EVALUATION OF RADIONUCLIDES SPECIFIC ACTIVITY OF GROUNDWATER RESOURCES. A CASE OF DIRE DAWA CITY, ETHIOPIA

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Abstract. The purpose of this work is to study specific activity of natural radioactivity concentrations of ^{226}Ra, ^{232}Th, ^{40}K in different water sample locations of Dire Dawa city. The samples were collected from groundwater resources of the city in two different seasons. High resolution gamma ray spectrometry was used to assess the natural radioactivity levels concentration of, and to measure the annual effective does, radium equivalent, absorbed dose rate, external hazard and internal hazard. The result revealed that the range of radioactivity level concentration in summer season of 4.30–6.01 Bq·L^{-1}, 3.75–12.74 Bq·L^{-1}, 3.04–122.48 Bq·L^{-1}, for ^{226}Ra ,^{232}Th and ^{40}K respectively. For the sample that was collected in winter season the range of radioactivity level concentrations of ^{226}Ra, ^{232}Th and ^{40}K are 0.70–6.02 Bq·L^{-1}, 1.11–9.72 Bq·L^{-1} and 12.2–134 Bq·L^{-1} respectively. The outdoor absorbed dose rate varies from 0.006 to 0.019 nGy·h^{-1} with an average of 0.014 ± 0.032. The effective radium equivalent specific activity concentration is peak in summer than in winter season in one of the groundwaters. The activity level of the radioactivity elements was lower than the world average values.

Key words: Specific radioactivity, radionuclides, groundwater, radium equivalent.

INTRODUCTION

Water is a very important element in environmental studies due to its daily use by humans, and the possibility of water-related infections [13]. Groundwater has natural radionuclides, such as ^{40}K, as well as natural decay chains of ^{226}Ra and ^{232}Th [13, 15]. The radionuclides are found everywhere around us; in the earth's crust, air, water, plants, and so on. They may naturally occur or they are artificially produced [9].

The presence of natural radionuclides in water depends on geological and geographical nature of the water's origin [8, 10, 13]. For groundwater (boreholes and wells), it depends on their presence and contents in lithologic of solids aquifer where water is stored [10, 12]. The dissolution and the amounts of natural radionuclides in groundwater system during water/rocks-soils interaction depends

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on the geochemical characteristics of rocks and soil as well. Because some dissolved radioactive elements from soils can leach through groundwater system during precipitation. Other factors that control their occurrence and distribution in groundwater are hydro-geological conditions of groundwater and geo-chemistry of each radionuclides [8].

The United Nations Scientific Committee on the Effects of Atomic Radiation has estimated the global average annual dose rate [19]. The amount of radiation per person in the environment is about 3.0 mSv/year. Of this, 80 % (2.4 mSv/year) is due to naturally occurring source of radiation. Whereas, 19.6 % (almost 0.6 mSv/year) is due to the use of radiation for medical diagnosis, and the remaining 0.4 % of (around 0.01 mSv/year) is from man-made source of radiation [19]. Researches done by [12, 14] show that the level of radioactivity in the ground water was aimed to assess the radiological risk of drinking water.

In Dire Dawa city, no research has been conducted to examine the radionuclides in groundwater of the city resources. The radiological purity of water supplies in the city is not only unknown, there is no baseline data of activity concentration of natural radionuclides in groundwater in the city. There was research done by Ayalew *et al*. [4] which shows the assessment of the background radiation of the soil in the city. This result revealed that the background radiation of the soil was medium. Radionuclides within the rock and sediments may have a great contribution to the radioactivity of groundwater. They may be dissolved or leached out of the source rocks sediment and remain in solution.

Therefore, the main purpose of this study is to measure the specific activity of radionuclide concentration in groundwater, drawn from local well waters, in the surrounding area of Dire Dawa city, used for drinking and other domestic activity.

MATERIALS AND METHODS

Dire Dawa is located at the eastern part of Ethiopia. It is found on the Harrar-Somali block to the south. The Precambrian metamorphic are horizontally overlain by thin local Mesozoic sandstones, followed by about 400 m of Jurassic limestones and an Upper Sandstone formation of variable thickness, locally exceeding 300 m [6]. The Mesozoic is followed disconformable by basalts of the Trap Series, up to about 1,000 m thick, which thin eastwards and die out in northern Somalia.

SAMPLING PROCEDURE

The first task of the experiment was collecting groundwater samples from the selected sites. To avoid contamination or removing unnecessary impurities, the container was drained. One percent above the top-level of the water in the plastic vessel was left for thermal expansion. The level of water in each plastic vessel was the same. 20 mL of diluted hydrochloric acid was added to the sample immediately after collection to reduce the pH and to minimize precipitation and absorption on container walls.

The groundwater uses by the inhabitants for several activities such as bathing, washing, cooking, and even being used as a reserve source of drinking water was chosen. The depth of ground surface of water varies from 104.4 to 361 m and its pH varies from 6.87 to 7.47, as shown in Table 1.

Table 1

Depth and pH of groundwater in each site

Site	**Depth (m)**	**pH**
Sabian	221	6.9
Genda Tesfa	190	7.11
Gende Gerada	359	7.43
Addisu Kela	150	7.14
Lege Hare	266	7.47
Tomme	332	6.87
Boren 2	104.4	7.42
Melka	356	7.12
Boren 4	361	7.41
Kezira	200	7.33

SAMPLE PREPARATION

Each sample was poured into a beaker and evaporated on a hot plate to 500 mL volume and then transferred to a clean dry planchet. The samples were evaporated to dryness on hot plate. The residues were transferred onto to a clean, dry, and weighed planchet. They were spread uniformly on the planchet by dropping a few drops of ethanol. The residues were allowed to dry and then covered with Mylar film ready for counting.

GAMMA SPECTROMETRY

Gamma ray spectroscopy is the most important tool to analyse properties of excited nuclei and to determine decay schemes and explore nuclei with respect to nuclear models. It is also an analytical technique used for the identification and quantification of gamma emitting isotopes in a variety of matrices. With little sample preparation, the spectrometry allows to detect several gamma-emitting radionuclei from the sample. The measurement gives a spectrum of lines, the amplitude of which is proportional to the activity of the radionuclide, and its position on the horizontal axis gives an idea on its energy.

SAMPLE COUNTING AND SPECIFIC ACTIVITY MEASUREMENT

Following samples preparation, each sample was placed in the device and was counted for 57,600 s. Measurement of the sample's spectrum was carried out using simple software (Accuspec software). Since, we need to extract the peak areas from these measurements. Later, the analysis of environmental samples spectra is performed using conventional Genie 2000 package software from Canberra). The specific activity of ^{238}U, ^{232}Th and ^{40}K, in Bq·L^{-1}, from the water samples was determined. Next, the ^{238}U activity was determined by taking the mean activity of the two separate photo peaks of the daughter nuclides: ^{214}Pb at 352.0 keV and ^{214}Bi at 609.3 keV; ^{232}Th specific activity was determined using photo peaks of ^{228}Ac at 911.1 keV and the photo-peak of ^{212}Pb at 583.1 keV; for ^{40}K we used directly the 1460.8 keV photo-peak [16].

The specific activity of the radionuclide in each water sample was calculated using the expression:

$$A = \frac{N}{\varepsilon(E) \times P \times t \times m} \quad (1)$$

where A = specific activity of the radionuclide in Bq·L^{-1}, N = net area count under the photo-peak of each radionuclide, m = mass of water sample, t = counting time, P = gamma yield or absolute probability of the specific gamma ray and $\varepsilon(E)$ = efficiency at specific gamma-ray energy, in Bq·L^{-1}.

RADIOLOGICAL PARAMETERS

Radium equivalent activity

The weighted sum of radium equivalent activity represents the natural radionuclides, and that is based on the approximation that 1 Bq·L^{-1} of ^{226}Ra, 0.7 Bq·L^{-1} of ^{232}Th, and 13 Bq·L^{-1} of ^{40}K yield the same radiation dose rates. The radium equivalent activity (Ra_{eq}) is defined mathematically by Eq. (2) [16].

$$Ra_{eq} = C_{\mathrm{Ra}} + 1.43\ C_{\mathrm{Th}} + 0.077\ C_{\mathrm{K}} \quad (2)$$

where C_{Ra}, C_{Th} and C_{K} are the specific activity, in Bq·L^{-1}, of ^{226}Ra, ^{232}Th and ^{40}K, respectively. The use of a material whose (Ra_{eq}) exceeds 370 Bq·L^{-1} is discouraged to avoid radiation hazards [9].

The absorbed dose rate in air

The absorbed dose rate is the amount of radiation energy absorbed per unit mass of the materials. Radionuclides background radiation absorption dose rate, measured one meter above the ground surface, expresses the received dose in the open-air radiation emitted from radionuclides concentrations in water. It is an important quantity to measure the radiation risk of a bio system, and it can be evaluated using Eq. 3 [3, 10].

$$AD = 0.461\, C_{\mathrm{Ra}} + 0.623\, C_{\mathrm{Th}} + 0.0414\, C_{\mathrm{K}} \tag{3}$$

where AD is the absorbed dose rate, 0.461, 0.623 and 0.0414 $(\mathrm{nGy \cdot h^{-1}})/(\mathrm{Bq \cdot L^{-1}})$ are the conversion factors for ^{226}Ra, ^{232}Th and ^{40}K, respectively.

Annual effective doses equivalent

Annual estimated average effective dose equivalent received by the people as calculated using a factor of 0.7 $\mathrm{Sv \cdot Gy^{-1}}$, which was used to alter the absorbed dose rate to human effective dose equivalent with an outdoor of 20 % and 80 % for indoor [4]. Annual estimated effective doses equivalent (*AED*) for outdoor and indoor are calculated based on conversion factors given by Eqs. 4 and 5 [5, 11].

$$AED_{\mathrm{outdoor}}(\mathrm{mSv \cdot y^{-1}}) = AD(\mathrm{nGy \cdot h^{-1}}) \times 8760\ \mathrm{h} \times 0.2 \times 0.7\ \mathrm{Sv \cdot Gy^{-1}} \times 10^{-6} \tag{4}$$

$$AED_{\mathrm{indoor}}(\mathrm{mSv \cdot y^{-1}}) = AD(\mathrm{nGy \cdot h^{-1}}) \times 8760\ \mathrm{h} \times 0.8 \times 0.7\ \mathrm{Sv \cdot Gy^{-1}} \times 10^{-6} \tag{5}$$

Determination of radiation hazard indices

Many of the radioactive materials decay naturally and produces external radiation field which exposes humans. Regarding of dose, the principal primordial radionuclides are ^{232}Th, ^{226}Ra and ^{40}K. Thorium and uranium series of radionuclides produce significant human exposure. The external hazard index (H_{ex}) is calculated by Eq. 6 [9].

$$H_{\mathrm{ex}} = \frac{C_{\mathrm{Ra}}}{370} + \frac{C_{\mathrm{Th}}}{250} + \frac{C_{\mathrm{K}}}{4810} \tag{6}$$

where C_{Ra}, C_{Th} and C_{K} are the specific radioactivity, in $\mathrm{Bq \cdot L^{-1}}$, of ^{226}Ra, ^{232}Th and ^{40}K, respectively. The external hazard index must be less than one for the radiation hazard to be negligible. H_{ex} equal to one corresponds to the upper limit of Ra_{eq} (370 $\mathrm{Bq \cdot L^{-1}}$) [10].

The internal hazard index (H_{in}) can be calculated by Eq. 7 [4].

$$H_{\mathrm{in}} = \frac{C_{\mathrm{Ra}}}{185} + \frac{C_{\mathrm{Th}}}{259} + \frac{C_{\mathrm{K}}}{4810} \tag{7}$$

where C_{Ra}, C_{Th} and C_K, are the specific radioactivity, in $Bq \cdot L^{-1}$ of ^{226}Ra, ^{232}Th and ^{40}K, respectively. The value of this index must be less than unity for the radiation hazard to be negligible. Both the external and internal hazard indices are pure numbers and they do not have dimensions.

ANALYSIS OF WATER SAMPLES

The samples were analysed using an n-type coaxial CANBERRA high-resolution gamma-spectrometry system. The spectrometer consists of a high purity germanium (HPGe) detector coupled to a desktop computer provided with Genie 2000 software for spectrum acquisition and evaluation. The detector crystal has a diameter of about 72.5 mm and a thickness of about 72.5 mm, with a relative efficiency of 70 %.

RESULTS AND DISCUSSION

One of the main purposes of this work was to determine the specific activity of groundwater selected from ten sites of Dire Dawa city. The samples were collected to identify the radiation exposure dose and to assess the impact on humans.

Table 2 shows the values of specific activity for ^{226}Ra, ^{232}Th and ^{40}K, as well as the value of specific activity in two different seasons, in drinking water samples collected from ten different sites at Dire Dawa City, using gamma-ray spectroscopy. The values were lower than the global mean values hazard specific activity indicated by UNSCEAR [22] (35, 30 and 400 $Bq \cdot L^{-1}$, respectively). Even if the specific activity of ^{40}K is less than the global limit, in some selected site of the groundwater, it was observed to be comparatively higher than that of both ^{226}Ra and ^{232}Th in all of the water sampling locations studied, as shown in Table 2.

Table 2

Spatial and temporal variation in specific activities of radionuclides in the groundwater of the city

Site	Nuclide	Specific Activity [$Bq \cdot L^{-1}$]			
		Winter	**Summer**	**Mean**	**SD**
Sabiyan	^{226}Ra	2.87	4.61	3.74	1.230
	^{232}Th	7.64	12.47	10.055	3.415
	^{40}K	132.49	122.48	127.485	7.078
Gende tesfa	^{226}Ra	4.95	5.71	5.33	0.537
	^{232}Th	9.72	11.39	10.555	1.181
	^{40}K	128	315	221.5	132.229
Gende Gerada	^{226}Ra	6.02	4.3	5.16	1.216
	^{232}Th	1.11	3.75	2.43	1.867
	^{40}K	125	135.7	130.35	7.566
Addisu kela	^{226}Ra	3.02	6.01	4.515	2.114
	^{232}Th	9.18	7.19	8.185	1.407
	^{40}K	122	115	118.5	4.950

Lege Hare	^{226}Ra	3.18	4.03	3.605	0.601
	^{232}Th	7.64	8.23	7.935	0.417
	^{40}K	134	125.5	129.75	6.010
Tomme	^{226}Ra	2.68	4.74	3.71	1.457
	^{232}Th	6.16	9.46	7.81	2.333
	^{40}K	109.15	109.38	109.265	0.163
Boren 2	^{226}Ra	4.59	3.79	4.19	0.566
	^{232}Th	9.22	12.5	10.86	2.319
	^{40}K	128.9	124.5	126.7	3.111
Melka	^{226}Ra	4.59	4.23	4.41	0.255
	^{232}Th	9.22	9.12	9.17	0.071
	^{40}K	110.5	118.75	114.625	5.834
Boren 4	^{226}Ra	4.94	4.65	4.795	0.205
	^{232}Th	4.2	7.4	5.8	2.263
	^{40}K	110.5	117.45	113.975	4.914
Kezira	^{226}Ra	7.01	5.74	6.375	0.898
	^{232}Th	1.4	9.54	5.47	5.756
	^{40}K	120	118.4	119.2	1.131

Table 3

The mean specific activities of radionuclides collected from groundwater (mean±SD)

Radionuclides	**Specific activity [Bq·L^{-1}]**	
	Winter	**Summer**
^{226}Ra	4.385±1.45	4.781±0.78
^{232}Th	76.55±3.26	9.11±2.68
^{40}K	122.05±9.31	140.22±61.81

Table 4

Absorbed dose rate, outdoor and indoor annual effective dose, radium equivalent activity and external and internal hazard index of the samples collected from the city groundwater during winter season

Site	***Ra$_{eq}$*** **(Bq·L^{-1})**	***AD*** **(nGy·h^{-1})**	***AED*** **(mSv·y^{-1})**		***H*$_{ex}$**	***H*$_{in}$**
			Outdoor	**Indoor**		
Sabiyan	23.997	11.568	0.014	0.056	0.066	0.073
Gende Tesfa	28.706	13.637	0.017	0.066	0.079	0.091
Gende Gereda	17.232	8.642	0.011	0.042	0.047	0.063
Addisu Kela	25.541	12.162	0.015	0.059	0.070	0.077
Tomme	19.893	9.592	0.012	0.047	0.055	0.061
Boren 2	27.700	13.197	0.016	0.064	0.076	0.087
Melka	26.283	12.435	0.015	0.060	0.072	0.083
Boren 4	19.455	9.469	0.012	0.046	0.053	0.066
Kezira	18.252	9.072	0.011	0.044	0.049	0.068

Lege Hare	24.423	11.773	0.014	0.057	0.067	0.075
Mean	23.148	11.155	0.014	0.054	0.063	0.074
Max	28.706	13.637	0.017	0.066	0.079	0.091
Min	17.232	8.642	0.011	0.042	0.047	0.061
SD	4.119	1.810	0.002	0.009	0.012	0.010

The mean specific activities of ^{226}Ra, ^{232}Th, ^{40}K were measured in winter and summer season in each groundwater, as shown in Table 3. The radium specific activities in each site are greater than the values obtained by Seghour [17].

The radium equivalent activity was calculated by equation (2) and shown in Table 3. According to reference [20], the threshold value of Ra_{eq} must be less than 370 Bq·L^{-1}. The mean value of radium equivalent activity in this study area is 23.148±4.12 Bq·L^{-1} and varies from 17.232 to 28.706 Bq·L^{-1} for ten sites during winter, as shown in Table 4. Similarly, the mean value of radium equivalent in summer is 28.60±7.07 Bq·L^{-1} and varies from 20.11 to 46.25 Bq·L^{-1}, as shown in Table 5. The highest radium equivalent was observed in Gende Tesfa site during winter and summer, whereas the lowest radium equivalent was observed in Gende Grada site in both seasons (Tables 4 and 5). This shows that the radium equivalent does not depend on season, but it depends on the site of groundwater.

The absorbed dose rate of the samples has been calculated from each site according to equation (4). The results are shown in Tables 4 and 5. They reveal that the absorbed dose rate due to the terrestrial gamma rays at 1 m above the ground were in the range of 8.64 to 13.64 nGy·h^{-1} for groundwater samples in winter. Its mean value is 11.12 nGy·h^{-1}, shown in Table 4. The mean value for groundwater samples of the city is significantly lower than the world average value of 55 nGy·h^{-1} [18]. Similarly, the absorbed dose rate during summer, shown in Table 5, ranged from 9.94 to 22.77 nGy·h^{-1} for groundwater samples and its mean value was 13.68 nGy·h^{-1}. The highest absorbed dose rate was detected in Gende Tesfa site in summer, but still this value is less than the standard limit given in reference [19].

The outdoor annual effective dose equivalent was calculated from the air absorbed doses using the relation given by Eq. (5) and shown in Tables 4 and 5. The values varied from 0.011 to 0.017 mSv·y^{-1} in winter shown in Table 4 and 0.012 to 0.028 mSv·y^{-1} in summer shown in Table 5 and with the mean 0.017 mSv·y^{-1}. The mean values in groundwater samples were lower than those of the worldwide average values for outdoor annual effective dose, of 0.07 mSv·y^{-1} [1, 21].

The indoor annual effective dose equivalent ranged from 0.042 to 0.066 mSv·y^{-1} with the average value of 0.054 mSv·y^{-1} in winter shown in Table 4, whereas in summer it ranged from 0.048 to 0.111 mSv·y^{-1} shown in Table 5. The result observed during summer was greater than the worldwide average values [1, 2].

The calculated values of hazard index (external and internal) for the groundwater samples have been obtained according to Eqs. 6 and 7, respectively, as shown in Tables 4 and 5. The value of the internal hazard index (H_{in}) ranged from 0.061 to 0.091, with a mean value of 0.074, as shown in Table 4, during winter. The highest value of external hazard index was (0.079) in Gende Tesfa site. Similarly, the highest value of the internal hazard index (0.091) was measured on the same site (Table 4). While, the lowest H_{ex} (0.047) was measured in Gende Gerada, and the lowest H_{in} (0.061) was measured at the Melka site during winter (Table 4).

Similarly, the results revealed that during summer (Table 5), the external hazard index of the groundwater samples in the city ranged from 0.055 to 0.126, with the mean value 0.078 ±0.019, smaller than the value reported in references [2, 3]. The highest H_{ex} and H_{in} were 0.126 and 0.140, respectively, and were measured at Gende Tesfa site, in the summer. Our results revealed that all the hazard indexes observed from all sites samples were ≤ 1, which is the threshold value recommended in references [1, 13]).

Table 5

Absorbed dose rate, outdoor and inner annual effective dose, radium equivalent activity (Ra_{eq}) and external and internal hazard index of the samples collected from the city groundwater during summer season

Site	Ra_{eq} (Bq·L^{-1})	*AD* (nGy·h^{-1})	*AED* (mSv·y^{-1})		H_{ex}	H_{in}
			Outdoor	**Indoor**		
Sabiyan	31.873	14.965	0.018	0.073	0.088	0.099
Gende Tesfa	46.253	22.769	0.028	0.111	0.126	0.140
Gende Gereda	20.111	9.937	0.012	0.048	0.055	0.066
Addisu Kela	25.147	12.011	0.015	0.058	0.069	0.084
Tomme	26.690	12.607	0.015	0.061	0.073	0.085
Boren 2	31.252	14.689	0.018	0.071	0.086	0.095
Melka	26.415	12.548	0.015	0.061	0.073	0.083
Boren 4	24.276	11.616	0.014	0.056	0.067	0.078
Kezira	28.499	13.491	0.017	0.066	0.078	0.092
Lege Hare	25.462	12.181	0.015	0.059	0.070	0.080
Mean	28.598	13.681	0.017	0.066	0.078	0.090
Max	46.253	22.769	0.028	0.111	0.126	0.140
Min	20.111	9.937	0.012	0.048	0.055	0.066
SD	7.074	3.513	0.004	0.017	0.019	0.020

CONCLUSIONS

With steadily growing pollution and ongoing climate change, the role of groundwater systems as a strategic resource of drinking water gains in weight. Nowadays, groundwater covers already 100 % of the drinking water needs of the city. As it was reported in reference [7], the natural radioactivity in groundwater has significant impact on overall effective dose received by inhabitants. Natural radioactivity levels in ten selected groundwater sites in Dire Dawa city have been assessed using gamma-ray spectrometry (HPGe). The calculated specific activity of ^{226}Ra, ^{40}K and ^{232}Th were compared with values published in other countries selected from the literature and observed to be within the same range [23]. The average values obtained were also below the guideline values recommended in reference [23]. The results obtained in this work were lower than the results shown in reference [7].

Moreover, the results indicate that the radium equivalent activity of the groundwater was site-dependent and showed a seasonal variation. The average annual effective dose rates in groundwater outdoor and indoor were 0.017 and 0.066 mSv·y^{-1}, respectively, in the summer season, lower than the average recommended in reference [23].

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