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NATURAL RADIOACTIVITY IN DRINKING WATER SOURCES IN KARGI AREA, MARSABIT – KENYA

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ABSTRACT: *Kargi is located between latitudes 2.476944 and 2.520833 N and longitudes 37.542778 and 37.601944 E covering an area of approximately 31.26 km^2. Health facilities in the area have reported a number of cancer disease cases and deaths annually thus prompting for the study. 14 number water samples were collected in Kargi, Marsabit with the aim of assessing the area's environmental radioactivity together with radiological health hazard. Radionuclides are considered as radiation exposure source which has been recorded to have damaging health effects for such populations living in high back-ground radiation areas. Natural radioactivity concentrations owing to ^{40}K, ^{232}Th and ^{226}Ra radionuclides were measured by using gamma ray spectrometry employing a high purity germanium (HPGe) type detector at University of Nairobi's Nuclear Science & Technology Institute, INST. To assess the natural radioactivity's radiological hazard, absorbed dose rate, radium equivalent together with effective dose have been computed and results compared with internationally accepted values. The mean values for activities for ^{40}K ranged from -0.88 to 81.22 Bql^{-1} (mean: 52.68±25.07 Bql^{-1}), ^{232}Th ranged from -2.00 to 7.99 Bql^{-1}(mean: 2.20±2.74 Bql^{-1}) and ^{226}Ra ranged from -0.25 to 7.84 Bql^{-1}(mean: 3.55±3.04 Bql^{-1}). Mean ^{232}Th and ^{40}K values were 2 times and 5 times higher than the limit values respectively, hence the area can be categorized a high back-ground radiation area, HBRA. The mean values with their standard deviations for calculated and measured dose rates below and above surface (1 m from the ground) were 5.37±2.93 and 77.76±27.82 nGyh^{-1} respectively. The computed radium equivalent values of all 14 samples were lower than accepted limit of 370 Bqkg^{-1} (Lu Xinwei et al., 2006). The mean elemental ratio for Th/U was lower than 3, an indication of thorium depletion or enrichment in the area. Investigated waters in this area are acceptable for life-long human consumption since their values of annual effective doses proved lower than the world-wide values.*

KEYWORDS: kargi-marsabit, nuclear science, gamma-ray spectrometry, lifetime cancer risk, activity.

INTRODUCTION

Water is considered a major ingredient of the human body hence crucial for life. Some natural phenomenon and various human activities do pollute, affecting the water quality. Some of the waste disposals associated with the human activities affecting water quality often contain

radioactive materials which remarkably contribute to the water bodies' background activity. Waters used for drinking normally contain a number of natural radionuclides including but not limited to radon, uranium isotopes, radium, tritium, etc. Their concentrations widely vary as they rely on the aquifer nature, called, the prevailing lithology and the presence or absence of air in it (Cristina *et al.*, 2012).

Sourced waters from deep wells and/or boreholes used for drinking are normally anticipated to have elevated concentration levels of radionuclides. This is due to it passing through bedrock fractures in or within the soil containing mineral deposits which might possess radioactive constituents and hence leaking into water ways (Njinga *et al*., 2015). Most radium salts are water solvents, thus drinking, together with mineral waters from the surface may be supplemented in radium and its heir radon.

Among the major ways in which radionuclides find their way into the human body is by radioactivity in drinking water, which might as a result lead to radiation-induced disorder (USEPA 2010). In his research, Otton, 1994 explains that at lower to moderate doses, radiation exposure in both human and animal may rise the long term occurrence of cancer disease and that genetic malformations rate may rise by exposure due to radiation. It is important therefore to find out the existing amount of radioactivity in water meant for drinking for every residential area in order to protect against its harmful effects (WHO, 2006). Drinking water guidelines by WHO suggest an indirect performance rating of individual dose criterion (IDC) of 0.1 $mSvy^{-1}$ by measurement of complete beta and alpha radioactivity and by checking the compliance of radionuclide activity concentration to obtained guidance levels (WHO, 2011).

Approximation of radiation dose diffusion is crucial in evaluating the risk of health to inhabitants, serving as a point of reference for documentation of changes existing in environmental radio-activity resulting from human impact on environment (Obed *et al*., 2005).Human beings are also unprotected by pollution of food chain occurring as a result of radionuclides deposition on leaves of plants, uptake by roots from polluted soil, water and/or sediment (Arogunjo *et al*., 2004). Again, human beings are unprotected from direct consumption of polluted waters (Avwiri and Agbalagba, 2007).

Health facilities in Kargi area have reported a number of cancer disease related cases and deaths. Animals have also been reported to have died from drinking of waters here. Taskin *et al.*, 2009, have it that radiation exposure could lead to kidney, lungs, bone, pancreas together with skin cancers, leukemia and cataracts. In studying the waters in the area, one gets to have knowledge of presence of natural radioactivity enbling us to assess any radiological hazard possible to humans and animals using such waters. The general intent is to determine the human exposure level to fount of radiation considered natural in Kargi area within Marsabit County, Kenya.

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MATERIALS AND METHODS

Study area

Figure 1.0 shows area under study. Kargi is in Marsabit county, Laisamis constituency, Loiyangalani sub-county and approximately 72 km from Marsabit town to the west. Manyattas, villages and Market place of Kargi were studied. Kargi borders Gabra, Samburu and Chalbi. The area under study, approximately 6.54 km by 4.78 km is located between latitudes 2°28´37´´ and 2°31´15´´ N and longitudes 37°32´34´´ and 37°36´07´´ E as seen from figure 1.0.

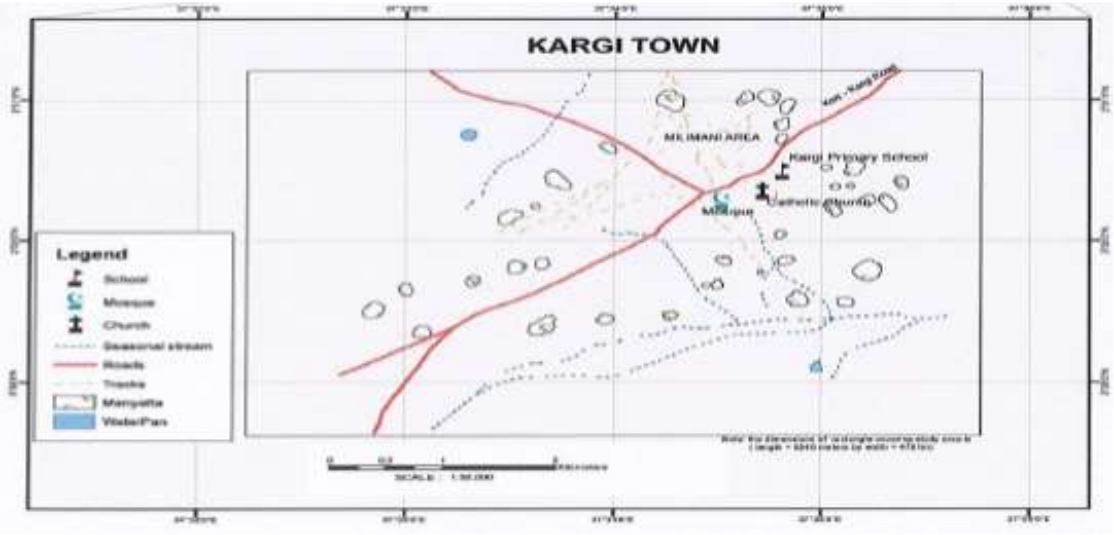

Figure 1.0: Kargi area map (Survey of Kenya, 2017, modified).

Sample collection and preparation.

Samples of water were collected from water sources in the area. These sources were categorised as boreholes, shallow wells which are shallow manually dug boreholes, Tap/Water kiosks and Dams/wells.

A handheld radiation survey meter was also used to monitor absorbed dose rates where the water samples were being collected. Table 1.0 shows water samples collection sources and their locations in the map.

Collection of 14 number water samples were done in standard (0.5 litre) polyethylene Marinelli containers, packed and clearly marked before transporting to Nairobi. These containers were clean washed and rinsed with diluted hydrochloric acid together with distilled water respectively before filling with water samples. 0.25 ml of concentrated Nitric acid (HNO_3) was added to the collected samples to help preventing any radium isotopes loss around the walls of the container and to avoid micro-organisms growth (Hany, E. and Abdallah, I. A., 2014). The containers were filled to brim not to allow any air inside and tightly closed before being transported.

In Nairobi, the samples were weighed, packed in special plastic containers (marinelli beaker) then closed tightly for 4-weeks, considered adequate duration needed to attain secular equilibrium state

after their progeny (Karahan and Bayulken, 2000). After the period, the samples were then taken to University of Nairobi's Nuclear Science & Technology Institute for spectroscopic analysis.

Natural radionuclide activity and absorbed doses

A high-purity germanium (HPGe) type γ-ray detector having 144 mm^3 active volume, 76 mm outside diameter with 1.8 keV and 31.6 % as resolution and efficiency respectively was used for measurement of activity concentrations. Each sample was put in a marinelli beaker, 500 cm^3, filled to a level similar as the IAEA certified standard of reference (IAEA-RGU-1, IAEA-RGTh-1 and IAEA –RGK-1) before placing in a lead-shielded detector. Each sample was run for between 16000 – 62000 seconds which was used as counting time. Sample reference standards from International Atomic Energy Agency, IAEA (IAEA-RGU-1, IAEA-RGTh-1 and IAEA –RGK-1) were used for the method validation and calibration of spectrometer (IAEA 2003) using Maestro software.

A comparative method was used to compute the activities of each radionuclide. Five significant gamma lines, ^{40}K line, $^{214}P_b$ and $^{214}B_i$ lines from ^{238}U and $^{212}P_b$ and $^{228}A_c$ lines from ^{232}Th for each sample existed. ^{40}K activity was evaluated from 1461 keV gamma-line, ^{238}U activity from 352 keV and 609 keV gamma lines of $^{214}P_b$ and $^{214}B_i$ respectively and that of ^{232}Th from 238 keV and 912 keV gamma-lines of $^{212}P_b$ and $^{228}A_c$ respectively.

Table 1: Sample code, Location, average activity concentration (Bql^{-1}) and Radium equivalent (Bql^{-1}).

Sample code	**LOCATION**		**Average activity Concentration, Bql^{-1}**			**Ra_{eq}, $Bqkg^{-1}$**
	(Longitude) Easting, E	**(Latitude) Northing, N**	**^{226}Ra**	**^{232}Th**	**^{40}K**	
W_1	37.591971	2.517675	3.63±2.79	0.12±2.12	71.33±10.81	9.29
W_2	37.596068	2.503742	4.55±1.81	2.64±2.14	54.63±8.67	12.53
W_3	37.583479	2.495335	7.84±5.05	1.71±6.03	36.01±28.50	13.06
W_4	37.58304	2.485123	2.08±2.19	0.98±3.16	68.92±9.92	8.79
W_5	37.549452	2.486624	3.39±4.39	7.99±4.61	57.72±24.74	19.26
W_6	37.553105	2.492507	6.04±2.18	3.94±3.76	77.16±14.58	17.62
W_7	37.568392	2.509391	5.66±2.40	-2.00±2.37	45.68±19.38	6.32
W_8	37.56921	2.501813	6.41±2.85	1.90±3.07	64.45±14.13	14.09
W_9	37.569227	2.501994	7.66±2.97	4.37±3.95	60.49±19.75	18.57
W_{10}	37.570479	2.492526	0.59±2.01	3.12±3.16	42.33±14.70	8.30
W_{11}	37.570588	2.499644	3.59±2.21	0.19±2.81	72.53±11.57	9.45
W_{12}	37.570606	2.499707	0.27±2.63	0.54±3.85	-2.36±13.22	0.84
W_{13}	37.576149	2.503919	-1.82±3.42	6.09±4.74	81.22±22.00	13.14
W_{14}	37.575952	2.503693	-0.25±2.70	-0.85±2.55	7.61±12.69	-0.88
Average			**3.55±3.04**	**2.20±2.74**	**52.68±25.07**	**10.74±6.03**

Absorbed gamma dose (D)

The absorbed dose (D) owing to gamma (γ) radiations in air, measured 1 m above the ground surface for continuous dissemination of the naturally occurring radionuclides (^{40}K, ^{232}Th, and ^{226}Ra) were computed based on UNSCEAR 2000 guidelines. Assuming that contributions coming from other naturally occurring radionuclides to be insignificant, then D can be computed from:

$$D\left(nGyh^{-1}\right) = \left(0.467C_{Ra} + 0.662C_{Th} + 0.043C_{K}\right) \quad \text{.....1}$$

Where,

C_k, C_{Th} and C_{Ra} are the average acivities of potassium, thorium and radium respectively in the sample and $nGyh^{-1}$ (nano Gray per hour) is the unit of the absorbed dose rates (D).

Radium equivalent activity (Ra_{eq})

Radium equivalent (Ra_{eq}) activity is a presentation of a mean sum of activities of K-40, Th-232 as well as Ra-226. Its approximation is established on the fact that 0.7 $Bqkg^{-1}$ of ^{232}Th, 1 $Bqkg^{-1}$ of ^{238}U together with 13 $Bqkg^{-1}$ of ^{40}K generate similar radiation dose rates. Alvwiri *et al.*, 2013 approximates radium equivalent activity as:

$$Ra_{eq}(Bqkg^{1}) = C_U + 1.43C_{Th} + 0.077C_K \quad \text{.....2}$$

Where,

$C_U, 1.43C_{Th}$ together with $0.077C_K \rightarrow$ activity concentrations in Bql^{-1} or $Bqkg^{-1}$ of ^{238}U, ^{232}Th as well as ^{40}K.

Annual effective dose (AED)

For a proper dose estimation caused by natural radionuclides as found in drinking water, several factors must be addressed. These factors include weighted mean activities in (Bql^{-1}), a yearly amount of water intake by a person, dose co-efficient ($mSv.Bq^{-1}$) for radionuclides ingestion by humans.

Equation 3 below estimates the annual effective dose resulting from water ingestion based on assumption that daily intake of water per person occur (EPA, 1999; Meltem and Gursel, 2010).

$$DR_W = A_W \times IR_W \times ID_F \quad \text{.....3}$$

Where DR_W is effective dose ($mSv.y^{-1}$), ID_F is dose equivalent ($mSv.Bq^{-1}$) conversion factor, A_W is activity (Bql^{-1}) and IR_W is a person's water intake in a year.

(WHO, 2008) estimates dose consumption rates for infants, children and adults as 0.5, 1.0 and 2.0 ld^{-1} respectively. Conversion factors for ^{226}Ra, ^{232}Th and ^{40}K for infants, children and adults are $1.4 \times 10^{-7}, 1.6 \times 10^{-6}\ and\ 5.2 \times 10^{-8}\ SvBq^{-1}, 6.8 \times 10^{-8}, 2.9 \times 10^{-7} and\ 1.3 \times 10^{-8}\ SvBq^{-1}, 4.5 \times 10^{-8}, 2.3 \times 10^{-7} and\ 6.2 \times 10^{-9}\ SvBq^{-1}$ respectively (ICRP, 2012).

Correlation between ^{232}Th and ^{40}K, ^{238}Uand ^{40}K and ^{238}U and ^{232}Th.

According to Avwiri *et al*., 2014, the elemental concentrations (ppm) of U-238, Th-232 together with percentage potassium can be computed from activities of Th-232, U-238 together with K-40 in $Bqkg^{-1}$ as measured using the below conversion factors:

1 ppm Th = 4.06 $Bqkg^{-1}$ (of Th-232)……4

1 ppm U = 12.35 $Bqkg^{-1}$ (of U-238) ……..5
1 % K = 313 $Bqkg^{-1}$ (of K-40) (IAEA Technical Report No. 1363)……6

RESULTS AND DISCUSSION

The specific activities of ^{40}K, ^{232}Th series (^{232}Th) as well as ^{238}U series (^{226}Ra), expressed in Bql^{-1} for samples obtained from Kargi area are as in table 1. From table 1 ^{226}Ra, ^{232}Th, ^{40}K and Ra_{eq} values range from -1.82 to 7.84 Bql^{-1}, -2.00 to 7.99 Bql^{-1}, -2. 63 to 81.22 Bql^{-1}, and -0.88 to 18.57 $Bqkg^{-1}$ with arithmetic mean and standard deviation as 3.55±3.04, 2.20±2.74, 52.68±25.07 and 10.74±6.03 respectively. The worldwide recommended values are respectively 10.0 Bql^{-1}, 1.0 Bql^{-1}, 10.0 Bql^{-1} (UNSCEAR, 2000 and WHO 2008 Standards) and 370 $Bqkg^{-1}$ as per Organization for Economic Cooperation and Development, OECD and (Lu Xinwei *et al*., 2006) for all the samples.
Except for ^{232}Th and ^{40}K average activity values, all other average activity values for ^{226}Ra and Ra_{eq} were less than the worldwide recommended values. Figure 2 shows average activities of the radionuclides ^{40}K, ^{226}Ra and ^{232}Th as found in the tested samples.

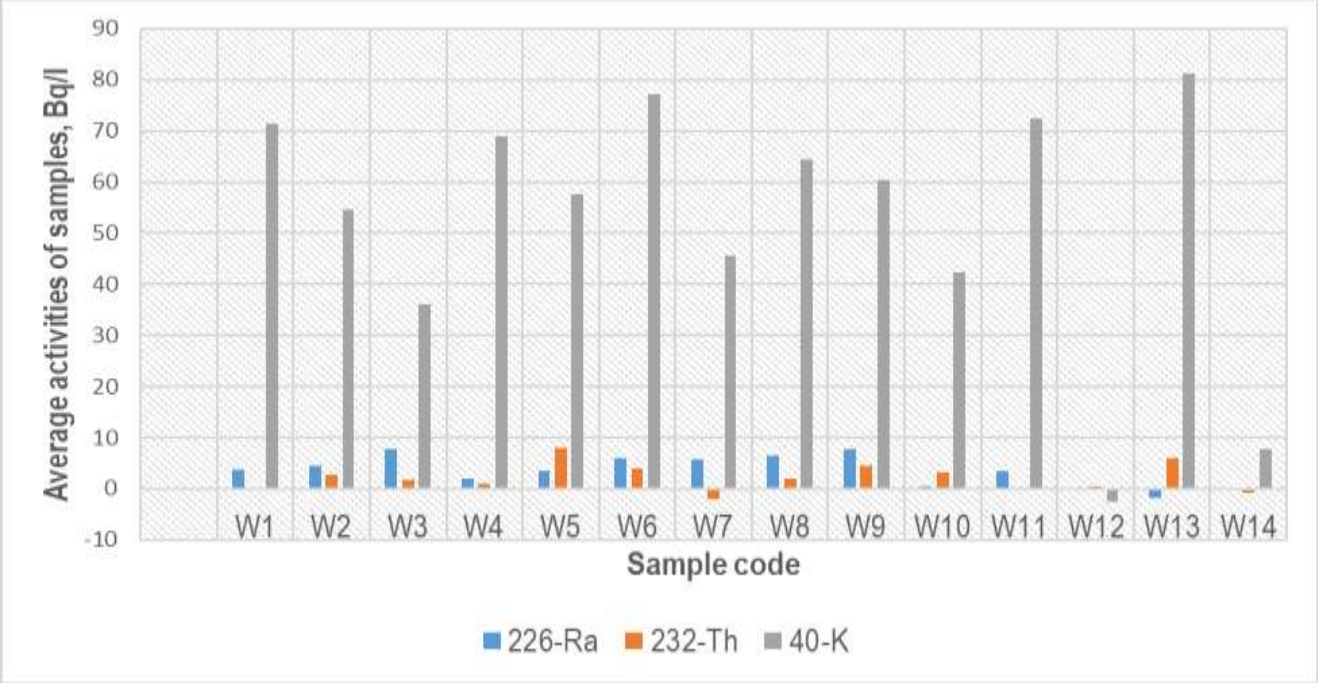

Figure 2: Activity values of samples due to ^{226}Ra, ^{232}Th and ^{40}K in investigated area.

Similar investigations as done in other countries were compared with the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in water samples from studied area and results summarised in table 2.

Table 2.0: Comparison of results of activity concentration in Bql^{-1} of ^{226}Ra, ^{232}Th and ^{40}K of tested samples with other countries

Country	**Activity concentration**			**Reference**
	^{226}Ra	**^{232}Th**	**^{40}K**	
Kenya, Kargi	Mean **3.55**	Mean **2.20**	Mean **52.68**	Present work
Egypt, A	0.20 (mean)	0.13 (mean)	5.29 (mean)	Hany, E and Abdallah, I. A. E., 2014
Egypt, B	1.83 – 18.53	1.47 – 12.78	8.09 – 64.64	
Sudan	0.007 – 0.014	0.001 – 0.039		Alfatih *et al.*, 2006
China	0.93 (max)			Zhuo *et al.*, 2001
Poland	0.001 – 0.049			Jankowski *et al.*, 2000

The values of measured absorbed doses as measured 1 m above ground surface at each water source are tabulated in table 3. Measured and calculated mean absorbed dose rates for all water samples was 77.76±27.82 and 5.37±2.93 **$nGyh^{-1}$** against the world's average value of 60 and 54 **$nGyh^{-1}$** respectively (UNSCEAR, 2000). The elevated measured absorbed dose rate could have been due to background radiation from the sun.
Figure 3.0 shows W_1 and W_9 having the highest measured together with calculated absorbed dose rates respectively. W_9 and W_{13} have the least measured and W_{14} has the least calculated absorbed dose rates.

Table 3.0: Calculated and measured (1 m above ground) absorbed dose of samples from sources.

Sample code	**Calculated**	**Measured**
	Absorbed dose, $nGyh^{-1}$	**Average (source) absorbed dose, $nGyh^{-1}$**
W_1	4.84	130.00
W_2	6.22	107.14
W_3	6.34	75.74
W_4	4.58	58.57
W_5	9.35	85.71
W_6	8.75	61.43
W_7	3.28	108.57
W_8	7.02	45.71
W_9	9.07	95.71
W_{10}	4.16	61.43
W_{11}	4.92	44.29
W_{12}	0.37	64.29
W_{13}	6.67	44.29
W_{14}	-0.35	105.71
Mean	**5.37±2.93**	**77.76±27.82**

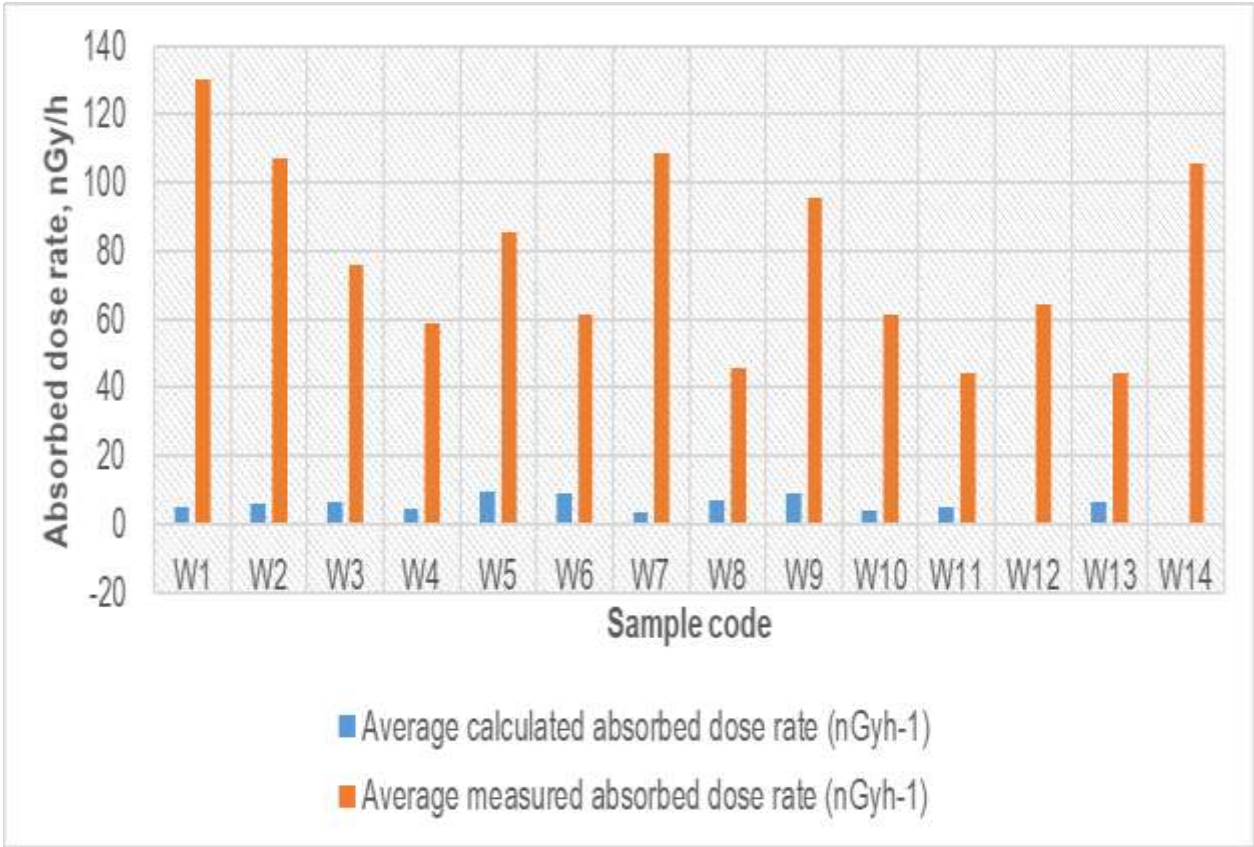

Figure 3: Mean absorbed doses as measured 1 m above ground surface and as calculated at each location.

Table 4 gives calculations for the AED values for adults, children and infants. From the table, the results are below the recommendation (WHO 2004, IAEA 1996, UNSCEAR 2000) of 0.1, 0.2 and 0.26 $mSvy^{-1}$ for effective doses for adults, children and infants respectively from a one-year consumption of drinking water.

Elemental radionuclide concentrations in the samples were computed from the activities in $Bqkg^{-1}$ employing conversion factors in equations 4 – 6. These results are presented in table 5. From the table, the computed elemental concentrations ranged from -0.020 to 0.685 ppm, -0.209 to 1.968 ppm and -0.209 to 0.259 % with arithmetic mean and standard deviation of 0.287±0.246 ppm, 0.541±0.674 ppm and 0.168±0.080 % for uranium, thorium and potassium respectively. Tzortzis and Tsertos (2004) together with Al-Hamarneh and Awadalla (2009) noted that high or low values of Th/U ratios may be an indication of a uranium depletion or thorium enrichment owing to natural processes alteration in that area. The theoretical values are expected to be 3.0 for normal continental crust.

Table 4.0. Annual effective doses ($mSvy^{-1}$) estimates caused by ingestion of ^{226}Ra, ^{232}Th and ^{40}K for different age groups as per sources.

Sample code	**^{226}Ra**			**^{232}Th**			**^{40}K**			**Total dose**		
	Infants	Children	Adults	Infants	Children	Adults	Infants	Children	Adults	Infants	Children	Adults
W_1	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000	0.000	0.001	0.000	0.000
W_2	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000	0.000	0.001	0.001	0.001
W_3	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.001	0.001
W_4	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000	0.000	0.001	0.000	0.001
W_5	0.000	0.000	0.000	0.000	0.001	0.001	0.001	0.000	0.000	0.001	0.001	0.002
W_6	0.000	0.000	0.000	0.000	0.000	0.001	0.001	0.000	0.000	0.001	0.001	0.001
W_7	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000	0.000
W_8	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000	0.000	0.001	0.001	0.001
W_9	0.000	0.000	0.000	0.000	0.000	0.001	0.001	0.000	0.000	0.001	0.001	0.001
W_{10}	0.000	0.000	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.001	0.001	0.001
W_{11}	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000	0.000	0.001	0.000	0.000
W_{12}	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
W_{13}	0.000	0.000	0.000	0.000	0.001	0.001	0.001	0.000	0.000	0.001	0.001	0.001
W_{14}	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Average	0.000	0.001	0.000	0.000	0.000	0.000	0.004	0.008	0.000	0.001	0.001	0.001
Global limit										0..26	0.2	0.1

Table 5.0. Elemental concentration of specific activity of ^{238}U, ^{232}Th and ^{40}K (ppm) in samples with their ratios.

Sample code	**^{238}U**	**^{232}Th**	**^{40}K**	**Th/U**	**K/U**	**K/Th**
W_1	0.294	0.030	0.228	0.101	0.775	7.710
W_2	0.368	0.650	0.175	1.765	0.474	0.268
W_3	0.635	0.421	0.115	0.663	0.181	0.273
W_4	0.168	0.241	0.220	1.433	1.307	0.912
W_5	0.274	1.968	0.184	7.169	0.672	0.094
W_6	0.489	0.970	0.247	1.984	0.504	0.254
W_7	0.458	-0.493	0.146	-1.075	0.318	-0.296
W_8	0.519	0.468	0.206	0.902	0.397	0.440
W_9	0.620	1.076	0.193	1.735	0.312	0.180
W_{10}	0.047	0.768	0.135	16.363	2.880	0.176
W_{11}	0.291	0.047	0.232	0.161	0.797	4.952
W_{12}	0.022	0.133	-0.008	6.084	-0.384	-0.063
W_{13}	-0.147	1.500	0.259	-10.179	-1.761	0.173
W_{14}	-0.020	-0.209	-0.209	10.342	-1.201	-0.116
Mean	**0.287±0.246**	**0.541±0.674**	**0.168±0.080**	**2.675±6.084**	**0.376±1.084**	**1.068±2.311**

CONCLUSIONS

From this study, the mean activities for the area studied is 5 times high, low and 2 times high against the worldwide accepted limit for ^{40}K, ^{226}Ra and ^{232}Th (UNSCEAR, 2000) respectively. This area can thus be classified as a High Back-ground Radiation Area (HBRA) and more precautions need to be taken when living in this area. Radium equivalent (Ra_{eq}) values for the studied waters are below global limit of 370Bqkg^{-1} as suggested by Lu Xinwei *et al*., 2006.

The mean absorbed dose as measured 1 m from the ground was high by 30%. This can be attributed to natural background from the sun. Mean calculated absorbed dose rate was below the recommended value. The mean measured absorbed dose was found higher than the mean calculated dose rate, this could be because the computed dose rates do not have cosmic rays. This area therefore may be considered a High Back-ground Radiation Area (HBRA). It is our recommendation that more research be done in this area, more so during rainy seasons. This will help know whether activities are carried from other places to this area.

The values of annual effective doses were lower in-comparison with global values, hence it can be of recommendation that waters in Kargi are acceptable for life-long human consumption.
The mean elemental ratio for Th/U was lower than 3, an indication of thorium depletion or enrichment in the area.

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