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Determination of natural radioactivity and hazards in volcanic soils of Kisoro district in south-western Uganda

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ABSTRACT

In this study, the activity concentrations of natural radionuclides U-238, Th-232, and K-40 were measured in soil samples collected from various spots where there are volcanic soils, in Kisoro district, in South Western Uganda. Then to assess the radiological hazard of the natural radioactivity, the absorbed dose rate (D_r), the radium equivalent activity (Ra_{eq}), the effective dose rate (E_r), the annual effective dose equivalent (E_y), Excess Lifetime Cancer Risk (*ELCR*), and the external (H_{ex}) and internal (H_{in}) hazard indices were calculated. NaI (*Tl*) was used to determine the activity concentration of these radionuclides in soils where NPK was applied and where it was not applied. The results were compared with standard values and it was concluded that no risk may be eminent to the residents in Kisoro district with an exception of areas near quarrying places and mines. This is because these places have high activity. The use of NPK only depletes potassium from the soil where it is applied. Hence, the probability of occurrence of any of the health effects of radiation is low. The measurements have been taken as representing baseline values of these radionuclides in the volcanic soils from the area under study.

Keywords: *Radionuclides, Volcanic Soils, Hazard indices*

Introduction

Natural radionuclides have existed on planet Earth for over 4.5 billion years. These natural radionuclides include; the radionuclides produced by cosmic-radiation interactions in space, their radioactive decay products, and radioactive elements in the earth's crust (Emelue and Eke 2014). Thus, they are distributed throughout the biosphere and accumulate in the atmosphere, soil, bottom sediments, water, and plants. Exposure to natural radiation energy has been identified as a threat to humanity. Geologically, uranium U, thorium, Th, radium, Ra, and radon, Rn occurs in very many rock types. Both thorium and uranium are concentrated in highly frachemated magmas, acidic igneous rocks like granite and hydrothermal solutions Killen and Heier (1975), while Radium and Radon as members of U-228 (Th-232) decay series are also found in rocks with high uranium content and in ground water of a fractured aquifer respectively (Asikainen and Kahlos 1979).

The exposure to these naturally existing radionuclides is responsible for 10% to 20% of lung cancer cases occurring annually in Norway, 400 to 1,100 deaths in Sweden, and 5,000 to 20,000 deaths in the U.S each year (Statens 1994). Mose *et al.* (1990) showed that the ingestion and inhalation of

radionuclides is more harmful than the external exposure to the naturally occurring radionuclides. The ingestion occurs through the food we eat and water we drink and results into stomach cancer while lung cancer results from the inhalation of radon. In view of increasing crop productivity, some farmers use inorganic fertilizers. These inorganic fertilizers affect both human and ecosystem health (Sunanda and Yajneshwar 2007). Land poisoning has occurs where inorganic fertilizers are being used (Sunanda and Yajneshwar 2007). This is caused by the run off of nitrogen and phosphorous into water, thus affecting fish and other aquatic life. Being soluble in water, fertilizers contaminate ground water with carcinogenic nitrates (Black, 1957).

The components of inorganic fertilizers applied to the volcanic soils in Kisoro District are radioactive and could have the potential of causing radiation related problems due to their ingestion and inhalation. The commonly used fertilizers include NPK (nitrogen, phosphorus and potassium), DAP (double ammonium Phosphate), urea, Rapid grows and Super grow, all of which have radioactive components (Baxter 1993). Kisoro District is the leading producer of Irish potatoes in Uganda, with some exports to neighboring Rwanda, DR Congo, and Burundi. Despite the high produce levels, information about the hazards of naturally existing radionuclides for the area is scanty. This study therefore, aimed at determining the activity levels of U-238, Th-232, and K-40 in soil and Irish potato samples.

Methods

Sites and Sample selection

The study sites were surveyed to establish the cropping patterns. The research sites were purposively divided into five (5) fields, which correspond to the sub counties with volcanic soils. The fields were divided into spots depending on the cropping pattern and where Irish potatoes were grown with or without NPK (inorganic fertilizer composed of Nitrogen, Phosphorous and Potassium). Seven spots were identified in Nyakabande sub-county compared to five spots in other sub counties because it is the largest sub-county. The choice of the number of spots in each sub-county also depended on the relative spacing between them. The inter spot spacing was estimated by considering the villages with the assumption that they were of the same size whereby no spots were taken from consecutive villages. The coordinates at each spot were recorded using Global positioning system (GPS). The coordinates and height above sea level of each spot were recorded. Soil and Irish potatoes were sampled from selected sub-counties where inorganic fertilizers were used and where they were not used. The coordinates of all spots where the samples were collected are presented in Tables 1 to 5.

Table 1: GPS coordinates of spots where soil samples were collected in Nyakabande sub-county. La=Latitude, Lo=Longitude and Al=Altitude

Spot	Village	La (°)	Lo(°)	Al (m)
S1	Muca	-1°13'28"	29°42'55"	1796
S2	Gikoro	-1°14^' 4.8"	29°43^' 7.9"	1826
S3	Butuga	-1°16^' 4.8"	29°42'19"	1787
S4	Mugombero	-1°15^' 7.2"	29°42'57"	1826
S5	Chuho	-1°15'56"	29°42'37"	1842
S6	Bugara	-1°15^' 8.2"	29°43'55"	1829
S7	Kanyabukungu	-1°15'58"	29°44'21"	1908

Table 2: GPS coordinates of spots where soil samples were collected in Chahi/Town Council. La=Latitude, Lo=Longitude and Al=Altitude.

Spot	Village	La (°)	Lo(°)	Al (m)
S1	Zindiro	-1°16'32"	29°42'12"	1894
S2	Kibaya	-1°16'58"	29°42'49"	1883
S3	Mudege	-1°16'46"	29°40'48"	1906
S4	Bihanga	-1°19^' 3.3"	29°44'44"	1933
S5	Chanaika	-1°20'21"	29°43'54"	2010

Table 3: GPS coordinates of spots where soil samples were collected in Nyarusiza sub-county. La=Latitude, Lo=Longitude and Al=Altitude.

Spot	Village	La (°)	Lo(°)	Al (m)
S1	Nyamushungwe	-1°18^' 8.0"	29°40'40"	1972
S2	Kararmbi	-1°19^' 40"	29°42'35"	2066
S3	Buhangura	-1°18^' 53"	29°40'54"	2008
S4	Condo	-1°19^' 48"	29°39'38"	2089
S5	Rugina	-1°20^' 27"	29°40'43"	2173

Table 4: GPS coordinates of spots where soil samples were collected in Muramba sub-county. La=Latitude, Lo=Longitude and Al=Altitude.

Spot	Village	La (°)	Lo(°)	Al (m)
S1	Maziba	-1°17^' 29"	29°37'26"	1865
S2	Kanyambiriko	-1°19^' 52"	29°36'18"	1926
S3	Gasuri	-1°19^' 52"	29°38'59"	2123
S4	Murinzi	-1°19^' 7.4"	29°39'36"	2076
S5	Kanyenka	-1°18^' 38"	29°37'55"	2027

Table 5: GPS coordinates of spots where soil samples were collected in Murora sub-county. La=Latitude, Lo=Longitude and Al=Altitude.

Spot	Village	La (°)	Lo(°)	Al (m)
S1	Gatete	-1°18^' 38"	29°23'15"	1900
S2	Gisha	-1°20^' 2.5"	29°46'39"	1889
S3	Nyabune	-1°20^' 7.6"	29°45'47"	1901
S4	Mupaka	-1°21^' 0.5"	29°46'12"	1906
S5	Karuzogero	-1°19^' 59"	29°45'15"	1921

One (1) kilogram of soil was sampled at each spot because after drying, and grinding, a sufficient amount of soil was left for analysis. Before sampling, the top soil was cleared in order to eliminate the deposition of atmospheric radionuclides from cosmic rays. A hole of 20cm by 20cm was dug using a pick axe. Soil samples were then collected at depths of 10cm, and 25cm. The 25 cm depth were chosen because most food crops root up to this depth (Carter 1993). Two depths were considered in order to study the variation of radionuclide concentration with depth.

Soil samples from each spot were labeled as fndpsm , where f, d, and s represented field, depth, and spot respectively; whereas n, p, and m are numbers ranging from 1 to 5. The samples from each spot at each

depth were pounded using mortar and pestle. The mortar and pestle were washed and left to dry in sunshine to reduce contamination before it could be used for another sample. The samples were sieved using 2 mm sieve to make them homogeneous and were dried using an oven set at 4500C to drive out any moisture. The samples were then left to cool in the oven. They were put on a clean cloth for re-sampling until about 0.25kg was left and sufficient to cover the detector crystal. They were then packed in Ziploc bags. All the prepared samples were packed in a wooden box and left for 30 days so as to reach secular equilibrium of thorium and its daughter nuclides.

The samples were placed in Marinelli beakers and were weighed. The mass of samples were calculated by subtracting the mass of empty beaker. The Marinelli beakers were then mounted on the NaI detector mounted in a lead shield. The program was started using "sta" command without any sample in order to determine the noise from the background. The data acquisition from the background was counted for 5000s and the spectrum was saved under the file name bk0612 using "sav" command. To prepare for the next data collection, the "zer" command was used. The samples were then loaded on the detector one after the other with an integration time ranging from 5000s to 5022s. Before any spectrum could be analyzed, the background count (noise) was subtracted to correct for the contribution of the noise from the background. This was done using "rsp" bk0612 command.

Using "τ" and "u" commands, and arrow keys, the markers were placed on the left and right of the peak. The command "cen" was used to determine the centroid position of the peak which was used to identify the energy of gamma radiation in that peak. Such a peak corresponds to a particular radionuclide present in the sample. The area under the peak, the standard deviation, and the count rate, P, which is the number of counts per second.

Activity Calculation

The specific activity (Bq/kg) was calculated using Equation (1) (Avwiri *et al*. 2012).

$$A = \frac{N}{mTC}, \qquad (1)$$

where N is the net area under the peak, m is the mass of the sample in kg, T is the lifetime and C is correction coefficient.

$$C = \eta k, \qquad (2)$$

where η is the branching ratio in which a gamma ray is emitted and k is the efficiency of the detector. The efficiency, k of the detector is given by Equation (3) (Avwiri *et al*. 2012).

$$k = \frac{P}{A}, \qquad (3)$$

where P is the count rate and A is the activity of each sample in each peak. The branching ratio of each radionuclide was obtained from the reactivity series. For example for K-40, η = 11% = 0.11 and k corresponding to 1.46 MeV is 2.13 % and C=0.11×0.0213=0.00234. The correction coefficients for other radionuclides are shown in Table 6.

Table 6: Correction coefficients for some radionuclides.

Energy (keV)	Radionuclide	Series	k	Correction Coefficient for NaI (TI) detector
186.2	Ra-226	U-236	9.00	0.00430
238.6	Pb-212	Th-232	8.37	0.06080
295.2	Pb-214	U-238	7.66	0.02370
351.9	Pb-214	U-238	6.99	0.03000
583.2	TI-208	Th-232	4.67	0.01010
609.3	Bi-214	U-238	7.77	0.02100
1173.2	60Co-60	None	3.22	0.02000
1460.8	K-40	None	2.04	0.00234

Hazards

The radium Equivalent (Ra_{eq}) which is an index that represents a weighted sum of activities of Ra-226, Th-232, and K-40 and is based on the estimation that 1 Bqkg^{-1} of Ra, 0.7 Bqkg^{-1} of Th-232 and 13 Bqkg^{-1} of K-40 produce the same gamma radiation dose rate (Emelue and Eke 2014). According to Avwiri *et al.* (2012); Shoeib and Thabayneh (2014), the index is given by

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_K, \qquad (4)$$

where C_{Ra}, C_{Th}, ^C_K are the average activity concentrations of Ra-226, Th-232 and K-40 respectively. This index was measured to determine the total gamma radiation dose rate a person receives from all the radionuclides in the environment.

The Absorbed dose rate (D_r) in air at an average height of 1m above the surface of the ground due to the radionuclides Ra-226, Th-232, and K-40 was calculated using the formula according to Shoeib and Thabayneh (2014).

$$D_r = DCR_{Ra} \times C_{Ra} + DCR_{Th} \times C_{Th} + DCR_K \times C_K, \qquad (5)$$

where DCR_{Ra}=*0.427* nSv/hr/Bqkg^{-1}, DCR_{Th} = 0.662 nS/hr/Bqkg^{-1}and DCR_K=0.043 nSv/hr/Bqkg^{-1} are dose conversion factors for the radionuclides Ra-226, Th-232 and K-40 respectively.

The effective dose rate in μSv/yr was calculated using Equation (6) Shoeib and Thabayneh (2014).

$$E_\gamma = D_r \times 0.2 \times 8760 \times 0.7 \times 10^{-3}, \qquad (6)$$

This equation takes into account that people spent 20% of their time outdoors and 80% indoors and 8760 × 0.7SvGy^{-1} is the conversion factor Avwiri *et al.* (2012).

The external hazard index, H_{ex} is used as a measure of the radiation exposure due to natural radioactivity in environmental materials. The value of this index must be less than one (unity) in order to keep the radiation dose to the admissible dose equivalent of 1.5 mSv per year (Shoeib and Thabayneh 2014). The value of the external hazard index, Hex was obtained using Equation (7).

$$H_{ex} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810}, \qquad (7)$$

The internal hazard index is used to measure the radiation exposure due to radon and its progeny in building materials. This is because, exposure to indoor radon increases internal exposure as it is inhaled. As it decays, the alpha particles it emits may inflame the internal body organs and the non-gaseous decay products may get trapped in the respiratory tract. These may cause cancer. The value of the internal hazard index was determined using Equation (8).

$$H = \frac{C_{Ra}}{185} + \frac{C_{Th}}{259} + \frac{C_K}{4810}, \quad (8)$$

The excess lifetime cancer risk is a measure of the probability of developing cancer over a lifetime at a given exposure to ionizing radiation. It represents the number of extra cancer cases expected in a given population of people when exposed to ionizing radiation. The Excess Lifetime cancer Risk, (ELCR) was estimated using the Equation (9) Avwiri *et al.* (2012).

$$ELCR = AED \times Averagelifetime \times Riskfactor \quad (9)$$

where AED is the annual effective dose rate in mSvy-1. For a Ugandan, the average life time is 50 years. For stochastic effects, the Risk factor per sievert is 0.05 (Avwiri *et al.* 2012). The world average of the excess lifetime cancer risk at 50 years is 0.290×10^{-3} (Emelue and Eke 2014).

Results and Discussion

Activity

The specific activity at depth of 10 cm and 25 cm were calculated and were found to be random due to mixing of soils during cultivation, otherwise the specific activity increases with depth (Carter 1993; Shashikumar et al. 2011). Due to the random variation, the average specific activity at both depths at each spot was calculated and the values are presented in Tables 7 to 11. The primed spots indicate where NPK was not applied and unprimed ones indicate where NPK was not applied.

Table 7: Average Activity $Bqkg^{-1}$ in soil samples at different spots from Nyakabande sub-county where NPK was and was not applied.

Spot	U	Th	K
S1	74.541	366.648	109.038
S2	60.446	259.549	203.794
S3	41.368	86.152	211.903
S4	55.427	218.138	106.351
S5	37.846	139.273	201.561
S6	54.308	174.057	185.143
S7	35.487	114.267	64.959

The average specific activity in $Bqkg^{-1}$ of uranium, thorium, and potassium where NPK was not applied were 57.946, 232.622, and 157.824 respectively while where it was applied were 42.673, 142.532, and 150.554 respectively. The concentration of thorium is highest at spot 1 (Muca) followed by spot 2 (Gikoro) because these spots are closer to the sand quarries at Rwingwe Hill (within a radius of about 1 km). According to Cember (2009), the abundance of thorium is highest in sandy soils and is generally more abundant than uranium. Thorium is widely distributed with rare localized deposits everywhere and

Uranium, thorium, and potassium concentrations in the soil samples were high at Mugombero, Gikoro, and Chuho spots that are closer to the wolfram mine at Mutolere (within a radius of about I km). Aguko *et al*. (2013) found that the concentrations of these radionuclides were higher than the world values (52Bqkg^{-1}, 110 Bqkg-1, and 440 Bqkg^{-1} respectively) around gold mine in Sakwa Wagusu in Kenya. This shows that there is some presence of these radionuclides around and near the mines.

Table 8: Average Activity in soil samples at different spots from Chahi/Town council where NPK was and was not applied.

Spot	U	Th	K
S1	55.684	156.388	163.978
S2	69.023	212.804	320.682
S3	49.197	163.569	233.979
S4	43.767	172.735	185.721
S5	59.498	233.439	252.404

The average value of the specific activity in Bqkg-1 of uranium, thorium, and potassium where NPK was not applied were 57.968, 177.587, and 239.546 respectively while where it was applied were 51.633, 203.087, and 219.063 respectively.

Table 9: Average Activity in soil samples at different spots from Nyarusiza where NPK was and was not applied

Spot	U	Th	K
S1	55.292	201.367	250.239
S2	41.242	207.318	202.569
S3	53.282	200.569	180.055
S4	54.163	206.216	220.56
S5	44.717	153.919	72.851

The average value of the specific activity in Bqkg^{-1} of uranium, thorium, and potassium where NPK was not applied were 48.938, 203.085, and 210.954 respectively while where it was applied were 49.440, 180.068, and 146.706 respectively.

Table 10: Average Activity in soil samples at different spots from Muramba where NPK was and was not applied.

Spot	U	Th	K
S1	69.605	238.463	143.251
S2	49.644	182.747	107.193
S3	48.622	188.378	153.955
S4	87.926	404.75	201.089
S5	41.411	159.287	148.555

The average value of the specific activity in Bq/kg of uranium, thorium, and potassium where NPK was not applied were 64.669, 282.019, and 174.822 respectively while where it was applied were 55.957, 203.196, and 134.780 respectively.

Table 11: Average Activity in soil samples at different spots from Murora where NPK was and was not applied.

Spot	U	Th	K
S1	63.805	229.721	353.642
S2	61.079	215.972	262.504
S3	63.683	240.887	200.752
S4	68.783	246.832	316.027
S5	43.205	166.472	149.027

The average value of the specific activity in Bq/kg of uranium, thorium, and potassium where NPK was not applied were 58.597, 241.341, and 272.899 respectively while where it was applied were 62.381, 228.430, and 231.628 respectively. The concentrations of all the radionuclides at spot 5 were comparatively low and the soils at the spot are not purely volcanic, there were some traces of papyrus peat soil that are not productive and swamps are reported to be good decontaminants (Kisoro-District Environment-Action-Plan 2008-2011).

Hazard

The radiological effects at all other spots were calculated and are conveniently summarized in the Tables (12) to (16).

Table 12: Radiological effects at each spot in Nyakabande sub-county.

Spot	Ra_{eq} (Bq/kg)	D_r (nSv/hr)	E_r (nSv/hr)	H_{ex}	$H_{\square}$	ELCR $\times 10^{-3}$
S1	607.243	274.593	336.760	1.63	1.84	0.84
S2	447.293	197.674	242.428	1.20	1.37	0.60
S3	180.881	74.739	91.660	0.48	0.60	0.23
S4	375.553	168.117	206.179	1.01	1.16	0.51
S5	252.526	108.402	132.944	0.68	0.78	0.33
S6	317.461	138.456	169.803	0.85	1.00	0.42
S7	203.890	90.840	111.407	0.55	0.64	0.27
Ave	340.692	150.800	184.941	0.94	1.05	0.46

Table 13: Radiological effects at each spot in Chahi/Town Council.

Spot	Ra_{eq} (Bq/kg)	D_r (nSv/hr)	E_r (nSv/hr)	H_{ex}	$H_{\square}$	ELCR $\times 10^{-3}$
S1	291.945	134.357	164.775	0.79	0.94	0.42
S2	398.025	184.138	225.827	1.07	1.26	0.56
S3	301.117	139.351	170.900	0.81	0.95	0.43
S4	305.079	141.025	172.953	0.82	0.94	0.43
S5	412.751	190.796	233.992	1.11	1.28	0.59
Ave	341.781	157.933	193.690	0.92	1.07	0.49

Table 14: Radiological effects at each spot in Nyarusiza sub-county.

Spot	Ra_{eq} (Bq/kg)	D_r (nSv/hr)	E_r (nSv/hr)	H_{ex}	$H_{\square}$	ELCR $\times 10^{-3}$
S1	359.515	166.394	204.065	0.97	1.11	0.51
S2	353.305	163.565	200.597	0.95	1.07	0.50
S3	353.960	163.270	200.235	0.96	1.10	0.50
S4	366.035	169.127	207.417	0.99	1.13	0.52
S5	270.431	124.121	152.222	0.73	0.85	0.38
Ave	340.985	157.295	192.907	0.92	1.05	0.49

The average radium equivalent value for all the spots was calculated and was found to be 353.145Bqkg^{-1}. According to UNSEAR (2000) report, the radium equivalent for the gamma radiation from various sources in an environment to pose limited risk should not be more than370 Bq/kg. From the results in this study, the average value lies below the threshold value and therefore the Ra_{eq} in the study area lies below the threshold value. The average value of the annual effective dose for all the spots was calculated and found to be0.195 mSv/y.

Table 15: Radiological effects at each spot in Muramba sub-county.

Spot	Ra_{eq} (Bq/kg)	D_r (nSv/hr)	E_r (nSv/hr)	H_{ex}	$H_{\square}$	ELCR $\times 10^{-3}$
S1	421.637	193.744	237.607	1.14	1.33	0.59
S2	319.226	146.786	180.018	0.86	1.00	0.45
S3	329.857	152.088	186.521	0.89	1.02	0.46
S4	682.202	314.136	385.256	1.84	2.08	0.96
S5	280.630	129.518	158.841	0.76	0.87	0.40
Ave	406.711	187.254	227.659	1.10	1.26	0.57

Table 16: Radiological effects at each spot in Murora sub-county.

Spot	Ra_{eq} (Bq/kg)	D_r (nSv/hr)	E_r (nSv/hr)	H_{ex}	$H_{\square}$	ELCR $\times 10^{-3}$
S1	419.536	194.527	238.567	1.13	1.31	0.59
S2	390.132	180.342	221.171	1.05	1.22	0.55
S3	423.609	195.292	239.506	1.14	1.32	0.60
S4	446.087	206.362	253.083	1.20	1.39	0.64
S5	292.735	135.061	165.639	0.79	0.91	0.41
Ave	394.420	182.317	223.593	1.07	1.23	0.56

According to IAEA (1994), the world range for annual effective dose rate ranges from 0.3-0.6 mSv/y and the maximum for safety is1 mSv/y. From the results of this study, the annual effective dose lies below the threshold limit. The average values of the external and internal hazard indices for all spots were calculated and were found to be 0.955 and 1.091 respectively.

According to UNSEAR (2000), the hazard indices, H_{ex} and $H_{\square}$ must be less than unity to minimize the radiological effects due to the exposure to the ionizing radiation. From the results of this study, the

average for H_{ex} is less than 1, while $H_{\square}$ is slightly above 1. Therefore, the external hazard index lies below the threshold value while the internal hazard index is slightly above the threshold value. This is mainly due to sand deposits in Nyakabande and the Kisoro Town council and the wolfram mine at Mutolere in Nyakabande sub-county where the concentrations of thorium were high. The internal hazard index is used to measure the radiation exposure due to radon gas and its progeny in building material (UNSEAR 2000).

Therefore, the sand from these quarries exposes people to slightly more dose than threshold value, though the risk of developing cancer at average age of 50 years of people from Kisoro Kisoro-District-Environment-Action-Plan (2008-2011), is still low. The average value of Excess Life Cancer Risk (ALCR) for all other spots were calculated and found to be0.497×10^{-3}. According to Ramasamy *et al.* (2009), the safe range for ALCR is (0.5-0.95) 10^{-3} for a life time of 50 years. This implies that the cancer risk due to radiation exposure for an average 50 yrs life time of a Ugandan is low in the area studied.

Conclusion

The activity concentrations of natural radionuclides and there potential radiological hazards has been determined for volcanic soils in Kisoro district. It was found out that for all spots, the average concentration of potassium was less in soil samples collected from where NPK was applied than where it was not applied. Thus, the radiological hazard of the natural radioactivity in the studied area is generally low. Consequently, no risk may be eminent to the residents in Kisoro district with an exception of areas near quarrying places and mines.

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