EISSN: 2707-0425

East African Journal of Science, Technology and Innovation, Vol. 4 (3): June 2023

Modeling Radionuclide Transfer from Pasture to Milk in Kisoro, South-Western Uganda

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Abstract

Determination of radionuclide transfer from animal feeds to animal products and ingestion transfer rates is important in assessing internal radiation risk to humans who consume the animal products. Seventy-nine (79) pasture samples and milk samples were collected from cattle farms in seven sub-counties. Activity concentrations of radionuclides in the samples were determined using gamma spectroscopy. Transfer ratios of radionuclides to from pasture to milk were calculated. Ingestion transfer rates of radionuclide from pasture to cow were also determined. Radionuclide accumulation to the body of the cow was modeled. Contributions to radiological effects due to accumulation of radionuclides in the body were estimated. Transfer ratios of uranium and thorium from pasture to milk varied from 0.07 to 0.17, and 0.05 to 0.17, respectively. The ingestion transfer rates of uranium varied from 3.1×10^{-6} to 7.6×10-6 y l^{-1} while for thorium, the ingestion transfer rates varied from 2.2×10^{-6} to 7.5×10^{-6} y l^{-1}. The contribution to radium equivalent (Bq l^{-1}), annual effective dose equivalent (mSv y l^{-1}), and excess lifetime cancer Risk due to the radionuclide transfer were 0.074, 0.002, and 0.0061 $\times10^{-3}$, respectively. Comparing with the safe values of annual effective dose equivalent in foodstuffs of 0.14 mSv y l^{-1} which translates to excess cancer risk of 0.42×10^{-3}, these contributions to radiological effects only account for about 1.5% of the total safe value of excess lifetime cancer risk, therefore, radionuclide transfer from pasture to milk causes a minimal radiation hazard to the milk consumers in Kisoro District.

Keywords: *Volcanic, Activity Concentration, Radionuclide Transfer, Milk*

Cite as: Habakwiha *et al.*, (2023). Modeling Radionuclide Transfer from Pasture to Milk in Kisoro, South-western Uganda. *East African Journal of Science, Technology and Innovation 4(3).*

Received: 05/04/2023
Accepted: 24/05/2023
Published: 30/06/2023

Introduction

The natural environmental radioactivity of an area depends mainly on local geological and geographical conditions of the area, and it is especially related to the rock types. Uranium (U), thorium (Th) and potassium (K) are the main elements that contribute to the natural terrestrial radioactivity (örgün *et al.,* 2007). Gamma Radiation Measurements and Dose Rates in the Coastal Areas of a Volcanic Island of Aegean Sea, Greece show elevated specific activities of the environmental abiotic materials, and consequently higher population dose rate, in comparison with other Greek regions (Florou and Kritidis, 1992). Surface gamma measurements in felsic volcanic rocks such as rhyolites and dacites, values occasionally exceed the background because these rocks break into pieces by the time of weathering under environmental conditions and spread over the environment (Tolluoglu et al., 2004). High activity concentration levels of pluton and volcanic rocks are consistent with their mineralogical composition especially high orthoclase and accessory mineral. The activity concentrations of naturally occurring radionuclides in the rocks are almost five and three times than that of the worldwide average value, respectively. (Org¨un et al., 2007. Concentrations of radionuclides in food depend on their concentrations in water and soil (and or sediments) (de Lurdes, 2007). Activity concentrations in milk samples from volcanic areas of Kisoro were found to be relatively higher than world average and hence milk consumption rate of 0.105 litres per day was estimated to in order to keep radiological effects low (Habakwiha *et al.,* 2023).

Mufumbiro ranges is a set of eight volcanic mountains consisting of Muhavura and Mgahinga along Uganda and Rwanda border; Sabyinyo along Uganda-Democratic Republic of Congo border; Nyiragongo, Nyamuragira and Mikemo in the Democratic Republic of Congo; and Karisimbi and Bisoke along Rwanda and DRC borders (Jack,1913; Habakwiha *et al.,* 2023). Muhavura, Mgahinga, Sabyinyo, Karisimbi, Bisoke, and Mikemo are dormant volcanoes, while Nyiragongo and Nyamuragira are active. Nyiragongo erupted in 2002 and 2021 and magma spewed in Lake Kivu and cultivation land (Tadesco *et al.,* 2007; Nyiragongo Eruptions, 2021). According to reports from Uganda Cancer institute, cancer cases are on rise both in young and old people. The studies above show that the activity concentrations hence dose rates in areas of volcanic origin are higher than word average values. In this study, radionuclide transfer from pasture to milk was modeled in order to assess the contribution to the radiological effects in particular the excess lifetime cancer risk. This study acts as a basis for assessing radionuclide transfer to other foodstuffs.

Materials and Methods

Research area and materials used to collect and prepare milk samples, and determination of activity concentrations in milk samples are shown in Habakwiha *et al.,* (2023). The following materials for pasture samples were used; (1) Global positioning system (GPS). This was used to locate the coordinates of the spots pasture samples were collected; (2) Air-tight polythene bags. These air-tight bags were used to keep pasture samples and ensure that there was no contamination after sampling from the surroundings; (3) Electric oven was used to dry pasture samples to constant weight to drive away moisture; (4) A grinder was used to grind pasture samples to finest samples and 2 mm sieves was used to sieve the samples for homogeneity. Before the next sample could be put in the grinder and sieve, the grinder and the sieve were washed to avoid contamination from previous sample; (5) 0.5 litre plastic bottles were used to keep milk samples and the bottles were hermetically sealed to avoid contamination; (6) 0.2 mill liters of formalin was added to each milk sample to preserve the samples until experimentation time; and (7) Marinelli beakers are beakers in the in which the samples were put in order to be mounted on the NaI detector for gamma spectrography. Pasture samples were collected from same farms where milk samples in Habakwiha *et al.,* (2023) were collected in order to study the specific transfer of the radionuclides on that very farm. Pasture samples were prepared in the form they were collected from the farms that is to say that they were not washed. The samples were purposely not washed because animals eat the pasture without washing them and we therefore expect elevated activity concentrations in pasture due to contamination from soil particles that stick onto the pasture. We also expect this contamination to have affected the activity concentrations in milk in the study by Habakwiha *et al.,* (2023). Each Pasture sample was oven dried at 90^0 C until constant weight was obtained, powdered and then hermetically sealed. All the samples were left for 30 days in order for the radionuclides to attain secular equilibrium with their daughters (Behinassi, 2014 and Al-Masri, 2000). The samples were prepared and stored in Mutolere SS laboratory for the 30 days and the experimentation was done in Makerere University Department of Physics radioisotope laboratory.

Determination of Activity Concentrations in pasture samples

The samples were put in Marinelli beakers and weighed. The Marinelli beakers were mounted on the NaI detector for gamma spectrography one after the other. The acquisition time of spectrum for each sample was between 7000 and 8000 s and this was initiated using Autodas software until a defined spectra were obtained for easy analysis. Background radiation was recorded for almost the same acquisition time as the samples (Habakwiha *et al.*, 2023). The area under each peak (N) was used to calculate the activity concentrations using Equation 1 for pasture samples (Avwiri, 2012).

$$C = \frac{N}{mTc} \quad \text{Bq kg}^{-1} \qquad (1)$$

where m is the mass of the sample in kg, T is the measuring time and $c = \eta \times k$ correction coefficient for each radionuclide that was used to calculate the specific activity; k is branching ratio of the radionuclides (Roca et al., 2004), while η is the efficiency of the detector that. The average specific activity for the radionuclides of the same series was calculated to get C_{Ra} or C_U and C_{Th}. The average activity concentrations per sub-county were used to determine transfer factors of the radionuclides from pasture to milk, and ingestion pathway transfer rates. Ingestion pathway transfer rates were used for modeling.

Transfer factors and Ingestion Pathway Transfer Rates

Transfer factors of radionuclides from pasture to milk were calculated using Equation 2 (Abdel-Rehman, 2017).

$$k = \frac{Activity\ Concentration\ in\ milk\ sample}{Activity\ concentraton\ in\ pasture\ sample} \qquad (2)$$

The ingestion pathway transfer rates of radionuclides from pasture (fodder) to cow hence milk was calculated using Equation 3 (Balonov et al., 2012).

$$k_{(p-m)}(t) = \frac{C_m}{C_{PO} \times Q_P} \qquad (3)$$

where $k_{(p-m)}$ (t) is the transfer rate of radionuclides from pasture to milk (year per litre), C_m (Bq l^{-1}) is activity concentration of radionuclide in milk, C_{po} is activity concentration of radionuclide in pasture (Bq kg^{-1}), and Q_p is amount of pasture consumed by a cow per year (kg y^{-1}). Activity concentration in Bq of the radionuclides is using Equation 4.

$$C_P = C_{PO} \times Q_P \qquad (4)$$

Modeling radionuclide transfer to milk

Balonov *et al.*, (2012), provides different models for radionuclide exposure and radionuclide transfer for non-human species. The models include the following;

FASTer model that was originally configured to consider a simple food chain consisting of vegetation-herbivore-carnivore in part to provide transfer parameters for organism-radionuclide. The model considers the daily intake of soil which was not considered in this study, daily intake of vegetation which we have considered in this study, fractional gut uptake which we have also not considered, and the live mass of the herbivore which we have also not considered in this study.

RESRAD model which is a computer code that was developed by Argonne National Laboratory for the United State Department of Energy to calculate site-specific residual radioactive material guidelines and radiation dose/risk to an exposed person (worker or resident) at a radionuclide contaminated site through direct exposure to external radiation from contaminated soil and internal exposure due to; inhalation of contaminated dust and radon, ingestion of plant food grown in the contaminated soil and irrigated by water drawn from the ponds, ingestion from meet got from livestock fed with fodder grown from the contaminated soil and water drawn from the ponds, consumption of milk from the livestock, consumption of aquatic foods from the ponds, rivers and lakes, drinking water from the ponds/wells, rivers and lakes. Some of the pathways of radionuclide transfer to human beings in this model were considered in this model but the model could not be used because it best suits artificially contaminated environment but not a natural one like the case is in this study. ECOMODE model that is suitable for radionuclide transfer to fish.

LAKECo-B model that is a dynamic model that describes the activity concentrations of radionuclides in aquatic organisms. The above models mainly address the radionuclide transfer after artificial radionuclide release to terrestrial and aquatic environments either by nuclear accident or mining. This study considers natural environment where radionuclides are transferred to humans and non-human species cumulatively through ingestion and some of the radionuclides leave the body through natural decay and excretion. Hence, using Malthus (1992), the simplest population model of single species and some ideas from Balonov *et al.,* (2012) models, transfer model from pasture to milk hence human beings through consumption of the milk was developed as discussed in the next subsections.

Single species exponential growth model

According to Malthus (1992), the simplest population model of single species is given by;

$$\frac{dx}{dt} = bx - dx = (b-d)x = rx, \tag{5}$$

With solution

$$x(t) = x_o e^{rt} \tag{6}$$

where b and d are per capita birth and death rate, x(t) is the population size of the species at time t, and r = b-d is growth rate. Comparing Model 6 with transfer of radionuclides from pasture to milk, since a cow accumulates radionuclides in its body hence dairy products like milk by consuming the pasture, $r \equiv k_{(p-m)}$ and $x \equiv C_P$. Hence Equation 5 was modified to suit radionuclide transfer through ingestion as shown in Equation 7.

$$\frac{dC_m}{dt} = k_{(p-m)} C_P \tag{7}$$

The solution of Equation 7 is;

$$C_m(t) = C_P e^{k_{(p-m)}t} \tag{8}$$

Substituting Equation 4 in Equation 8, gives Equation 9.

$$C_m(t) = C_{Po} Q_p e^{k_{(p-m)}t} \tag{9}$$

Radionuclides are removed naturally removed from the body of the cow through decay and excretion (Olos, 2022). The rate of removal of the radionuclides from the body was modeled using Equation 10.

$$\frac{dC_m}{dt} = -\lambda C_P e^{k_{(p-m)}t} - C_m k_e \tag{10}$$

where λ is decay constant and k_e is excretion coefficient. The decay constant λ is given by Equation 11.

$$\lambda = \frac{\ln 2}{T_{eff}} \tag{11}$$

where T_{eff} is effective half-life. Effective half-life is related to Physical half-life T_P, and biological half-life T_B, of a radionuclide by Equation 12 (Olos, 2022).

$$T_{eff} = \frac{T_P \times T_B}{T_P + T_B} \tag{12}$$

A radionuclide whose physical half-life is too long compared to its biological half-life; its effective half-life tends to biological half-life. The physical half-life, biological half-life and effective half-life for selected radionuclides are shown in Table 1 (Olos, 2022). ^{232}Th main pathway into a body is through inhalation of suspended soil particles, although some ^{232}Th is ingested in food and water. The biological half-life of ^{232}Th is 8,000 days in bones and 700 days in tissues (Olos, 2022). In this study, the average value of 4,350 days was used for the whole body. The average effective half-lives of radionuclides in the same series were used in modeling. The average effective half-lives of radionuclides in the same series were used in modeling. Table 1 shows the physical, biological and effective half-lives of selected radionuclides.

Tables

Physical, Biological, and Effective half-lives of selected radionuclides in humans

Radionuclide	Physical half life	Biological half -life	Effective half- life
^{210}Po	138 days	60 days	42 days
^{226}Ra	1620 years	45 years	44 years
^{210}Pb	22 years	10 years	6.9 years
^{235}U	700 mega years	15 days	15 days
^{232}Th	1.4×10^{10} years	4,350 days	4,350 days

The solution of Equation 10 is given by Equation 13,

$$C_m(t) = -\frac{\lambda \times C_{po} \times Q_P e^{k_{(p-m)}t}}{k_{(p-m)} + k_e} + \left(C_{mo} \times Q_m + \frac{\lambda \times C_{po} \times Q_P}{k_{(p-m)} + k_e} \right) e^{-k_e t} \qquad (13)$$

where $C_m(t)$ is the radionuclide concentration in milk consumer after time t, and C_{mo} is initial concentration in milk, Q_m is milk consumption rate. The net accumulation model of the radionuclides to a milk consumer is Equation 9 minus Equation 13. According to Yu *et al.,* (2001), the milk consumption rate is 92 litres per year, and livestock daily fodder intake rate in environmental pathway p=4, is 68 kg and for p=5, is 55 kg. According to RPA (2011), the fodder consumption rate is 65 kg per day. In this study, the average value of the consumption rate of 62.7 kg per day was used as fodder intake rate since the environmental pathway index was not identified. This study used the sum of excretion rates through urine and feces from the study by Schafer *et al.,* (2006) titled 'Investigation of excretion rates the radionuclides ^{230}Th, ^{226}Ra, ^{210}Pb, and ^{210}Po of persons of the general population and of workers in selected regions in Germany. Keith et al., (2019), states that the excretion rate of ^{232}Th is 2.5% of the body burden and that the body burden of ^{232}Th is 0.4 Bq d^{-1}, implying that the excretion rate for ^{232}Th is 10 mBq d^{-1}. The sum of the excretion rates through feces and urine used for this study are shown in Table 2. BDL stands for 'Below Detectable Limit'.

Since the values of human gastrointestinal absorption rates f_1 in humans were assumed to be equal to those in animals (Linsalata, 1994), in this study, we have also assumed that the excretion rates for the radionuclides and effective half lives in humans are the same in animals. The average excretion rates and effective half-lives of radionuclides in the same series were used in Modeling.

Table 2

Sum of excretion rates through feces and urine for general public

Radionuclide	Excretion Rates (mBq d^{-1})		
	Urine	Faeces	Sum
^{210}Po	8.1	21	29.1
^{226}Ra	6.3	30	36.3
^{210}Pb	3.5	BDL	3.5
^{235}U	BDL	4.1	4.1
^{232}Th	-	10	10

Contribution to Radiological Effects from Radionuclide Transfer to milk

Contribution to Radiological Effects; Radium Equivalent Activity (Ra_{eq}), Annual Effective Dose Equivalent (AEDE), and Excess Lifetime Cancer Risk (ELCR) as defined in Habakwiha et al., were calculated using accumulated radionuclides in milk using similar Equations. Potassium is very essential in plants and animals for their growths and therefore, causes limited Radiation risk (Ibikunle, 2018) and therefore, its contribution to radiological effects was not added. The accumulated radionuclides in Bq were converted to Bq l^{-1} by dividing the activity in Bq by milk consumption rate of 62 liters per year in order to calculate the contribution to the radiological effects.

Results

Activity Concentrations, Transfer Factors, and Ingestion Transfer Rates

The activity concentrations of ^{226}Ra, and ^{232}Th in milk samples used in this study shown in Habakwiha *et al.,* (2023). Table 3 shows the activity concentrations of ^{226}Ra and ^{232}Th in pasture from different spots in the Sub-counties while Table 4 shows the average activity concentrations of the radionuclides for the whole volcanic area in pasture, and milk. Figure 1 compares the average concentrations in pasture, and milk for the whole volcanic area.

Table 3

Activity Concentrations in Pasture from all Sub-counties

	Activity Bq kg^{-1}		Spot	Activity Bq kg^{-1}		Spot	Activity Bq kg^{-1}	
	^{226}Ra	^{228}Th		^{226}Ra	^{228}Th		^{226}Ra	^{228}Th
		NYAKABANDE	12	9.61 ± 0.85	28.61 ± 2.08	2	2.01 ± 0.01	23.54 ± 1.90

1	9.82 ± 0.72	23.13 ± 0.91	13	9.10 ± 1.00	27.10 ± 1.97	3	8.23 ± 0.60	2.51 ± 0.07
2	10.61 ± 1.06	24.50 ± 1.46			NYARUSIZA	4	7.61 ± 0.61	68.72 ± 4.60
3	9.99 ± 1.16	23.59 ± 1.56	2		24.11 ± 1.46	5	11.32 ± 0.83	3.74 ± 0.09
4	15.92 ± 1.11	24.50 ± 1.16	3	13.18 ± 1.07	23.16 ± 1.73	6	10.50 ± 0.66	24.11 ± 1.90
5	6.76 ± 0.96	10.01 ± 0.67	4	13.62 ± 1.01	23.73 ± 2.01	7	10.26 ± 0.64	25.50 ± 1.91
6	7.45 ± 0.75	24.67 ± 1.56	5	10.89 ± 0.73	25.97 ± 1.73	8	15.74 ± 0.93	91.13 ± 5.17
7	9.17 ± 0.96	21.16 ± 2.56	6	8.22 ± 0.62	18.55 ± 0.59	9	6.62 ± 0.43	14.46 ± 1.10
8	7.84 ± 0.76	5.80 ± 0.86	7	12.16 ± 1.01	21.52 ± 1.17	10	9.18 ± 0.77	10.52 ± 0.98
9	12.65 ± 0.97	18.72 ± 2.36	8	10.21 ± 0.77	24.87 ± 1.46	11	12.58 ± 0.90	26.98 ± 2.11
10	9.85 ± 0.96	21.62 ± 1.46	9	7.35 ± 0.07	17.98 ± 0.94	12	9.64 ± 0.69	28.81 ± 2.21
11	13.31 ± 1.16	20.70 ± 1.11	10	5.48 ± 0.08	13.15 ± 0.81	13	10.49 ± 0.70	47.78 ± 3.61
12	13.43 ± 1.16	22.57 ± 1.76	11	18.41 ± 1.41	29.31 ± 1.24	14	3.59 ± 0.03	10.86 ± 1.92
13	8.45 ± 0.96	21.07 ± 1.56	12	9.95 ± 0.90	22.77 ± 1.31	15	7.29 ± 0.41	41.12 ± 3.01
14	8.77 ± 0.87	20.24 ± 1.87	13	16.14 ± 1.02	22.94 ± 1.40			NYARUBUYE
15	7.73 ± 0.76	11.44 ± 0.52			MURORA	1	ND	3.69 ± 0.15
16	7.28 ± 0.97	17.16 ± 1.51	1	9.69 ± 0.84	23.19 ± 1.81	2	ND	2.85 ± 0.18
		CHAHI AND K M	2	10.96 ± 0.91	25.44 ± 1.90	3	ND	37.19 ± 2.15
1	10.05 ± 0.92	17.36 ± 1.41	3	10.01 ± 0.82	23.14 ± 1.81	4	2.52 ± 0.21	5.48 ± 0.45
2	4.51 ± 0.92	12.61 ± 0.91	4	13.33 ± 0.90	36.32 ± 2.01	5	1.91 ± 0.11	4.87 ± 0.35

3	14.51 ± 1.21	19.97 ± 2.10	5	7.92 ± 0.65	10.01 ± 1.10	NYAKINAMA		
4	13.93 ± 0.86	18.51 ± 1.82	6	6.74 ± 0.55	18.40 ± 1.78	1	8.23 ± 0.61	9.10 ± 0.866
5	17.54 ± 1.62	34.47 ± 3.91	7	13.85 ± 0.81	18.40 ± 1.78	2	2.79 ± 0.06	9.52 ± 0.80
6	15.29 ± 1.12	23.98 ± 1.92	8	2.06 ± 0.14	16.89 ± 1.41	3	9.33 ± 0.56	18.68 ± 1.26
7	19.92 ± 1.65	33.04 ± 2.63	9	3.00 ± 0.09	14.30 ± 1.12	4	8.14 ± 0.45	9.00 ± 0.76
8	4.31 ± 0.90	12.06 ± 0.99	10	6.02 ± 0.54	19.00 ± 1.50	5	2.77 ± 0.07	3.09 ± 0.05
9	13.47 ± 1.16	18.53 ± 1.33	11	13.20 ± 0.71	9.76 ± 0.91	6	10.37 ± 0.86	20.76 ± 1.06
10	13.85 ± 1.23	18.40 ± 1.37	MURAMBA					
11	13.12 ± 1.21	31.68 ± 1.41	1	8.85 ± 0.64	26.38 ± 1.66			

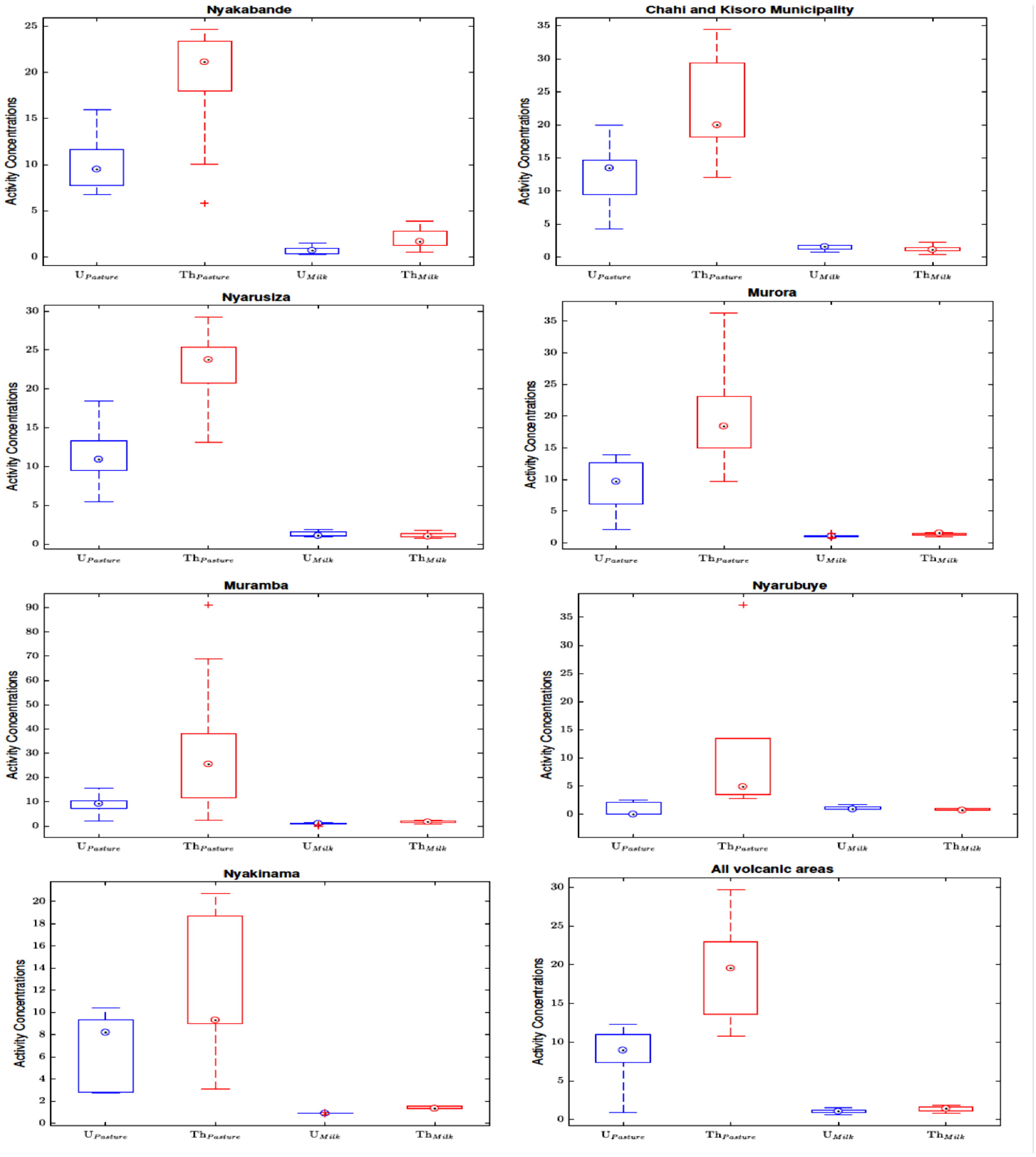
Table 4

Average Activity Concentrations in Pasture and Milk samples from all the Volcanic areas

S/County	Pasture (Bq kg^{-1})		Milk (Bq l^{-1}) (Habakwiha et al., 2023)	
	^{226}Ra	^{228}Th	^{226}Ra	^{228}Th
Nyakabande	9.94 ± 0.96	19.43 ± 1.41	0.66 ± 0.08	1.88 ± 0.41
Chahi+ KM	12.25 ± 1.12	22.86 ± 1.60	1.49 ± 0.25	1.10 ± 0.15
Nyarusiza	11.34 ± 0.84	22.94 ± 1.40	1.24 ± 0.05	1.15 ± 0.03
Murora	8.80 ± 0.63	19.53 ± 1.54	1.06 ± 0.02	1.42 ± 0.02
Muramba	8.93 ± 0.61	29.74 ± 2.15	0.95 ± 0.01	1.74 ± 0.04
Nyarubuye	0.88 ± 0.06	10.81 ± 0.66	1.108 ± 0.01	0.80 ± 0.01
Nyakinama	6.94 ± 0.44	11.69 ± 0.78	0.90 ± 0.01	1.402 ± 0.01
Average	8.44 ± 0.59	19.57 ± 1.36	1.06 ± 0.06	1.37 ± 0.10

Figure 1

Comparison of activity concentrations in pasture and milk in the whole volcanic area.



The activity concentrations of ^{226}Ra, and ^{232}Th in pasture (Bq kg-1), and milk (Bq l-1) were compared as shown in Figure 1 to explain the transfer of the radionuclides from pasture to milk.

In Nyakabande Sub-county, the activity concentration of ^{226}Ra in pasture and milk varied from 6.76±0.96 at spot 5 whose coordinates are 1.251^0 S, 29.7^0 E in Rambura village and 0.27 in sample NDEM1 from Butuga village to 15.92± 1.11 at spot 4 whose coordinates are 1.246^0 S, 29.712^0 E in Gikoro village and 1.48 in sample NDEM6 from Bihuru village with an average of 9.94 ±0.96 and 0.66 respectively while that of ^{232}Th varied from 5.80±0.86 at spot 8 whose coordinates are 1.2390 S, 29.6980 E in Gasaro village and 0.51 in sample NDEM11 from Bugara village to 24.67± 1.56 at spot 6 whose coordinates are 1.257^{0} S, 29.716^{0} E in Bihuru village and 3.19 in sample NDEM6 from Bihuru village with averages of 19.43 ± 1.41 and 3.19, respectively.

In Chahi Sub-county and Kisoro Municipality, the activity concentration of ^{226}Ra in pasture and milk varied from 4.31 ± 0.90 at spot 8 with coordinates 1.316^{0} S, 29.734^{0} E in Rubagabaga and 0.69 in sample CHIM11 from Ndimiro village to 19.92± 1.65 at spot 7 whose coordinates are 1.304^{0} S, 29.724^{0} E in Buhinga and 1.79 in sample CHIM10 from Kabara village with an average of 12.25± 1.12 and 1.49, respectively while that of ^{232}Th varied from 12.06±0.99 at spot 8 whose coordinates are 1.316^{0} S, 29.734^{0} E in Rubagabaga and 0.42 in sample CHIM2 from Nyaruyaga village to 34.47 ± 3.91 at spot 5 whose coordinates are 1.343^{0} S, 29.709^{0} E in Nyamigenda village and 2.3 in sample CHIM13 from Bihanga village with averages of 22.86 ±1.60 and 1.20, respectively.

In Nyarusiza Sub-county, the activity concentration of ^{226}Ra in pasture and milk varied from 5.48 ± 0.08 at spot 10 whose coordinates are 1.341^{0} S, 29.683^{0} E in Bitongo village and 1.00 in sample NZAM13 from Chondo village to 18.41 ± 1.41 at spot 11 whose coordinates are 1.323^{0} S, 29.706^{0} E in Karambi village and 1.88 in sample NZAM1 from Nyamushungwa village with an average of 11.34 ± 0.84 and 1.24, respectively while that of ^{232}Th varied from 13.15 ± 0.81 whose at spot 10 whose coordinates are1.341^{0} S, 29.683^{0} E in Bitongo village and 0.80 in sample NZAM12 from Buhangura village to 29.31 ± 1.24 at spot 11 whose coordinates are 1.323^{0} S, 29.706^{0} E in Karambi village and 1.80 in sample NZAM3 from Nturo village with averages of 22.94±1.40 and 1.15, respectively.

In Murora Sub-county, the activity concentration of ^{226}Ra in pasture and milk varied from 2.06 ± 0.14 at spot 8 whose coordinates are 1.319^{0} S, 29.771^{0} E in Gatete village and 0.80 at in MRAM4 village to 13.85 ± 0.81 at spot 7 whose coordinates are 1.317^{0} S, 29.779^{0} E in Chibumba village and 1.55 in MRAM1 village with an average of 8.80 ± 0.63 and 1.06, respectively while that of ^{232}Th varied from 9.76 ± 0.91 at spot 11 whose coordinates are 1.327^{0} S, 29.753^{0} E in Karuzogero village and 1.05 in sample MRAM1 from Nyabune village to 36.32± 2.01 at spot 4 whose coordinates are 1.341^{0} S, 29.778^{0} E in Nyabitare village and 1.58 in sample MRAM5 from Gisha village with averages of 19.53 ± 1.54 and 1.42, respectively.

In Muramba Sub-county, the activity concentration of ^{226}Ra in pasture and milk varied from 2.01 ± 0.01 at spot 2 whose coordinates are 1.297^{0} S, 29.607^{0} E in Bunagana village and non-detectable value in sample MBAM3 from Gakoro village to 12.58± 0.90 at spot 11 whose coordinates are 1.315^{0} S, 29.674^{0} E in Gasarara village and 1.33 in sample MBAM11 from Gasarara village with an average of 8.93±0.61 and 0.95, respectively while that of ^{232}Th varied from 2.51±0.07 at spot 3 whose coordinates are 1.308^{0} S, 29.612^{0} E in Gakoro village and 0.96 in sample MBAM4 from Ruhango village to 91.13 ± 5.17 at spot 8 whose coordinates are 1.325^{0} S, 29.647^{0} E in Bukazi village and 2.39 in sample MBAM1 from Maziba village with averages of 29.74 ± 2.15 and 1.74, respectively

In Nyarubuye Sub-county, the activity concentration of ^{226}Ra in pasture and milk varied from non-detectable values at spots 1, 2, and 3 whose coordinates are 1.266^{0} S, 29.650^{0} E, 1.270^{0} S, 29.645^{0} E, and 1.263^{0} S, 29.641^{0} E in Gakere, Rutundwe 1, and Rutundwe 2 villages and 0.86 in sample NYEM3 from Rutundwe 2 village to 2.52 ± 0.21 at spot 4 whose coordinates are 1.267^{0} S, 29.626^{0} E in Kinyababa village and 1.72 in sample NYEM2 from Rutundwe 1 village with an average of 0.88 ± 0.06 and 1.11, respectively while that of ^{232}Th varied from 2.85± 0.81 at spot 2 whose coordinates are 1.270^{0} S, 29.645^{0} E in Rutundwe 1 village and 0.69 in samples NYEM2 and NYEM3 from Rutundwe 1 and 2 villages to 37.19 ± 2.15 at spot 3 whose coordinates are 1.263^{0} S, 29.641^{0} E in Rutundwe 2 village and 0.95 in sample NYEM1 from Gakere village with averages of 10.81 ± 0.66 and 0.80, respectively.

In Nyakinama Sub-county, the activity concentration of ^{226}Ra in pasture and milk varied from 2.79±0.06 at spot 2 whose coordinates are 1.244^{0} S, 29.691^{0} E in Bugwene village and 0.80 at in sample NMAM1 from Bugwene village to 10.37 ± 0.86 at spot 6 in Gitebe village and 1.00 in sample NMAM6 from Gitebe village with an average of 6.94±0.44 and 0.90, respectively while that of ^{232}Th varied from 3.09± 0.05 at spot 5 whose coordinates 1.263^{0} S, 29.680^{0} E in Gifunzo village and 1.31 in sample NMAM5 from Gifunzo village to 20.76 ± 1.06 at spot 6 whose coordinates are 1.268^{0} S, 29.655^{0} E in Gitebe village and 1.57 in samples NMAM4 and NMAM6 fom Kanyogo and Gitebe villages with averages of 11.69±0.78 and 1.40, respectively.

The average activity concentration of ^{226}Ra in pasture and milk for the whole volcanic area was

8.43 ± 0.59 and 1.06, respectively while that of 232Th 19.57 ± 1.36 and 1.06, respectively.
The errors in activity concentrations in milk samples were negligible to 2 decimal places are were therefore omitted.

In Nyakabande Sub-county, the transfer factors of the radionuclides from pasture to milk and ingestion transfer rates for ^{226}Ra varied from 0.027 and 1.2×10^{-6} to 0.200 and 8.7×10^{-6} with averages of 0.071 and 3.1×10^{-6}, respectively while those for ^{232}Th varied from 0.036 and 1.1×10^{-6} to 0.479 and 7.6×10^{-6} with averages of 0.120 and 5.2×10^{-6}, respectively.

In Chahi Sub-county and Kisoro Municipality, the transfer factors of the radionuclides from pasture to milk and ingestion transfer rates for ^{226}Ra varied from 0.052 and 1.7×10^{-6} to 0.393 and 8.6×10^{-6} with averages of 0.155 and 4.4×10^{-6}, respectively while those for ^{232}Th varied from 0.027 and 1.2×10^{-7} to 0.116 and 3.6×10^{-6} with averages of 0.057 and 2.5×10^{-6}, respectively.

In Nyarusiza Sub-county, the transfer factors of the radionuclides from pasture to milk and ingestion transfer rates for ^{226}Ra varied from 0.062 and 3.3×10^{-6} to 0.186 and 8.2×10^{-6} with averages of 0.119 and 5.2×10^{-6}, respectively while those for ^{232}Th varied from 0.029 and 1.3×10^{-6} to 0.078 and 3.4×10^{-6} with averages of 0.051 and 2.3×10^{-6}, respectively.

In Murora Sub-county, the transfer factors of the radionuclides from pasture to milk and ingestion transfer rates for ^{226}Ra varied from 0.059 and 2.5×10^{-6} to 0.505 and 2.2×10^{-6} with averages of 0.171 and 7.5×10^{-6}, respectively while those for ^{232}Th varied from 0.037 and 1.6×10^{-6} to 0.158 and 6.9×10^{-6} with averages of 0.085 and 3.7×10^{-6}, respectively.

In Muramba Sub-county, the transfer factors of the radionuclides from pasture to milk and ingestion transfer rates for ^{226}Ra varied from non-detectable values to 0.588 and 1.2×10^{-6} with averages of 0.141 and 6.2×10^{-6}, respectively while those for ^{232}Th varied from 0.014 and 6.1×10^{-7} to 0.791 and 1.2×10^{-5} with averages of 0.141 and 6.2×10^{-6}, respectively.

In Nyarubuye Sub-county, the transfer factors of the radionuclides from pasture to milk and ingestion transfer rates for ^{226}Ra varied from non-detectable values to 0.465 and 2.0×10^{-5} with averages of 0.162 and 7.1×10^{-6}, respectively while those for ^{232}Th varied from 0.019 and 8.1×10^{-7} to 0.258 and 1.3×10^{-5} with averages of 0.168 and 7.3×10^{-5}, respectively.

In Nyakinama Sub-county, the transfer factors of the radionuclides to milk and ingestion transfer rates for ^{226}Ra varied from 0.009 and 4.2×10^{-6} to 0.322 and 1.4×10^{-5} with averages of 0.174 and 7.6×10^{-6}, respectively while those for ^{232}Th varied from 0.070 and 3.1×10^{-7} to 0.423 and 1.9×10^{-5} with averages of 0.171 and 7.5×10^{-6}, respectively.

The average transfer factor and ingestion transfer rate in whole volcanic area for ^{226}Ra were 0.142 and 5.9×10^{-6} while for ^{232}Th, they were 0.113 and 4.9×10^{-6}, respectively. Table 5 shows the average transfer factors and ingestion transfer rates of the radionuclides per Sub- County and for the whole volcanic area.

Table 5

Average Radionuclide Transfer Factors and Ingestion Transfer Rates to Milk in Volcanic areas

S/County	Pasture to milk (kg l^{-1})		Ingestion Transfer Rate (y l^{-1})	
	^{226}Ra	^{228}Th	^{226}Ra	^{228}Th
Nyakabande	0.07	0.12	3.1×10^{-6}	5.2×10^{-6}
Chahi+ KM	0.16	0.06	4.4×10^{-6}	2.5×10^{-6}

Nyarusiza	0.12	0.05	5.2×10^{-6}	2.2×10^{-6}
Murora	0.17	0.09	7.5×10^{-6}	3.7×10^{-6}
Muramba	0.14	0.14	6.2×10^{-6}	6.2×10^{-6}
Nyarubuye	0.16	0.17	7.1×10^{-6}	7.3×10^{-6}
Nyakinama	0.17	0.17	7.6×10^{-6}	7.5×10^{-6}
Average	0.14	0.11	5.9×10^{-6}	4.9×10^{-6}

Using average values of activity concentration and average ingestion transfer factors for the whole volcanic area in Tables 4 and 5 respectively, and Equation 13, the corresponding net accumulation transfer models for ^{238}U, and ^{232}Th daughters are given by Equations 14 and 15 respectively.

$$C_U(t) = 198710.5e^{6.2\times10^{-6}t} - 5590.6e^{-0.041t} \quad (14)$$

$$C_{Th}(t) = 3047908.2e^{4.9\times10^{-6}t} - 2599866.4e^{-0.01t} \quad (15)$$

Potassium is very essential in plants and animals for their growths and therefore, causes limited Radiation risk (Ibikunle, 2018) and therefore, its net accumulation was not modeled. Model Equations 14 and 15 are shown in Figures 2 and 3, respectively.

Figure 2

Average transfer models of uranium from pasture to milk for the whole volcanic area.

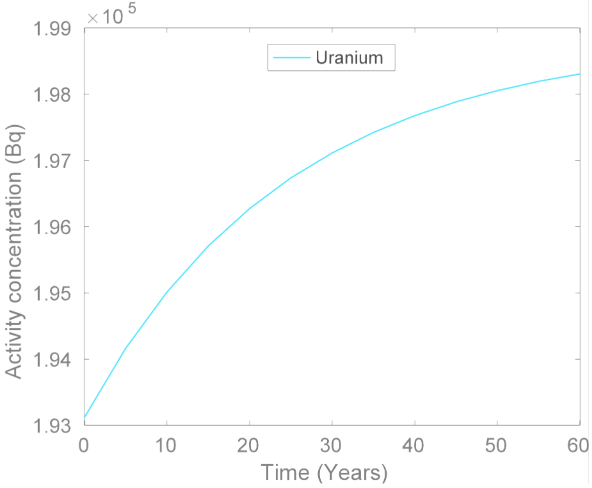
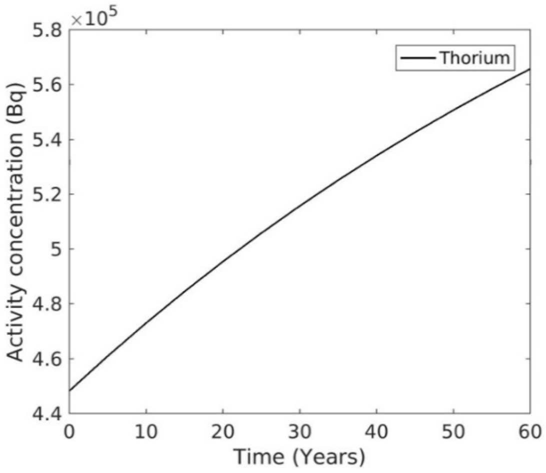


Figure 3

Average transfer models of thorium from pasture to milk for the whole volcanic area



From Figures 2, and 3 the net accumulation of the radionuclides ^{238}U, and ^{232}Th in milk in 60 years are 5100 Bq and 11700 Bq, respectively. The daily average net accumulation activity concentrations that contribute to radiological effects are 0.0025 Bq l^{-1} and 0.05 Bq l^{-1} for ^{238}U, and ^{232}Th, respectively. The corresponding Raeq (Bq l^{-1}), AEDE (mSv y^{-1}), and ELCR) are 0.074, 0.002, and 6.1×10^{-6}, respectively.

Discussion

The average activity concentrations of ^{232}Th were higher than that of ^{238}U for all spots. The average activity concentrations of ^{232}Th were highest in Muramba and least in Nyarubuye. The descending order of average activity concentrations was; Muramba (29.74±1.40 Bq kg^{-1}), Nyarusiza (22.94± 2.15 Bq kg^{-1}), Chahi and Kisoro Municipality (22.86±1.60 Bq kg^{-1}), Murora (19.53 ± 1.54 Bq kg^{-1}), Nyakabande (19.43 ± 1.41 Bq kg^{-1}), Nyakinama (11.69 ± 0.78 Bq kg^{-1}), and Nyarubuye (10.81 ± 0.66 Bq kg^{-1}). The trend of the average concentration of ^{238}U was almost the same in the descending order of Chahi and Kisoro Municipality (12.25±1.12 Bq kg^{-1}), Nyarusiza (11.34 ± 0.84 Bq kg^{-1}), Nyakabande (9.93 ± 0.96 Bq kg^{-1}), Muramba (8.93 ± 0.61 Bq kg−1), Murora (8.80 ± 0.63 Bq kg^{-1}), Nyakinama (6.94 ± 0.44 Bq kg^{-1}), and Nyarubuye (0.88 ± 0.06 Bq kg^{-1}).

The Sub-counties of Muramba, Chahi and Nyarusiza border the volcanic mountains of Muhabura, Mgahinga, and Sabyinyo while Murora, Kisoro Municipality and Nyakabande border the Sub-counties of Muramba, Nyarusiza, and Chahi. This could be the reason why the radionuclide concentrations are higher in these Sub-counties. Nyarubuye and Nyakinama are surrounded by block hills and therefore the volcanic soils in the Sub-counties have some clay and loam soils and this is probably why the activity concentrations are relatively lower. It was also noticed that activity concentrations in samples collected from relatively flat areas and lower altitudes were relatively higher than those from sloping ones probably due to erosion. Since the average activity concentrations in milk are almost constant in all Sub-counties ranging from 0.66 to 1.88 Bq l^{-1}, the transfer ratios of the radionuclides from pasture to milk are higher in Nyarubuye and Nyakinama. The Ingestion transfer rates of pasture to cow is almost constant to the order of 10^{-6} for all the Sub-counties.

According to Ganz (2011) and FAO and WHO (2016), the weighted Annual Effective dose from radionuclides of uranium and thorium series is 0.14 mSv y^{-1} which translates to 0.42×10^{-3} excess lifetime cancer risk for the whole diet. Comparing the contribution of milk consumption to the lifetime excess cancer risk of 0.0061×10^{-3} with the safe value of 0.42×10^{-3} for the whole diet, milk contributes about 1.5% which is very small.

Conclusion

The activity concentrations of the radionuclides were found out to be relatively distributed with some exceptions of some spots where uranium was below detectable limits in Nyarubuye. This may be due to soils in Nyarubuye that are a mixture of volcanic and non-volcanic due soil erosion from surrounding non volcanic hills. Spots from relatively flat have relatively higher radionuclide concentrations than relatively sloping ones. This is an agreement with the findings by Kapanadze (2019). Transfer ratios of uranium and thorium from pasture to milk varied from 0.07 to 0.17, and 0.05 to 0.17, respectively. The ingestion transfer rates of uranium varied from 3.1×10^{-6} to 7.6×10^{-6} y l^{-1} while for thorium, the ingestion transfer rates varied from 2.2×10^{-6} to 7.5×10^{-6} y l^{-1}. The transfer factors and ingestion transfer rates were uniformly distributed like the radionuclide concentrations. Contribution to radium equivalent (Bq l^{-1}), annual effective dose equivalent (mSv y^{-1})), and excess lifetime cancer Risk) due to the radionuclide transfer were 0.074, 0.002, and 0.0061×10^{-3}, respectively. These contributions to radiological effects are low since the milk alone accounts for about 1.5% of the total safe value of excess lifetime cancer risk; therefore, radionuclide transfer from pasture to milk causes a minimal radiation hazard.

Acknowledgement

We acknowledge the Radioisotope Laboratory of Makerere University for analyzing our samples. We highly thank the Physics laboratory of St. Paul's Mutolere Secondary School where the samples were prepared and stored.

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