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Environmental Impact Assessment of Natural Radioactivity, Heavy and Major Metals in Primary Schools' Drinking Water

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Abstract

Measurement of natural radioactivity levels of major and heavy metals concentration was carried out for water samples collected from primary schools in Jeddah city, Saudi Arabia. The estimations were done for two different age groups, children (students 7 - 12 y) and adults (teachers and workers > 17 y) at the schools. The chemical analysis was performed by an Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES) and a high-resolution HPGe detector was used for the natural radionuclides measurement. The obtained results for the heavy metals (Fe, Cu, and Zn) and major and heavy elements (Na, K, Ca and Mg) concentrations in all water samples did not exceed the limits of WHO, EPA and TSE-266 guidelines for drinking water. Only Pb exceeded the safe limit. Generally, heavy metals and major elements' concentrations in water samples were found decrease in sequence of Ca > Na > Mg > K > Zn > Pb > Cu > Fe. The radioactivity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in all studied samples were below the permissible safe limit value. The total average annual effective doses of (^{226}Ra + ^{232}Th + ^{40}K) radionuclides were 0.259 mSv·y^{-1} for the children and 0.112 mSv·y^{-1} for adults, which are below the recommended annual dose level 1.0 mSv·y^{-1} as reported by WHO (2006). The present drinking waters are high quality waters and safe in terms of natural radioactivity and the results of the HQ values for the individual heavy metals showed that there was no health risk for humans due to consumption of these waters.

Keywords

Heavy Metals, Major Elements, Radioactivity

1. Introduction

Water is an imperative matrix in environmental studies for its daily use for human consumption and its ability to transport pollutants (Degerlier & Karahan, 2010). The natural radionuclides (^{226}Ra, ^{232}Th and ^{40}K) and heavy elements concentrations in drinking water are significant for human health. Radionuclides are present in the form of dust, particles, or molten minerals in drinking water. The existence of radionuclides in drinking water causes human internal exposure, caused by the decay of radionuclides taken into the body through the ingestion pathway (Fatima et al., 2006). The major sources of heavy metals contamination in drinking water were iron pipes used in distribution systems and the geological pollution of the region that water was originated from some of which elements have been widely implicated in human health (Öztürk & Yilmaz, 2000). Therefore, it is important to determine background radiation levels and amounts of metal accumulation in the environment to assess the hazards associated with their intake to prevent possible risks. Radioactivity, major and heavy element rates should not exceed the permissible limits for drinking water. For this reason, the metals and natural radioactivity concentration in water have been studied by many investigators in several countries (Ali et al., 2016; Radulescu et al., 2017; Ghaderpoori et al., 2018; Parhoudeh et al., 2019). In Saudi Arabia, there are limited studies on drinking water (Al-Ghamdi, 2014; Al-Zahrani, 2016; Althoyaib & El-Taher, 2016; Alseroury et al., 2018). According to our literature survey conducted by the researcher, there are no data found on the quality of schools' drinking water supplies in the city of Jeddah. So, this is the first study to assess the levels of background radiation, major, and heavy metals in drinking water of this area. The aim of the study is to measure the concentration of heavy metals (Fe, Cu, Pb and Zn), major elements (Na, K, Ca, Mg) and natural radioactivity of ^{226}Ra, ^{232}Th and ^{40}K in the schools' drinking water and to estimate the radiological risk resulting in the population using these waters. The estimations were done for two different age groups, children (7 - 12 y) and adults (>17 y), these age groups represent the student (7 - 12 y), teachers and workers (>17 y) at the schools. The data produced in this study will provide a reference point levels of natural radioactivity and chemical elements in the drinking water of the study area and help in setting the background information for future research on drinking water for radiological safety of humans.

2. Material and Methods

2.1. Study Area

Jeddah city is located on the Red Sea coast in the west of Saudi Arabia, 949 kilometres from the capital, Riyadh. It is situated between the latitude of (21˚32'32) N, and longitude (39˚11'52) E, within an area 5460 km^2. It has a population of 3,456,259 inhabitants (**Figure 1**).

2.2. Preparation of Samples

The water samples were collected from various primary schools in all parts of Jeddah city; stretching from its north to south and from east to west. The water

Figure 1. Locations of water samples in Jeddah city.

desalination is the source of schools' water, which is used for drinking and washing. The water samples were taken directly from the drinking water cistern and were acidified with the solution HCl to prevent the absorption of radionuclides on the walls of the containers (IAEA, 1989). In the laboratory, all samples were evaluated by using inductively coupled plasma optical emission spectrometry (ICP-OES) to determine the metals' (Na, K, Ca, Mg, Fe, Cu, Pb and Zn) concentrations. For the gamma measurement, Marinelli beakers were washed with distilled water. About 0.5 litres of each sample was put into a dry beaker which was sealed and stored for four weeks to ensure that the secular equilibrium had reached before the measurement was done.

2.3. Gamma-Ray Measurements

The gamma-ray spectra of the samples were measured using a hyper-pure germanium detector (HPGe) with 25% efficiency and 2 keV resolution at 1332 keV gamma line of ^{60}Co were employed for all the measurements. The system was calibrated for energy and efficiency (IAEA, 1989). Genie 2000 computer software performed the spectrum analysis. Each sample after equilibrium was positioned on top of the HPGe detector and counted for 36000 s. The background radiation was measured every week under the same conditions as the sample. The gamma-ray transitions of energies 351.9 keV (^{214}Pb) and 609.3 keV (^{214}Bi) were used to determine the concentration of the ^{226}Ra series. The gamma-ray lines at the 911.1 Kev (^{228}Ac), 238.63 keV (^{212}Pb) and 583.1 keV (^{208}Tl) were used to verify the concentration of the ^{232}Th series. ^{40}K activities were estimated from gamma-peaks 1460.8 keV.

The activity levels for the natural radionuclides in the measured samples were computed using the following relation (El-Taher, 2012):

$$A\left(\mathrm{Bq}\cdot\mathrm{L}^{-1}\right)=C_a/\varepsilon P_r V \tag{1}$$

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 a litre.

3. Results and Discussion

3.1. Major and Heavy Metal Concentrations in Water Samples

Table 1 contains the heavy metal's concentration results and the comparison with the recommended values of the World Health Organization (WHO, 2006), United States Environmental Protection Agency (EPA, 2002), and Turkish Standards Annual Progress Report (TSE-266, 1997). The elemental concentrations are expressed in Milligrams per litre. For the major elements, the results indicate that Na varied from 3.96 mg·L^{-1} in sample w14 to 64.3 mg·L^{-1} in sample w5 with a mean value of 10.48 mg·L^{-1}. The maximum values of K and Ca were found in the sampling code w5 (3.39 mg·L^{-1}and 25.4 mg·L^{-1}), while their minimum values were measured in the sampling code w7 as 0.361 mg·L^{-1} and 7.9 mg·L^{-1} in the sample code w15, respectively, with the corresponding mean values of 0.74 mg·L^{-1} and 16.26 mg·L^{-1} for K and Ca, respectively. Although the concentration of Mg varied from 0.672 mg·L^{-1} in sample w6 to 2.9 mg·L^{-1} in sample w9 with a mean value of 1.57 mg·L^{-1}, the results were much lower than the maximum acceptable concentrations of Na (175 mg·L^{-1}), K (12 mg·L^{-1}), Ca (200 mg·L^{-1}) and Mg (50 mg·L^{-1}), reported by TSE-266 as shown in **Table 1**. For heavy elements Fe, Cu and Zn ranged between 0.0153 - 0.0216 mg·L^{-1}, 0.0525 - 0.0656 mg·L^{-1} and 0.0466 - 0.139 mg·L^{-1}, respectively. The maximum and minimum values of both Fe and Cu, for the water samples were found in the samples w5 and w15, respectively.

Table 1 shows that the mean elemental contractions in the water samples were 0.018, 0.059 and 0.089 mg·L^{-1} for Fe, Cu and Zn, respectively. These results were below the guideline values as tabulated in **Table 1**. The highest concentration of heavy metals was found for Pb element, it ranged from 0.039 to 0.081 mg·L^{-1} with a mean value (0.061 mg·L^{-1}). All the samples' concentration and the mean value were exceeding the recommended values of (WHO, 2006; EPA, 2002). The concentration of lead in drinking waters rises mainly through anthropogenic activities, and it accumulates in the body mostly in the bones. For that fact, it may cause damage to nervous system and intellectual performance for children and it can also escalate blood pressure and the cardiovascular diseases in adults (EEC, 2001). Therefore, the investigated water samples are unsafe for drinking purposes and are not suitable for life-long human consumption. Heavy metals concentration and major elements [mg·L^{-1}] in water samples under investigation were shown in **Figure 2** and **Figure 3**, respectively.

Table 1. Major and heavy metal concentrations in water samples with the guidelines limits.

Sample code	Major metal concentration [mg·L^{-1}]				Heavy metals concentration [mg·L^{-1}]			
	Na	K	Ca	Mg	Fe	Cu	Pb	Zn
W1	7.11	0.451	15.3	0.818	0.0179	0.0619	0.0811	0.137
W2	9.54	0.62	14.7	1.38	0.0214	0.0633	0.0731	0.139
W3	5.4	0.438	21.6	0.971	0.021	0.0649	0.075	0.118
W4	5.47	0.436	25.4	1.24	0.02	0.0632	0.0711	0.12
W5	64.3	3.39	22.1	7.33	0.0216	0.0656	0.0801	0.0466
W6	5.86	0.551	19.7	0.672	0.0176	0.0595	0.0616	0.0803
W7	4.57	0.361	18.2	0.986	0.0174	0.0596	0.0592	0.0808
W8	5.08	0.447	22.2	0.995	0.0185	0.0592	0.061	0.0737
W9	22.7	1.55	9.79	2.9	0.0178	0.0583	0.0609	0.0776
W10	6.34	0.516	17.1	1.17	0.0168	0.0575	0.0562	0.0735
W11	4.71	0.387	22	1.01	0.0168	0.0572	0.0561	0.0698
W12	5.14	0.488	8.9	1.07	0.0159	0.0548	0.0478	0.073
W13	4.6	0.543	9.84	0.905	0.016	0.0555	0.0488	0.0705
W14	3.96	0.384	9.06	0.887	0.0157	0.0538	0.0461	0.0747
W15	7.74	0.549	7.9	1.14	0.0153	0.0525	0.0392	0.114
Mean	10.84	0.74	16.26	1.57	0.018	0.059	0.061	0.089
WHO2006	200	-	-	-	0.3	2	0.01	--
EPA2002	-	-	-	-	0.3	1.3	0.015	5
TSE-266 1997	175	12	200	50	0.2	3	-	5

3.2. Hazard Quotient

The Risk of the heavy metals through ingestion may be characterized using the following equation (Zhuang et al., 2009):

$$HQ = ADD/RfD \tag{2}$$

where RfD (milligrams per kilogram per day) is the reference dose, its values of all the measured heavy metals were taken from US-EPA (1993). ADD (milligrams per kilogram of body weight per day) is the average daily dose and it was determined by the following equation (Zhuang et al., 2009):

$$ADD = C_{\text{metal}} \times W/m \tag{3}$$

where C_{metal} is the concentration of heavy metals in water sample; W is the average daily consumption of water (2 litres for adults), as given by UNSCEAR (2000); m is the body weight of 70 kg for adults. If $HQ > 1.00$, there is a highly possible risk associated with that metal.

Result of health risk assessments (HQ) of the various heavy metals considered in this study is presented in **Table 2**. The HQ ranges from 0.001 (Fe) to 0.486

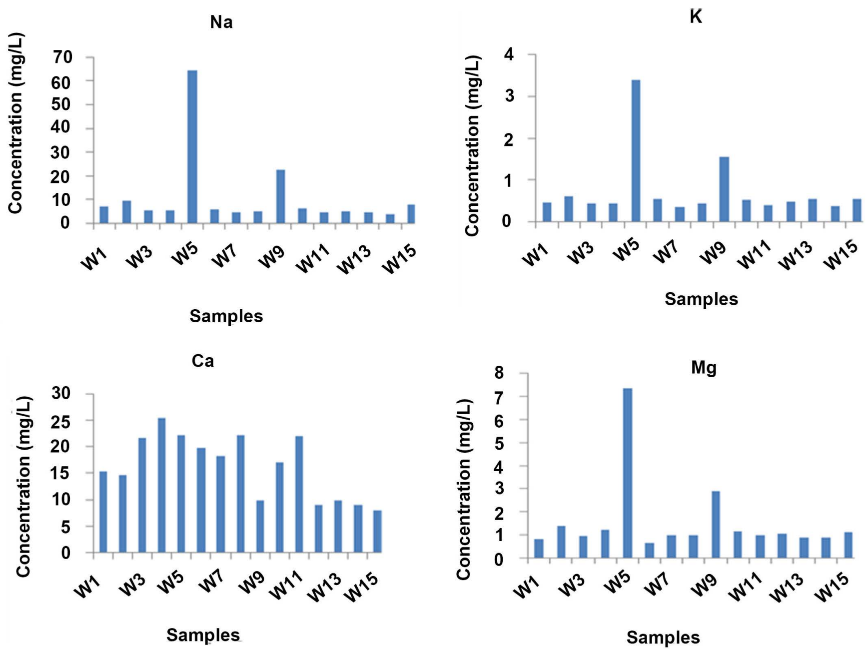

Figure 2. Major metal concentration [mg·L^{-1}] in water samples under investigation.

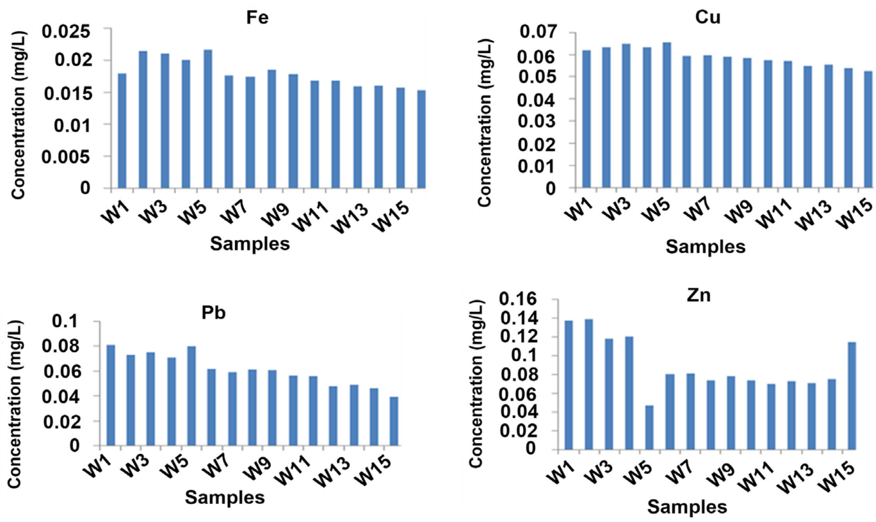

Figure 3. Heavy metal concentration [mg·L^{-1}] in water samples under investigation.

(Pb). All the HQ values were below the critical value of 1 as reported by the US-EPA. Even though the concentration of Pb in the drinking water samples was found above the limit set by (WHO, 2006; EPA, 2002). This metal does not pose a risk to human health, where the HQ was calculated for Pb was 0.486, below the recommended limit. But there is the need for a continuous monitoring of contamination level of this metal in schools' drinking water since it can accumulate to toxic level. It will help to detect any change in it accumulation pattern that could become a hazard to human safety. Generally, HQs were less than 1, indicating that there is no significant potential health risk associated with the consumption of these waters.

Table 2. ADDs and HQs of heavy metals in drinking water samples.

Metals	*RfD* (mg/kg body weight/day)	*ADD* (mg/kg body weight/day)	*HQ* (Hazard Quotient)
Fe	7×10^{-1}	0.0005	0.0007
Cu	4.0×10^{-2}	0.0017	0.0049
Pb	3.5×10^{-3}	0.0017	0.486
Zn	3.0×10^{-1}	0.0025	0.0085

3.3. Activity Concentrations

The activity concentration of the three radionuclides ^{226}Ra, ^{232}Th and ^{40}K for the investigated drinking water were reported in Table 3. As shown, the activity concentrations of ^{226}Ra ranged from 0.087 ± 0.03 to 0.434 ± 0.05 Bq·L^{-1} with an average of 0.28 Bq·L^{-1}, it was not detected in three samples (W3, W7 & w13). ^{232}Th concentration ranged from 0.038 ± 0.02 to 0.186 ± 0.05 Bq·L^{-1} with an average 0.085 Bq·L^{-1} and for ^{40}K concentrations changed from 1.43 ± 0.073 to 12.19 ± 0.586 Bq·L^{-1} with an average 5.24 Bq·L^{-1}.

^{40}K is the most abundant concentration in the water samples, it can be noted that, potassium-40 is an isotope of critical element; it is controlled by the human cells. So, the body content of ^{40}K is verified largely by its physiological characteristics rather than its intake (Jibiri et al., 2007). It is obvious through the present work that all results are less than the limit given in the report WHO (2006), as shown in Table 3 and Figure 4. A comparison between the measured activity concentration values of ^{226}Ra, ^{232}Th and ^{40}K in the present work with the results for local and other countries was tabulated in Table 4. We can indicate that the obtained average concentrations of ^{226}Ra (0.32 Bq·l^{-1}), ^{232}Th (0.12 Bq·l^{-1}) and ^{40}K (5.24 Bq·l^{-1}) are comparable with the reported values of drinking water from various locations in the world, taking into account the data of the countries of different geographical locations differ regarding specific mineral.

3.4. Effective Dose

Effective doses due to ingestion of waters were determined using the following equation (UNSCEAR, 2000):

$$E_d = A_c A_i C_f \tag{4}$$

where E_d the effective dose (mSv·y^{-1}), A_c is the activity concentration (Bq·L^{-1}), A_i is the consumption rate of water (l/year). According to WHO (2006), the dose was estimated by considering a consumption rate is 730 l/year for adults and 512 l/year for children. The dose conversion factors C_f for adults and children were (2.8×10^{-7}, 2.3×10^{-7}, 6.2×10^{-9} and 1.5×10^{-6}, 2.5×10^{-7}, 7.6×10^{-9} Sv·Bq^{-1}) for ^{226}Ra, ^{232}Th and ^{40}K respectively (WHO, 2006; ICRP-60, 1990).

Table 5 shows the maximum and minimum values of effective dose of ^{226}Ra, ^{232}Th, and ^{40}K for children and adults were below the reported range (0.2 - 0.8 mSv·y^{-1}) of effective dose from ingestion of drinking water reported by

Table 3. Activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K radionuclides from drinking water samples collected from primary schools in Saudi Arabia, Jeddah city.

Sample code no.	Activity concentration (Bq·L^{-1})		
	^{226}Ra	^{232}Th	^{40}K
W1	0.283 ± 0.03	0.076 ± 0.04	2.38 ± 0.133
W2	0.194 ± 0.02	0.124 ± 0.03	8.54 ± 287
W3	0.292 ± 0.02	N.D	1.43 ± 0.073
W4	0.501 ± 0.03	0.095 ± 0.02	2.24 ± 0.709
W5	0.213 ± 0.05	0.172 ± 0.02	1.90 ± 0.105
W6	0.358 ± 0.03	0.211 ± 0.03	4.23 ± 0.341
W7	0.087 ± 0.03	N.D	3.32 ± 0.175
W8	0.434 ± 0.05	0.142 ± 0.02	5.38 ± 0.276
W9	0.302 ± 0.02	0.023 ± 0.04	5.07 ± 0.266
W10	0.236 ± 0.04	0.186 ± 0.05	6.17 ± 0.323
W11	0.087 ± 0.03	0.075 ± 0.02	12.19± 0.586
W12	0.162 ± 0.02	0.068 ± 0.02	10.92 ± 0.510
W13	0.324 ± 0.14	N.D	7.44 ± 0.368
W14	0.361 ± 0.12	0.062 ± 0.02	2.16 ± 0.073
W15	0.357 ± 0.03	038 ± 0.02	4.91 ± 0.246
Range	0.194 ± 0.02 - 0.434 ± 0.05	0.023 ± 0.04 - 0.186 ± 0.05	1.43±0.073 - 12.19 ± 0.586
Average	0.28	0.085	5.24
WHO2006	1	0.1	10

N.D.: Not detected.

Table 4. Comparison of measurement results of activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in drinking water from various countries.

Country	Water source	Activity concentration (Bq·L^{-1})			Reference
		^{226}Ra	^{232}Th	^{40}K	
Saudi Arabia	Drinking water	1.94 - 0.434 (0.28)	0.023 - 0.186 (0.085)	1.43 - 12.19 (5.24)	Present work
Saudi Arabia	Bottled drinking water	0.21 - 2.25 (0.77)	0.37 - 0.232 (1.3)	0.24 - 33.74 (11.1)	(Al-Ghamdi, 2014)
Saudi Arabia	Drinking water	0.105 - 0.568 (0.32)	0.016 - 0.382 (0.12)	2.16 - 18.84 (10.96)	(Al-Zahrani, 2016)
Turkey	Bottled drinking water	0.517 - 1.22 (0.78)	0.232 - 1.87 (1.05)	1.54 - 2.57 (2.19)	(Kabadayi & Gümüş, 2011)
Malaysia	Bottled mineral water	0.41 - 0.56 (0.46)	0.34 - 0.99 (0.54)	2.88 - 7.65 (4.05)	(Priharti et al., 2015)
Pakistan	Drinking water	1.09	0.55	16.17	(Mashiatullah et al., 2016)
Malaysia	Bottled mineral water	1.45 - 3.30	0.65 - 3.39	21.12 - 25.31	(Khandaker et al., 2017)
Egypt (Assiut city)	Drinking water	0.019 - 0.492 (0.203)	0.015 - 0. 351 (0.081)	0.050 - 2.255 (0.688)	(El-Gamal et al., 2019)
Iran	Drinking water	(0.57)	(0.98)	(6.42)	(Parhoudeh et al., 2019)

Table 5. Annual effective dose due to the intake of natural radionuclides ^{226}Ra, ^{232}Th and ^{40}K in the present drinking water.

Radionuclide		Annual effective dose (mSv/y)	
		Children	*Adults*
^{226}Ra	Minimum	0.067	0.018
	Maximum	0.275	0.102
	Average	0.215	0.057
^{232}Th	Minimum	0.005	0.004
	Maximum	0.027	0.035
	Average	0.024	0.031
$^{40}K^{22}$	Minimum	0.007	0.006
	Maximum	0.529	0.616
	Average	0.020	0.024
Total average annual effective dose		0.259	0.112

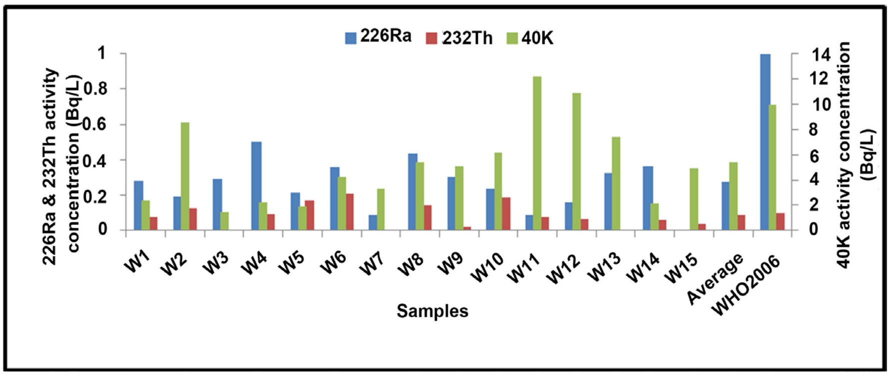

Figure 4. Activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in water samples, Saudi Arabia.

UNSCEAR (2000). The total average annual effective doses of (^{226}Ra + ^{232}Th + ^{40}K) radionuclides were 0.259 mSv·y^{-1} for the children and 0.112 mSv·y^{-1} for adults, which are below the recommended annual dose level 1.0 mSv·y^{-1} as reported by WHO (2006). It turned out that the overall dose consumed by children is higher than that received by adults and 226 Ra causes the major dose. In fact, ^{226}Ra is known as an extremely radiotoxic radionuclide; the ingestion radium is saved in growing bones, therefore children have a higher risk factor because of their intensive bone growth during these years, and action must be taken to restrict their ingestion (Fatima et al., 2006). The inputs of average dose of ^{226}Ra, ^{232}Th and ^{40}K to the total annual effective dose due to the intake of the drinking water samples were for children (82/97%, 9.15% & 7.88%) and (51.06%, 27.74% & 21.21%), respectively for adults as illustrated in **Figure 5**. The total effective dose from a year's consumption of water samples by the children and adults were estimated, respectively, to be 25.9.2 % and 11.2% of the WHO limit value 1 mSv·y^{-1} Consequently, it can be recommended that the present drinking water samples will not cause any radiological health detriment and be acceptable for human consumption.

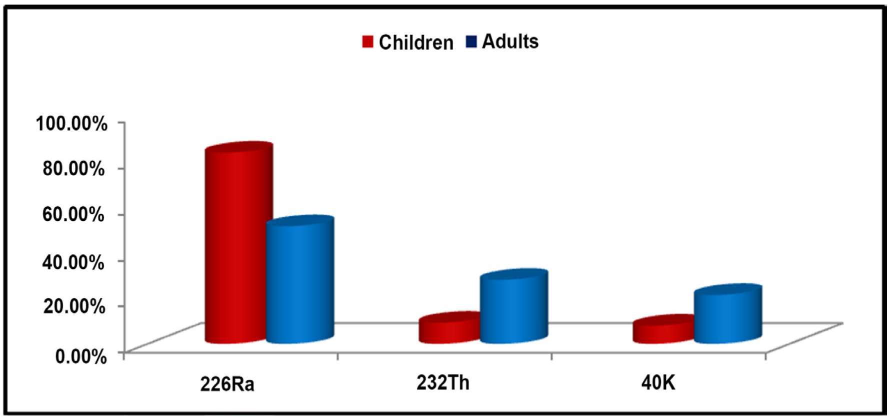

Figure 5. Percentage contribution dose of ^{226}Ra, ^{232}Th and ^{40}K to the total effective dose in the water samples.

4. Conclusion

The natural radioactivity levels of ^{226}Ra, ^{232}Th and ^{40}K and some chemical metals (Na, K, Ca, Mg, Fe, Cu, Pb and Zn) were determined in drinking water collected from primary schools in Jeddah city, Saudi Arabia. The activity concentrations of the three radionuclides in the water samples were found to be within the recommended values. It is found that total averages of effective doses due to all radionuclides were 0.259 and 0.112 $mSv \cdot y^{-1}$ for the children and adults, respectively, which were 25.9% and 11.2% of the WHO accepted limit values of 1.0 mSv from one year's consumption of drinking water for children and adults, respectively. As a result, it can be stated that none of the radioactivity is supposed to cause major health problems to human beings. The achieved outcomes showed that the heavy and major metals (Fe, Cu, Pb, Zn, Na, K, Ca and Mg) concentrations in all water samples did not exceed the WHO, EPA and TSE-266 standards. Only Pb concentration in all water samples exceeded both the limit of (WHO, 2006; EPA, 2002) guidelines. In general, the obtained HQ values for the individual metals showed that there was no health risk for humans due to the consumption of these waters. Therefore, it can be recommended that the present study discloses the fact that the investigated waters can be used as drinking water. However, water-treatment is highly suggested in order to minimize the Pb concentration in water sources to make the water drinkable and used directly. Regular monitoring of the radionuclides and the metals in drinking water is crucial to avoid the excessive build-up of the metals in these waters. Finally, the handiness of data from this study is very valuable as it serves as critical information concerned with the quality of drinking water. It complements data needed for setting of guidelines on radiological safety for drinking water globally.

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Conflicts of Interest

The author declares no conflicts of interest regarding the publication of this paper.

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