Assessment of Radioactivity Levels and Risks Due To Different Rock Types from the KERIO Valley "High Background Radiation Area (HBRA)" of Kenya

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Abstract: The goal of this study was to analyse the natural radioactivity levels and assess the contribution of different rock types to environmental dose in the Kerio Valley region of Kenya, a suspect high background radiation area (HBRA). The activity concentrations of 238 U, 232 Th and 40 K in granite, sandstone, tuff, limestone, quartzite, gneiss, marble and conglomerate rocks were found to vary from 72.70±8.47 Bqkg⁻¹-116.15±11.46 Bqkg⁻¹, 40.32±14.48 Bqkg⁻¹- 83.65± 9.91 Bqkg⁻¹ and 427.41±48.23 Bqkg⁻¹-1397.24±65.27 Bqkg⁻¹ respectively. Radium equivalent activity and external hazard index show that all the rock types do not exceed the recommended limit of 370 Bqkg⁻¹ and 1, respectively. Calculated outdoor absorbed dose rate ranged from 90.585 nGyh⁻¹ (limestone) to 159.085 nG/h (quartzite) while the indoor absorbed dose rate ranged from 66.696 nGyh⁻¹(limestone) to 157.442 nGyh⁻¹ (quartzite), which are above the world average value of 60nGy⁻¹. The annual effective dose rate analysis is also presented.

Keywords: Natural radioactivity; rock types; gamma spectrometry; radium equivalent activity; gamma index; dose rates.

1. INTRODUCTION

Natural radionuclides are the largest contributor to external radiation sources of the world population ⁽¹⁾. Radon is the main contributor of environmental radiation ⁽²⁾ hence the leading in human exposure. The radionuclides can spread to the environment through building materials sourced from areas with elevated levels of radioactivity or even infiltrate water sources such as rivers and streams that pass through such areas. In all the cases the main concern is how much is the radiation exposure.

The radioactivity level in any place depends on the soil type and mineral uptake ⁽³⁾. High concentration of ²³⁸U, ²³²Th and ⁴⁰K are associated with soils that developed from acidic magmatic rocks (igneous rocks) and clay. Therefore, natural environmental radioactivity depends on the geology of the place ^(4, 5). Rocks such as granite, tuffs, gneisses, phonolites and limestone are used as building materials ⁽⁶⁾. Given that these rocks are common in the Kerio Valley and are practically used for building and construction, it is important to measure the concentration of radionuclides in these rocks that are used and those that have a potential of being used as building materials to assess the possible radiological hazards to human health and to develop standards and guidelines for the use and management of these materials ^(7, 8).

Previous study carried out in Kerio Valley ⁽⁹⁾ show that there is uranium in various ore bodies of fluorite samples with concentrations ranging from 34ppm to 983 ppm while thorium which occurred in several samples in varying concentrations ranged from 23 ppm to 166 ppm. Therefore, uranium and thorium are abundant in the ore bodies of fluorite. However, there is hardly any study to determine the level of indoor as well as the outdoor radiation due to rocks

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and soil in that area. Investigation of trace element profiles of geothermal field matrices in Kerio Valley using chemometric assisted XRF spectroscopy has been done ⁽¹⁰⁾. The high background radiation in the area was found not to be associated with geothermal field.

There has been increasing interest in the study of radioactivity in various building materials. Few data is available about specific activity of ²²⁶U, ²²⁶Th and ⁴⁰K in raw material and products ⁽⁷⁾. Gamma ray spectroscopy provides a reliable method for measuring environmental radiation from naturally occurring radionuclides. The study reported in this work is important because it will provide data for environmental radiation risk assessment. The data obtained will also provide reference levels against which the extent of future enhancement in the radioactivity level of Kerio Valley can be assessed.

The radiation exposure due to building materials can be divided into two: external and internal exposure ⁽¹¹⁾. The external exposure is caused by direct gamma radiation while internal exposure is caused by inhalation of inert gas of ²²²Rn and its short lived decay products. External exposure is further divided into: outdoor and indoor external exposure. Indoor rexposure is caused by materials used in a building but at the same time these materials used in a building act as a shield of outdoor radiation into the house ⁽¹²⁾. In a massive house made of different building materials such as stones, concrete and cement, the factor that mainly affects the indoor activity concentration is the natural radionuclides in those materials. Indoor dose rates are elevated depending on the activity concentration of the materials used ⁽²⁾. The worldwide average radionuclide activity concentration in building materials are ²²⁶U (33 Bqkg⁻¹), ²³²Th (45 Bqkg⁻¹) and ⁴⁰K (400 Bqkg⁻¹) ⁽²⁾. Some countries have laws that govern control of building material to protect the public from elevated indoor exposure.

2. EXPERIMENTAL

2.1 Study area

Kerio Valley lies between the Tugen hills and the Elgeyo escarpment in Kenya, in a narrow strip of about 80 km by 10 km, $35^{0}18-36^{0}20$ E and $0012-0^{0}56$ N. This region is rocky and is characterized by quarrying activities of the various rocks for construction purposes both in the Valley and neighbouring towns. The area has different rock types of different geological formation. Majority of them are volcanic rocks and sedimentary rocks of volcanic origin. Also, various types of metamorphic rock have formed from both sedimentary and igneous rocks ⁽¹³⁾. The bulk of metamorphic rocks are crystalline gneiss. A fluorite ore body occurs at the foot of Elgeyo escapement and stretches from Kimwarer ridge to Muskut; in the southern part of the valley. There is a close association between fluorite mineralization and the uranium anomalies ⁽¹³⁾. Generally, granite igneous rocks are known to have high level of primordial radionuclides - meaning high activity concentration ⁽⁶⁾.

2.2 Sample collection and preparation

Thirty six rock samples were collected from different geological setting characterizing Kerio Valley. The rocks were chiseled out while others were randomly handpicked from the surface and quarries. The rocks were sampled randomly. The geographic coordinates of the sampling points were determined by the Global Positioning System (GPS). The samples were kept in polythene bags to avoid contamination and were numbered for identification. The identification and geological classification of the rock samples was done at Kenyatta University. The rock samples were collected from Twakeu, Tambach, Kitanyi, Kabiemit and Soy administrative sub regions. The rock samples included: granite, tuff, conglomerate, sandstone, limestone, marble, gneiss and Quartizite. The rocks were crushed, sieved and dried to remove moisture. The dried samples were weighed, sealed in radon impermeable plastic containers and stored for four weeks to allow the parent radionuclides in the sample to reach secular equilibrium between ²²⁶Ra and its short-lived decay products ⁽¹⁴⁾. A NaI (TI) - based gamma ray spectrometer was used to measure the activity of radionuclides in the samples.

2.3 Calculation of radionuclide activity concentration

The activity concentration for the natural radionuclides in the samples was computed after spectral decomposition using the stripping off method $^{(15)}$. Activity concentration was calculated using the relation $^{(16)}$;

$$\frac{A_S M_S}{l_S} = \frac{A_R M_R}{l_R} \tag{1}$$

where A_S is the activity concentration of a radionuclide in the sample, M_S is the mass of the sample, I_S is the peak intensity of the radionuclide in the sample, A_R is the activity of the reference standard sample, M_R is the mass of the reference standard sample and I_R is the peak intensity of the radionuclide in the standard sample.

Vol. 3, Issue 1, pp: (40-47), Month: April 2015 - September 2015, Available at: www.researchpublish.com

2.4 Radium equivalent activity

The radiation index is used to assess the suitability of a material for building based on the estimation that 370 Bqkg⁻¹ of ²²⁶Ra, 259 Bqkg⁻¹ of ²³²Th and 4810 Bqkg⁻¹ of ⁴⁰K produce the same gamma dose rate. The recommended maximum value of radium equivalent in building materials and products must be less than 370 Bqkg⁻¹ for safe use. Radium equivalent activity was calculated using the formula ⁽¹⁷⁾;

(2)

(4)

(5)

(6)

(7)

$$Ra_{Eq} = QC_U + RC_{Th} + SC_K$$

where Q, R, and S are conversion factors and they take value; 1, 1.43 and 0.077 respectively ⁽¹⁷⁾.

2.5 External hazards index

This index is used to estimate the radiological hazard of environmental samples. The hazard is defined in terms of external radiation hazard index and is denoted by H_{ex} ; it was calculated using the equation ⁽¹⁷⁾;

$$H_{ex} = \frac{C_U}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \tag{3}$$

where C_U , C_{Th} and C_K are the activity concentration of ²³⁸U, ²³²Th and ⁴⁰K respectively, in Bq/kg. The value of this index must be less than a unit in order for the radiation hazard to be considered acceptable. The maximum acceptable value of H_{ex} corresponds to the upper limit of Ra_{eq} 370 Bq/kg.

2.6 Outdoor gamma radiation dose rate

The calculated activity concentrations in Bqkg⁻¹ of 40 K, 238 U and 232 Th were used to determine the absorbed dose rate 1m above the ground using the formula ${}^{(18)}$;

$$D = 0.417C_{Ra} + 0.604C_{Th} + 0.0417C_{K}$$

where 0.417 nGyh⁻¹/Bqkg⁻¹, 0.604 nGyh⁻¹/Bqkg⁻¹ and 0.0417 nGyh⁻¹/Bqkg⁻¹ are conversion factors for radium, thorium and potassium respectively ⁽²⁾.

2.7 Indoor gamma absorbed dose rate

The indoor absorbed dose rate in air in a room can be calculated by ⁽¹¹⁾;

$$D = \alpha C_{Ra} + \beta C_{Th} + \gamma C_K$$

where α , β and γ are the indoor dose rate per unit activity concentration of ²³⁸ Ra, ²³²Th and ⁴⁰K (nGyh⁻¹/Bqkg⁻¹), respectively. The values of α , β and γ were taken to be 0.67, 0.78 and 0.057 respectively ⁽¹¹⁾. The conversion factors are chosen based on the model house typical to the building constructed using rock materials in Kerio Valley. This is a house in which the floor and the walls are made of radioactive material while the ceiling is absent or made of wood. This is not however, the excess exposure from building materials because concrete structure shield against gamma radiation from undisturbed Earth's crust. Using the world average value of 60 nGyh⁻¹⁽²⁾ for the background, the excess dose rate in the room is therefore calculated by subtracting this background from the calculated value of the indoor dose rate.

2.8 Annual effective dose

To estimate the annual effective dose, the conversion factors from absorbed dose rate in air to effective dose and occupancy time were used. The accepted value of conversion factor from absorbed dose in air to effective dose received by human beings is 0.7SvGy^{-1 (2)}.

The outdoor annual effective dose in mSvy⁻¹ was calculated using ⁽¹⁹⁾;

$$H_{E_o} = D_o T_o f_c$$

where D_o is the outdoor absorbed dose rate in nGyh⁻¹, T_o is the outdoor occupancy time and f_c is the conversion factor. For outdoor an occupancy time of 20 % was considered on the basis that the children who form majority of the population spend more time indoor than outdoor.

The indoor annual effective dose in mSvy⁻¹ was calculated using ⁽¹⁹⁾.

$$H_{E_I} = D_I T_I f_c$$

where D_I is the indoor absorbed dose rate in nGyh⁻¹, T_I is the indoor occupancy time and f_c is the conversion factor. Indoor an occupancy time of 80 % was used.

Vol. 3, Issue 1, pp: (40-47), Month: April 2015 - September 2015, Available at: www.researchpublish.com

3. RESULTS AND DISCUSSION

3.1 Activity concentration of naturally occurring radionuclides

For all the rock types ²³⁸U has higher (72.707-116.149 Bqkg⁻¹) activity concentration than ²³²Th (40.317-83.653 Bqkg⁻¹) with an exception of gneiss in which the activity concentration of ²³²Th (83.653 Bqkg⁻¹) is greater than that of ²³⁸U (77.574 Bqkg⁻¹). This is in agreement with the study ⁽⁹⁾ in which uranium anomalies in various ore bodies of fluorspar was reported in Kerio Valley.

Table 1 presents a summary of statistical analysis for the measured activity concentration in all rock samples. 232 Th indicates asymmetrical distribution with the tail extending towards high concentration (positive skewness) while 40 K and 238 U have negative skewness. However the three radionuclides show a strong evidence of normal distribution since the values for kurtosis and skewness are within the range of normal distribution for the measured samples. The high value of the standard deviation (Table 1) for all the radionuclides means that there are no regular trends in the variation of radioactivity for the different radionuclides for the rock types from Kerio Valley which indicates a strong diversity of the rock types in the study area. The diversity may be as a result of non-uniformity in chemical and geochemical properties in the rocks.

The results obtained in this work are compared with results reported in other parts of Kenya (Machakos -Thika area) for similar types of rocks ⁽⁷⁾. Machakos is a potential area for building material from rocks and the aim of the comparison is to establish trends in variation of activity concentration between Machakos and Kerio Valley. From the comparison, the concentration of ⁴⁰K and ²³⁸U for the all rocks types collected from Kerio Valley exceed the activity concentration of the rock types reported in Machakos -Thika area.

The results of mean activity concentration of rock type obtained in this work were also compared with the results obtained from other countries for similar types of rocks (Table 2) in areas suspected to be high background radiation area (HBRA). The compared values for mean activity concentration are above the world average for background radiation of 33 Bqkg⁻¹, 45 Bqkg⁻¹ and 400 Bqkg⁻¹ for ²³⁸U, ²³²Th, and ⁴⁰K respectively ⁽²⁾. It is noted that there is no great variation of concentration of the naturally occurring radionuclides between the reported values in other countries and the values reported in this work.

3.2 Radiation hazard indices

The radiation hazard indices Ra_{eq} ranged from 207.524 Bqkg⁻¹ (for limestone) to 350.453 Bqkg⁻¹ (for quartzite). Are below the recommended maximum value of 370 Bqkg⁻¹. Therefore none of the rock types pose significant hazard to human exposure. However, gneiss and quartzite which have indices close to the maximum acceptable limit may be hazardous for occupancy time greater than 20% and 80% for outdoor and indoor respectively considered in this work. When compared with the indices reported in other countries for some similar rock types (Table 3), the indices are greater except for granite in Egypt and Nigeria.

Radium equivalent activity mean values ranged from 207 $Bqkg^{-1}$ (limestone) to 280 $Bqkg^{-1}$ (quartzite). These values are below the recommended maximum value of 370 $Bqkg^{-1}$. The high Ra_{eq} in quartzite can be attributed to high activity concentration of ²³⁸U (Table 1). This hazard index conforms to external and internal hazard indices which indicate that the radiation from rocks in the indoor environment Kerio Valley are within the acceptable limit.

3.3 Absorbed dose rate and annual effective dose rate

For all rock types, the indoor gamma dose rate was found to be greater than the outdoor dose rate. Quartzite had the highest dose (163.518 nGyh⁻¹) and 251.772 nGyh⁻¹ for outdoor and indoor respectively. Limestone had the least dose (95.918 nGyh⁻¹) and 125.747 nGyh⁻¹ for outdoor and indoor respectively (Table 4). The values are above the world average of 60 nGyh⁻¹ and 70 nGyh⁻¹ for indoor and outdoor dose rate respectively. When compared with values from high background radiation areas from other parts of the world ⁽²⁾, the values fall in the same range (Table 5). It is noted that igneous rocks (granite) and sedimentary rocks of the igneous origin (quartzite and gneiss) have the highest dose rates. This is as a result of their high abundance of potassium as seen from the activity concentration (Table 1) and relatively high concentration of both uranium and thorium.

The annual effective dose rates due to indoor exposure are greater than the outdoor dose rate for all rock types (Table 4). This is as a result of the high indoor absorbed dose rate observed in all the rock types. Quartzite had the highest annual effective dose rate of 0.195 mSvy^{-1} and 0.772 mSvy^{-1} for outdoor and indoor respectively. Limestone had the least with 0.111 mSvy^{-1} and 0.327 mSvy^{-1} for indoor and outdoor respectively. Indoor annual effective dose rate and the outdoor annual effective dose rate for all rock types are above the world average of 0.4 mSvy^{-1} and 0.07mSvy^{-1} (2) with the exception of sandstone and limestone which have external effective dose rate below the world average.

Vol. 3, Issue 1, pp: (40-47), Month: April 2015 - September 2015, Available at: www.researchpublish.com

4. CONCLUSION

The activity levels of natural terrestrial radionuclides of ²³⁸U, ²³²Th and ⁴⁰K in rocks sampled from Kerio Valley region of Kenya, have been measured using NaI (Tl) gamma ray spectrometry. The radiological parameters have been estimated in different types of rocks based on the activity concentration. The activity concentration varied from 72.707 \pm 58.470 Bqkg⁻¹ (sandstone) to 116.149 \pm 31.463 Bqkg⁻¹ (marble) for ²³⁸U; 40.317 \pm 34.481 Bqkg⁻¹ (limestone) to 83.652 \pm 59.912 Bqkg⁻¹ (gneiss) for ²³²Th and 427.408 \pm 408 Bqkg⁻¹ (marble) to 1397.238 \pm 265.265 Bqkg⁻¹ (granite) for ⁴⁰K. It was found that present in the igneous rock (granite and tuff) and metamorphic rocks of the igneous origin (quartzite and gneiss) have high activity concentrations. This may be attributed to their high silica (SiO₂) content (over 70%) and mineral components such as quartz and feldspar which have the ability of absorbing ²³⁸U, ²³²Th and ⁴⁰K from circulating solutions. Marble which is a sedimentary rock formed from limestone has the highest (116.16 Bqkg⁻¹) ²³⁸U activity concentration while limestone has the lowest (427.42 Bqkg⁻¹) activity concentration for potassium.

The calculated hazard indices; radium equivalent activity and external hazard were found to range (207.104 - 350.453) Bqkg⁻¹ and (0.599 - 0.946) Bqkg⁻¹ respectively. This indicates that the rocks from Kerio Valley are fit to be used as building material and do not pose any risk to the inhabitants. The indoor and outdoor absorbed dose rates ranged from (66.696 - 157.443) nGyh⁻¹ and (90.585 - 159.085) nGyh⁻¹ respectively; which are above the world average of 60 nGyh⁻¹ and 70 nGyh⁻¹ (²⁾ for indoor and outdoor dose rates respectively. The effective dose rates for outdoor radiation and the indoor radiation ranged from (0.111 - 0.195) mSvy⁻¹ and (0.327 - 0.77207) mSvy⁻¹. These values are above the world average 0.07 mSvy⁻¹ and 0.41 mSvy⁻¹ respectively for all the rock types. However, sum of the indoor dose rate and the outdoor dose rate are below the accepted limit of 1 mSvy⁻¹ (²⁷). It is noted that some rocks contribute more to the environmental dose than others implying that not all rocks contribute more to the high background radiation in Kerio Valley.

Rock type		²³⁸ U	²³² Th	⁴⁰ K
	Mean	99.364±51.45	47.644±29.21	1397.238±265.35
	Skewness	1.048	0.571	-0.965
	kurtosis	-0.950	-1.615	-1.044
Granite	Std Dev.	51.453	29.205	265.346
	Mean	96.069±30.78	71.259±57.14	1192.457±214.96
	Skewness	-0.234	0.633	-0.134
	kurtosis	-1.743	-1.526	-1.491
Tuff	Std Dev.	31.293	62.286	214.962
	Mean	72.707±58.47	64.180±44.68	677.387±200.49
	Skewness	0.100	0.291	-0.430
	kurtosis	-2.036	-2.214	-1.671
Sandstone	Std Dev.	58.470	44.684	200.487
	Mean	95.879±38.37	74.240±62.32	980.691±157.14
	Skewness	-0.0272	0.225	0.268
	kurtosis	-2.175	-2.145	-2.2.131
Conglomerate	Std Dev.	38.372	62.316	157.137
	Mean	104.785±68.61	40.317±34.48	427.408±165.76
	Skewness	0.080	0.573	0.186
	kurtosis	-2.346	-1.787	-2.021
Limestone	Std Dev.	68.614	34.481	165.759
	Mean	116.149±31.46	53.577±21.61	576.461±116.35
	Skewness	-0.016	0.271	-0.381
	kurtosis	-0.233	-2.333	-2.333
Marble	Std Dev.	31.463	21613	31.463
	Mean	77.574±24.61	83.653±59.91	1294.353±319.57
	Skewness	0.231	0.241	-0.575
	kurtosis	-2.108	-2.165	-1.834
Gneiss	Std Dev.	24.608	72.199	319.570
	Mean	101.730±25.92	44.317±34.48	1224.669±124.50
	Skewness	0.125	0.000	0.094
	kurtosis	-1.870	-1.437	-2.329
Quartizite	Std Dev.	25.923	23.405	124.498

Table.1 Average activity concentration of natural radionuclides in different rock types

Country	²³⁸ U (Bqkg ⁻¹)	²³² Th (Bqkg ⁻¹)	⁴⁰ K (Bqkg ⁻¹)	Reference
Yemen	46.9±13	90.1±34	1424.9±640	(20)
Egypt	51.3±20	46.9±23	1213.0±561	(21)
Nigeria	69.4 <u>±</u> 30	63.4±23	487.0±169	(17)
Turkey	47.5±11	43.7±25	1018.3±436	(22)
Kenya work (Kenya)	94.47±31	65.0±23	1003.9±217	Present study

Table.2 Comparison of mean activity concentration of rock types reported by other researchers in other countries

Table.3 Dose rate (nGyh⁻¹), radium equivalent activity, hazard indices and annual effective dose rate reported in other countries verses those obtained in this work.

Country	Rock type	Dose rate (nGyh ⁻¹)	Ra _{eq}	H _{ex}	Iy	HE (mSvy ⁻¹)	Reference
Egypt	Gneiss	92.7	191.0	0.5	-	-	(21)
0.71	Granite	201.2	417.4	1.1	-	-	× /
	Sandstone	22.0	45.7	0.1	-	-	
Nigeria	Granite	42.0	384	1.0	1.4	0.2	(17)
_	Sandstone	17.0	173	0.4	0.6	0.1	
	Limestone	17.0	157	0.4	0.7	0.1	
	Quartzite	18.0	170	0.5	0.6	0.1	
Turkey	Sandstone	71.3	139.6	0.3	-	0.3	(22)
	Limestone	52.2	54.0	0.1	-	0.3	
	Marble	30.1	254.2	0.7	-	0.2	
Kenya	Granite	132.9	285.4	0.8	1.1	0.2	Present work
(Kerio	Tuff	137.2	304.4	0.8	1.1	0.2	
Valley)	Sandstone	100.6	228.1	0.6	0.8	0.1	
-	Conglomerate	130.0	290.5	0.8	1.1	0.2	
	Limestone	90.6	207.1	0.6	0.7	0.1	
	Marble	110.6	232.1	0.6	0.8	0.1	
	Gneiss	128.8	333.7	0.9	1.2	0.2	

Table.4 Absorbed dose rates and annual effective dose rate due to different rocks from Kerio Valley

Rock type	$D_o(nGyh^{-1})$	$D_i(nGyh^{-1})$	$H_{E_o}(mSvy^{-1})$	$H_{E_i}(mSvy^{-1})$
Granite	132.95	124.06	0.16	0.61
Tuff	137.15	128.60	0.17	0.63
Sandstone	100.60	78.07	0.12	0.38
Conglomerate	130.03	118.73	0.16	0.58
Limestone	90.59	66.70	0.11	0.33
Marble	110.06	93.15	0.14	0.46
Gneiss	128.75	116.71	0.15	0.57
Quartzite	159.09	157.44	0.20	0.77

Table.5 Exposure rate	e from high 🛛	background	radiation	areas in	different	countries
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Region	Area	Exposure rate in air (nG	Reference	
		Outdoor dose rate	utdoor dose rate Indoor dose rate	
Czech	Central Bohemic	90-170	119	(23)
Italy	Lazio	175 (120-270)	250 (105-440)	(24)
Italy	Orvieto	560		(1)
Italy	Southern Tuscany	150-300	190 (40-350)	(25)
India	Kerala Madras	1500 (845-5270)	110 (66-157)	(26)
Kenya	Kerio Valley	124 (91-158)		Present work

Vol. 3, Issue 1, pp: (40-47), Month: April 2015 - September 2015, Available at: www.researchpublish.com

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APPENDIX - A

Figure.1 A map of Kerio Valley showing the sampling sites. The circles represent sites collected from different specific administrative sub regions. The administrative sub regions are: 1-Twakeu, 2-Tambach, 3-Kitanyi, 4-Kabiemit and 5-Soy.