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# Activity concentrations in bottled drinking water in Saudi Arabia and consequent dose estimates

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**Abstract**

The natural radioactivity concentrations of $^{226}Ra$, $^{232}Th$ and $^{40}K$ were determined in different brands of bottled drinking water samples collected from the local market of Saudi Arabia. The average measured activity concentrations of the nuclides were found to be 0.77, 1.3 and 11.1 Bq $l^{-1}$, respectively. The total average annual effective doses due to all three natural radionuclides on different age groups of infants, children and adults were estimated. The measured activity concentrations were compared with similar studies and the activity concentrations of natural radionuclides in bottled drinking water were found to be lower than most of these values. Also, the effective doses resulting from the consumption of bottled drinking water estimated for the three age groups were found to be below the WHO recommended limit of 0.1 mSv $y^{-1}$. The results can serve as baseline levels of activity concentration in drinking water in Saudi Arabia.

Keywords: Natural radioactivity, effective dose.

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## 1. Introduction

The most common radionuclides found in water are $^{222}$Rn, $^{226}$Ra and $^{234}$U from the radioactive decay of $^{238}$U, and $^{228}$Ra of $^{232}$Th decay series. The most radiotoxic and hazardous among them are $^{226}$Ra and $^{228}$Ra, which behave like calcium when absorbed into the body. Internal exposure of humans to high levels of radium for a long time may produce bone and sinus cancers [2]. The accurate measurement of the activity concentration of naturally occurring radionuclides in drinking water is useful for determining human exposure to ionising radiation by ingestion and domestic uses because the doses from these pathways are strongly related to the amount of radionuclides present. It is also an important parameter for the radiological protection of the population from bottled drinking water [7, 13].

The United Nations Scientific Committee on the Effects of Atomic Radiation [15] has estimated that exposure to natural sources contributes >70% of the population radiation dose and the global average human exposure from natural sources is 2.4 mSv y$^{-1}$ (cosmic ray 0.4, terrestrial gamma ray 0.5, radon 1.2, and food and drinking water 0.3).

The World Health Organization (WHO) has recommended safe values for various drinking water quality parameters in its general guidelines [19]. These guidelines have been used by different countries to formulate their own national waterquality standards. Unsafe drinking water is the cause of many diseases in many underdeveloped and developing countries of the world.

In Saudi Arabia, consumption of bottled drinking water has increased in recent years due to practicality and availability of this water distribution method and due to the public’s conception of the importance of mineral water in the human diet and the medicinal benefits of the dissolved mineral components for the human body. For this reason, mineral water suppliers have achieved rapid growth; most of them are installed near the source where the water is bottled and distributed to markets mainly, in Saudi Arabia’s larger cities. The quality of bottled drinking water varies from place to place, depending on the condition of the source water from which it is drawn and the treatment it receives. Therefore, public health hazard assessment is essential due to consumption of these bottled waters.

The occurrence of natural radionuclides in drinking water has been extensively studied with the objective to assess the safety of drinking water with respect to its radionuclide content [1, 2, 4 7, 11, 12, 16, and 20].

The aim of this study is to determine natural radionuclide’s activities in the most-consumed bottled drinking waters (imported and exported) in Saudi Arabia (Jeddah city) from various manufacturers. The measurement results found in this study can thus be used to estimate the effective doses for different age categories of the public. The data generated in this study may contribute to determine the baseline levels of natural radioactivity in drinking bottled water and help in the development of future guidelines in the country for radiological protection of the population.

## 2. Materials and methods

### *2.1. Samples*

Twenty two samples of bottled water were collected randomly from local stores located in Jeddah city, Saudi Arabia. The date of purchase was March 2014. About 0.4 L of each sample was filled in a Marinelli beaker and sealed air tight to ensure that $^{222}$Rn was not lost during transfer, all the prepared samples were stored for at least 1 month, enough time to restore secular equilibrium between $^{222}$Rn and its radioactive descendants.

### *2.2. Gamma-ray spectroscopy*

The activity concentrations of water samples were measured using hyper pure germanium detector (HPGe) vertical coaxial detectors (Ortec) with 25% efficiency and 2 keV resolution at 1332 keV gamma line of $^{60}Co$ employed for all the measurements. The detector is housed inside a thick lead shield to reduce the background of the system. The system was calibrated for energy and efficiency, where the energy calibration was carried out by counting the radioactive standards of known energies such as $^{60}Co$, for Eɣ (1332.5 and 1173.2 keV), and $^{137}Cs$, for Eɣ (661.6 keV). The efficiency calibration was performed using the point source of $^{152}Eu$ IAEA [9]. The background radiation measurement was performed every week under the same conditions of sample, where an empty plastic bottle washed with dilute HCl with distilled water was counted for this purpose. Each sample after equilibrium was placed on top of the HPGe detector and counted for 828000 sec. The detector was coupled to a Canberra Multichannel Analyzer computer system.

The characteristic gamma peaks selected for the determination of the different radionuclides were 295.1 and 351.9 keV ($^{214}Pb$) and 609.3 keV ($^{214}Bi$) for $^{226}Ra$, 911.1 and 968.97 keV ($^{228}Ac$) and 583.2 keV ($^{208}Tl$) for $^{232}Th$, while the $^{40}K$ activity was determined from the 1460.7 keV emission [4].

### *2.3. Natural specific activity measurement*

The activities of the radionuclides were calculated using the following equation [5]:

$$A\,(\mathrm{Bq\,l^{-1}}) = \frac{Ca}{\varepsilon P_r} \tag{1}$$

where A is the activity of the radionuclide in Bq $l^{-1}$, $C_a$ is the counts per second, $\varepsilon$ is the detection absolute efficiency at a specific γ–ray energy and $P_r$ is the emission probability of Gamma-decay and **V** is the volume of the water sample in liters.

### *2.4. Estimation of annual effective doses*

The annual effective dose to an individual due to intake of natural radionuclides from the bottled drinking water is estimated using the following relationship [3, 15]:

$$E_d = A_c A_i C_f \tag{2}$$

where $E_d$ is the annual effective dose (Sv $y^{-1}$) to an individual due to the ingestion of radionuclides from drinking water, *Ac* is the activity concentration of the ingested drinking water (Bq $l^{-1}$), $C_f$ is the ingested dose conversion factor for radionuclides (Sv $Bq^{-1}$), which varies with both radioisotopes and the age of the individual as listed in Table 1 [8], *Ai* is the annual intake of bottled drinking water (l $y^{-1}$), which is calculated for different age groups of population : for infants (1–2 y), children (age from 2–7 y, 7–12 and 12–17 y) and adults (age from 17 y and above). Table 1 shows the age-dependent annual consumption per year from the Risica and Grande [14].

The total effective dose D (Sv $y^{-1}$) to an individual was established by summing contributions from all radionuclides present in the water samples as

$$D = \sum A_c A_i Cf \tag{3}$$

**Table 1. Dose conversion factors for ingestion of radionuclides in water and water consumption for different age groups (l $y^{-1}$)**

| Age groups (year) | Dose conversion factors (Sv $Bq^{-1}$) $^{226}Ra$ | $^{232}Th$ | $^{40}K$ | Water consumption ($ly^{-1}$) |
|---|---|---|---|---|
| 1–2 y | $9.6 \times 10^{-7}$ | $4.5 \times 10^{-7}$ | $4.2 \times 10^{-8}$ | 250 |
| 2–7 y | $6.2 \times 10^{-7}$ | $3.5 \times 10^{-7}$ | $2.1 \times 10^{-8}$ | 350 |
| 7–12 y | $8.0 \times 10^{-7}$ | $2.9 \times 10^{-7}$ | $1.3 \times 10^{-8}$ | 350 |
| 12–17 y | $1.5 \times 10^{-6}$ | $2.5 \times 10^{-7}$ | $7.6 \times 10^{-9}$ | 550 |
| >17 y | $2.8 \times 10^{-7}$ | $2.3 \times 10^{-7}$ | $6.2 \times 10^{-9}$ | 750 |

## 3. Results and discussion

### *3.1. Activity concentrations of analysed radionuclides in bottled water samples*

Measured activity concentrations of natural radionuclides of $^{226}Ra$, $^{232}Th$ and $^{40}K$ in bottled drinking water samples are shown in Table 2. The activity concentration results show that the $^{226}Ra$ concentration ranged from LDL in samples 18 – 2.25 Bq $l^{-1}$ for sample $^{232}Th$, the concentration is found to be LDL Bq $l^{-1}$, for sample 17 and 3.0 Bq $l^{-1}$ for sample 4. $^{40}K$ concentration is found to be ranged from 0.24 in sample 22 to 33.74 Bq $l^{-1}$in sample 1. The mean specific activities of $^{226}Ra$, $^{232}Th$ and $^{40}K$ in the bottled drinking water samples are 0.77, 1.3 and 11.1 Bq $l^{-1}$, respectively. The WHO current Guidelines 1993 and the United States Environmental Protection Agency recommendations [15] allow a maximum $^{226}Ra$ concentration in drinking water of 1000 and 185 mBq $l^{-1}$, respectively. The measured mean value of $^{226}Ra$ concentration in drinking bottled water samples in this study appears within these two limits.

Figure 1 gives the comparison between the activity concentration of $^{226}Ra$, $^{232}Th$ and $^{40}K$ in bottled water under investigation.

**Table 2. Activity concentration (mBq $l^{-1}$) of $^{226}Ra$, $^{232}Th$ and $^{40}$ in bottled drinking water samples**

| Sample code nos. | Origin of sample | Activity concentration range (Bq $l^{-1}$) $^{226}Ra$ | $^{232}Th$ | $^{40}K$ |
|---|---|---|---|---|
| W1 | K.S.A | 1.15 ± 0.001 | 1.50 ± 0.004 | 33.74 ± 0.003 |
| W2 | K.S.A | 0.96 ± 0.001 | 1.72 ± 0.002 | 12.31 ± 0.003 |
| W3 | K.S.A | 0.61 ± 0.001 | 0.37 ± 0.002 | 10.62 ± 0.003 |
| W4 | K.S.A | 2.55 ± 0.001 | 3.00 ± 0.004 | 12.17 ± 0.003 |
| W5 | K.S.A | 0.90 ± 0.006 | 1.38 ± 0.007 | 03.29 ± 0.003 |
| W6 | K.S.A | 0.40 ± 0.001 | 1.13 ± 0.002 | 16.31 ± 0.001 |
| W7 | France | 0.70 ± 0.005 | 1.81 ± 0.002 | 15.85 ± 0.001 |
| W8 | France | 0.95 ± 0.006 | 0.88 ± 0.003 | 05.17 ± 0.004 |
| W9 | Switzerland | 0.94 ± 0.006 | 0.93 ± 0.004 | 03.38 ± 0.001 |
| W10 | K.S.A | 0.72 ± 0.002 | 2.37 ± 0.002 | 02.73 ± 0.003 |
| W11 | France | 0.52 ± 0.006 | 0.83 ± 0.003 | 2.32 ± 0.001 |
| W12 | Scotland | 1.11 ± 0.005 | 1.01 ± 0.002 | 9.54 ± 0.002 |
| W13 | K.S.A | 0.75 ± 0.006 | 1.56 ± 0.001 | 12.22 ± 0.001 |
| W14 | K.S.A | 1.20 ± 0.001 | 1.77 ± 0.002 | 16.07 ± 0.003 |
| W15 | K.S.A | 0.41 ± 0.004 | 0.77 ± 0.003 | 17.34 ± 0.001 |
| W16 | K.S.A | 0.49 ± 0.001 | 1.31 ± 0.003 | 27.86 ± 0.001 |
| W17 | K.S.A | 0.42 ± 0.005 | LDL | 11.79 ± 0.002 |
| W18 | Syria | LDL | 1.14 ± 0.003 | 19.03 ± 0.003 |
| W19 | Lebanon | 0.34 ± 0.001 | 0.53 ± 0.004 | 0.85 ± 0.002 |

| | | | | |
|---|---|---|---|---|
| W20 | K.S.A | 0.64 ± 0.006 | 1.05 ± 0.003 | 06.30 ± 0.001 |
| W21 | K.S.A | 0.40 ± 0.001 | 0.49 ± 0.004 | 05.05 ± 0.004 |
| W22 | K.S.A | 0.86 ± 0.007 | 1.77 ± 0.002 | 00.24 ± 0.003 |
| Minimum | | LDL | 0.37 | 33.74 |
| Maximum | | 2.55 | 3.00 | 0.24 |
| Average | | 0.77 | 1.3 | 11.1 |

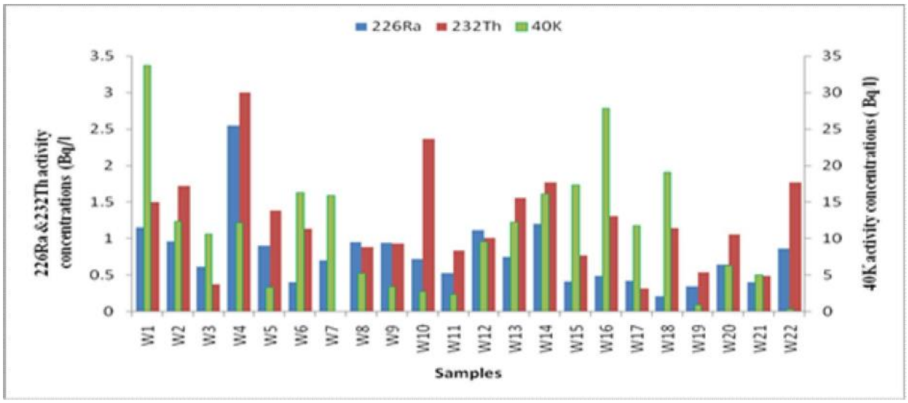


**Figure 1. Comparison between the mean activity concentration of $^{226}$Ra, $^{232}$Th and $^{40}$K in bottled water, Saudi Arabia**

### *3.2. Comparison of results with similar works in other countries*

The activity concentrations of these nuclides from various countries have been compiled in Table 3, together with the values obtained in this work. Measured concentrations of $^{226}$Ra are found to be less than the reported values in drinking water from Turkey (0.517 – 1.22 Bql$^{-1}$), Egypt (1.6 – 0.97 Bql$^{-1}$), Nigeria (0.57 – 26.86 Bql$^{-1}$) and Yemen (2.25 – 3.45 Bql$^{-1}$). Measured $^{232}$Th concentration in drinking water samples is also found below the reported concentration in drinking water of Turkey (0.676 – 0.232 Bq l$^{-1}$), Algeria (0.004 – 0.006 Bq l$^{-1}$), Pakistan (0.004 – 0.006 Bql$^{-1}$), Egypt (0.2 – 1.13 Bq l$^{-1}$), Serbia (0.2 – 1.13 Bql$^{-1}$), Nigeria (0.20 – 60.06 Bql$^{-1}$) and Yemen (0.3 – 1.37 Bql$^{-1}$). The presented results show that $^{40}$K concentration is somewhat lower than the data presented from Nigeria and is found to be greater than the values in drinking water from the other parts of the world as shown in Table 3.

The comparison of results with data from different countries shows that the results of this study are consistent with result from different locations in the world. The data presented in Table 3 varies by country since geographical locations differ in terms of characteristic mineral.

**Table 3. Comparison of measurement results of activity concentrations of $^{226}$Ra, $^{232}$Th and $^{40}$K from various countries.**

| Country | Water source | Activity concentration (Bq l$^{-1}$) $^{226}$Ra | $^{232}$Th | $^{40}$K | Reference |
|---|---|---|---|---|---|
| Saudi Arabia | Bottled drinking water | 0.21 – 2.25 | 0.37 – 0.232 | 0.24 – 33.74 | Present study |
| Algeria | Bottled drinking water | 0.013 – 0.148 | 0.018 – 0.055 | <0.07 – 2.19 | Seghour and Seghourbn [12] |
| Pakistan | Bottled drinking water | 0.008 – 0.015 | 0.004 – 0.006 | 0.092 – 0.216 | Fatima *et al.* [7] |
| Turkey | Bottled drinking water | 0.517 – 1.22 | 0.676 – 0.232 | 1.54 – 2.57 | Onder Kabadayi and Hasan [11] |
| Egypt | Natural water | 1.6 – 0.97 | 0.21 – 1.1 | 9.7 – 23.0 | El Arabi *et al.* [4] |
| Serbia | Drinking water | 0.01 – 0.530 | 0.2 – 1.13 | | Tanaskovic |

| | | | | | |
|---|---|---|---|---|---|
| | | | | | *et al.* [20] |
| Nigeria | Drinking water | 0.57 – 26.86 | 0.20 – 60.06 | 0.35 – 29.01 | Ajayi and Owolabi [2] |
| Yemen | Ground drinking water (Juban) | 2.25 – 3.45 | 0.3 – 1.37 | 26.73 | Abdallah Ibrahim *et al*. [1] |

### *3.3. Annual effective dose for different age groups*

As shown in Table 4, the results of the calculated age-dependent annual effective dose (μSv $y^{-1}$) indicate that:-

1) The annual effective dose due to the intake of $^{226}Ra$ varies from the minimum value 0.051 μSv $y^{-1}$ to the maximum value 0.587 μSv $y^{-1}$ with an average 0.18 μSv $y^{-1}$ for infants (1–2 y). For children age groups (2–7, 7–12 and 12–17 y), $^{226}Ra$ varies from 0.046 to 0.553 μSv $y^{-}$ with an average value of 0.17 μSv $y^{-}$, from 0.059 to 0.714 μSv $y^{-}$ with an average value 0.22 μSv $y^{-}$ and from 0.173 to 2.103 μSv $y^{-}$ with an average value of 0.64 μSv $y^{-}$ respectively. For adults (age >17 y), the minimum annual effective value is 0.044 μSv $y^{-}$ and the maximum value is 0.536 μSv $y^{-1}$ with an average 0.16 μSv $y^{-1}$. $^{226}Ra$ is a highly radiotoxic radionuclide; when humans ingest radium, 20% is absorbed into the bloodstream, the absorbed radium is initially distributed to soft tissues and bones, but its retention is mainly in the growing bones [4].
2) The annual effective dose due to the intake of $^{232}Th$ varies from 0.042 μSv $y^{-1}$ to 0.338 μSv $y^{-1}$, with an average value of 0.15 μSv $y^{-1}$ for infants (1–2 y) and varies from 0.045 to 0.368 μSv $y^{-1}$ with an average of 0.16 from 0.038 to 0.305 with an average of 0.13 μSv $y^{-1}$ and from 0.054 to 0.13 with average 0.19 for children age groups (2–7, 7–12 and 12–17 y) respectively. For adults, $^{232}Th$ varies from a minimum value of 0.064 to a maximum value of 0.518 with an average value of 0.22 μSv $y^{-1}$.
3) The annual effective dose due to the intake of $^{40}K$ has been estimated ranging from 0.003 to 0.354 μSv $y^{-1}$ with average 0.12 μSv $y^{-1}$ for infants (1–2 y). For children, the minimum annual values are 0.002, 0.001 μSv $y^{-1}$ for age (2–7 y, 7–12 y & 12–17 y) and the maximum values 0.25 μSv $y^{-1}$ (2–7 y), 0.153 μSv $y^{-1}$ (7–12 y) and 0.14 μSv $y^{-1}$ (12–17 y) with averages of 0.08 for (2–7 y) and 0.05 μSv $y^{-1}$ for(7–12 and 12–17 y). For adults the annual effective dose varies from a minimum value of 0.001 μSv $y^{-1}$ to the maximum value 0.157 μSv $y^{-1}$ with an average of 0.05 μSv $y^{-1}$. It appears that the average annual dose from $^{40}K$ is below the reference value of 1.0 $mSvy^{-1}$ recommended by ICRP in all ages.
4) The total average annual doses received from the intake of $^{226}Ra$, $^{232}Th$ and $^{40}K$ due to the ingestion of bottled drinking water were (0.45 μSv $y^{-1}$) for infants, (0.41, 0.4 & 0.87 μSv $y^{-1}$) for children age groups and 0.43 μSv $y^{-1}$ for adults, as shown in Table 4. This indicates that doses received by children aged 12–17 y are higher than that received by other ages of children, infants and adults. The main dose contribution is caused by $^{226}Ra$ in the bones, so the children age groups have a higher risk factor because of their intensive bone growth during these years and action should be taken to restrict their intake. Also, it can be seen that the total annual effective doses due to natural radioactivity in bottled drinking water samples calculated for different age groups are much below the recommended reference level of 0.26, 0.2 and 0.1 mSv $year^{-1}$ for effective dose for infants, children and adults, respectively, published by WHO [18], IAEA [10] and UNSCEAR [15].
5) The results in Table 4 give the contributions of $^{226}Ra$ to the total average annual effective dose due to the intake of bottled water; 40% for infants, (41%, 55% and 73%) for children and (37%) for adults. For the natural radionuclide $^{232}Th$, the results showed that it is the second largest contributor to the total annual effective dose, contributing 33% for infants, children (39%, 33% and 22%) and the largest for adults 51%. The lowest contributions due to the intake of $^{40}K$ to the total annual effective dose are 27%, (0%, 12% and 5%) and 12% for infants, children and adults, respectively.

A comparison of the average annual effective dose (µSv $y^{-1}$) for all age groups in this study are summarised in Figure 2. and the total average of effective dose for all age groups are presented in Figure 3.

**Table 4. Annual effective dose (mSv $y^{-1}$) due to the intake of natural radionuclides of $^{226}Ra$, $^{232}Th$ and $^{40}K$ from bottled drinking water.**

| Radionuclide | | Effective dose (µSv/y) | | | | > 17 |
|---|---|---|---|---|---|---|
| | | Infants | | Children | | Adults |
| | | 1–2 y | 2–7 y | 7–12 y | 12–17 y | y |
| $^{226}Ra$ | Minimum | 0.051 | 0.046 | 0.059 | 0.173 | 0.044 |
| | Maximum | 0.587 | 0.553 | 0.714 | 2.103 | 0.536 |
| | Average | 0.18 | 0.17 | 0.218 | 0.64 | 0.16 |
| $^{232}Th$ | Minimum | 0.042 | 0.045 | 0.038 | 0.054 | 0.0638 |
| | Maximum | 0.338 | 0.368 | 0.305 | 0.435 | 0.518 |
| | Average | 0.15 | 0.16 | 0.132 | 0.19 | 0.22 |
| $^{40}K$22 | Minimum | 0.354 | 0.002 | 0.153 | 0.001 | 0.0011 |
| | Maximum | 0.003 | 0.248 | 0.001 | 0.141 | 0.157 |
| | Average | 0.12 | 0.08 | 0.051 | 0.05 | 0.05 |
| Total Average | | 0.45 | 0.41 | 0.40 | 0.87 | 0.43 |

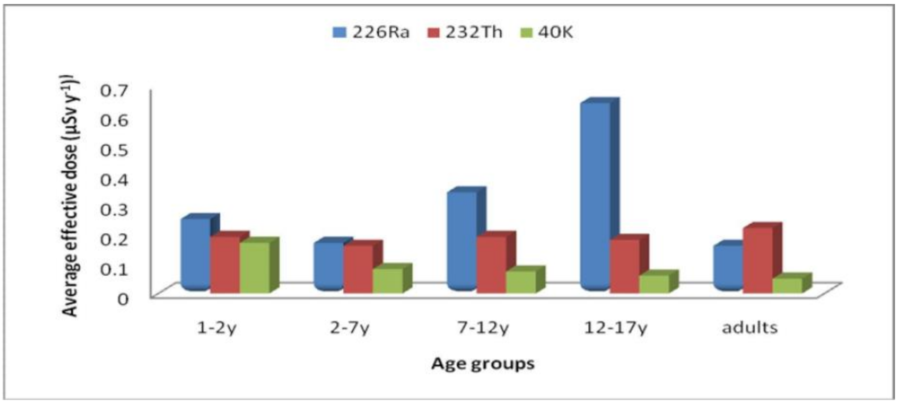


**Figure 2. Comparison between average annual effective dose rate (µSv$y^{-1}$) of natural radioactivity in bottled drinking water consumed by different age groups in Saudi Arabia**

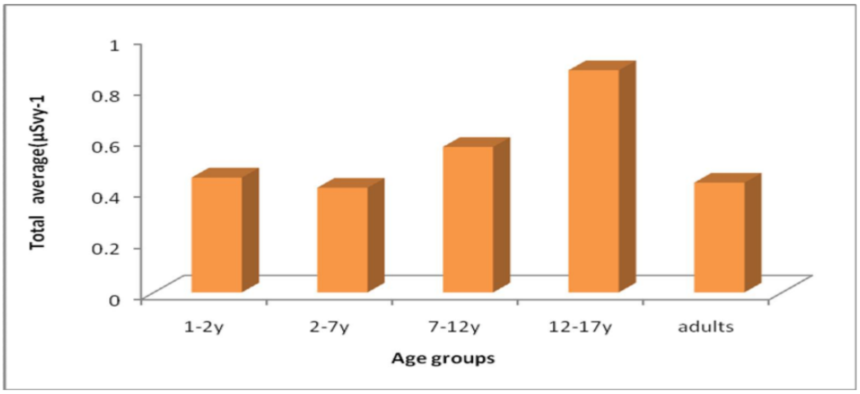


**Figure 3. Total average annual effective dose for all age groups due to the intake of bottled water**

## 4. Conclusion

Activity concentrations of $^{40}$K, $^{238}$U and $^{232}$Th nuclides were measured in bottled drinking water collected from markets in Jeddah city, Saudi Arabia. The mean concentrations of $^{226}$Ra, $^{232}$Th and $^{40}$K are found to be 0.77, 1.3 and 11.1 Bq $l^{-1}$, respectively. These values are found below the reported concentration values in drinking water of countries worldwide reported in the literature and WHO current Guidelines and the United States Environmental Protection Agency.

Annual effective doses for consumption of water for infants (1–2 y), children (2–7 y, 7–12 y and 12–17 y) and adult were calculated for ingestion of radionuclides $^{226}$Ra, $^{232}$Th and $^{40}$K. The total average annual estimated effective doses for $^{226}$Ra, $^{232}$Th and for $^{40}$K are found to be (0.45 µSv $y^{-1}$) for infants (0.41, 0.40 & 0.87 µSv $y^{-1}$) for children age groups and 0.44 µSv $y^{-1}$ for adults, which are below the average limits (0.1 mSv $y^{-1}$) reported by UNSCEAR [15] and WHO [17]. So, the present results for investigated radionuclides in bottled drinking water in Saudi Arabia are below the limit values and pose no detrimental health effects.

The data generated in this study will provide a good baseline for setting the standards quality in bottled drinking water in Saudi Arabia, which can be used to evaluate possible future changes.

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