



Natural Radio Activity Levels in Water and Soil at Kemessie Hot Spring, North-Eastern Ethiopia

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Abstract: Natural Radioactivity levels in soil and water supplies remain of interest because of the radiation-induced public health hazards. A large part of the Ethiopian population relies on springs for their drinking water. The present work investigated radioactivity level in soil and water of 12 samples collected from different locations of Kemessie. Concentrations of radionuclides in soils and water samples were determined by gamma-ray spectrometer using a NaI (TI) detector. The mean activity level of the natural radionuclides ^{238}U , ^{232}Th and ^{40}K are 248.71 ± 20.8 , 60.1 ± 3 and $576.46 \pm 39.86 \text{ Bqkg}^{-1}$ respectively from soil at Kemessie. The mean activity level of the same radionuclides from water samples are 2.30 ± 0.05 , 1.70 ± 0.53 and $17.70 \pm 1.53 \text{ BqL}^{-1}$. Also radium equivalent activity, total dose rates, internal and external hazard indices of the soil and water samples under consideration were calculated. The results showed that the average radium equivalent activity, total absorbed dose rate, internal and external hazard indices from soil samples at Kemessie are higher than recommended values. However for water samples all the hazard indices are within safety limits. The paper recommends further studies to estimate internal and external doses from other suspected radiological sources to the population in the region. Such studies can further be extended to many hot springs in this country.

Keywords: Natural Radioactivity, Absorbed Dose, Annual Effective Dose, Water, Soil

1. Introduction

One of the most important aspects of radioecological studies is to provide the scientific basis and awareness to mankind due to impact of different radionuclides. The natural radioactivity in our environment has been investigated intensively in the last years due to public concern of radiation induced health hazard. Natural environmental radioactivity and the associated external exposure due to gamma radiation depend mainly on the local geological and geographical conditions and appear at different levels in each region in the world [1]. A hot spring brings dissolved solids especially a very high mineral content, containing everything from simple calcium to lithium, and even radium, radon, ^{40}K [2, 3] etc which are radioactive materials. The knowledge of natural radioactivity present in soil and water enables one to assess any possible radiological hazard to mankind by uses of such material. There are quite number of hot springs existing in Ethiopia and they attract tourists from within the country and

abroad. Many people do take bath in the hot springs and using the water for cooking purpose as well. Local people believe the water is used to cure some kind of skin diseases. But scientific assessment of natural activities had not been done in many areas in this country. The aim of this study is to estimate the radioactivity concentrations as well as the environmental outdoor (observed) gamma dose rates. The Radium Equivalent, absorbed dose rate, internal and external hazard indices, the annual effective dose have been assessed. This study is vital in assessing the health risk to the population and serves as a reference for changes in environmental radioactivity due to anthropogenic activities as no such study has been carried out before. This study assesses the specific activities and examines some of the radiation hazard indices of these naturally occurring radionuclides ^{238}U , ^{232}Th , and ^{40}K in soil and water samples from Kemessie hot spring, using gamma-ray spectrometry.

2. Materials and Methods

2.1. Description of Study Areas

The study was carried at hot spring Kemessie found in the main East Africa Rift valley at Ethiopia nearby the main road from Addis Ababa to Mekelle. Figure 1 shows the location of study area on Ethiopia map.

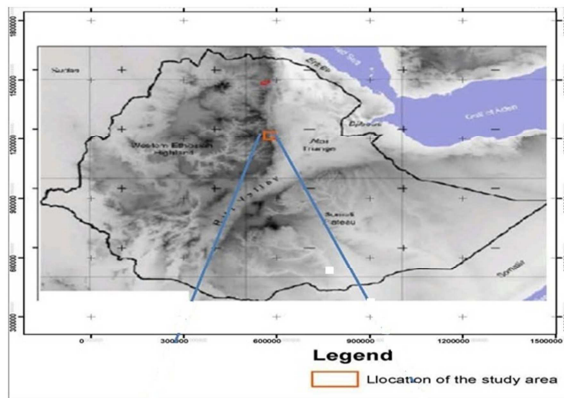


Figure 1. Location of the study area on Ethiopia map.

2.2. Kemissie Hot-Spring

It is found near to Kemissie town at a place known as Chefa wet land. This hot spring can be found at $10^{\circ}38'06.57''\text{N}$, $39^{\circ}55'21.95''\text{E}$ at elevation of 4656 ft from sea level. Figure 2 depicts satellite picture of Kemessie hot spring.



Figure 2. Location of Kemessie hot spring.

3. Sampling and Samples Preparation

3.1. Soil Sampling

We collected samples at three different locations of the

site. Location-0 is the place where water percolates from the Earth while Location-1 and Location-2 are the places where people use water for bathing and they are a few meters away from the main source. Total 6 soil samples (2 at location-0 + 2 at location-1 + 2 at location-2) were collected. Codes KL_s-0 are given for the samples collected at the Origin of the water. Similarly KL_s-1 and KL_s-2 are labeled for the soil samples collected at the prescribed locations respectively. The surface soil samples were collected at a depth of 10-15 cm in high quality plastic bottles and mixed thoroughly. After removing the stones and inorganic materials, the samples were oven dried at $80^{\circ}\text{C} - 90^{\circ}\text{C}$ for about 24 hours. The dried samples were grinded with mortar and pestle and then allowed to pass through a 1 mm-mesh sieve. In order to maintain radioactive equilibrium between parent nuclides and its daughters, the soil samples were stored for a period of one month for equilibrium.

3.2. Water Sampling

Water samples were collected at the same locations where soil samples were collected and the respective codes are $KLw-0$ at the Origin, $KLw-1$ and $KLw-2$ at Locations 1 and 2 respectively. Before collecting the water samples the containers were washed with dilute hydrochloric acid and rinsed with distilled water. Each beaker was filled up to brim and a tight cap was pressed on so that the air was completely removed from it. The collected water samples were left for an overnight period in polyethylene containers to allow setting of any suspended solid materials and for each sample a clear supernatant was separated by decantation. The clear solution was acidified by adding 0.5 ml of concentrated HCl per liter, to prevent any loss of radium isotopes around the container walls, and to avoid growth of microorganisms. The water samples were then homogenized well by shaking. The final acidity of water samples reaches pH-2. The samples were stored for over 30 days to reach secular equilibrium before radiometric analysis [4, 5].

4. Experimental Setup

After 30-days of sample collection, the experiments for radioactivity measurement of the spring water and soil samples were carried out at the Ethiopian Radiation Protection Authority (ERPA) using a thallium activated Canberra vertical high purity $4'' \times 4''$ Sodium iodide [NaI (TI)] detector connected to ORTEC 456 amplifier. The detector was connected to a computer program that matched gamma energies to a library of possible isotopes. The detector was shielded by 15cm thick lead on all four sides and 10 cm thick on top.

4.1. Methodology for Gamma Spectrometry

Each sample was measured with a gamma-ray spectrometer consisting of a NaI (TI) setup and multichannel analyzer 8192 channel, with the following specifications: resolution (FWHM) at 1.33MeV ^{60}Co is 60keV, relative

efficiency at 1.33MeV ^{60}Co is 7.5 %. The sample was placed over the detector for at least 10 hours. The spectra were evaluated with the computer software program GEMEI 2000 based on MCA with spectra display on monitor and by identifying the specific isotope from gamma energy and concentration determined by comparing the net areas of the

photo-peaks with those of standards. Before taking measurements on the gamma spectrometer, the NaI (TI) was calibrated using standard sources. After ensuring the linearity between channels versus energy, measurements have been taken for the samples. Table 1 represents the spectroscopic parameters employed for quantification.

Table 1. Spectroscopic parameters employed for quantification.

Element	Emitter Nuclide	Half life	Gamma ray energy (KeV)	Absolute emission probability of gamma Decay (%)
^{238}U (^{226}Ra)	^{214}Pb	1600 y	241.75	7.43
^{232}Th	^{212}Pb	10.64 h	238.63	44.60
^{40}K	-	1.3×10^9 y	1460.81	10.67

^{226}Ra activity of the samples was determined via its daughter nuclide (^{214}Pb) through the intensity 241.75 keV. ^{232}Th activity of the sample was determined from the daughter (^{212}Pb) through the intensity 238.63 gamma-line and ^{40}K activity determined from the 1460.8 keV emissions gamma-lines.

4.2. Radiation Hazard Indices Calculations

It is justifiable to exploit as many as possible of the known radiation health hazard indices to arrive at a safe conclusion on the health status of an exposed person or environment. The indices estimated in this report are as follows. To represent the activity levels of ^{238}U , ^{232}Th and ^{40}K by a single quantity, which takes into account the radiation hazards associated with each component, a common radiological index has been introduced [6, 7]. This index is called the Radium Equivalent activity (Ra_{eq}) and is mathematically defined by [1]:

$$\text{Ra}_{\text{eq}} (\text{Bq. Kg}^{-1}) = (1.43 \times A_{\text{Th}}) + A_{\text{Ra}} + (0.077 \times A_{\text{K}}) \quad (1)$$

Where A_{Ra} , A_{Th} and A_{K} are the activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K respectively. In the above relation, it has been assumed that 10Bq kg^{-1} of ^{226}Ra , 7Bq kg^{-1} of ^{232}Th and 130Bq kg^{-1} of ^{40}K produce equal gamma dose.

The total air absorbed dose rate (nGy h^{-1}) due to the mean activity concentrations of ^{238}U , ^{232}Th and ^{40}K (Bq kg^{-1}) can be calculated using the formula [8].

$$D (\text{nGy. h}^{-1}) = 0.462A_{\text{Ra}} + 0.621A_{\text{Th}} + 0.0417A_{\text{K}} \quad (2)$$

where A_{Ra} , A_{Th} and A_{K} are the mean activity concentrations of ^{238}U , ^{232}Th and ^{40}K , respectively, in (Bq kg^{-1}). Beck et al. (1972) [9] derived this equation for calculating the absorbed dose rate in air at a height of 1 m above the ground from measured radionuclides concentrations in environmental materials.

To estimate annual effective dose rates, the conversion coefficient from absorbed dose in air to effective dose (0.7Sv Gy^{-1}) and an outdoor occupancy factor (0.2) proposed by UNSCEAR (2000) are used. Therefore, the annual effective dose rate (mSv y^{-1}) was calculated by the formula [1]:

$$\text{Effective dose rate (m Sv y}^{-1}) = D (\text{nGy h}^{-1}) \times 8760 \text{ h} \times 0.8 \times$$

$$0.7 \text{ Sv Gy}^{-1} \times 10^{-6} \quad (3)$$

External hazards arise when radiation from a source external to the body penetrates the body and causes a dose of ionizing radiation. A widely used hazard index (reflecting external exposure) called the external hazard index H_{ex} is defined as follows [1]:

$$H_{\text{ex}} = (A_{\text{Ra}}/370 + A_{\text{Th}}/259 + A_{\text{K}}/4810) \leq 1 \quad (4)$$

In addition to the external hazard index, radon and its short lived progeny are also hazardous to the respiratory organs. The internal exposure due to radon and its daughter progenies is quantified by the internal hazard index H_{in} [1], which is given by the equation:

$$H_{\text{in}} = (A_{\text{Ra}}/185 + A_{\text{Th}}/259 + A_{\text{K}}/4810) \leq 1 \quad (5)$$

The values of the indices (H_{ex} , H_{in}) must be less than unity for the radiation hazard to be negligible [7].

5. Results and Discussion

Various spectra were generated from various samples to identify various radio nuclides present in the samples using gamma spectrometric analysis. Three naturally occurring radionuclides ^{238}U , ^{232}Th and ^{40}K were determined in the soil and water samples. Table 2 represents the results of activities concentrations of the soil samples from Kemessie hot spring. Among the three selected locations at Kemessie, the activity of ^{238}U ranges from 150.3-353.54Bqkg $^{-1}$ with mean value 248.71 \pm 20.8Bqkg $^{-1}$ while ^{232}Th ranges from 22.4-95.6Bqkg $^{-1}$ with mean value 60.1 \pm 3.3 Bqkg $^{-1}$. The activity concentration of ^{40}K was observed to be relatively higher than that of both ^{238}U and ^{232}Th in all the soil-sampling locations studied. Its value ranged from 452.4-682.5 Bqkg $^{-1}$ with mean value 576.46 \pm 39.86 Bqkg $^{-1}$.

Table 2. Activity of the radionuclides from soil samples at Kemessie.

S. No	Location	Sample code	238U BqKg-1	232Th Bq Kg-1	40K BqKg-1
1	At the Origin	KLS-0	353.54 \pm 28.3	95.6 \pm 5.0	682.5 \pm 57.4
2	Location-1	KLS-1	242.3 \pm 20.9	62.3 \pm 2.9	594.5 \pm 40.9
3	Location-2	KLS-2	150.3 \pm 13.4	22.4 \pm 2.1	452.4 \pm 21.3
	Average		248.71 \pm 20.8	60.1 \pm 3.3	576.46 \pm 39.86

This clearly shows that ^{226}Ra and ^{232}Th exhibits higher concentration than the limit ($10\text{-}50\text{BqKg}^{-1}$) prescribed by UNSCEAR (2000) [1].

Table 3 represents activity of radio nuclides from water samples at Kemessie. ^{238}U ranges from 1.21-3.85 with the mean value $2.30 \pm 0.05\text{BqL}^{-1}$ while ^{232}Th ranges from 0.74-

2.65BqL^{-1} with the mean value $1.70 \pm 0.53\text{BqL}^{-1}$. The activity concentration of ^{40}K was observed to be comparatively higher than that of both ^{238}U and ^{232}Th in all the water-sampling locations studied. It ranges from $10.63\text{-}24.33\text{BqL}^{-1}$ with the mean value $17.70 \pm 1.53\text{BqL}^{-1}$.

Table 3. Activity of radionuclides from water samples at Kemessie.

S. No	Location	Sample code	$^{238}\text{U}\text{BqL}^{-1}$	$^{232}\text{Th}\text{BqL}^{-1}$	$^{40}\text{K}\text{BqL}^{-1}$
1	At the Origin	KL _w -0	3.85 ± 0.03	2.65 ± 1.50	24.33 ± 2.15
2	Location-1	KL _w -1	1.85 ± 0.07	1.72 ± 0.07	18.15 ± 1.15
3	Location-2	KL _w -2	1.21 ± 0.05	0.74 ± 0.03	10.63 ± 1.30
	Average		2.30 ± 0.05	1.70 ± 0.53	17.70 ± 1.53

Table 4. Estimated Health Hazard indices of soil at Kemessie.

S. No.	Code	Radium Equivalent BqKg^{-1}	Absorbed dose rate nGyh^{-1}	Internal Hazard Index, H_{in}	External Hazard Index, H_{ex}	Annual effective dose mSvy^{-1}
1	KL _s -0	542.80 ± 39.87	256.16 ± 18.57	2.42	1.45	1.26 ± 0.09
2	KL _s -1	377.17 ± 28.20	175.42 ± 13.16	1.67	1.02	0.86 ± 0.06
3	KL _s -2	230.04 ± 18.47	107.80 ± 8.57	1.03	0.62	0.53 ± 0.04
	Average	383.34 ± 28.85	179.79 ± 13.43	1.70	1.03	0.88 ± 0.06

Table 5. Estimated Health Hazard indices of water at Kemessie.

S. No.	Code	Radium Equivalent BqL^{-1}	Absorbed dose rate nGyh^{-1}	Internal Hazard Index, H_{in}	External Hazard Index, H_{ex}	Annual effective dose mSvy^{-1}
1	K _w -0	9.51 ± 2.34	4.44 ± 1.04	0.036	0.026	0.022
2	KU _w -1	5.71 ± 0.26	2.68 ± 0.12	0.020	0.015	0.013
3	KU _w -2	3.09 ± 0.19	1.46 ± 0.1	0.012	0.008	0.007
	Average	6.10 ± 0.93	2.86 ± 0.42	0.023	0.016	0.014

The overall results show that the ^{238}U , ^{232}Th and ^{40}K are not uniformly distributed in soil, but the radioactivity varies, often greatly, over a distance of some meters. The radium equivalent activities due to the presence of naturally occurring terrestrial radionuclides ^{238}U , ^{232}Th and ^{40}K were calculated based on their activities in soils using equation (1). This is due to higher concentration of ^{238}U (^{226}Ra). Average Req at Kemessie is above 370Bqkg^{-1} which is recommended maximum levels of radium equivalents in soil [6]. The external gamma dose rates were calculated based on their activities in soil and water samples using equation (2). At all the locations external gamma dose rates due to soil are higher than recommended values (58nGyh^{-1}) reported by UNSCEAR (2008) [10]. It is also observed that the index H_{in} due to soil is more than 1 at all study locations of Kemessie site which is above the safety limit recommended by UNSCEAR (2008)[10]. It is observed the respective values are decreasing as we move away from Origin which may be due to other factors.

R_{eq} for water varies from minimum (3.09) BqKg^{-1} to maximum (9.51) BqL^{-1} with the mean value $6.10 \pm 0.93\text{BqL}^{-1}$. The absorbed dose rate varies from minimum (1.46 to 4.44) nGyh^{-1} with the mean value $2.86 \pm 0.42\text{nGyh}^{-1}$ which is within the range of values given in UNSCEAR (2008) report $\{(18\text{-}93)\text{mGyh}^{-1}\}$ [10]. The indices H_{in} , H_{ex} and annual effective dose due to water at all study locations are below the safety limits recommended by UNSCEAR (2008) [10]. However annual effective dose due to soil and water were found to be below the

maximum limit of 1.5mSvy^{-1} recommended by UNSCEAR (2000) [1].

6. Conclusions

The level of natural radioactivity in soil and water collected from Kemessie hot springs has been evaluated using high resolution gamma-ray spectrometry. These data show that the activity concentration of naturally occurring radionuclides in soil samples from Kemessie hot spring are above the world average ranges which are $248.71 \pm 20.8\text{Bqkg}^{-1}$ (7-50) for ^{226}Ra , $60.1 \pm 3.3\text{Bqkg}^{-1}$ (7-50) for ^{232}Th and $576.46 \pm 39.86\text{Bqkg}^{-1}$ (100-700) for ^{40}K . This clearly shows that ^{226}Ra and ^{232}Th exhibits higher concentration than the limit ($10\text{-}50$) prescribed by UNSCEAR (2000). Among the three nuclides, ^{40}K has the highest activity concentration. Similarly activity of the same nuclides for water is $2.30 \pm 0.05\text{BqL}^{-1}$, $1.70 \pm 0.53\text{BqL}^{-1}$ and $17.70 \pm 1.53\text{BqL}^{-1}$. The soil samples show higher concentrations than the water samples. Also radium equivalent activity, total dose rates and external and internal hazard indices for those samples under consideration were calculated. The average radium equivalent activity (R_{eq}), the average absorbed dose rate due to soil samples at Kemessie hot spring are higher than recommended values (370BqKg^{-1} , 58nGh^{-1}) reported by UNSCEAR (2008). The average internal hazard index (H_{in}) and the average external hazard index (H_{ex}) crossed the value one which is above the safety limit. However radium

equivalent activity, total dose rates and external and internal hazard indices for water are within the recommended values by UNSCEAR (2008). It is observed the respective values are decreasing as we move away from Origin which may be due to other factors. However annual effective dose due to soil and water were found to be below the maximum limit of 1.5 mSv y^{-1} recommended by UNSCEAR (2000).

The paper recommends further studies to estimate internal and external doses from other suspected radiological sources to the population in the region. Such studies can further be extended to many hot springs in this country. The total absorbed dose rate from soil samples at the Origin of water and Location-1 of Kemessie are higher than recommended values. The data derived from this study represent a baseline database of dose levels that can serve as a reference point for future studies.

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