# ASSESSMENT OF THE NATURAL RADIOACTIVITY OF BOTTLED DRINKING WATER PRODUCED IN DIRE DAWA, ETHIOPIA

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Abstract. Measurements of natural radioactivity in drinking water have been evaluated in most parts of the world to assess the dose rate and irradiation risk. This work aims to determine the natural radioactivity of bottled drinking water produced in Dire Dawa City and the surrounding area. Natural radioactivity water from nine different brands is produced and commonly sold in bottles. The measurement was done by gamma-ray spectroscopy using a hyper-pure germanium detector. The water specific activity in <sup>238</sup>U ranged from 2.91 ± 0.42 to 4.04 ± 0.42 Bq L<sup>-1</sup>; in <sup>226</sup>Th, it ranged from 4.66 ± 2.48 to 13.31 ± 0.2.48 Bq L<sup>-1</sup>, and in <sup>40</sup>K, it ranged from 106.52 ± 8.71 to 136.52 ± 8.71 Bq L<sup>-1</sup>. These results were compared with the reported values from other countries. Annual estimated effective doses from the intake of natural radionuclides in bottled drinking water were found to be below the limit of 0.1 mSv y<sup>-1</sup> recommended by World Health Organization (WHO). The cumulative average annual effective doses of <sup>238</sup>U for different age groups of (0 – 1), (1 – 2), (2 – 7), (7 – 12), (12 – 17) years, and above 17 years were estimated to be 3.01, 0.30, 0.60, 0.90, 2.88, and 0.66 mSv y<sup>-1</sup>, respectively.

Key words: radioactivity, specific activity, bottled water, annual effective dose.

# **INTRODUCTION**

Humans are exposed to ionizing radiation sources that include cosmic rays and natural radionuclides present in the air, food, and drinking water [20]. This natural radioactivity (*i.e.*, the background radiation) is the source of radiation exposure of human body constituents [1]. Ionizing radiation, which has always existed naturally, is constantly irradiating all living species. The sources of that exposure are cosmic rays from outer space and the Sun's surface, terrestrial radionuclides found in the Earth's crust, construction materials, air, water, and foods, and the human body itself. According to UNSCEAR, exposure to natural resources is expected to affect more than 70 % of the population [35–37]. The global average exposure of humans to natural sources is  $2.4 \text{ mSv y}^{-1}$  [35, 36]. The estimated dose for the human body due to natural radionuclides is  $0.3 \text{ mSv y}^{-1}$  [35–37].

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According to WHO guidelines, the contribution to the radiation exposure rate of drinking water is approximately 0.01 mSv  $y^{-1}$ , as reported in [39]. The specific activity of drinking water can increase the overall absorbed dose. The specific concentration of radionuclides in drinking water has been extensively studied by measuring the level of specific activities and the annual effective absorbed dose [3, 12–13, 16, 19, 24, 29, 32, 38].

Guidelines issued by WHO for the quality of drinking water show that the dose rate is below the recommended level (0.1 mSv  $y^{-1}$ ), and water is believed to be safe for human consumption [40–41].

Groundwater contains various dissolved radioactive materials [1, 9, 17]. Radionuclides presence in groundwater depends on the physical, chemical, and geological properties of the aquifer components [1, 9, 17, 42]. The water radioactivity distribution depends on the geography, soil geology, rock, and other parameters [1, 9, 30, 37].

Assessment of natural radioactivity in drinking water has been done in different areas to determine the risk of water consumption [1, 3, 12]. Radionuclides affect human health since they are deposited in the human body through water drinking. They radionuclides dissolved in water are emitting radon and gradually affect living tissues because water penetrates the whole body [1, 5, 12].

The consumption of bottled mineral water in Dire Dawa City is steadily increasing. This study aims to characterize the quality of bottled mineral waters regarding the concentration of soluble radionuclides and to assess the health impact and effects connected to long-term consumption of water containing <sup>238</sup>U, <sup>232</sup>Th, and <sup>40</sup>K. Moreover, the nature of the drinking water in Dire Dawa City has been classified as hard water for drinking purposes [1].

## MATERIALS AND METHODS

### STUDY AREA

The Dire Dawa City is situated in eastern Ethiopia, surrounded by highlands. It has 280,000 inhabitants. The ecological environment of this area and its geology provide good opportunities for industrial development and tourism activities to stimulate and accelerate the economic growth of the city and the country. It is located at the latitude of  $9^{\circ}36'$  N and longitude of  $41^{\circ}52'$  E.

### SAMPLING AND CHEMICAL CHARACTERISTICS OF BOTTLED WATER

For this investigation, samples of bottled mineral water are typical of those consumed by the population of Dire Dawa City. This water was available on the market for over 13 years and was analyzed in this research. There are nine bottled water samples whose bottle sizes or volumes are similar (Aqua Dire, Aqua Uno, Vita spring, Ayaan spring, Efftin spring, Free spring, Liban spring, Dal-son spring, and Aqua souls spring). The samples were labeled using an identification code (ID) consisting of letters and numbers such as: SBWi (i = 1, 2,...9). They were commercially available for human consumption in the local markets as shown in Table 1. They came from wells and boreholes, and they were spread out in various locations. It can help us to look at the wider geographical distribution to identify the potential impact of regional heterogeneity on water quality [1, 9, 25].

The sites of industries were found at different locations. It may help us to take into account of wider geographical distribution to capture any potential effects of the variability of the underlying geology of the regions to the quality of water [1, 9, 25].

Bottle water brands	Sample codes	Latitude (N)	Longitude (E)
Aqua Dire Mineral	SBW1	9.54	41.53
Aqua Uno spring water	SBW2	9.62	41.54
Vita spring water	SBW3	9.61	41.23
Ayan spring	SBW4	9.60	41.87
Efftin spring	SBW5	9.58	41.76
Free spring	SBW6	9.36	41.52
Liban spring	SBW7	9.45	41.44
Dal-son spring	SBW8	9.43	41.45
Aqua souls spring	SBW9	9.52	41.23

#### Table 1

Name of the mineral bottled waters with their geographical locations

#### Sample preparation

The sample was poured into a beaker and evaporated to a volume of 500 mL on a hot plate. After that, it was transferred to a dry planchet and evaporated on a hot plate. The residues were weighed and transferred to a clean, dry slab. They were spread uniformly by drooping ethanol. The deposits were allowed to dry and then covered with Mylar film for radiation counting [1]. Samples were analyzed for radioactivity in the Environmental Protection Agency of Ethiopia's radiation monitoring laboratory.

#### **Gamma spectrometry**

Gamma-ray spectroscopy is a vital tool to analyze the properties of excited nuclei and determine their decay schemes. The analytical technique is used for the identification and quantification of gamma-emitting isotopes in a variety of matrices. Sample preparation and spectrometry allow the detection of several gamma-emitting radionuclides from the sample. The measurement gives the spectrum of the line's amplitude, which is proportional to the specific activity of the radionuclide. The line/peak position on the horizontal axis gives an idea of gamma radiation energy [1].

#### Sample counting and specific activity measurement

Following the preparation, the sample was placed in the device and counted for 57,600 s. The sample spectrum was carried out using simple software (Accuspec software) helping to extract the peak areas. The environmental sample spectra are analyzed using conventional Genie 2000 package software from Canberra (Industries, Inc., USA) to calculate the natural radioactivity [14].

The specific activities of <sup>238</sup>U, <sup>232</sup>Th, and <sup>40</sup>K from the water samples were determined. The specific activity of <sup>238</sup>U was determined by taking the mean of the two photo peaks of the daughter nuclides: <sup>214</sup>Pb at 352.0 keV and <sup>214</sup>Bi at 609.3 keV. <sup>232</sup>Th specific activity was determined using photo peaks of <sup>228</sup>Ac at 911.1 keV and of <sup>212</sup>Pb at 583.1 keV, while the specific activity of <sup>40</sup>K was determined at 1,460.8 keV photo-peak [1, 29]. Because <sup>226</sup>Ra is one important offspring of <sup>238</sup>U with a high lifetime, they will be used alternatively throughout the paper.

The specific activity of the radionuclide in each water sample was calculated using Eq. 1 [8]:

$$A = \frac{\frac{N_{\rm s}}{t_{\rm s}} - \frac{N_{\rm b}}{t_{\rm b}}}{\varepsilon(E_i l_{\rm y} M_{\rm s})} \tag{1}$$

where *A* is the specific activity of the radionuclide, in Bq·L<sup>-1</sup>,  $N_s$  the net counts of the radionuclide in the samples,  $N_b$  the net counts of radionuclide in the background,  $I_{\gamma}$  the gamma emission probability (gamma yield),  $\varepsilon(E_i)$  the peak efficiency of the detector at energy  $E_i$ ,  $t_s$  is counting time,  $t_b$  is background measuring time and  $M_s$  the mass of the sample (kg) [1, 29]. The radium equivalent activity ( $Ra_{eq}$ ) is defined mathematically by Eq. (2) [1, 18, 28]:

$$Ra_{eg} = C_{\rm U} + 1.43C_{\rm Th} + 0.077C_{\rm K} \tag{2}$$

where  $C_U$ ,  $C_{Th}$ , and  $C_K$  are the specific activities of <sup>226</sup>Ra, <sup>232</sup>Th, and <sup>40</sup>K, respectively. Radium equivalent activity ( $Ra_{eq}$ ) more than 370 Bq L<sup>-1</sup> is prohibited in order to prevent the radiation risks [1, 23, 29, 37].

Many radioactive materials decay naturally and produce external radiation exposure of human beings, the principal radionuclides being <sup>232</sup>Th, <sup>226</sup>Ra, and <sup>40</sup>K. The U and Th series of produced radionuclides cause a significant human exposure. The external hazard index ( $H_{ex}$ ) is calculated by Eq. (3), reported in [1, 29]:

$$H_{\rm ex} = \frac{C_{\rm U}}{370} + \frac{C_{\rm Th}}{259} + \frac{C_{\rm K}}{4810} \le 1 \tag{3}$$

This index must be less than unity to make the radiation hazard insignificant, and the geographic area safe for humans to live in.

The internal hazard index  $(H_{in})$  can be determined by Eq. (4), mentioned in [1, 8, 29]:

$$H_{\rm in} = \frac{c_{\rm U}}{185} + \frac{c_{\rm Th}}{259} + \frac{c_{\rm K}}{4810} \le 1 \tag{4}$$

The value of this index must be less than unity to make negligible the radiation hazard. Both indices are pure numbers and therefore, do not have dimensions [1, 8, 28].

The outdoor absorbed dose rate, *AD*, reported in [37] has an average value of 51 nGy  $h^{-1}$ . The rate is given by Eq. (5) [37–38]:

$$AD = 0.461C_{\rm U} + 0.623C_{\rm Th} + 0.0417C_{\rm K} \tag{5}$$

where the coefficients: 0.461, 0.623, and 0.0414  $(nGy \cdot h^{-1})/(Bq \cdot L^{-1})$  are the conversion factors for <sup>238</sup>U, <sup>232</sup>Th, and <sup>40</sup>K, respectively.

The annual estimated average effective dose received by the people was calculated using a factor of  $0.7 \text{ Sv} \cdot \text{Gy}^{-1}$ , which was used to alter the absorbed dose rate to the human effective dose equivalent with an outdoor 20 % and an indoor of 80 % [1, 8]. Annual estimated effective dose equivalents (*AED*) for outdoor and indoor use are calculated based on conversion factors given by Eqs. (6) and (7) [8, 23]:

$$AED_{\text{outdoor}}(\text{mSv y}^{-1}) = AD(\text{nGy} \cdot \text{h}^{-1}) \times 8,760\text{h} \times 0.2 \times 0.7\text{SvGy}^{-1} \times 10^{-3}(6)$$

$$AED_{indoor}(mSv y^{-1}) = AD(nGy \cdot h^{-1}) \times 8,760h \times 0.7 \times 0.7Sv Gy^{-1} \times 10^{-3} (7)$$

The gamma radiation hazard index  $(I\gamma)$  is another radiation index (called also representative level index) defined by the following formula:

$$I_{\rm v} = 0.0067C_{\rm U} + 0.01C_{\rm Th} + 0.00067C_{\rm K} \le 1 \tag{8}$$

where  $C_{\rm U}$ ,  $C_{\rm Th}$ , and  $C_{\rm K}$  have the same meaning as in Eq. (5)

The estimation of annual effective doses to an individual, due to intake of natural radionuclides from bottled drinking water, is estimated using the following relationship.

$$D_{\rm w} = C_{\rm w} C R_{\rm w} D_{\rm cw} \tag{9}$$

where  $D_w$  is the annual effective dose (mSv  $y^{-1}$ ) due to ingestion of radionuclides from the consumption of bottled water,  $C_w$  is the specific concentration of radionuclides in the ingested drinking water (Bq L<sup>-1</sup>),  $CR_w$  is the annual intake of drinking water (L  $y^{-1}$ ),  $D_{cw}$  is the ingested dose conversion factor for radionuclides provided by ICRP (Sv Bq<sup>-1</sup>) [36–37]. The conversion factor varies with age for different age groups: (0-1), (1-2), (2-7), (7-12), (12-17) years, and  $4.7 \times 10^{-6}$ ,  $9.6 \times 10^{-7}$ ,  $6.2 \times 10^{-7}$ ,  $8.0 \times 10^{-7}$ ,  $1.5 \times 10^{-6}$ , and  $2.8 \times 10^{-7}$  Sv. Bq<sup>-1</sup> respectively [41].

The annual effective dose from bottled drinking water has been calculated keeping in view the different age groups: (0-1), (1-2), (2-7), (7-12), (12-17) years, and above 17 years. For this study, the annual average intake of bottled drinking water is considered to be 200, 260, 300, 350, 600, and 730 liters for the age groups of (0-1), (1-2), (2-7), (7-12), (12-17) years, and adults of age above 17 years [42].

The radiological risk related to the radium isotope can be evaluated by calculating the lifetime cancer risk (R) using the following equation [11, 14]:

$$R = MCL \times RC \times TWI \tag{10}$$

where *MCL* is the maximum contaminant level (Bq L<sup>-1</sup>), *RC* is the mortality risk coefficient (7.17 × 10<sup>-9</sup> and 2.0 × 10<sup>-8</sup> per Bq for <sup>238</sup>U and <sup>232</sup>Th, respectively), and *TWI* is the total water intake (2 L d<sup>-1</sup> × 365.4 d y<sup>-1</sup> × 70 ys) [10].

#### Analysis of water samples

The samples were analyzed using an n-type coaxial CANBERRA highresolution gamma-spectrometry system. The spectrometer consists of high purity germanium (HPGe) detector coupled to a desktop computer provided with Genie 2000 software for spectrum acquisition and determination.

### **RESULTS AND DISCUSSIONS**

#### RESULTS

The outcome of the radioactive background radiation from the radionuclides in the sample of mineral bottled water in Dire Dawa City, Ethiopia. Drinking bottled water contains several natural radionuclides. This work helps to determine the specific activity of <sup>238</sup>U, <sup>232</sup>Th, <sup>40</sup>K, radium equivalent, radiation hazard indices, or public health hazards, and annual effective doses of inhalation and ingestion of water.

Table 2 shows the chemical data of the water samples given by the producer list. The pH measurement is the most important and frequently used test in water chemistry, as shown in Table 2.

The Ethiopian drinking water standard has set an allowable pH value ranging from 6.5 to 8.5 [4, 16]. The mineral bottled water produced in Dire Dawa City has a pH of 7.0 to 7.1, which is below the country standard limit set by [16, 39].

### Table 2

The chemical parameters of bottled drinking water brands. TDS = total dissolved solid in one liter of water

Bottled water brands	pН	$TDS (mg L^{-1})$
SBW1	7.1	23.0
SBW2	7.1	27.0
SBW3	7.1	20.0
SBW4	7.0	35.0
SBW5	7.2	80.0
SBW6	7.1	85.0
SBW7	7.0	8.7
SBW8	7.1	10.0
SBW9	7.1	21.0
Ethiopian standard	6.5-8.5	1,000
WHO standard [38]	6.5-8.0	500

*TDS* is the total dissolved solids in 1 L of water, which describes all solids (including mineral salts) dissolved in water. Recent works show that *TDS* found in drinking water ranged from 105 to 478 mg L<sup>-1</sup> [30]. The *TDS* values of mineral water in Dire Dawa City ranged from 8.7 to 80 mg L<sup>-1</sup> below the mentioned results [15, 30].

The chemical properties of various brands of bottled drinking water in Dire Dawa City are shown in Table 3. Drinking water hardness ranged between 10 and 500 mg of CaCO<sub>3</sub> per liter, according to WHO recommendations [39–41]. The CaCO<sub>3</sub> content in all mineral bottled water is less than the limit recommended by WHO [15]. As a result, the bottled water produced and consumed by inhabitants is deemed soft and safe to drink.

Chemical	SBW1	SBW2	SBW3	SBW4	SBW5	SBW6	SBW7	SBW8	SBW9
components	(mg/L)								
$Ca^{2+}$		3.20	1.8	1.80	4.8	1.80	0.80	2.85	3.3
$Mg^{2+}$	0.10	1.30	0.8	0.38	1.1	0.81	0.10	0.65	1.1
Na <sup>+</sup>	1.50	2.70	1.4	3.00	3.0	0.90	0.50	1.20	2.4
Cl−	2.84	3.24	5.4	32.26	10.0	0.30	2.41	1.40	3.2
$SO_4^{2-}$		3.20		0.004	3.4	2.02	0.20	1.20	
CaCO <sub>3</sub>	5.00	6.28		4.80	18.2	10.30	1.00	6.80	4.2
NO <sub>3</sub>		0.80				0.01			
$\mathbf{K}^+$		0.60	1.6		2.5	0.32	0.20	2.40	0.8

Table 3 Chemical parameters of different brands of bottled drinking water samples (mg  $L^{-1}$ )

given by the processors

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Drinking water in the United Kingdom has a mean  $K^+$  value of 2.5 mg L<sup>-1</sup> and an upper 90<sup>th</sup> percentile concentration of 5.2 mg L<sup>-1</sup>, according to research [27]. Its value is equal to that of the bottled water with code of SBW5 which is greater than the recommended value of the Ethiopian standard. The K<sup>+</sup> concentration in some water brands is lower than the standard values, which does not pose health concerns

The activity concentrations of natural radionuclides <sup>238</sup>U, <sup>232</sup>Th, and <sup>40</sup>K in different bottled water samples available in Dire Dawa City are shown in Table 4. It is observed that the specific activity in bottled drinking water samples due to <sup>238</sup>U varies from  $2.91 \pm 0.42$  Bq L<sup>-1</sup> to  $4.04 \pm 0.42$  Bq L<sup>-1</sup> with an average of  $3.50 \pm 0.42$  Bq L<sup>-1</sup>, for <sup>232</sup>Th, varies from  $4.66 \pm 2.48$  Bq L<sup>-1</sup> to  $13.31 \pm 2.48$  Bq L<sup>-1</sup>, and for <sup>40</sup>K, ranges from  $106.38 \pm 8.71$  to  $136.52 \pm 8.71$  Bq L<sup>-1</sup> with an average of  $121.24 \pm 8.31$  Bq L<sup>-1</sup>. Table 4 shows that the specific activity of <sup>238</sup>U, <sup>232</sup>Th, and <sup>40</sup>K in all samples are varied, which may be attributed to the mineral contents of the drinking water. The result revealed that the highest specific activity of <sup>40</sup>K was obtained in this work.

#### Table 4

The specific activity of natural radionuclides  $^{238}\text{U},\,^{232}\text{Th},$  and  $^{40}\text{K}$  sampled bottled water in Dire Dawa.

Name	<sup>238</sup> U (Bq L <sup>-1</sup> )	<sup>232</sup> Th (Bq L <sup>-1</sup> )	<sup>40</sup> K (Bq L <sup>-1</sup> )
SBW1	3.07	12.49	124.58
SBW2	3.99	12.20	118.58
SBW3	1.89	9.55	3.00
SBW4	3.21	10.53	124.83
SBW5	3.17	10.47	106.38
SBW6	3.68	10.85	117.81
SBW7	3.76	11.21	122.90
SBW8	2.91	13.31	112.60
SBW9	3.70	11.18	126.94
Average	3.26	11.31	106.40
Max	3.99	13.31	126.94
Min	1.89	9.55	3.00
Std	0.63	1.16	39.33

Table 5 shows the radioactive equivalent, rate of absorption, external and internal annual effective dose, and external and internal risk level. The radium equivalent of bottled water varies from  $15.78 \pm 4.67$  to  $30.61 \pm 4.67$  Bq L<sup>-1</sup>, with an average of  $27.63 \pm 4.67$  Bq L<sup>-1</sup>. The radium equivalent has a peak in SBW8-code bottled water. The absorbed dose rate of bottled water produced and distributed in Dire Dawa City and the surrounding was varied from  $6.95 \pm 2.36$  to  $14.39 \pm 2.36$  nGy h<sup>-1</sup> with an average of  $12.99 \pm 2.36$  nGy h<sup>-1</sup>. The absorbed dose rate has its peak in SBW1 code.

The annual effective dose for the residence consumer due to intake of radon from bottled water was evaluated using Eqs. 6 and 7. The result shows that the annual

effective dose rate for the outdoors varied from 0.01 to 0.02 mSv  $y^{-1}$  with an average of 0.02 mSv  $y^{-1}$  whereas, the indoors, it varied from 0.03 to 0.07 mSv  $y^{-1}$  with an average value of 0.06 mSv  $y^{-1}$ .

The internal and external hazard indexes have been determined, where the internal hazard index was  $0.05 \pm 0.01$  to  $0.09 \pm 0.01$  while the external hazard index was  $0.04 \pm 0.01$  to  $0.08 \pm 0.01$ , which is below the maximum hazard index according to UNSCEAR [36–37]. The external radiation hazard index is less than 1, and makes bottled water safe to consume.

#### Table 5

Samples	Raeq	AD	$AED [mSv y^{-1}]$		Hex	$H_{ m in}$	Iγ
codes	$(\operatorname{Bq} L^{-1})$	$(nGy h^{-1})$	Outdoor	Indoor			
SBW1	30.52	14.39	0.02	0.07	0.08	0.09	0.23
SBW2	30.57	14.38	0.02	0.07	0.08	0.09	0.23
SBW3	15.78	6.95	0.01	0.03	0.04	0.05	0.11
SBW4	27.88	13.25	0.02	0.06	0.08	0.08	0.21
SBW5	26.33	12.42	0.02	0.06	0.07	0.08	0.20
SBW6	28.27	13.37	0.02	0.07	0.08	0.09	0.21
SBW7	29.25	13.84	0.02	0.07	0.08	0.09	0.22
SBW8	30.61	14.33	0.02	0.07	0.08	0.09	0.23
SBW9	29.46	13.96	0.02	0.07	0.08	0.09	0.22
Average	27.63	12.99	0.02	0.06	0.08	0.08	0.21
Max	30.61	14.39	0.02	0.07	0.08	0.09	0.23
Min	15.78	6.95	0.01	0.03	0.04	0.05	0.11
Std	4.61	2.36	0.00	0.01	0.01	0.02	0.04

The radium equivalent activity, absorbed dose rate, outdoor and indoor annual effective dose, and external and internal hazard index of the samples collected from the city's local market.



Fig. 1. The age-dependence of the annual effective dose of <sup>238</sup>U due to ingestion.



Fig. 2. The age-dependence of the annual effective dose of <sup>40</sup>K radionuclide.

The annual effective dose of  ${}^{40}$ K is shown in Fig. 2. The results revealed that for adults, the annual effective dose varied from 0.01 to 0.07 mSv y<sup>-1</sup> for ages between 12 and 17 years. It varied from 0.01 to 0.58 mSv y<sup>-1</sup> for ages between 7 and 12 years. It was varied from 0.01 to 0.58, for ages 2–7 years. For ages, 1–2 years varied from 0.02 to 0.60 mSv y<sup>-1</sup>. For ages 1–2 years varied from 0.03 to 0.80 mSv y<sup>-1</sup> and for infants, ages 0–1 years varied from 0.04 to 1.57 mSv y<sup>-1</sup>. The minimum annual effective dose of  ${}^{40}$ K was observed in SBW3 sample codes at all ages.



Fig. 3. Age-dependence of the annual effective dose of <sup>232</sup>Th radionuclide.

The annual effective dose of  $^{232}$ Th is shown in Fig. 3. The results revealed that for adults, it varied from 17.5 to 17.25 mSv y<sup>-1</sup>. The range for ages 12 to 17 years varied from 16.20. to 14.75 mSv y<sup>-1</sup>. For ages between 7–12 varied from





Fig. 4. Cancer risk due consumption of bottled water contaminated with <sup>238</sup>U and <sup>232</sup>Th.

The cancer risk due to bottled water is shown in Fig. 4. The cancer risk associated with bottled drinking water for <sup>238</sup>U varied from  $6.44 \times 10^{-4}$  to  $1.36 \times 10^{-3}$  and mean value of  $1.11 \times 10^{-3}$ . Similarly, <sup>232</sup>Th cancer risk varied from  $9.08 \times 10^{-3}$  to  $1.26 \times 10^{-2}$  with a mean value of  $1.07 \times 10^{-2}$ . The results of this study showed that the estimates of the effective annual dose and the risk of cancer in humans based on the concentration of <sup>238</sup>U and <sup>232</sup>Th differ from the previous results [11].

#### DISCUSSION

The specific activities of <sup>238</sup>U <sup>232</sup>Th, and <sup>40</sup>K levels were reported in other studies carried out, especially when compared with bottled water studies from Bangladesh, Saudi Arabia, Ghana, Pakistan, Iran, and Dire Dawa shown in Table 6. The result revealed that the specific activity of <sup>238</sup>U is lower than the value obtained in Pakistan. The present finding of radionuclide specific activities is moderate relative to other countries mentioned in Table 6.

The average <sup>232</sup>Th concentration obtained in this work is higher than that in other countries, as shown in Table 6. When compared to the well water from the other cities (Table 6), the bottled water had a higher specific activity. In addition, one of the samples had a higher specific activity of  $^{40}$ K than the water from the other countries (Table 6).

As one can see in Table 5, the  $Ra_{eq}$  (radium equivalent activity) is lower than the international limit mentioned in [2]. Moreover, the result obtained by [1] in groundwater, in the same city and in the summer season, was higher than that in the current study.

The quantities commonly used for estimating the exposure of the population to terrestrial radionuclides are the absorbed dose rate and the annual effective dose. The calculated average absorbed dose rate was 12.99 nGy  $h^{-1}$  which was lower than the average value of 60 nGy  $h^{-1}$  [36]. It could not pose any health risk to the people staying in the area, as they will receive low doses of these harmful radionuclides.

This study shows that the total annual effective dose in the bottled water sample for outdoor and indoor exposures is much smaller than the internal exposures shown in [36] and reported an average value of  $0.12 \text{ mSv y}^{-1}$ , which is lower than the limit of  $0.1 \text{ mSv y}^{-1}$  [41] and lower than the ICRP preference limit of  $1.0 \text{ mSv y}^{-1}$  [24, 39–41].

The average external hazard index  $(H_{ex})$  values were 0.08, and the internal hazard index  $(H_{in})$  was 0.08. These values were much less than unity and are considered insignificant and safe for humans consuming bottled drinking water.

The cancer risk for adults was shown in Fig. 5 and compared to [11]. The estimation coefficients for <sup>238</sup>U and <sup>232</sup>Th in samples 3 and 8 were compared with the coefficients mentioned in [14]. The findings of the remaining are higher than the values listed in [11, 14].

Country	Types of	Parameters	<sup>226</sup> Ra	<sup>232</sup> Th	<sup>40</sup> K	Refs	
	water		$(Bq L^{-1})$	$({\rm Bq} {\rm L}^{-1})$	$(Bq L^{-1})$		
Bangladesh	Bottled water	Range	1.89-4.97	1.42-9.72	10.88-32.24	[20]	
		Average	3.28	6.40	18.26	[28]	
Saudi	Crowndwyster	Range	0.24-1.52	0.15-0.81	0.66-5.64	[6]	
Arabia	Groundwater	Average	0.85	0.43	2.84		
Ghana	Bottled water	Range	0.00-0.53	0.30-0.56	3.57-5.74	[27]	
		Average	0.14	0.43	4.87	[20]	
Saudi	Bottled water	Range	0.21-2.25	0.37-3.00	0.24-33.74	[5]	
Arabia		Average	0.77	1.30	11.10	[5]	
Delaistan	Dettledeneter	Range	8.00-15.00	4.00-6.00	92.00-216.00	[17]	
Pakistan	Bottled water	Average	11.30	5.20	140.90	[10]	
		Range	0.12-2.84	0.39-7.47	2.93-7.17	[10]	
Iran	Groundwater	Average	0.67	1.65	4.72	[19]	
Ethiopia,	thiopia,		2.07-6.38	2.43-10.56	109.27-221.50	[1]	
Dire Dawa	Groundwater	Average	4.36	7.81	131.26		
Ethiopia,		Range	1.89-3.99	9.55-13.35	3.00-126.94	This	
Dire Dawa	Bottled water	Average	3.26	11.31	106.40	study	
Nigeria Well	337 11 4	Range	1.47-7.28	1.67-3.36	46.93-97.62	[22]	
	wen water	Average	3.61	2.26	60.84	[33]	

#### Table 6

Comparison of specific activity naturally occurring of radionuclides in bottled and groundwater in the area (range and average values)

#### CONCLUSIONS AND RECOMMENDATIONS

The most popular bottled mineral waters or springs in Dire Dawa were examined in terms of their radioactive background radiation, radiation equivalent, absorbed volume, and potential health hazards. The selected analytical approach ensures low detection limits and precise results.

A total of nine brands of mineral bottled water samples were investigated for the content of <sup>238</sup>U, <sup>232</sup>Th, and <sup>40</sup>K. The different brands had different concentrations of radionuclides, and the corresponding average annual effective doses did not exceed the recommended WHO limit.

The average specific activities of  $^{238}$ U,  $^{232}$ Th, and  $^{40}$ K are 3.26, 11.31, and 106.40 Bq L<sup>-1</sup>, respectively. These values are below the current WHO guideline recommendations and the reported concentrations in drinking water in different countries.

In comparison to  $^{238}$ U and  $^{232}$ Th, the specific activity of  $^{40}$ K, a naturally occurring radionuclide in the Earth's crust and the human body, this one is the primary contributor in all of the chosen bottled water samples. The  $^{40}$ K average specific activity was higher than those in some works in Table 6.

The data generated in this study will provide a solid baseline for setting the standard quality of bottled drinking water in Ethiopia.

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### REFERENCES

- ABAS, E. B. SITOTAW, E. MENGISTU, Evaluation of radionuclides specific activity of groundwater resources. A case of Dire Dawa City, Ethiopia, *Romanian J. Biophys.*, 2021, 31(1), 1–11.
- ABBADY, A.G.E., Estimation of radiation hazard indices from sedimentary rocks in Upper Egypt, *Appl. Radiat. Isot.*, 2004, 60(1), 111–114.
- AJAYI, O.S., T.P. OWOLABI, Determination of natural radioactivity in drinking water in private Dug Wells in Akure, Southwestern Nigeria, *Radiation Protection Dosimetry*, 2008, 128(4), 477–484.
- ALEMU, Z.A., K.T. TEKLU, A.A. TSIGEREDA, K.H. BALCHA, S.D. MENEGESHA, Physicochemical quality of drinking water sources in Ethiopia and its health impact: a retrospective study, *Environ. Syst. Res.*, 2015, 22(4), 1-8, DOI 10.1186/s40068-015-0049-7.
- AL-GHAMDI, A.H., Activity concentrations in bottled drinking water in Saudi Arabia and consequent dose estimates, *Life Science Journal*, 2014, 11(9), 771-777.
- AL-GHAMDI, A.H., Radioactivity measurements and radiation dose assessments in ground water of Al-Baha region, Saudi Arabia, *Journal of Geoscience and Environment Protection*, 2019, 7(10), 112–119, https://doi.org/10.4236/gep.2019.710009.

- AL-SALEH, F.S., B. AL-BERZAN, Measurements of natural radioactivity in some kinds of marble and granite used in Riyadh region, *Journal Nuclear Radiation Physics*, 2007, 2(1), 25–36.
- AYALEW, D., B. SITOTAW, E. MENGISTU, Assessment of natural radioactivity levels in the soil of Dire Dawa city, Ethiopia, *Romanian J. Biophys*, 2019, 29(4), 113–122.
- 9. DE OLIVERIA, J., B. PACI MAZZILLI, P. DA COSTA, P.A. TANJGAVA, Natural radioactivity in Brazilian bottled mineral waters and consequent doses, *Journal of Radioanalytical and Nuclear Chemistry*, 2001, **249**(1), 173–176.
- EHSANPOU, E., M.R. ABDI, M. MOSTAJABODDAVATI, H. BAGHERI, <sup>226</sup>Ra, <sup>232</sup>Th, and <sup>40</sup>K contents in water samples in part of central deserts in Iran and their potential radiological risk to human population, *Journal of Environmental Health Science & Engineering*, 2014, **12**(1), 1–7.
- EL-GAMAL, H., A. SEFELNASR, G. SALAHEDIN, Determination of natural radionuclides for water resources on the West Bank of the Nile River, Assiut Governorate, Egypt, *Water*, 2019, 11, 311–344
- 12. EL ARABI, A.M., N.K, AHMED, D.K. SALAHEL, Natural radionuclides and dose estimation in natural water resources from Elba Protective Area, Egypt, *Radiation Protection Dosimetry* 2006, **121**(3), 284–292.
- 13. EL-MAGEED, A.I.A., A.E.H. EL-KAMEL, A.E.B. ABBACY, S. HERB, I.I. SALE, Natural radioactivity of ground and hot spring water in some areas in Yemen, *Desalination*, 2013, **321**, 28–31.
- 14. ENVIRONMENTAL PROTECTION AGENCY (EPA), Radon in drinking water health risk reduction and cost analysis, *Fed. Reg.*, 1999, **64**, 9560–9599.
- ETHIOPIAN STANDARD (ES), Drinking Water Specification, 5<sup>th</sup> ed., Quality and Standards Authority of Ethiopia, Addis Ababa, 2013.
- FATIMA, I., J.H. ZAIDI, M. ARIF, S.N.A. TAHIR, Measurement of natural radioactivity in bottled drinking water in Pakistan and consequent dose estimates, *Radiat. Prot. Dosim.*, 2006, 123(2), 234–240.
- 17. FONOLLOSA, E., A. PENALVER, F. BORRULL, C. AGULAR, Radon in spring waters in the south of Catalonia, *Journal of Environmental Radioactivity*, 2016, **151**, 275–281.
- HAMZA, Z.M., S.A. KADHIM, H.H. HUSSEIN, Assessment of the norms for agricultural soils in Ghammas town, Iraq, *Plant Archives*, 2019, **19**(1), 1483–1490.
- KABADAYI, O., H. GU'MU'S, Natural activity concentrations in bottled drinking water and consequent doses, *Radiation Protection Dosimetry*, 2012, 150(4), 532–535.
- KINAHAN, A., M. HOSODA K. KELLEHER, T. TSUJIGUCHI, N. AKATA, S. TOKONAMI, L. CURRIVAN, L. VINTRÓ, Assessment of radiation dose from the consumption of bottled drinking water in Japan, *International Journal of Environmental Research and Public Health*, 2020, 17(14), 1–12, doi:10.3390/ijerph17144992.
- 21. KOCHER, D.C., A.L. SJOREEN, Dose-rate conversion factors for external exposure to photon emitters in soil, *Health Phys.*,1985, **48**, 193–205.
- 22. MONICA, S., A.K. VISNU PRASAD, S.R. SONIYA, P.J. JOJO, Estimation of indoor and outdoor effective doses and lifetime cancer risk from gamma dose rates along the coastal regions of Kollam district, Kerala, *Radiat. Prot. Environ.*, 2016, **39**, 38–43.
- MOUNTFORD, P.J, D.H. TEMPERTON, Recommendations of the International Commission on Radiological Protection (ICRP) 1990, *Eur. J. Nucl. Med.*, 1992, 19(2), 77–79, doi: 10.1007/BF00184120. PMID: 1563443.
- NIKOLOPOULOS, D., E. PETRAKI, A. MAROUSAKI, S. POTIRAKIS, G. KOULOURAS, Environmental monitoring of radon in the soil during a very seismically active period occurred in South West Greece, *Journal of Environmental Monitoring*, 2012, 14, 564–578.
- PORTUPHY, M.O., A. FAANU, A. SAWYERR, Radiological risk assessment due to ingestion of some bottled drinking water on the Ghanaian market, *Ghana J. Sci.*, 2018, 59, 93–102.
- POWELL, P., R.J. BAILEY, P.K JOLLY, Trace elements in British tap-water supplies, Swindon, WRc (Report PRD 706-M/1), 1987.

- RAHMAN, M.M., A. MONDAL, M.A. KABIR, K. ASADUZZAMAN, Assessment of natural radioactivity levels and radiological significance of bottled drinking water in Bangladesh, *American Journal of Physics and Applications*, 2015, 3(6), 203–207, doi: 10.11648/j.ajpa.20150306.13.
- SAM, A.K., N. ABBAS, Assessment of radioactivity and associated hazards in local and imported cement types used in Sudan, *Radiation Protection Dosimetry*, 2010, 93(1), 275–277.
- SEGHOUR, A., F.Z. SEGHOUR, Radium, and <sup>40</sup>K in Algerian bottled mineral waters, and consequent doses, *Radiation Protection Dosimetry*, 2009, **133**(1), 50–57.
- SHIVAKUMARA, B.C., M.S. CHANDRASHEKARA, E. KAVITHA, L. PARAMESH, Studies on <sup>226</sup>Ra and <sup>222</sup>Rn concentration in drinking water of Mandya Region, Karnataka State, India, *Journal of Radiation Research and Applied Sciences*, 2014, 7(4) 491–498.
- 31. SIVAPAN, C.H.O.O., The relationship between the total dissolved solids and the conductivity value of drinking water, surface water, and wastewater, *International Academic Research Conference in Amsterdam*, 2019.
- TANASKOVIC, I., M. ERENIC-SAVKOVIC, L.J. JAVORINA, Radioactivity of spa water in Serbia, *The Symposium of Society for Radiation Protection of Serbia and Montenegro*, *Proceedings*, 12–14 October, Tara, Belgrade, Serbia, 2011, 137–140.
- TCHOKOSSA, P., J. OLOMO, F. BALOGUN, Assessment of radionuclide concentrations and absorbed dose from consumption of community water supplies in oil and gas producing areas in Delta State, Nigeria, *World Journal of Nuclear Science and Technology*, 2011, 1(3), 77-86, doi: 10.4236/wjnst.2011.13012.
- 34. TOLA, F.B., G.T. ANBESSA, B.B. YIKNA, Anti-tuberculosis commodities management performance and factors affecting it at public health facilities in Dire Dawa city administration, Ethiopia, *Journal of Multidisciplinary Healthcare*, 2020, **13**, 1677–1691.
- 35. UNITED NATIONS SCIENTIFIC COMMITTEE ON THE EFFECTS OF ATOMIC RADIATION (UNSCEAR), Sources, and Effects of Ionizing Radiation. Report to the General Assembly, United Nations, New York, 1993.
- 36. UNITED NATIONS SCIENTIFIC COMMITTEE ON THE EFFECTS OF ATOMIC RADIATION (UNSCEAR), *Report to the General Assembly, with Scientific Annexes*, Volume I: SOURCES; United Nations, New York, NY, USA, 2000.
- UNITED NATIONS SCIENTIFIC COMMITTEE ON THE EFFECTS OF ATOMIC (UNSCEAR), *Radiation Sources, and Effects of Ionizing Radiation*, Report to the General Assembly, with Scientific Annexes, United Nations, New York, 2008.
- WALSH, M., G. WALLNER, P. JENNINGS, Radioactivity in drinking water supplies in Western Australia, *Journal of Environmental Radioactivity*, 2014, 130, 56-62.
- 39. WORLD HEALTH ORGANIZATION (WHO), *Management of Radioactivity in Drinking Water*, World Health Organization, Geneva, Switzerland, 2018.
- WORLD HEALTH ORGANIZATION (WHO), Management of Radioactivity in Drinking Water, World Health Organization, Geneva, Switzerland, 2017, 203–218.
- WORLD HEALTH ORGANIZATION (WHO), *Guidelines for Drinking-Water Quality*, 2<sup>nd</sup> ed. Vol. 2, Health criteria and other supporting information., Geneva, 1996.
- 42. YOUSEF, H.A, Assessment of the annual effective dose of bottled mineral waters using closed can technique, *Journal of Advances in Physics*, 2018, **4**(3), 5696–5707.