaries of the Kafue River. The Kafue River is a major source of domestic water for Kitwe town. The Fikondo Stream, for example, receives most of the Nkana Mine wastewater draining

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tailings dam TD15A [2], while the Mwambashi River drains the Chambishi-Nkana Basin, which receives runoff from the Chambishi Mine north of Kitwe and from the large tailing facilities of the Nchanga Mine via the Muntimpa Stream and eventually drains into the Kafue River [3].

No studies have been conducted to date to measure natural radionuclides in the Fikondo Stream, Mindolo Stream, Kitwe Stream and Mwambashi River, which are tributaries of the Kafue River. The main objective of this project was to measure the levels of natural radionuclides in the sediments and water of the Fikondo and Mindolo streams as well as those of the Mwambashi River as a potentially polluted freshwater course receiving industrial wastewater and the Kitwe Stream as a control with non-waste-bearing water around Kitwe Town on the Copperbelt. The evidence of potential for natural radionuclides in the study area is as follows: In the 1950s, there was uranium processing near Mindolo Shaft of the Nkana Mine [4], whose waste was a concern of the Copperbelt Environment Project (CEP) during the privatization of the Zambian mining industry at the beginning of the 21st century [5]. Concerns about natural radioactivity in CEP were emboldened by the National Institute for Scientific and Industrial Research (NISIR) studies of naturally occurring radioactive materials (NORM) [6] and [7].

For example, in 1997, for the first time in Zambia, research was conducted to measure radon levels in several underground mines on the Copperbelt. This preliminary radon survey, which was concluded in 1998, showed that at least 30% of the forty-two randomly sampled sites in eight underground mines had radon levels greater than 1000 Bq/m^3 [8]. Furthermore, between 2008 and 2010, NISIR conducted research to assess uranium levels in the drinking water of three Copperbelt towns, namely, Chambishi, Chingola and Kitwe. The survey results showed that the activity concentration of uranium isotopes in drinking water varied over a wide range, and the activity concentration in underground mine water was higher than that in surface water, particularly for Chambishi Town, whose values ranged from 57.5 to 340 mBq/l with a committed effective dose of 20.28 µSv/yr [9].

Method and Instrumentation

On 27 November 2015, a total of twelve water and sediment samples were collected from the Fikondo, Mindolo, and Kitwe streams and the Mwambashi River. The samples consisted of four sediment, six stream water and two river water samples. Water samples were collected in 2.5 L high-density polyethylene containers, while sediments were collected in low-density polyethylene bags. Global positioning system (GPS) coordinates were recorded immediately after sampling at each site. Water samples were preserved by adding 1 ml of concentrated nitric acid to a pH of 2 and transported to the laboratory. The sediment samples were air-dried to avoid the loss of radionuclides. The dried samples were thoroughly ground to a pore size of 75 µm. The moisture content of the dried, homogenous samples was determined using the gravimetric drying oven method with Equation 1.

$$\text{Moisture Content} = \frac{W_2(g) - W_3(g)}{W_3 - W_1(g)} * 100\% \qquad \text{Eq: 1}$$

where W_1 is the weight of the container, W_2 is the weight of the container plus wet soil and W_3 is the weight of the container plus dry soil.

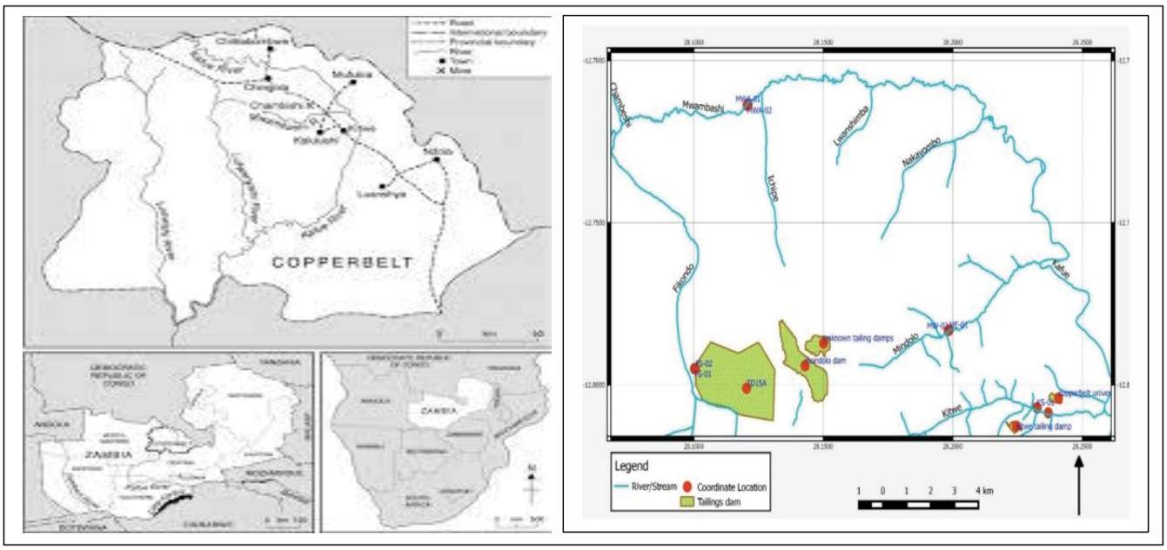

Figure: 1 Location map showing Zambia and (Von Der Heyden C.J., 2003)

Figure: 2 Topographical map of the study Copperbelt Province area (Kitwe and Kalulushi areas).

Source: Researchers field work

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Sample Preparation

The samples were prepared according to the International Atomic Energy Agency (IAEA) guidebook for the measurement of radionuclides in food and the environment [10]. Acidified 500 ml water samples were evaporated at 105°C to a volume of approximately 60 ml using an airflow-equipped oven. The samples were placed on Petri dishes, sealed with Parafilm and left for one month to reach radioactive secular equilibrium. The method was modified to suit the use of Petri dishes instead of Marinelli beakers. The sample geometry and sample weight and/or volume were validated by the use of three IAEA reference materials.

Quality Control

Prior to use, energy and efficiency calibration of the gamma spectrometer was performed, and the results were validated using reference materials, namely, IAEA-RGK-1, IAEA-RGTh-1, and IAEA-RGU-1. These reference materials were also measured together with the samples as part of an internal QC/QA programme in line with ISO 17025 for radio-analytical laboratories [11] and [12]. The calibrated gamma-ray spectrometer was checked for changes in efficiency at low, medium and high energies using standard point sources of ^{133}Ba, ^{57}Co and ^{137}Cs. The background radioactivity distribution in the environment around the detector was checked by counting an empty sealed petri dish with the same geometry as the samples. The background measurements were performed at regular intervals, and the results were plotted on a control chart.

Gamma Ray Spectrometry Measurement

The prepared sediment and water samples were analyzed using a Canberra hyperpure broadband germanium detector (BEGe) connected to a digital spectrum analyzer (DSA-1000) with a DC voltage of +4000 V and 68% detector efficiency. The detector was connected to the Genie 2000 computer program, ver. 3.3. The window for spectral acquisition and analysis software matched gamma energies to a library of possible isotopes. The detector was shielded by a thick lead with a copper lining inside the counting chamber. The activity concentrations of radionuclides in Bq/l for water and Bq/kg dry sediment samples were calculated (after background decay correction) using Equation 2.

$$A = \frac{N}{\varepsilon \times m \times t \times \gamma} \qquad \text{Eq: 2}$$

where N is the net peak area of a peak at a characteristic energy, ε is the detector efficiency, m is the sample volume or mass in either liters or kilograms, t is the total counting time and γ is the probability of the emission of the radionuclide of interest.

Theoretical calculation of radiological effects

The radiation equivalent (Ra_{eq}eq), absorbed dose rate (D), annual effective dose equivalent (AEDE) and external radiation hazard index (H_{ex}) were used to assess the primordial nuclides of naturally occurring radioactive materials, namely, ^{226}Ra, ^{232}Th, and ^{40}K [13]

Radium equivalent (Ra_{eq})

The exposure to γ-rays of materials that contain ^{238}U, ^{232}Th, and ^{40}K is defined in terms of the radium equivalent values given by Equation 3 [32].

$$Ra_{eq}\left(\frac{Bq}{kg}\right) = A_{Ra} + 1.43A_{Th} + 0.077A_k \leq 370 \qquad \text{Eq: 3}$$

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

Absorbed dose rate (D)

The absorbed dose rate in air outdoors at 1 m above the ground surface due to the activity concentrations of ^{238}U, ^{232}Th, and ^{40}K is defined by Equation 4 [31].

$$D\left(\frac{nGy}{h}\right) = (DCF_K \times A_K) + (DCF_U \times A_U) + (DCF_{Th} \times A_{Th}) \qquad \text{Eq: 4}$$

where DCF_K, DCF_U and DCF_{Th} are the dose conversion factors in nGy/h/Bq/kg, whose values are 0.0417, 0.462 and 0.604, respectively, and A_K, A_{Ra} and A_{Th} are the potassium, uranium and thorium activity concentrations in Bq/kg, respectively.

Annual effective dose equivalent (AEDE)

The annual effective dose equivalent due to the activity in sediments was calculated by Equation 5.

$$AEDE\left(\frac{mSv}{y}\right) = D\left(\frac{nGy}{h}\right) \times \frac{24h}{day} \times \frac{365.25days}{y} \times 0.2 \times 0.7\frac{Sv}{Gy} \times 10^{-6} \qquad \text{Eq: 5}$$

where 0.7 Sv/Gy is the conversion coefficient from the absorbed dose in the air to the effective dose received by adults, 0.2 represents the outdoor occupancy factor [13], and D is the absorbed dose rate.

The maximum permissible limit of Ra_{eq} activity is 370 Bq/kg. This corresponds to an annual effective dose equivalent (AEDE) of 1 mSv/yr for the general public. For radiation workers, the five-year average is 100 mSv [14].

The annual effective dose (AED) resulting from the ingestion of water was estimated based on the assumption that the daily intake of water per person is 2 L per day [15] from the expression given in Equation 6 [16].

$$AED\left(\frac{mSv}{y}\right) = \sum_i I \times A \times C \times 365 \qquad \text{Eq: 6}$$

where I is the water intake per day (L/day), A is the daily intake of radionuclides (Bq/L) and C is the ingestion coefficient of individual radionuclides (Bq/L).

The effective doses resulting from the intake of ^{226}Ra, ^{40}K

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and ^{232}Th are determined directly from water. The AED is calculated with the ingestion of individual radionuclides and the ingestion dose coefficient reported by the International Commission on Radiological Protection (ICRP) as 4.5×10^{-7}, 6.2×10^{-9} and 2.3×10^{-7} Sv/Bq, respectively [17].

External radiation hazard index (H_{ex})

The external hazard index (H_{ex}) was calculated from Equation 7 [18].

$$H_{ex} = \left(\frac{A_{Ra}}{370}\right) + \left(\frac{A_{Th}}{259}\right) + \left(\frac{A_K}{4810}\right) \leq 1 \quad \text{Eq: 7}$$

where A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{238}U (or ^{226}Ra), ^{232}Th and ^{40}K, respectively.

Internal hazard index (H_{in})

The internal hazard index is less than unity for the radiation hazard to be negligible. Internal exposure to radionuclides is very hazardous, can lead to diseases such as cancer, and can be evaluated using equation 8 [19].

$$H_{ex} = \left(\frac{A_{Ra}}{185}\right) + \left(\frac{A_{Ta}}{259}\right) + \left(\frac{A_K}{4810}\right) \quad \text{Eq: 8}$$

where A_{Ra} or A_{Ra} and A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively.

Excess lifetime cancer risk (ELCR)

The excess lifetime cancer risk (ELCR) was calculated using Equation 9 [20].

$$ELCR = AED\left(\frac{mSv}{y}\right) \times DL \times RF \quad \text{Eq: 9}$$

where AED is the annual equivalent dose, DL is the average duration of life (estimated to be 70 years), and RF is the risk factor (Sv^{-1}), i.e., fatal cancer risk per sievert for stochastic effects. The ICRP has an RF of 0.05 for the public.

Correlation studies

To determine the extent of the presence of radioactive nuclides at a particular location, correlation studies were performed between combinations of the (^{226}Ra/^{232}Th), (^{226}Ra/^{40}K) and (^{232}Th/^{40}K) radionuclides using Microsoft Office Excel 2016. Excel software was also used to calculate the coefficient of variability, $R^{2,}$ which is a measure of the proportion of variability in a dataset [21]. The elemental concentration of the radionuclide in the samples was calculated from the activity concentrations in Bq/kg using the conversion factor in Equations 10 to 12, and correlation studies were performed on the K/Th, Th/Ra and Ra/K pairs [22].

$$1\%\ K = 313\left(\frac{Bq}{kg}\right)\ ^{40}K \quad \text{Eq: 10}$$

$$1\ ppm = 12.35\ \left(\frac{Bq}{kg}\right)\ ^{238}U \quad \text{Eq: 11}$$

$$1\ ppm = 4.06\ \left(\frac{Bq}{kg}\right)\ ^{232}Th \quad \text{Eq: 12}$$

Results and Discussion

The ^{238}U activities for samples in radioactive equilibrium were evaluated using the 351.92 keV line of ^{214}Pb and the 609.31 keV line of ^{214}Bi. The gamma-ray energies of 728 keV, 239 keV and 912 keV for ^{212}Bi, ^{212}Pb and ^{228}Ac, respectively, were used to estimate the activity of ^{232}Th. However, the activity concentrations of ^{40}K were measured directly using its own gamma ray line (1460.81 keV). The study's evaluation data for the natural decay series of ^{226}Ra, ^{235}U and ^{232}Th are shown in Table 1 and Table 2.

Table 1: Activity (dry mass) in Bq/kg for ^{226}Ra, ^{232}Th and ^{40}K in Sediment samples

Sample Site	Sample ID	^{40}K	^{232}Th	^{226}Ra
			(Bq/kg)	
Fikondo	S01 - FS	574.2 ± 56	72.2 ± 7	62.9 ± 6
Mwambashi	S02 – MR	724.5 ± 163	53.7 ± 12	51.8 ± 12
Mindolo	S03 – MS	780.7 ± 62	56.5 ± 6	60.8 ± 6
Kitwe	S04 – KS	906.8 ± 66	43.1 ± 4	104.6 ± 17
Mean		746.5 ± 87	56.4 ± 7	70.0 ± 10
Range		574.2– 906.8	43.1 – 72.2	51.8 – 104.6
UNSCEAR		500	50	50

Source: Researchers field work, 2015

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Table 2: Activity in (Bq/L) for ^{226}Ra, ^{232}Th and ^{40}K in water samples (ND* - Not Detected)

Sample Site	Sample ID	^{40}K	^{232}Th	^{226}Ra
			(Bq/L)	
Fikondo	FS-01	45.0 ± 7	ND	ND
	FS-02	43.4 ± 6	ND	ND
Mwambashi	MWA-01	42.6 ± 6	ND	17.9 ± 4
	MWA-02	37.9 ± 6	1.3 ± 0.5	2.0 ± 0.5
Mindolo	ME-01	39.5 ± 6	ND	ND
	MW-02	45.2 ± 6	ND	ND
Kitwe	KS-01	40.8 ± 6	ND	28.5 ± 6
	KS-02	46.3 ± 6	ND	1.7 ± 0.6
Mean		42.6 ± 6	1.3 ± 0.5	12.5 ± 3
Range		37.9 - 46.3	0 – 1.3	1.7– 28.5
WHO Limit		10.0	1.0	1.0

Source: Researchers field work, 2015

The ranges of activity concentrations of ^{40}K, ^{232}Th and ^{226}Ra in the sediments presented in Table 1 were 574.2 ± 56 to 906.8 ± 66, 43.1 ± 4 to 72.2 ± 7 and 51.8 ± 12 to 104.6 ± 88 Bq/kg, respectively, while the range of ^{40}K in the water was 39.6 ± 46.3 ± 6. ^{226}Ra and ^{232}Th were not detected in most water samples. The activity concentrations of ^{40}K in the sediment and water samples were all above the world permissible and WHO limits. ^{232}Th was below the world permissible limit in the S04 – KS sediment sample, while in the MWA-02 stream sample, it was above the WHO limit. The activity concentration in the sediment samples was also above the UNSCEAR 2000 world permissible limit. In the water samples, only the ^{238}U values of MWA-01 and KS-01 were above the WHO limit, while they were not detected in the ME-01, MW-02, FS-01 and FS-02 water samples. The elemental concentration ranges in the sediment samples were calculated to be 1.83 – 2.90% for ^{40}K, 10.63 – 17.77 ppm for ^{232}Th and 4.19 – 8.47 ppm for ^{226}Ra, as shown in Table 3.

The correlation coefficients between the activity concentrations of ^{238}U/^{232}Th, ^{226}Ra/^{40}K and ^{232}Th/^{40}K were $R^2 = 0.71$, $R^2 = 0.0$ and $R^2 = 0.80$, respectively. This shows that there is a good correlation between ^{226}Ra/^{232}Th and ^{232}Th/^{40}K. The correlation of the elemental concentrations was also determined as K/Th ($R^2 = 0.86$), Th/Ra ($R^2 = 0.85$) and Ra/K ($R^2 = 0.05$). According to Tzortzis and Tsertos (2004) as well as Al-Hamameh and Awadallah (2009), the theoretical values of U/Th are expected to be approximately 3.0 for normal continental crust [23] and [24].

Figure 3 shows a comparison of the activity values of a control stream (Kitwe Stream) against those of suspected streams (Fikondo, Mindolo Streams and Mwambashi River). The control contained higher activity values of ^{226}Ra and ^{40}K with a lower value of ^{232}Th in comparison to the susceptible streams.

Table 3: Elemental Concentration of ^{226}Ra, ^{232}Th and ^{40}K in Sediment samples with their ratios

SAMPLE CODE	^{238}U (ppm)	^{232}Th(ppm)	^{40}K (%)	Th/U	K/U	K/Th
S01 - FS	5.09	17.77	1.83	3.49	0.36	0.10
S02 – MR	4.19	13.16	2.31	3.14	0.55	0.17
S03 – MS	4.92	13.83	2.49	2.81	0.51	0.18
S04 – KS	8.47	10.63	2.90	1.25	0. 54	0.34
Mean	5.67 ± 2	13.85 ± 3	2.38 ± 0.4	2.67 ± 1	0.49 ± 0.09	0.20 ± 0.10
Range	4.19-8.47	13.16-17.77	1.83-2.90	1.25-3.49	0.36-0.55	0.10-0.34
IAEA	2 – 3	8 – 12	2 – 2.5			

Source: Researchers field work, 2015

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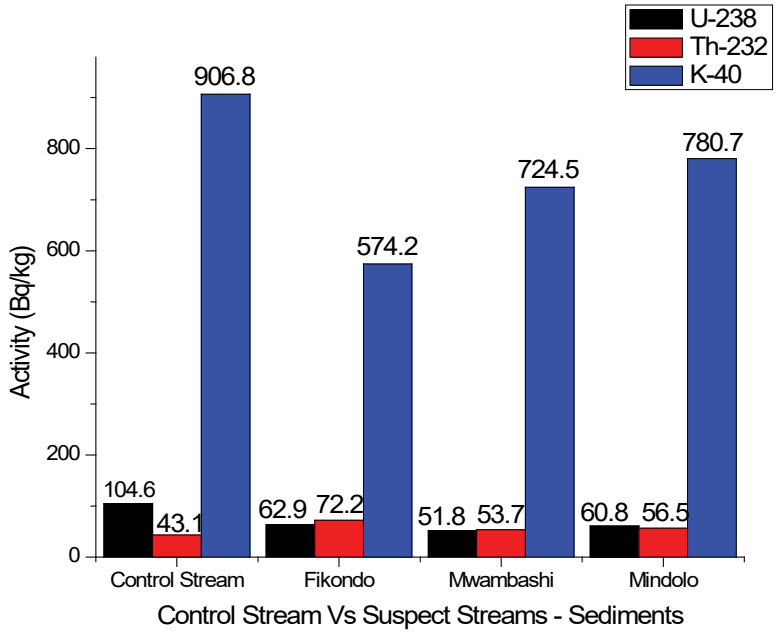

Figure 3: Comparative activity (Bq/kg) values of suspect streams and control stream
Source: Researchers field work, 2015

The radiological hazard to the public, due to radioactivity arising from radionuclides contained in all sediments and water samples collected from the study area, was assessed. The estimated absorbed dose rates based on the radioactivity in the sediment and water samples ranged from 86.4 to 112.2 nGy/hr and from 1.7 to 14.9 nGy/h, respectively. The ADR was higher than the world permissible limit in sediments and below the limit in water samples. The annual effective doses for the different sediment and water locations ranged from 0.11 to 0.14 mSv/yr and from 1.18 to 11.2 mSv/yr, respectively, which are higher than the worldwide effective dose of 0.07 mSv/yr [25]. The Ra_{eq} activity ranged from 183.9 to 236.1 Bq/kg and 3.04 to 31.6 Bq/l for sediment and water, respectively. The Ra_{eq} reported in this study was lower than the value of 370 Bq/kg recommended by the Organization for Economic Cooperation and Development (OECD). The calculated values of H_{ex}, H_{in} and ELCR ranged from 0.50 to 0.64 in the sediments; 0.01 to 0.09; 0.01 to 0.16 in the streams; 0.37 to 0.48; and 0.63 to 39.2, respectively. These results show that H_{ex} and H_{in} are below the limit of unity in both sediments and water, which means that the radiation dose is below the permissible limit of 1 mSv/yr recommended by the IAEA [26]. The ELCR for sediments is below the maximum limit, while in water samples, it is above the limit. The activity concentrations of ^{238}U, ^{232}Th and ^{40}K are comparable to the world average values and those of other African countries [13].

Conclusion

Natural radioactivity in sediment and water samples from the Kitwe and Kalulushi streams was evaluated using gamma ray spectrometry. This study revealed the presence of ^{226}Ra,

Table 4: Comparative average values for ^{238}U, ^{232}Th and ^{40}K

	^{226}Ra	^{232}Th	^{40}K	
LOCALITY		(Bq/kg)		REFERENCE
Algeria	23 ± 2	18 ± 2	310 ± 3	(Amrani & Tahtat, 2001)
Cameroon	8 ± 2	0.35	19 ± 2	(Ngachin N, 2005)
Egypt	40 ± 8	20 ± 3	157 ± 7	(Abbady, Uosif, & El-Taher, 2005)
Nigeria	2	1	7	(Ademola, Hammed, & Adejumobi, 2008)
Worldwide Average	77	84	1200	(UNSCEAR, 2008)
This Study	70 ± 10	56.4 ± 7	746.6 ± 87	

Source: Researchers field work, 2015

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^{232}Th and ^{40}K in sediments and ^{226}Ra and ^{40}K in water. The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the sediments ranged from 51.8 ± 12 to 104.2 ± 17, 43.1 ± 4 to 72.2 ± 7, and 574.2 ± 56 to 906.8 ± 66 Bq/kg, respectively, while ^{40}K in the stream water samples ranged from 39.6 ± to 46.3 ± 6. ^{226}Ra and 232Th were not detected in most water samples.

The excess lifetime cancer risk and other radiological hazard indices were also evaluated. Although the data for the measured hazard indices reported in this study were lower than the world limit values, since only a few sites were measured in this cross-sectional study, more measurements are recommended (see Figure 9). These follow-up measurements include additional streams and soil sites to determine, if any, the sources, speciation and enrichment of ^{226}Ra or ^{232}Th due to mining activities or from natural geological formations. The values obtained in this study showed slightly higher activity concentrations, ADR and ELCR in sediment samples and ADR, AED and ELCR in stream samples than the world average values, but the measured average levels were unlikely to portend radiological health risks to people living around the study area. The data produced in this work, if augmented, with other natural radioactivity environmental measurements could be used as baseline radiological data for future studies of the study area.

Competing Interests

The authors declare that there are no competing interests.

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