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Assessment of Natural Radioactivity Level and Associated Radiological Risk in Ground-water samples from Hakim Gara Quarry Area, Ethiopia

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Abstract

Radiological hazards could arise from high amount of radionuclides in food and water when ingested unless investigation is conducted. Radioactivity levels in water needs investigation especially if the groundwater supply is used for domestic consumption. In this study, radioactivity levels of long-lived naturally occurring Radionuclides ^{226}Ra, ^{232}Th and ^{40}K were measured from water samples of six groundwater collected from areas closer to Harar town around Hakim Gara quarrying site. Gamma spectroscopic system of High Purity Germanium (HPGe) detector was employed to determine radioactivity level and to assess the radiological impacts imposed to the public. The measured values were ranging from 1.9 Bq.L^{-1}to 3.0 Bq.L^{-1} with mean of 2.7±0.4 Bq.L^{-1} for ^{238}U, from 7.7 Bq.L^{-1} to 9.3 Bq.L^{-1} with mean of 8.1±0.9 Bq.L^{-1} for ^{232}Th and from 71.2 Bq.L^{-1} to 104.9 Bq.L^{-1} with mean of 87.9±11.4 Bq.L^{-1} for ^{40}K. The radiological health risk assessments H_{ex}, H_{in}, D_{γ}, Annual Effective Dose Equivalent (AEDE) and Excess Lifetime Cancer Risk (ELCR) were calculated and their mean values were 0.057±0.002 for H_{ex}, 0.064±0.003 for H_{in}, 9.76±0.35 nGyh^{-1} for D_{γ}, 0.06±0.002 mSv.y^{-1} for AEDE and 0.021±0.008 x10^{-3} for ELCR. From radiological point of view, based on the findings of measurements analyzed in samples of this study, the groundwater of the study area is safe to use for domestic purpose. The results obtained in this study could contribute as input for the national database and enable to conduct further studies in the future.

1. Introduction

Due to the occurrence and distribution of natural radionuclides with different types and concentrations everywhere, radioactivity and radiation exist in different types and amounts. A review by Zapecza and Szabo (1986) indicated that radionuclides are found as trace elements in most rocks and soils and are formed mainly by the radioactive decay of long-lived parent elements ^{238}U and ^{232}Th. A study by Van *et al.* (2023) indicates that some radionuclides in the deep soil and rock can be released into the surrounding environments such as surface soil, surface water, groundwater and air, mining activities and ultimately get into plants and animals (An et al., 2023).

The occurrence and distribution of the natural radionuclides in groundwater depends primarily by the local geology and geochemistry of the bed rock (Zapecza & Szabo, 1986; Walencik *et al.*,

2010). The dominant radionuclides in ground and surface water that can pose a potential health hazard under natural conditions are ^{210}Po, ^{210}Pb, ^{210}Bi, ^{222}Rn, ^{226}Ra and ^{228}Ra in the ^{232}Th series (Atlas & Radiation, 2020). A review by Otto and Szabo (1986) has also shown that ^{238}U with its daughters (^{234}U, ^{226}Ra, ^{222}Rn) and ^{232}Th with its daughter (^{228}Ra) are the most common radionuclides in groundwater. The health hazards from uranium isotopes are due more to chemo- than radiotoxicity. Other isotopes in these series are generally not present in groundwater in significant quantities because of low presence, low solubility in water and very short half-lives (Atlas & Radiation, 2020).

The impact of radiation emitted from radionuclides on human beings is great. As it is noted in the European Atlas of Natural Radiation, the activity of ^{40}K in the environment is not considered as a hazard since it is an essential element in human metabolism, and K metabolism is strongly self-regulated (Atlas & Radiation, 2020). The World Health Organization has mentioned in its guideline to water quality (WHO, 2011) that except in extreme conditions, the risk of radiation dose from the ingestion of radionuclides in drinking-water is much lower than risks from microorganisms and chemicals that could be present in water (Herschy, 2012).

It is also estimated by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) that the total radiation exposure from all natural and anthropogenic sources is 3 mSvy^{-1} of which 2.4 mSvy^{-1} is from naturally occurring sources of radiation (UNSCEAR, 2008). Out of the 2.4 mSvy^{-1} dose, water and food contribute about 0.3 mSvy^{-1}. Of the 0.3 mSvy^{-1} dose received by people from ingestion of naturally occurring radionuclides, drinking water contributes 0.05 mSv (WHO, 2022). The WHO guideline also recommends maximum value of 0.1 mSv/y as safe water for human consumption (Kinahan et al., 2020).

One of the most essential natural resources for living organisms is water. Around 70% of the Earth's surface is covered with water. A study by UNESCO has shown that, from this total of water body on earth, about 2.5% is fresh water and found in the ground (IAEA, 2011). With population increase freshwater withdrawals from surface water and groundwater sources have also increased (Alfstad et al., 2024).

Increasing the excavation of surfaces in the terrestrial could easily increase the mobility of the exposed radionuclides by wind, flood and other mechanisms. Human activities of removal of the earth surface for different purposes such as stone quarrying could enhance the mobility of the naturally occurring radionuclides to other nearby surface or groundwater bodies. Radionuclides have the property of solubility and potential to migrate with the help of moving water within and on the surface of soil. This mobility of radionuclides on the surface enables them to mix with surface water or infiltrate into the groundwater. This could lead to radiation exposure by consuming water from the vicinity of quarrying activity. Hence, there is a need for assessing the radioactivity concentrations from naturally occurring radionuclides that could exist in groundwater. The aim of this study was thus to identify gamma radiation emitting long-lived naturally occurring radionuclides in surface and groundwater and to determine the level of radioactivity concentration and assess the associated radiological risks while using water for domestic purposes. The results obtained from this study could be an input to the national baseline data.

2. Materials and Methods

2.1. Description of the Study area

Quarrying areas are generally hilly or mountainous and it is difficult to dig wells to find groundwater. However, there is a possibility of contamination of ground or surface water lying at lower areas due to movement of radionuclides with runoff. The water samples used in this investigation were obtained from mount Hakim Gara quarry sites located in Harari region, Ethiopia. Locations of the sampling sites are shown in Fig. 1.

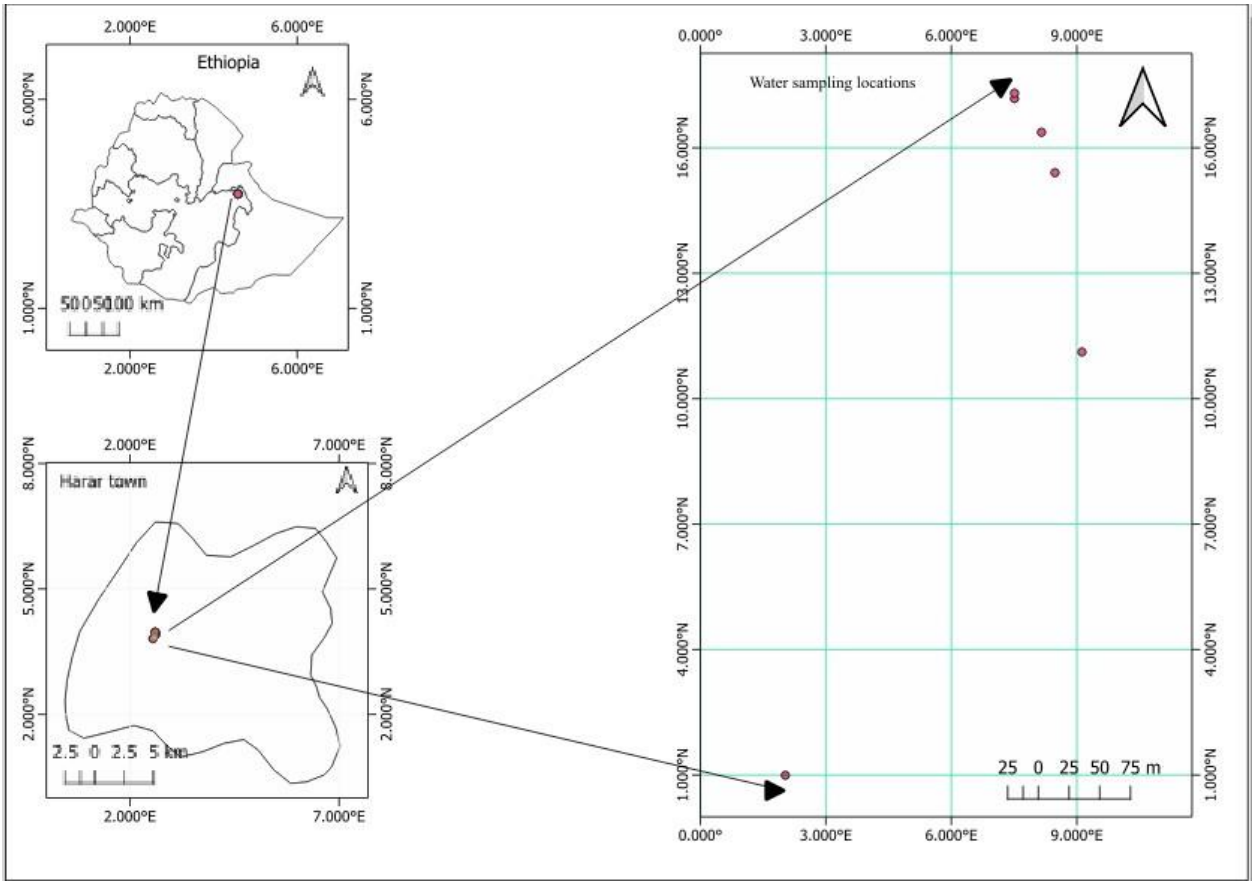

Figure 1. Map of study area location

The water wells were found at the bottom of the hills and mountains found on the edges of the Harar town, capital of the Harari region. The samples were taken during the months of December and February of 2022. The geographical coordinates of groundwater locations obtained using Global Positioning System (Model: Garmin - GPS 72H) are shown in Table 1. The geological formation of the area underlay large deposits of limestone (Haileyesus Walle et al., 2000).

Table 1. Geographical coordinates of water sampling locations.

Sample Code	Lat. (N)	Long. (E)	Alt. (m)
TS1	9.3036	42.1316	1874.1
TS2	9.30493	42.1314	1859
TS3	9.30523	42.1313	1856
TS4	9.30548	42.1311	1856.2
TS5	9.30552	42.1311	1855.6
TS6	9.30047	42.1294	1907.4
Ave.			1868.05
stdev			18.74

2.2. Sampling and Preparation of Samples

2.2.1. Water Sampling

From six water wells, a total of six water samples in each round (first round February 2022 and second round November 2022) were collected randomly. New and clean containers were used for collection. The samples were then transported to the Radio-analytical laboratory of Ethiopian Technology Authority - ETA (formerly known by Ethiopian Radiation Protection Authority – ERPA) for the measurement and analysis of gamma emitting radionuclides from the samples.

2.2.2. Sample Preparation

In the laboratory the water samples were transferred to Marinelli beakers (model 538G-E). The edges of the top cover of the samples were sealed

using Teflon tape by leaving a small space to prevent expansion. Each beaker was labeled and placed on shelf to wait for more than thirty days to achieve secular equilibrium due to the constant production of new radon by the presence of ^{226}Ra in the samples (Atlas & Radiation, 2020).

2.3. Measurements and Data Collection

Measurement of radioactivity concentration from water samples were performed using High Purity Germanium (HPGe) gamma-ray detecting system (Model B13010). The system consists of an n-type coaxial high-purity germanium detector having a crystal of diameter 72.5 mm with thickness of 72.5 mm and Active Volume of 239 cm³ (Canberra, Czech Republic). The relative efficiency, energy resolution, FWHM and peak-to-Compton ratio of the detector are (70%, 1.90 keV (FWHM) at 1333 KeV, 1.05 KeV (FWHM) at 122 keV and 70:1 at 1332.5 KeV), respectively. The detector is surrounded by copper plate of 6 mm thickness. Lead of thickness 4 cm and Cadmium shield of 1 mm thickness were used to reduce fluorescence X-rays to minimize the background gamma radiation emitted from the surrounding building materials and cosmic rays.

Prior to counting of gamma radiation in the water samples, efficiency calibration of the measuring system was carried out using calibration sources having standard mixed radionuclides (prepared by Czech Metrology Institute). The background level of gamma radiation in the laboratory was measured for 56 hours before any measurement on the samples and its value was deducted from gamma radiation measured of each sample. The water samples in each container were placed in the HPGe gamma spectrometry system and measurement of water samples were carried out by setting 16 hours of counting time for each sample. The data of each sample was collected from the energy spectrum formed using the Genie 2000 software.

2.4. Data Analysis

The data analysis of the water sample in each container was performed based on the spectra obtained from each sample. Determination of specific activity of radionuclides of interest (^{238}U, ^{232}Th and ^{40}K) in the water samples were performed by taking their activity from the photopeaks in the spectra of their daughter radionuclides obtained by the data acquisition and analysis software, Genie 2000 utilized by the spectroscopy system.

2.4.1. Calculation of activity concentration

The radioactivity concentration C (in Bq.kg^{-1}) of radionuclides were determined for the different gamma transitions using (Altıkulaç et al., 2015).

$$C = \frac{N_{cps}}{\varepsilon(E_i) I_i t_c V_s} \quad (1)$$

Where; N_{cps} is the net counts per second, $\varepsilon(E_i)$ is the peak efficiency of the detector at energy E_i, I_i is the probability of gamma emission, t_c is counting time and V_s is volume of the sample. The mean detectable Activity (MDA) was calculated using

$$MDA = \frac{LLD}{\varepsilon(E_i) I_i V_s}. \quad (2)$$

The lower limit detection (*LLD)* is given by Curie (1968) equation

$$LLD = 2.61 + 4.65\sqrt{B}$$

Where B is the background count in the spectrum. The other variables in Eq. 2 are as explained in Eq. 1.

2.4.2. Radiation hazard assessment

Hazard indices is determined using radium equivalent activity (Ra_{eq}), which is related to external dose from gamma radiation. It helps to assess the probability of radiological hazard occurrence. Internal dose from alpha radiation that originates from ^{222}Rn and its progeny or decay of radium. It serves as a relative measure of the gamma ray exposure associated with Ra, Th and K. Ra_{eq} is obtained from the concentrations of the three radionuclides as (Barbosa da Silva et al., 2024),

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_K \quad (3)$$

External hazard index (H_{ex})

Gamma radiation emitted from cosmic and the terrestrial sources are of external radiation that contribute to the radiation dose (Wouter Schroeyers, 2017). But the main contribution to this dose depends on the radionuclide concentration in the water. The External hazard indices (Hex) for water sample is calculated as (Abdullah et al., 2019)

$$H_{ex} = \frac{A_U}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810}. \quad (4)$$

A_U, A_{Th} and A_K are activity concentrations of ^{238}U, ^{232}Th and ^{40}K, respectively.

Internal hazard index (H_{in})

The internal hazard index (H_{in}) for the water sample is calculated as (Ademila, 2018),

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810}. \quad (5)$$

A_{Ra} is activity concentration of ^{226}Ra and explanations of the others are as explained in Eq. 4.

Absorbed Gamma Dose Rate (D_γ)

Radiation energy could be absorbed by a body while they encounter the body. The absorbed energy of the irradiated body per unit mass of the body determine the absorbed dose. Activity concentrations of radionuclides measured are converted to doses by applying dose conversion factors 0.462, 0.604 and 0.0417 for A_u, A_{Th} and A_K, respectively and the maximum permissible limit is 55 $nGyh^{-1}$ (UNSCEAR, 2000). Assessment of radiological impact of radionuclides in the measured water samples, needed the estimation of γ- radiation doses, which is calculated as (Auwal, 2020),

$$D_\gamma(nGyh^{-1}) = 0.462A_U + 0.604A_{Th} + 0.0417A_K. \quad (6)$$

D_γ is the absorbed gamma dose rates, A_U, A_{Th}, and A_K are activity concentrations of ^{238}U, ^{232}Th, and ^{40}K, respectively.

Annual Effective Dose (AEDE)

Estimation of the *annual effective dose equivalent* received by a person can be calculated from the absorbed dose rate using (Abdullah et al., 2019)

$$AEDE(outdoor)\ (mSvy^{-1}) = (0.2)D_{\gamma}T\ (7\ x\ 10^{-4})\ = 1.226\ D_{\gamma} \qquad (7a)$$

$$AEDE(indoor)\ (mSvy^{-1}) = (0.8)D_{\gamma}T\ (7\ x\ 10^{-4}) = 4.901\ \ D_{\gamma} \qquad (7b)$$

D_{γ} is the absorbed dose rate in μSv/h, T is the total number of hours annually (8760 h), and 0.2 and 0.8 are the outdoor and indoor occupancy factors; respectively, and the 7 x 10^{-4} is conversion factor 0.7 $SvGy^{-1}$ into mSv and it is to convert the absorbed dose in air to human effective dose in adults (UNSCEAR, 2008). Summing up the two above equations, gives

$$AEDE\ (mSvy^{-1}) = 6.132\ D_{\gamma}$$

The worldwide average AEDE value for adult is 0.48 $mSvy^{-1}$ (UNSCEAR, 2008) and this value is higher by 10% and 30% for children and infants, respectively.

Excess Lifetime Cancer Risk (ELCR)

The ELCR tool is used to assess and predict the probability of risk of developing cancer from low-dose radiation exposure in the lifetime of a person (Ugbede & Benson, 2018; Agbalagba *et al.*, 2016; Jindal *et al.*, 2018). Equation (7) was applied to estimate and assess the cancer risk (ELCR) of the public and workers and it is calculated as,

$$ELCR = (AEDE)(DL)(RF). \qquad (8)$$

AEDE is the annual effective dose equivalent; DL is the average duration of lifetime of a person (70 years). RF is the cancer risk factor. Low-dose background radiation is considered as one of the causes of long-term effects on public. As per the ICRP recommendation, the value of cancer risk factor for radiation exposure of the general public, is 0.05 $Sv{\cdot}y^{-1}$ (ICRP, 2007; Agbalagba *et al.*, 2016; Ugbede & Benson, 2018).

3. Results and Discussion

To assess the radiological risks in six water samples collected form Hakim Gara stone quarrying area near Harrar town, the radioactivity level from long-lived radionuclides were determined using the gamma spectrometric technique. The results, obtained from quantitative and qualitative analysis of spectral data collected, is presented in Table 2.

Table 2. Mean activity concentrations of radionuclides and the radiological hazard parameters

Sample ID	Activity concentrations (Bq L^{-1})				Hazard index						
	^{238}U	^{232}Th	^{40}K	Ra_{eq}	H_{ex}	H_{in}	Dγ (nGyh^{-1})	AEDE (In-door) (mSvy^{-1})	AEDE (Out-door) (mSvy^{-1})	AEDE (To-tal) (mSvy^{-1})	ELCR (x10^{-3})
TW-S1	3	8.1	95.3	21.93	0.059	0.067	10.25	0.05	0.013	0.063	0.22
TW-S2	2.7	6.7	104.9	20.46	0.055	0.063	9.71	0.048	0.012	0.06	0.208
TW-S3	2.8	9.3	77.4	22.07	0.06	0.067	10.14	0.05	0.012	0.062	0.218
TW-S4	2.9	7.2	94.3	20.38	0.055	0.063	9.59	0.047	0.012	0.059	0.206
TW-S5	1.9	8.9	71.2	20.05	0.054	0.059	9.2	0.045	0.011	0.056	0.197
TW-S6	2.7	8.1	84.5	20.86	0.056	0.064	9.7	0.048	0.012	0.059	0.208
Min.	1.9	6.7	71.2	20.046	0.054	0.059	9.2	0.045	0.011	0.056	0.197
Max.	3	9.3	104.9	22.07	0.06	0.07	10.25	0.05	0.013	0.063	0.22
Ave.	2.7	8.1	87.9	20.98	0.057	0.064	9.76	0.048	0.012	0.06	0.21
stdev	0.4	0.9	11.4	0.78	0.002	0.003	0.35	0.002	0	0.002	0.008
Permissible (WHO, 2022)	**10.0**	**1.0**	**n/a**	**370**							
UNSCEAR (2008)					**1.0**	**1.0**	**55**			**0.48**	**0.29**

From the view point of activity concentration of ^{238}U, all wells exhibited values which are lower than the recommended limits of 10 BqL^{-1}. In terms of ^{232}Th, all the wells showed higher than the recommended value of 1 Bq L^{-1}. Thorium isotopes emit alpha and beta particles with or without gamma radiations (ATSDR, 2019). Thorium is not readily absorbed by the lungs of gastrointestinal tract since absorption depends on solubility of the substance and particle size. But it can be retained in the lung following inhalation exposure. Most of the inhaled thorium is excreted in the feces within several days following ciliary clearance from the lungs to the gastrointestinal tract. Most of the ingested thorium is not absorbed and excreted in the feces (US EPA, 2024). It primarily distributes to lymph nodes and bone surface.

The calculated activity concentrations of the Radium equivalent ranged from 20.05 Bq.L^{-1} to 22.07 Bq.L^{-1} with mean values of 20.98±0.78 Bq.L^{-1}. Minimum and maximum Radium equivalent values measured in water samples of TW-S2 and TW-S3. In all the water samples, the values of the radium equivalent has shown less than the recommended threshold value of 370 Bq.L^{-1} and poses no threat to the quarry workers or the residents at this time. The annual effective doses of radionuclides in all the wells is below the recommended level of 0.48 (UNSCEAR, 2008). All the wells have comparable values and it seems that they are not yet polluted to the level that poses danger to the community.

Since there was no similar study performed earlier in the study area, the findings of this study were compared with similar study performed in groundwater of the nearest town Dire Dawa, about 56 Km from Harar. In the study of Abas (2021), the measured activity concentrations were ranged from 4.385±1.45 to 4.78±0.78 Bq.L^{-1} for ^{238}U, 9.11±2.68 to 76.55±3.26 Bq.L^{-1} for ^{232}Th and 122.05±9.31 to 140.22±61.81 Bq.L^{-1} for ^{40}K, respectively. All the mean values of activity concentrations of ^{238}U, ^{232}Th and ^{40}K of this study were found less than the values from the Dire Dawa groundwater study (Abas et al., 2021).

As shown in Table 2, both the calculated mean external and internal hazard indices of this study vary from 0.054 to 0.060 and from 0.059 to 0.070, respectively. Comparing these values with the reference limits set by UNSCEAR for hazard indices of unity, they are far below unity (UNSCEAR, 2000).

The calculated values of absorbed dose rate (Dγ) are ranging from 9.20 nGh^{-1} to 10.25 nGh^{-1} with mean value of 8.55± 9.76±0.35 nGh^{-1}. The mean absorbed dose rate of the water samples has shown too far below the maximum permissible limit of 55 nGh^{-1}.

The result of health hazard parameters obtained

in this study for water samples collected from mean altitude of about 1867.8.m was compared with the results of our previous study on the soils collected from quarry sites at mount Hakim Gara with mean altitude of about 2050 m (Regassa et al., 2023) as shown in Table 3. On average, the soil and water sampling points are separated by a distance of about 1.8 Km. The ratios of all the parameters of soil to water has shown big difference between the values considered showing the gamma radiation from the analysis of water samples are relatively insignificant. All the health hazard parameters for water samples were smaller as compared with parameters for soil samples.

Table 3. Comparison values of radiation health hazard parameters in water and soil of the study area.

Parameter measured	Soil Sample	Water Sample	Soil/water
H_{ex}	0.44	0.057	7.72
H_{in}	0.49	0.064	7.66
D_{γ}	65.55	9.76	6.72
AEDE	0.08	0.06	1.33
ELCR	0.28	0.021	13.33

The results of this study was compared with other studies as presented in Table 4, and the ^{238}U concentration level of this study was only higher than the Bangladesh study (Banik et al., 2021) and Nigerian study by Auwal (Auwal, 2020) but lower than the rest. The ^{232}Th value was higher from studies by the Vietnam study (An et al., 2023) and the Dire Dawa study (Abas et al., 2021) and lower than the others studies (Nguelem *et al.*, 2013; Oyebanjo & Magbagbeola, 2015; Auwal, 2020; Banik *et al.*, 2021). Similarly the ^{40}K value is found lower than the studies from Vietnam (An et al., 2023) and Ethiopia Dire Dawa (Abas et al., 2021). But it is higher than values of studies from Bangladesh (Banik et al., 2021), Nigeria (Auwal, 2020), Ghana (Nguelem et al., 2013) and (Oyebanjo & Magbagbeola, 2015)].

Table 4. Relating average activity concentrations of ^{238}U, ^{232}Th and ^{40}K values in groundwater with other studies.

Country	Activity concentration (Bq L^{-1})			Ref.
	^{238}U	^{232}Th	^{40}K	
Bangladesh	2.59	2.45	32	(Banik et al., 2021)
Nigeria -Buni Gari	0.0538	0.327	6.2755	(Auwal, 2020)
Vietnam	49 ± 3	77 ± 4	528 ± 25	(An et al., 2023)
Ghana-Adentan	-	0.57±0.29	3.67±2.46	(Nguelem et al., 2013)
Nigeria-Lagos	6±1.4	4.5±1.2	13.4±10.8	(Oyebanjo & Magbagbeola, 2015)
Ethiopia-Dire Dawa	4.78±0.78	9.11±2.68	140.22±61.81	(Abas et al., 2021)
This study	2.7±0.4	8.1±0.9	87.9±11.4	
WHO	10	1	n/a	(Herschy, 2012)

Based on the values given in Table 4, in terms of ^{238}U our result is closer to the results of Bangladesh and Dire Dawa-Ethipia and interms of ^{232}Th our results are closer to the results of Dire Dawa-Ethiopia which is the nearest position to the quarry area relative to the others and this showed foundation rocks of the two areas are similar.

4. Conclusion

The level of natural radioactivity in water samples collected from areas nearby Hakim Gara quarry in Harar town was carried out using high-resolution gamma-ray spectrometry (HPGe) system. The result of the health hazard parameters analysis based on the obtained data of activity

concentration of naturally occurring gamma emitting radionuclides ^{238}U, ^{232}Th and ^{40}K measured in all the water samples were within the recommended limits. Thus, from the findings of this study the quarry activity on Hakim Gara has no direct impact on the gamma radioactivity concentration level of natural radionuclides to the available groundwater in the town. From the radiological perspective the groundwater is safe for domestic activities, but it is recommended to conduct further radiological monitoring and on all available groundwater supply in and around the study area to ascertain the safety of using water by the public. Similar studies shall further be extended to areas of quarrying activities all over the country.

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Disclosure statement

No potential conflict of interest was reported by the author(s).

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