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 22different brands o f commonly bottled drinking water samples collected from local market of Saudi Arabia (Jeddah city), measured using high-resolution HPGe detector system. The average measured activity concentrations of the nuclides 226Ra, 232Th and 40K are 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 for different age groups of Infants (1–2 y), children (2-7y, 7–12 y) and adults (> 17 y) were estimated to be 0.487, 0.516, 0.629, 0.865 and 0.43 μSv y^{-1}, respectively.The measured activity concentrations have been compared with similar studies from different locations. it was observed that measured activity concentrations of natural radionuclides in the bottled drinking water were lower than most of these values. Also,the effective doses resulting from the consumption of the bottled drinking water were estimated for all three age groups and were found below the World Health Organisation (WHO 2008) recommended limit of 0.1 mSv y^{-1} as well as the average radiation dose of 0.29 mSv y^{-1} received per head worldwide due to ingestion of natural radionuclides assessed by UNSCEAR (2000). The resulting data may serve as base-line levels of activity concentration in drinking waters in Saudi Arabia.

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1. Introduction

The most common radionuclides found in water are 222Rn, 226Ra and 234U from the radioactive decay of 238U; and 228Ra of 232Th decay series. The most radiotoxic and hazardous among them are 226Ra and 228Ra, 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 (Ajayi and Owolabi; 2008). The accurate measurement of the activity concentration of naturally occurring radionuclides in drinking water is useful for determining human population exposure to ionising radiation by ingestion and domestic uses because the doses from these pathways are strongly related to the amount of radionulcides present. It is also an important parameter for the radiological protection of the population from bottled drinking water (Remy and Lemaitre; 1990, Fatima *et al*; 2006)

The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR; 2000) 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 (WHO; 1993). These guidelines have been used by different countries to formulate their own national water quality 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 (El Arabi et al; 2006, Fatima *et al*; 2006, Ajayi and Owolabi; 2008, Seg hour and Seghour; 2009, Tanaskovic et al.; 2011, Onder Kabadayi and Hasan; 2012, Abdallah Ibrahim *et al* 2013 and Walsh *et al* 2014).

The aim of the present 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 base-line 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 Liter of each sample was filled in a Marinelli beaker and sealed air tight to ensure that 222Rn was not lost during transfer, all the prepared samples were stored for at least 1 month, enough time to restore secular equilibrium between 222Rn and its radioactive descendants.

2.2Gamma-ray spectroscopy

The activity concentrations of water samples were measured using hyper pure germanium detector (HPGe) vertical co-axial detectors (Ortec) with 25% efficiency and 2 keV resolution at 1332 keV gamma line of ^{60}Co were 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 60Co, for Eɣ (1332.5 and 1173.2 keV), and 137Cs, for Eɣ (661.6 keV). The efficiency calibration was performed by using point source of 152Eu IAEA (1989).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 828000s. The detector coupled to a Canberra Multichannel Analyzer (MCA) computer system.

The characteristic gamma peaks selected for the determination of the different radionuclides were 295.1 and 351.9 keV (214Pb) and 609.3 keV (214Bi) for 226Ra, 911.1 and 968.97 keV (228Ac) and 583.2 keV (208Tl) for 232Th, while the 40K activity was determined from the 1460.7 keV emission (El Arabi *et al* 2006).

2.3 Natural specific activity measurement

The activities of the radionuclides were calculated using the following equation (El-Taher, 2011):

$$A\ (Bq\ l^{-1}) = Ca / \varepsilon\ P_r\ V \quad \ldots\ldots\ldots\ldots (1)$$

Where A is the activity of the radionuclide in Bq l^{-1}, C_a the counts per second, ε the detection absolute efficiency at a specific γ–ray energy and P_r 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 (Alam *et al*., 1999, UNSCEAR; 2000):

$$E_d = A_c\ A_i\ C_f \quad \ldots\ldots\ldots\ldots (2)$$

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 in 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 are listed in Table (1) (ICRP 1996), Ai is the annual intake of bottled drinking water (l y^{-1}) which is calculated for different age groups of population : for infants (1-2y), children (age from 2-7y, 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 &Grande, 2000.

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:-

$$D = \sum Ac\ Ai\ Cf \quad \ldots\ldots\ldots (3)$$

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})			Water consumption(ly^{-1})
	^{226}Ra	^{232}Th	^{40}K	
1-2y	**9.6×10^{-7}**	**4.5×10^{-7}**	**4.2×10^{-8}**	**250**
2-7 y	**6.2×10^{-7}**	**3.5×10^{-7}**	**2.1×10^{-8}**	**350**
7-12y	**8.0×10^{-7}**	**2.9×10^{-7}**	**1.3×10^{-8}**	**350**
12-17y	**1.5×10^{-6}**	**2.5×10^{-7}**	**7.6×10^{-9}**	**550**
>17 y	**2.8×10^{-7}**	**2.3×10^{-7}**	**6.2×10^{-9}**	**750**

3. Results and Discussion

3.1 Activity concentrations of analyzed radionuclide's in bottled water samples

Measured activity concentrations of natural radionuclides of 226Ra, 232Th and 40K in bottled drinking water samples are shown in Table 2. The

Activity concentrations results show that 226Ra concentration ranged from LDLin sample no 18 to 2.25 Bq l^{-1} for sample no 4. 232Th concentration is found to be LDL Bq l^{-1} for sample no 17 and 3.0Bq l^{-1} for sample no 4. 40K concentration is found to be ranged from 0.24 in sample no22 to 33.74 Bq l^{-1}in sample no 1. The mean specific activities of 226Ra, 232Th and 40K in the bottled drinking water samples are 0.77, 1.3 and 11.1 Bq l^{-1}, respectively. WHO current Guidelines (1993) and the United States Environmental Protection Agency recommendations (UNSCEAR 2000) allow a maximum 226Ra concentration in drinking water of 1000 and 185 mBq l^{-1}, respectively. The measured mean value of 226Ra concentration of in drinking bottled water samples in this study appears within these two limits.

Figure 1 shows 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 226Ra, 232Th and 40 in the bottled drinking water samples

Samples code no.	Origin of sample	Activity concentration range (Bq l^{-1})		
		226Ra	232Th	40K
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

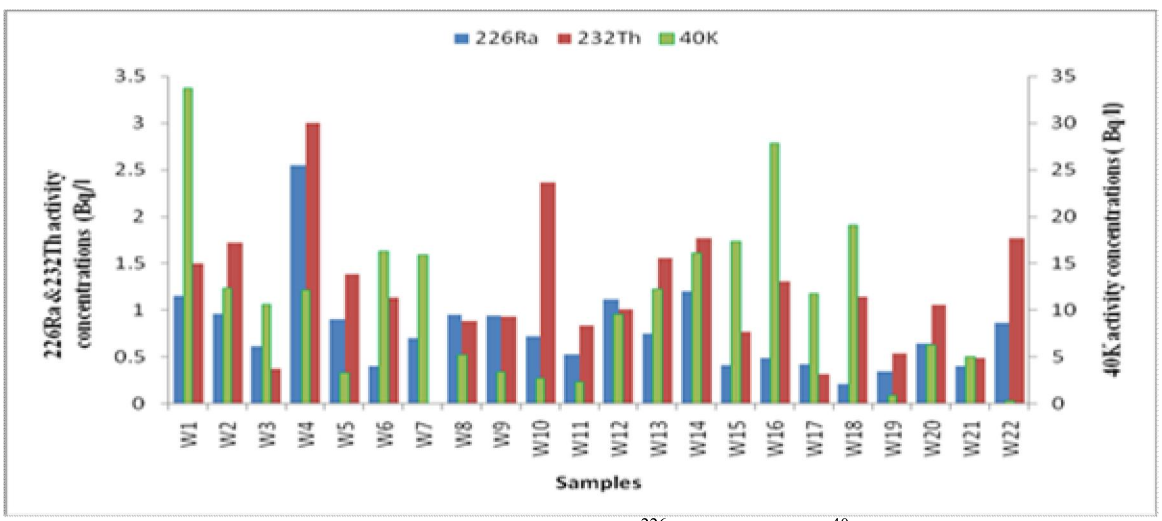

Fig.1: Comparison between the mean activity concentration of ^{226}Ra, 232Th and ^{40}K in bottled water, Saudi Arabia

3.2 Comparison of results with similar in other countries

The activity concentrations of these nuclides from various countries have been compiled in Table 3 together with values obtained in the present work. Measured concentrations of 226Ra 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}), Negeria (0.57-26.86 Bql^{-1}) and Yemen(2.25-3.45 Bql^{-1}). Measured 232Th 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 – 0.006Bql^{-1}), Egypt (0.2- 1.13 Bq l^{-1}), Serbia (0.2- 1.13 Bql^{-1}), Nigeria (0.20- 60.06Bql^{-1}) and Yemen(0.3- 1.37Bql^{-1}). The presented results show that 40K 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 226Ra, 232Th and 40K from various countries.

Country	Water source	Activity concentration (Bq l^{-1})			Reference
		226Ra	232Th	40K	
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	0.01 8 – 0.055	<0.07- 2.19	Seghour A. and Seghour (2009)
Pakistan	bottled drinking water	0.008- 0.015	0.004 – 0.006	0.092 - 0. 216	Fatima et al (2006)
Turkey	bottled drinking water	0.517 - 1.22	0.676- 0.232	1.54 - 2.57	Onder Kabadayi and Hasan (2012)
Egypt	Natural water	1.6–0.97	0.21–1.1	9.7–23.0	El Arabi et al (2006)
Serbia	Drinking water	0.01- 0.530	0.2- 1.13	--------	Tanaskovic et al. (2011)
Nigeria	Drinking water	0.57-26.86	0.20- 60.06	0.35-29.01	Ajayi and Owolabi (2008)
Yemen	Ground drinking water(Juban)	2.25 – 3.45	0.3- 1.37	26.73	Abdallah Ibrahim et al 2013

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 226Ra 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 (1-2y). For children age groups (2-7y, 7-12y &12-17y), 226Ra varies from 0.046 to 0.553 µSv y^{-} with average value 0.17 µSv y^{-}, from 0.059 to 0.714 µSv y^{-} with average value 0.22 µSv y^{-} and from 0.173 to 2.103 µSv y^{-} with average value 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}. 226Ra is a highly radiotoxic radionuclide, when humans ingest radium, 20% is absorbed into the blood stream, the absorbed radium is initially distributed to soft tissues and bones, but its retention is mainly in growing bones (El Arabi et al ;2006).

(2). The annual effective dose due to the intake of 232Th varies from 0.042 µSv y^{-1} to 0.338 µSv y^{-1}, with an average values of 0.15 µSv y^{-1} for Infants (1-2y) and varies from 0.045 to 0.368 µSv y^{-1} with average 0.16, from.038 to 0.305 with average 0.13 µSv y^{-1} and from 0.054 to 0.13 with average 0.19 for children age groups (2-7y,7-12y and 12-17y) respectively. For adults 232Th varies from minimum value 0.064 to maximum value 0.518 with an average value 0.22 µSv y$^{-1.}$

(3). The annual effective dose due to the intake of 40K has been estimated ranging from 0.003 to 0.354 µSv y^{-1} with average 0.12 µSv y^{-1} for Infants (1-2y). For children the minimum values of annual are 0.002,0.001 µSv y^{-1} for age (2-7y, 7-12y &12-17y) and the maximum values 0.25 µSv y^{-1} (2-7y), 0.153 µSv y^{-1} (7-12y) and 0.14 µSv y^{-1} (12-17y) with averages of 0.08 for(2-7y) and 0.05 µSv y^{-1} for(7-12y&12-17y). For adults the annual effective dose varies from minimum value 0.001 µSv y^{-1} to maximum value 0.157 µSv y^{-1} with an average 0.05 µSv y^{-1}. It appears that the average annual dose from 40K 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 226Ra 232Th and 40K due to the ingestion of the 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 age (12-17y) is higher than that received by the other ages children, infants and adults. The main dose contribution is caused by 226Ra 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 1996, IAEA 2002 and UNSCEAR 2000.

(5) The results in Table 4, gives the contributions of 226Ra to the total average annual effective dose due to the intake of the bottled water ; 40% for infants, (41%, 55% &73%) for children and (37 %) for adults. For the natural radionuclide 232Th, the results showed that it is the second largest contributor to the total annual effective dose contributing, 33% for infants, children (39 %, 33% & 22%) and the largest for adults 51 %. The lowest contributions due to the intake of 40K to the total annual effective dose are 27%, (0%, 12% & 5 %) and 12 % for the infants, children and adults, respectively.

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

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

Radionuclide		Effective dose (µSv/ y)				
		Infants	Children			*Adults*
		1-2y	2-7y	7-12y	12-17y	> 17 y
^{226}Ra	Minimum	0.051	0.046	0.059	0.173	.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

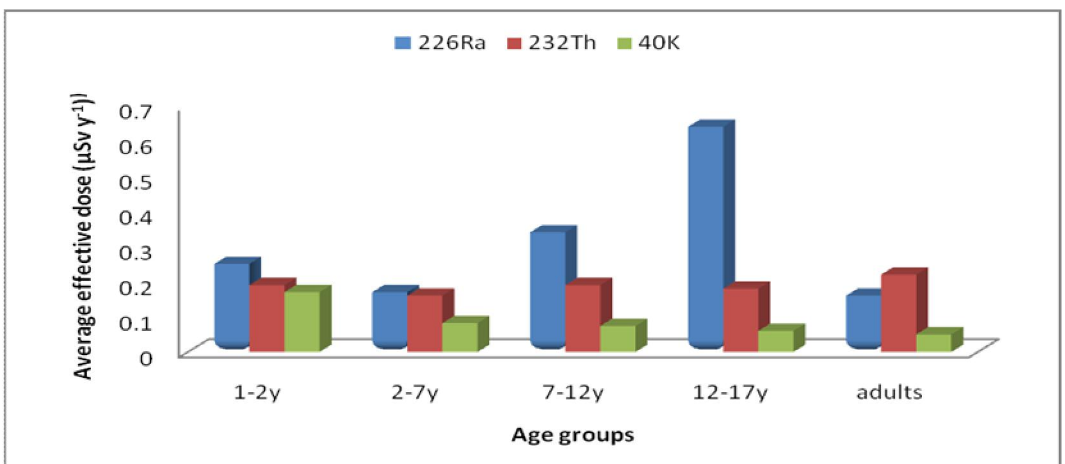

Fig 2: Comparison between average annual effective dose rate (µSvy^{-1}) of natural radioactivity in bottled drinking water consumed by different age groups in Saudi Arabia

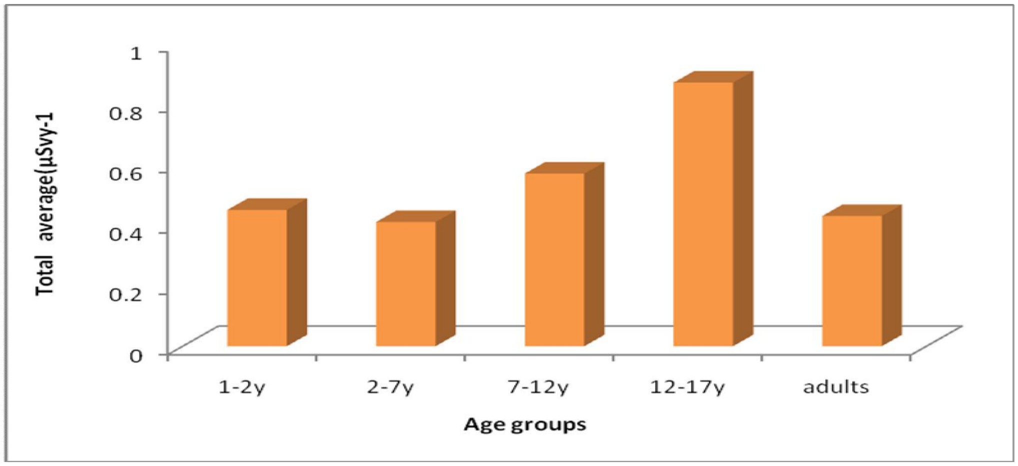

Fig 3: Total average annual effective dose for all age groups due to the intake of the bottled water

4. Conclusion

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

2- Annual effective doses for consumption of water for infants (1-2y), children(2-7y, 7-12y and 12-17y) and adult were calculated for ingestion of radionuclides 226Ra, 232Th and 40K. The total average annual estimated effective doses for 226Ra, 232Th and for 40K 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 2000 and WHO(2008). So, the present results for investigated radionuclides in bottled drinking water in Saudi Arabia are below limit values and pose no detrimental health effect.

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

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