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DETERMINATION OF NATURAL RADIOACTIVITY LEVELS OF ^{238}U, ^{232}TH AND ^{40}K IN ROCKS AROUND GONDAR CITY, NORTHWEST ETHIOPIA

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ABSRACT

The aim of the present work was to assess the terrestrial radionuclides distribution and their radiological health related risks in rocks around Gondar city using NaI (Tl) detector gamma-ray spectrometry. The samples were dried, pulverized, homogenized and then on average 300g from each rocks were packed in polyethylene containers. The polyethylene containers were tightly sealed for 4 weeks until the parent radionuclides ^{238}U, ^{232}Th and their daughter products in the rock samples attained in secular equilibrium. Based on the characteristic spectral peaks produced by daughter nuclides of ^{208}Tl (in $^{23}8U$ series) at 2614.4kev, ^{214}Bi (in ^{232}Th) at 1764.4kev and the singly decaying ^{40}K at 1460.8kev, the activity concentrations were determined for ^{226}Ra (^{238}U) from 18.06 ± 5.59 $BqKg^{-1}$ to 27.05 ± 4.60 $BqKg^{-1}$ with an average value of 21.36 $BqKg^{-1}$; for ^{232}Th from 54.57 ± 1.42 $BqKg^{-1}$ to 106.32 ± 1.65 $BqKg^{-1}$ with mean activity 59.60 ± 2.34 $BqKg^{-1}$ and for ^{40}K from 152.68 ± 14.10 $BqKg^{-1}$ to 873.01 ± 13.08 $BqKg^{-1}$. The calculated absorbed dose rate and the annual effective dose rates were varied from 49.98±1.99 to102.26±2.84 and from 0.061to 0.126 μSvy $^{-1}$ respectively. The result of the finding showed that the activity concentration, radium equivalent activity, absorbed dose rate, annual effective equivalent dose, and external and internal hazard indices of the rocks were obtained below the world recommended values. However, higher activity concentrations of ^{232}Th and ^{40}K above the world range and mean values were absorbed in rock areas of Dabecha Rufauel and Gondar air port.

Key words: Natural radionuclides; gamma-ray spectrometer, activity concentration; radiation hazard indices

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Introduction

The sources of background radiation from radioactive materials present in the atmosphere, marine and terrestrial environments (Gafver& Faerevik, 2004) can be categorized into three categories depending on their origins: primordial, Cosmogenic and Anthropogenic (UNSCEAR, 1993). About 98 % of the background radiation in the natural environment is usually contributed from cosmogenic radionuclides such as ^{3}H, ^{7}Be, ^{14}C and ^{22}Na created in the earth's atmosphere due to the interaction of high energy cosmic rays from the outer space with the atomic nuclei in the atmosphere (NCRP, 1975, Malain et al., 2012, Al-Beddri et al., 2014) and due to primordial radioactive elements that were produced since the creation of earth. In particular, long-lived naturally occurring radioactive materials (NORMs) such as uranium-238 (^{238}U), uranium-235 (^{235}U) and thorium-232 (^{232}Th) and their serial decays, and the singly decay of potassium-40 (^{40}K) represent for the major cause of radiation field in the natural environment (IAEA, 2003).

On the other hand, radiation emissions due to anthropogenic (or manmade) radionuclides that base from nuclear power plants, medical centers, industrial plants, and research facilities are very small under normal circumstances. However, sometimes unwise or inappropriate utilizing of such radiation sources can be devastating and could be the cause of radiological hazards to human being and all other living things in the surroundings. Basically, the major ways by which human exposed to irradiation are radiation from sources outside the body (external exposure), radionuclides that are ingested through consumption of food and water or as inhaled in radioactive gases (internal exposure)(UNSCEAR, 2000).

In general short and long lived radioactive isotopes are widely distributed and present at different trace levels in various geological and biological formations of such as soils, rocks, building materials, ground water, air, food items, plants and tissue of living things and give rise to external and internal exposures due to gamma radiation. Thus, in managing the effects of radiation exposure due to both natural and man-made sources, the knowledge of radionuclides distribution and the radiation levels in the environment we live in is an indisputable fact (Avwiri et al., 2012).

This is, therefore; the present study was carried out with the objective to assess and radiation levels of rocks around Gondar city using the technique of NaI (Tl) detector gamma-ray

spectrometer as these rocks are commonly used for construction purpose in the city. According to (Fasae, 2013) building materials such as soil, cements and rocks can cause substantial radiation exposure if the materials contain elevated levels of natural radionuclides compared with that of undisturbed earth crust. In this assessment the radiological effects of the rocks in terms of radium equivalent activities, absorbed dose rate, external and internal radiation hazard indexes were determined and compared whether the activity levels are within the scope of various worldwide permissible or acceptable values as stated in UNSCEAR-1988, ICRP-1990, UNSCEAR- 2000,) and also with other reported values. The result of this finding can be served as a base line value of activity concentration for further assessing the radiation pollution of environment.

MATERIALS AND METHOD

Samples collection and preparation

A total of five rock samples were collected from five different locations around Gondar city in Northwest Ethiopia. The rocks of the selected locations are commonly used for house and road constructions in the city in the form of fine gravels of different sizes and the form of rocks as they are naturally as shown in figure 1. About 400g of rock sample from each sampling sites was collected, packed in polythene and labeled SR1, SR2, SR3, SR4 and SR5 for easy identification as shown in table 1. The samples were sun dried in an open air for a week. After the samples were dried they were crushed, pulverized and passed through a 100 mesh to get a very fine powder in a homogeneous state. The powered form of the rock samples was heated again in a standard oven at 100 ^{o}C for 24h to avoid moisture contents.

Table 1: Sample code, sampling site and GPS coordinate of the study areas

Sample code	Name sampling site	GPS Coordinate
SR1	Arno Seneba	Lat $12^{0}21'45.62''$N, Long. $37^{0}33'55.19''$E
SR2	Dngay Gores	Lat $12^{0}34'18.84''$N, Long. $37^{0}25'31.40''$E
SR3	Srur Damaza	Lat $12^{0}33'0.18''$N, Long. $37^{0}23'8.06''$E
SR4	Dabecha Rufauel	Lat $12^{0}32'29.80''$N, Long. $37^{0}21'54.94''$E

SR5	Gondar Airport	Lat $12^0 31' 25.39'' N$, Long. $37^0 25' 43.58'' E$

(a) **(b)**

Figure 1 Crashed rocks of in the form of gravel for building and road constructions collected from (a) Gondar airport and (b)Suru Damaza sampling sites

Finally, a representative mass on average 300g from each sample was weighed and packed in a cylindrical plastic container of 170 cm^3 (i.e 7cm height by 6 cm diameter) volume (Umar et al., 2012). The polyethylene container was tightly sealed and wrapped using thick collotype and keep for more than 4 weeks until the radionuclides ^{238}U, ^{232}Th and their daughter products in the rock samples attain secular equilibrium.

Gamma-ray Spectrometric Analysis

The activity concentration levels of ^{238}U, ^{232}Th and ^{40}K in the rock samples were measured using the gamma-ray spectrometric counting system that consisted of NaI (Tl) scintillation detector with dimensions of 7.62 cm by 7.62 cm, preamplifier, amplifier and a Canberra multichannel analyzer (MCA) connected via the associated electronics. The scintillation detector was housed in a 6 cm thick lead shielding and lined with cadmium and copper sheets (CERT Manual, 1999, Umar et al., 2012) to reduce background radiation entering it from the laboratory of measurement. For accurate measurement, the gamma-ray spectrometric system was calibrated and tested its linearity using Geological Certified Reference Material (GCRM) samples known as potassium (RGK-1), Thorium (RGTh-1) and Uranium (RGU-1) obtained from International Atomic energy Agency (IAEA), Vienna (IAEA 1987). Equal quantities of these standard

sources of known energy were mixed together, packed for about a month to attain a secular equilibrium. As per the established protocol of the laboratory (at the Center for Energy Research and Training (CERT), Zaria, Nigeria), the configuration and geometry of the system was maintained throughout the analysis.

Specific Activity Concentration (A)

After four weeks later each sample of rock including the GCRM sample was placed on the detector surface and counted for 29000 seconds. The net numbers of counts for each photo peak were obtained by subtracting the background counts from the total counts in the same photo peak using an advanced ORTEC made MAESTRO multi-channel analyzer (MCA) emulation software. The MAESTRO program is a software packages that was developed for data acquisition, storage display and data analysis of the acquired gamma spectra (Auwal et al., 2010). The net area under each photo-peak of counts of the spectrum was determined for each of the radionuclide of interest in the rock sample could be calculated by the following expression (UNSCEAR, 2000).

$$A = \frac{C}{\varepsilon(E) \times I_{\gamma}(E) \times m} \quad (1)$$

Where A is activity of the radionuclide of interest; C is the net peak count rate ($C = \frac{PA_{totalcount} - PA_{bgrcount}}{t_c}$) in count per second, $PA_{total\ count}$ is the total peak area count, $PA_{bgr\ count}$ is the background peak area count, t_c is the counting time in second, ε (E_γ) is the efficiency of the detector at energy E, I_γ (E_γ) is the number of gamma per disintegration of the radionuclides of interest for transmission at energy E and m is the mass of the sample under investigation.

However, as per the protocol established in CERT, Zaria, Nigeria, the activity concentrations (A in $BqKg^{-1}$) of the natural radionuclides (^{40}K, ^{226}Ra & ^{232}Th) in the rock samples were determined by dividing the measured net count rates (C in count per second) of the photo-peak in equation (1) to the calibration factors K as shown in equation (2).

$$A(BgKg^{-1}) = \frac{C}{K} \quad (2)$$

Where, K is a constant calibration factor representing the denominator expression in equation (1) for each radionuclide determined by the detector's efficiency at constant geometry. Thus the K calibration factors for ^{40}K, ^{226}Ra and ^{232}Th have been determined as 0.000643, 0.000863 and 0.000877 respectively in CERT laboratory.

The specific activity concentration of ^{40}K in the rocks was determined at gamma transmission line 1460.8 kev. About 98% of the external gamma radiation from ^{238}U series is derived by ^{226}R subseries (Kinsara et al., 2014). This implies that the activity concentration of ^{226}Ra (in ^{238}U decay series) was measured at the secular equilibrium with decay product of ^{214}Bi at gamma transmission lines 351.93 and 1764.49 keV, and with decay product of ^{214}Pb at gamma transmission line 583.19 keV. Similarly, the activity concentration of ^{232}Th was also measured at the secular equilibrium with decay product ^{208}T at gamma transmission lines of 510, 583.19 kev and 2614.4 keV (El-Mamoney et al., 2004) as shown figure 1. Since radium (Ra) and its daughter products produces 85% of the radiological effects in the Uranium series, the activity concentration of ^{238}U was represented by activity concentration of ^{226}Ra. The typical gamma spectrum as shown in figure 2 for the rock of Suru Damaza is an illustrative example of characteristic spectral peaks produced for ^{238}U, ^{232}Th and ^{40}K.

Radium Equivalent Activity (Ra_{eq})

Since the distribution of natural radionuclides cannot be uniform throughout in soils, rocks and building materials, radium equivalent activity (Ra_{eq}) was introduced to define the uniformity in respect to exposure of radiation arising from ^{238}U, ^{232}Th and ^{40}K and widely used index in the assessment of radiological hazards. It is a weighted sum of the activities of ^{238}U, ^{232}Th and ^{40}K based on the estimation that 370 BqKg^{-1} of ^{226}Ra, 260 BqKg^{-1} of ^{232}Th and 4810 BqKg^{-1} of ^{40}K produce the same gamma ray dose rate (UNSCER, 2000). Radium equivalent activity is calculated using the following equation (Beretka and Mathew, 1985, Oyebanjo et al., 2012).

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \leq 370 \quad (3)$$

Where, A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K respectively in BqKg^{-1}. The maximum value of Ra_{eq} in building materials should not be beyond 370 BqKg^{-1} for radiological safety (Abdulrahman et al., 2013).

Absorbed dose Rate in Air (D)

The assessment of radiological hazard due to exposure of external terrestrial gamma-ray radiation from ^{226}Ra, ^{232}Th and ^{40}K in rocks, soils and building materials can be measured using Absorbed Dose Rate in Air (ADRA) at about 1m above the ground. Based on the conversion factors 0.462, 0.604 and 0.0417 for ^{226}Ra, ^{232}Th and ^{40}K respectively as set by UNSCER, (2000), the activity concentration of each natural radionuclide can be converted to absorbed dose rate in air (Oyebanjo et al., 2012) using in equation (4):

$$D(nGyh^{-1}) = 0.462A_{Ra} + 0.604A_{Th} + 0.0417A_{K} \quad (4)$$

Where, D is the total absorbed dose rate in air in nGyh^{-1} and A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in BqKg^{-1} respectively.

Radiological hazard indices

The radiological effects due exposure of radiation arising from natural radionuclides of ^{226}Ra, ^{232}Th and ^{40}K can also be evaluated through external hazard radiation index (H_{ex} and internal hazard radiation index (H_{in}). An external or internal hazard index which is greater than unity can be significantly radiation hazard to human body. Thus, external and internal hazard radiation indices can be estimated using radiation index expressions given in equations (5) and (6) as follow. Similar, building materials such as rocks, bricks, cements or concrete which are used massively for building construction can be the sources of indoor radiation due to the accumulation of naturally occurring radioactive elements ^{238}U, ^{232}Th and ^{40}K. Thus, activity concentration index ($I_{\gamma r}$) of these radioactive elements can be calculated using the following expression in equation (7) (Murugesan et al. 2011).

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (5)$$

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (6)$$

$$I_{\gamma r} = \frac{A_{Ra}}{150} + \frac{A_{Th}}{100} + \frac{A_K}{1500} \quad (7)$$

Where, A_{Ra}, A_{Th} and A_K are the specific activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in $BqKg^{-1}$ respectively.

Annual Effective Radiation Dose Rate

The total annual effective dose rate due to the natural activity of ^{226}Ra, ^{232}Th and ^{40}K in the rocks was calculated using equation (8) as follow

$$E\ (mSvy^{-1}) = D(nGyh^{-1}) \times 87640h \times 0.2 \times 0.7 SvGy^{-1} \times 10^{-6} \quad (8)$$

Where, E is the total effective dose rate ($mSvy^{-1}$), D is the absorbed dose rate in air in $nGyh^{-1}$, 0.2 outdoor occupancy factors, 0.7 $SvGy^{-1}$ is a conversion factor from absorbed dose in air to effective dose received by adults as recommended by UNSCEAR, 2000 and 87640h is the time in hours in a year.

RESULTS AND DICUSSIONS

The activity concentrations of terrestrial radionuclides ^{238}U, ^{232}Th and ^{40}K were investigated in the rock samples. In so doing the accuracy, precision and energy calibration of the gamma spectrometer were also evaluated using the activity concentrations of RGK -1, RGU-1 and RGTH-1 with same procedure as done for the analysis of rocks samples.

Table 2: Activity concentrations of natural radionuclides in this work compared with the activity concentration of IAEA certified.

Sam. Code	Activity measured in this work	Mass concentration in this work	Activity IAEA certified	Mass concentration IAEA certified	Activity STD (%)
RGU-1	4971.95±4.90	403.0±0.4	4940±30	400±2	0.6
RGTh-1	2937.286±28.19	723.5±7.0	3250±40	800±10	9.6
RGK-1	13450.55±30.48	(43±0.1)%	14000±400	(44.8±0.03)%	3.96

From the results obtained it was observed that the measured activity concentrations of the RGK - 1, RGU-1 and RGTH-1 in this work were in good agreement with IAEA certified values as presented in table 2. Similarly the percentage relative standard deviation (RSD %) between the IAEA certified values and the measured values of this work was obtained less than 10 % which indicated the gamma rays spectrometry used in this study was well calibrated.

The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the rocks were determined by equations (2) and the results obtained are presented in table 3. Along all rock samples, the measured activity concentration of ^{226}Ra was found in the range between 18.06 ± 5.59 BqKg^{-1} and 27.05 ± 4.60 BqKg^{-1} with a mean value of 21.36 BqKg^{-1}. The range and mean activity concentrations of ^{226}Ra were obtained below the worldwide activity concentration range 17-60 BqKg^{-1} and mean concentration of 35 BqKg^{-1} as reported in UNSCEAR, 2000.

Table 3: Activity concentrations of ^{226}Ra (in ^{238}U series), ^{232}Th and ^{40}K in different rock samples

Sample ID	Location	Specific Activity (in BqKg^{-1})		
		^{226}Ra	^{232}Th	^{40}K
SR_1	Arno Seneba	19.38±2.80	56.66±0.31	160.99±12.28
SR_2	Dngay Gores	18.90±4.19	57.00±1.21	152.68±14.10
SR_3	Srur Damaza	18.06±5.59	54.57±1.42	231.94±12.28
SR_4	Dabecha Rufauel	23.42±2.40	84.14±1.26	873.01±13.08
SR5	Gondar Airport	27.05±4.60	106.32±1.65	226.95±10.83
Mean this work		21.36	59.60	282±12.51
Range this work		18.06 -27.05	54.57-1 06.32	152.68-873.01
World Median		35	30	400
Worldwide Range		17-60	11-64	140-850

The activity concentration of ^{232}Th was varied from 54.57 ± 1.42 to 106.32 ± 1.65 $BqKg^{-1}$ with a mean activity of 59.60 ± 2.34 $BqKg^{-1}$. Even though the activity of ^{232}Th in rocks of Arno Seneba, Dngay Gores and Surur Dmaza were obtained within worldwide permissible range but in rocks of Dabecha Rufauel and Gondar Airport, the activity concentrations of ^{232}Th were found above world range (UNSCEAR 2000) as shown in table 3.

Table 4: Radium equivalent (Ra_{eq}), Absorbed dose rate (D in $nGyh^{-1}$), External hazard index (H_{ex}) and equivalent (in $mSvy^{-1}$) and Annual Effective dose equivalent (E in $mSvy^{-1}$)

Sample. ID	Ra_{eq}	D	H_{ex}	H_{in}	E	$I_{\gamma r}$
Arno Seneba	112.32	49.98±1.99	0.305±0.053	0.358±0.058	0.061	0.803
Dngay Gores	57.69	52.93±3.26	0.588±0.019	0.639±0.03	0.065	0.805
Srur Dmaza	119.32	54.22±3.98	0.308±0.042	0.357±0.072	0.067	0.821
Dabecha Rufauel	219.77	102.26±2.84	0.570±0.015	0.634±0.021	0.126	1.579
Arno Seneba	114.47	91.05±3.57	0.532±0.20	0.604±0.033	0.112	1.801
Range In this work	57.69 - 219.77	49.98±1.99 - 102.26±2.84	0.305-0.570	0.358-0.639	0.061-0.126	0.803-1.801
Mean In this work	124.45	70.09±3.13	0.522±0.13	0.518±0.04	0.086	1.162
Worldwide Range	-	18-93	-	-	20-100	-
World Recommended value	< 370	55	< 1	< 1	1	≤ 3

Basically natural potassium has three main isotopes known as ^{39}K, ^{40}K and ^{41}K. Among the three isotopes, ^{40}K is the only natural radioactive isotope with abundance of 0.012%. The activity

concentration of ^{40}K in the present study was found in the range from 152.68 ± 14.10 to 873.01 ± 13.08 $BqKg^{-1}$ with a mean value of 282±12.51 $BqKg^{-1}$.

As can be seen in table 3 and Figure 3 as well , the highest activity concentration of ^{40}K was recorded in the rock of Debecha Rufauel (873.01 ± 13.08 $BqKg^{-1}$), which is beyond the worldwide mean and range values. This indicates that the rock of Debecha Rufauel accumulates high contents of ^{40}K.

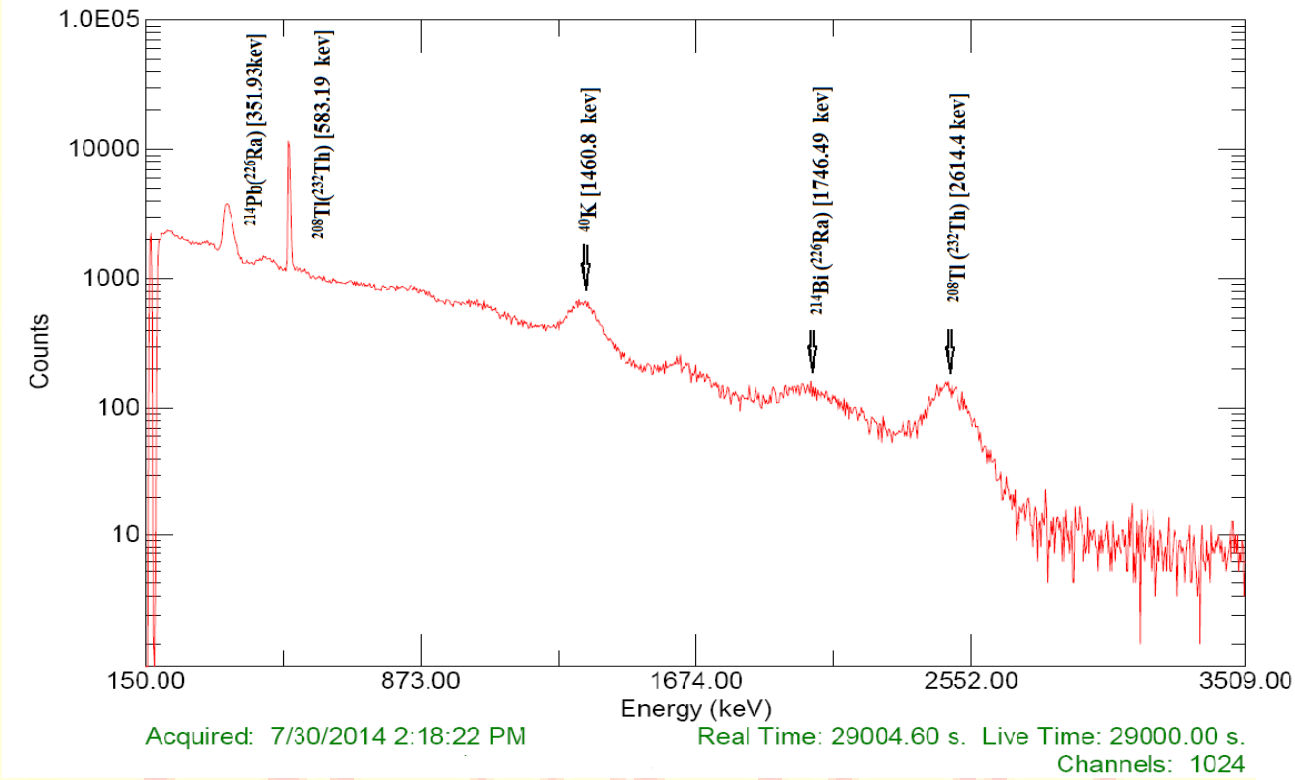

Figure 2 the typical gamma-ray spectrum obtained by the NaI (Tl) detector gamma spectrometry for rock of Surur Damaza area

The estimated radium equivalent (Ra_{eq}) activity of ^{238}U, ^{232}Th and ^{40}K in all rocks of the present study was varied from 57.69 to 219.77 $BqKg^{-1}$ with the mean value of 124.45 $BqKg^{-1}$ as shown in table 4 which was obtained less than 370 $BqKg^{-1}$ as recommended by UNSCER, 2000. With regard to the total terrestrial gamma dose rate 1m above the ground, it was obtained in the range between 49.98±1.99 and 102.26 ±2.84 $nGyh^{-1}$ with a mean value 70.09 ± 3.13 $nGyh^{-1}$. A maximum dose rate 102.26 ± 2.84 $nGyh^{-1}$ which is above the worldwide range (18-93 $nGyh^{-1}$)

was observed in rock of Debecha Rufauel due to the higher activity concentrations of ^{232}Th and ^{40}K as shown in table 3.

The annual effective equivalent dose rate was found in the range from 0.061 to 0.126 SvGy^{-1} along all rocks which is below world average dose rate 1 mSvGy^{-1}. According to ICRP, 1990, the maximum permissible annual effective equivalent dose for general public is 1 mSvGy^{-1}. Similarly, as tabulated in table 4 the calculated external and internal radiation hazard indices for all rocks were obtained less than unity (< 1).

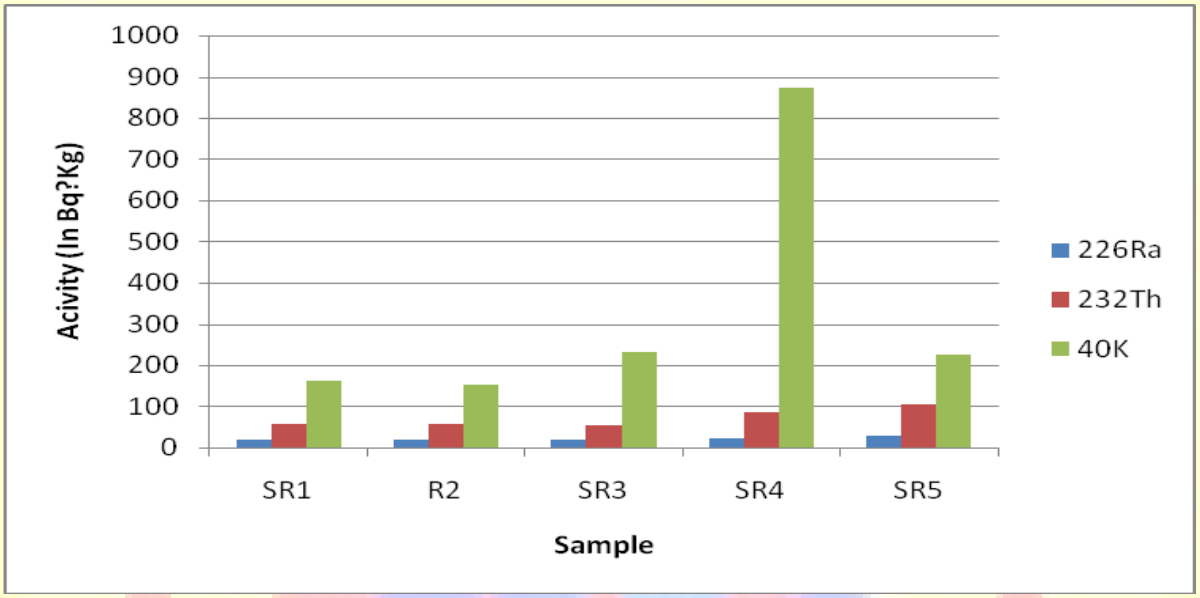

Figure 3: A graph of activity concentration distribution of ^{226}Ra (^{238}U), ^{232}Th and ^{40}K in rock samples

The estimated activity concentration index was also obtained between 0.803 and 1.801 which is below recommend values $I_{\gamma r} \leq 3$. The activity concentration index $I_{\gamma r} \leq 1$ corresponds to 0.3mSvy^{-1}and $I\gamma r \leq 3$ corresponds to 1mSvy^{-1} indicating that the terrestrial gamma radiation emanating from the natural radionuclides in rocks used for building construction is as such insignificant to cause radiological related health risk.

Conclusion

In the present investigation, the activity concentrations of terrestrial radionuclides ^{238}U (^{226}Ra), ^{232}Th and ^{40}K and their associated radiological hazards were assessed in rocks around Gondar city in Northwest Ethiopia by means of NaI (Tl) detector gamma spectrometer. The results of the study showed that activity concentration of ^{238}U (^{226}Ra) in all rocks was obtained lower than worldwide range and median. The activity concentration of ^{232}Th was obtained higher than world median in all rocks. However, even though the range of ^{232}Th in rocks of Dabecha Rufauel and Gondar airport was found above the worldwide range, in rocks of Arno Snaba, Dngy gores, Surur Damaza, the range of ^{232}Th was measured lower than the worldwide range. Higher activity concentration of ^{40}k above the worldwide range and median was observed only in rock of Dabecha Rufauel while in all other rocks the activity of ^{40}k was obtained below the worldwide range and median.

With regard to the radiological hazards, the radium equivalent activity, absorbed dose rate, annual effective dose rate, active concentration index, the external and internal radiation hazard indices of all rocks were obtained below the world recommended values except that of the absorbed dose rate in rocks of Dabecha Rufauel and Gondar airport obtained above the world recommended values. In general, the result of the find indicates that rocks of the study areas are still in the normal radiation level and do not pose radiological health related risks with the exception of the rock of Dabecha Rufauel which scored high activity of ^{40}K above the worldwide median and range.

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