



## Full Length Review Article

### RISK ASSESSMENT DUE TO INGESTION OF NATURAL RADIONUCLIDES AND HEAVY METALS IN DRINKING WATER

\*Al-Zahrani, J.H.

Department of Physics, Faculty of Science, King AbdulAziz University, Jeddah, Saudi Arabia

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#### ABSTRACT

The concentrations of natural radioactivity of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  and some heavy elements (Zn, Mn, Fe, Mn, Cu, Ni, and Pb) were measured in drinking water samples collected from different mosques in Makkah city, Saudi Arabia. A high-resolution HPGe detector was used for the natural radionuclides measurement. The average activity concentrations were 0.32, 0.12 and 10.96 Bq l<sup>-1</sup> for  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$ , respectively. The cumulative annual effective dose due to ingestion of water samples was estimated to be 0.832  $\mu\text{Sv y}^{-1}$  for children and 0.369  $\mu\text{Sv y}^{-1}$  for adults, which are below the limit of 1.0 mSv y<sup>-1</sup> for individual public exposure. These water samples were also analyzed for concentration of some heavy elements (Fe, Zn, Cu, Mn, Ni and Pb) using the inductively coupled plasma optical emission spectrophotometer (ICP-OES). The obtained results established that the heavy elements concentration in the drinking water samples did not exceed the recommended limits WHO and EPA guidelines for drinking water.

#### INTRODUCTION

The presence of natural radionuclides as  $^{238}\text{U}$ - and  $^{232}\text{Th}$  series and  $^{40}\text{K}$  "in water causes health hazards owing to the human internal exposure from the absorbed into the body through ingestion. Thus, measurement of natural radioactivity in drinking water is necessary for public health studies, which allows the evaluation of population exposure to radiation by the consumption of water" (Fatima *et al.*, 2007). "The radioactivity in drinking water may be man-made, resulting from waste discharges and atomic bomb fallout, or produced naturally by the dissolution of gasses and rock minerals" (Todorović *et al.*, 2012). Also, Heavy metals such as Cd, Cu, Mn, Ni, Pb and Zn are essential at very low level of intake and are potentially harmful to live organisms at some level of exposure and absorption (Kalay and CANLI 2000). The daily consumption of water is indubitable and cannot be deferred by humans. As a consequence, monitoring the quality of drinking water is essential to secure the public health. Natural radionuclides and heavy elements in drinking water have been widely studied to estimate the safety of drinking water on its radionuclide and metal's content. "e.g., (Davutluoglu *et al.* 2011; Almayahi, Tajuddin, and Jaafar 2012; Abdul *et al.* 2012; El-Gamal *et al.* 2013; Aytas *et al.*

2014; El-Gamal and El-Mageed 2014; Gorur and Camgoz 2014; Walsh, Wallner, and Jennings 2014; Saleh *et al.* 2015; Darko *et al.* 2015; Calin, Ion, and Radulescu 2015; Yang, Ha, and Jin 2015; Mohapatra *et al.* 2015). This work is necessary due to the require of the database available on the waters and is highly required to keep human drinking water standards. Therefore, this study seeks to determine the activity concentration of the radionuclides as  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  and heavy elements as (Fe, Zn, Cu, Mn, Ni and Pb) in water samples, which were collected from mosques in Makkah city, Saudi Arabia, to estimate the associated effective doses resulting from ingestion of such waters by the local public and the pilgrims. Nuclear spectrometry techniques and a mass spectrometry method were applied to obtain the presently required data. The current results were compared with the recommended values and the reported data from other countries. The obtained data in this study may afford baseline levels of natural radioactivity and heavy elements in such water and provide background information for future research on drinking water for radiological protection of the human.

#### MATERIALS AND METHODS

##### Preparation of samples

The water samples were collected from various mosques resting stations on the highways of Makkah city, Saudi Arabia. The selection of sampling locations was based on the

\*Corresponding author: Al-Zahrani, J.H.

Physics Department, Faculty of Science, King AbdulAziz University, Jeddah, Saudi Arabia

importance of them for the local inhabitants and the pilgrims that they gather from different countries for the pilgrimage to Makkah city, which all of them are using these waters for drinking and bath. The samples were taken directly from the water supply and drinking water cistern without filtering 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 the samples were analyzed by using Inductively coupled plasma optical emission spectrometry (ICP-OES) to determine the heavy metals (Fe, Zn, Cu, Mn, Ni and Pb) concentrations. For the gamma measurement, Marinelli beakers were washed with distilled water and then about 0.5 liters of each sample were put into a dry Marinelli beaker, sealed and stored for four weeks before the measurement was done, to ensure secular equilibrium is reached.

### Gamma-ray measurements

The gamma-ray spectra of the samples were measured using a hyper-pure germanium detector (HPGe) with 25% efficiency and 2keV resolution at 1332 keV gamma line of  $^{60}\text{Co}$  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 placed on top of the HPGe detector and counted for 36000 s. The background radiation was measured every week under the same conditions of the sample. The gamma-ray transitions of energies 351.9 keV (214Pb) and 609.3 keV (214Bi) were used to determine the concentration of the  $^{226}\text{Ra}$  series. The gamma-ray lines at the 911.1 Kev ( $^{228}\text{Ac}$ ), 238.63 keV(212Pb) and 583.1 keV (208Tl) were used to determine the concentration of the  $^{232}\text{Th}$  series.  $^{40}\text{K}$  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 2015*):

$$A (\text{Bq l}^{-1}) = C_a / \varepsilon P_r V \quad \dots \dots \dots (1)$$

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

### Estimation of annual effective dose

Annual radionuclide intakes and effective doses due to ingestion of waters were calculated using the equation (*UNSCEAR 2000*):

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

Where  $E_d$  the effective dose ( $\text{mSv y}^{-1}$ ),  $A_c$  is the activity ( $\text{Bq l}^{-1}$ ),  $A_i$  is the consumption rate of water ( $\text{l/y}$  year). The dose was estimated by considering a consumption rate is 730  $\text{l/year}$  and 512  $\text{l/year}$ , for adults and children, respectively, (*WHO 2003*). The dose conversion factors  $C_f$  for adults and children, as reported by *WHO 2006* and *ICRP 1996*, were ( $2.8 \times 10^{-7}$ ,  $2.3 \times 10^{-7}$ ,  $6.2 \times 10^{-9} \text{ Sv Bq}^{-1}$ ) and ( $1.5 \times 10^{-6}$ ,  $2.5 \times 10^{-7}$ ,  $7.6 \times 10^{-9} \text{ Sv Bq}^{-1}$ ) for  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  respectively.

## RESULTS AND DISCUSSION

### Activity concentration results

The activity concentration measurement of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in water samples together with their corresponding total uncertainties are shown in Table 1. The activity concentrations for  $^{226}\text{Ra}$  ranged from  $0.105 \text{ Bq l}^{-1}$  (sample W1) to  $0.568 \text{ Bq l}^{-1}$  (sample W14)  $\text{Bq l}^{-1}$ , while,  $^{232}\text{Th}$  concentrations ranged from  $0.016$  (W2) to  $0.382$  (W10)  $\text{Bq l}^{-1}$ . The concentration of  $^{40}\text{K}$  was found in the range between  $2.16$  (W11) and  $18.84$  (W2)  $\text{Bq l}^{-1}$ . It can be seen, from Table 1, there is a variation of the concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  from one sample to another, because the sources of these waters may be distillation waters or groundwaters at different depths with various minerals derived from aquifer rocks.

**Table 1. Activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  radionuclides from drinking water samples**

Sample code no.	Activity concentration ( $\text{Bq l}^{-1}$ )		
	$^{226}\text{Ra}$	$^{232}\text{Th}$	$^{40}\text{K}$
W1	$0.105 \pm 0.02$	$0.101 \pm 0.06$	$13.36 \pm 0.34$
W2	$0.462 \pm 0.18$	$0.106 \pm 0.04$	$18.84 \pm 0.61$
W3	$0.105 \pm 0.02$	$0.171 \pm 0.01$	$17.31 \pm 0.65$
W4	$0.389 \pm 0.24$	$0.296 \pm 0.02$	$14.42 \pm 0.47$
W5	$0.213 \pm 0.08$	$0.049 \pm 0.001$	$10.09 \pm 0.34$
W6	$0.274 \pm 0.04$	$0.039 \pm 0.007$	$17.33 \pm 0.59$
W7	$0.077 \pm 0.05$	$0.016 \pm 0.003$	$14.83 \pm 0.28$
W8	$0.511 \pm 0.15$	$0.056 \pm 0.26$	$7.21 \pm 0.24$
W9	$0.456 \pm 0.03$	$0.203 \pm 0.05$	$9.14 \pm 0.25$
W10	$0.17 \pm 0.02$	$0.382 \pm 0.04$	$4.21 \pm 0.22$
W11	$0.076 \pm 0.03$	$0.031 \pm 0.04$	$2.16 \pm 0.07$
W12	$0.216 \pm 0.16$	$0.086 \pm 0.02$	$10.09 \pm 0.34$
W13	$0.378 \pm 0.24$	$0.115 \pm 0.03$	$8.74 \pm 0.05$
W14	$0.568 \pm 0.04$	$0.026 \pm 0.002$	$7.31 \pm 0.06$
W15	$0.341 \pm 0.02$	$0.053 \pm 0.02$	$9.43 \pm 0.06$
Range	$0.105 \pm 0.02$ $0.568 \pm 0.04$	$0.016 \pm 0.00$ $0.382 \pm 0.04$	$2.16 \pm 0.07$ $18.84 \pm 0.61$
Average	0.32	0.12	10.96
WHO2006	1	0.1	10

The results indicate that the concentration values of  $^{226}\text{Ra}$  are higher than those of  $^{232}\text{Th}$ , this may because the radium is more soluble in water (*Kitto and Kim 2005*). According to UNSCEAR2000, the maximum acceptable value of  $^{226}\text{Ra}$  concentration in drinking water is  $1000 \text{ mBq l}^{-1}$ , the measured values of  $^{226}\text{Ra}$  concentration in drinking water samples appear lower than this limit. Whereas, the maximum concentration values in drinking water of  $^{232}\text{Th}$  ( $1.0 \text{ Bq kg}^{-1}$ ) and  $^{40}\text{K}$  ( $10 \text{ Bq kg}^{-1}$ ) as given by (*WHO2006*), in this study, the average concentration values of  $^{232}\text{Th}$  ( $0.12 \text{ Bq l}^{-1}$ ) and  $^{40}\text{K}$  ( $10.69 \text{ Bq l}^{-1}$ ) were in the same order. It should be noted that, in the present results,  $^{40}\text{K}$  is the most abundant, about 96% of the total ( $^{226}\text{Ra} + ^{232}\text{Th} + ^{40}\text{K}$ ) concentrations. “Potassium-40 is an isotope of an essential element; it is controlled largely by its physiological characteristics rather than its intake” (*Jibiri, Farai, and Alausa 2007*).

Table 2, shows a comparison between the measured activity concentration values in drinking water with those obtained in the last five years ago in other countries. Where, the present average concentrations of  $^{226}\text{Ra}$  ( $0.32 \text{ Bq l}^{-1}$ ) and  $^{232}\text{Th}$  ( $0.12 \text{ Bq l}^{-1}$ ) are comparable to Egyptian results and less than

the reported values from Saudi Arabia (bottled water), Yemen, Malaysia, Turkey, and Ghana. The present results show the average of 40K concentration is nearly the same values in Saudi Arabia (bottled water) and Malaysia, lower than Yemen but greater than the observed values in Egypt, Turkey, and Ghana. Therefore, we can indicate that the obtained results are consistent with the data obtained from various locations in the world, taking into account the data of the countries of different geographical locations differ regarding specific mineral.

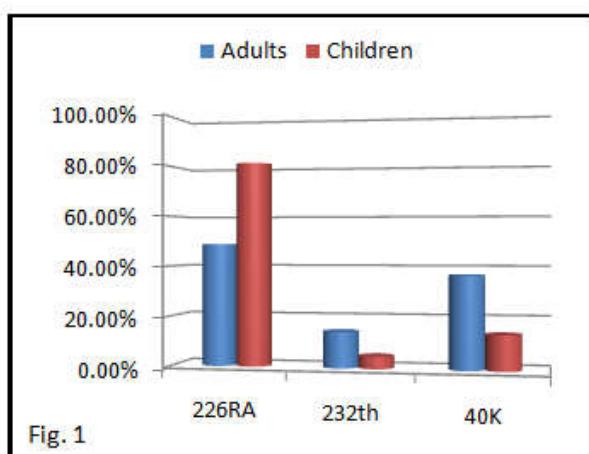
- The maximum and minimum values of effective dose for children and adults with average values were found below the reported range ( $0.2 - 0.8 \text{ mSv}^{-1}$ ) and the average value ( $0.3 \text{ mSv}^{-1}$ ) of effective dose from ingestion of drinking water reported by UNSCEAR2000.
- The cumulative average annual effective doses of ( $^{226}\text{Ra} + ^{232}\text{Th} + ^{40}\text{K}$ ) radionuclides were  $0.832 \mu\text{Sv y}^{-1}$  for the children and  $0.369 \mu\text{Sv y}^{-1}$  for adults, which are

**Table 2. Comparison of water measurement results of activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  with those obtained in other parts of the world**

Country	Water source	Activity concentration ( $\text{Bq l}^{-1}$ )			Reference
		$^{226}\text{Ra}$	$^{232}\text{Th}$	$^{40}\text{K}$	
Saudi Arabia	Drinking water	0.105- 0.568 (0.32)	0.016 - 0.382 (0.12)	2.16 - 18.84 (10.96)	Present study
Ghana	Well water	0.38	0.54	5.05	(Darko et al. 2015)
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)
Egypt (Assiut city)	Drinking water	0.07- 0.54 (0.20)	0.05-0.5 (0.13)	3.25- 8.72 (5.29)	(El-Gamal and El-Mageed 2014)
Yemen	Groundwater	2.25-3.45 (2.95)	0.3-1.37 (0.72)	26.73-43.7 (34.9)	(El-Mageed et al. 2013)
Malaysia	Drinking water	0.70-7.03 (2.86)	0.55-8.64 (3.78)	22- 53 (15.2)	(Almayahi, Tajuddin, and Jaafar 2012)
Turkey	Bottled drinking water	0.517 - 1.22 (0.78)	0.232 -1.87 (1.05)	1.54 - 2.57 (2.19)	(Kabadayı and Gümuş 2011)

**Table 3. Annual effective dose due to the intake of natural radionuclides  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in the present drinking water**

Radionuclide	Annual effective dose ( $\mu\text{Sv/y}$ )	
	Children	Adults
$^{226}\text{Ra}$	Minimum	0.159
	Maximum	1.231
	Average	0.675
$^{232}\text{Th}$	Minimum	0.011
	Maximum	0.134
	Average	0.040
$^{40}\text{K}^{22}$	Minimum	0.023
	Maximum	0.200
	Average	0.117
Cumulative average annual effective dose	0.832	0.369



**Fig.1. Percentage contribution to the total absorbed dose of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$**

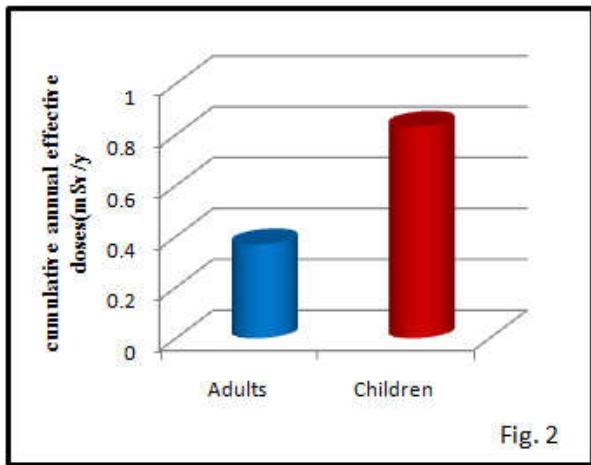
#### Annual effective dose results

The values of an annual effective dose of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  for children and adults with corresponding average values were tabulated in Table 3. It can be indicated that:

much below the recommended annual dose level 0.2 and  $1.0 \text{ mSv}^{-1}$ , for children and adults, respectively, published by WHO 2006, ICRP(1996).

- The cumulative dose received by children is higher than that received by adults and  $^{226}\text{Ra}$  causes the major dose. In fact, “ $^{226}\text{Ra}$  is an extremely radiotoxic radionuclide; the ingestion radium is saved in growing bones, the children have a higher risk factor because of their intensive bone growth during these years, and action should be taken to limit their ingestion” (Fatima et al. 2007; Al-Ghamdi 2014).
- The contributions of average dose of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  to the cumulative annual effective dose due to the intake of the drinking water samples were for children (81.13 %, 4.85%, 14.02%, respectively), and (48.78%, 14.37%, 36.85%, respectively) for adults.
- The contributions of the cumulative effective dose from a year’s consumption of water samples by the children and adults were estimated, respectively, to be 83.2 % and 36.9% of the WHO limit value  $100 \mu\text{Sv}^{-1}$ . Consequently, it can be recommended that the present drinking water samples not be creating any health effect and be acceptable

for lifelong human consumption. The percentage contribution to the total absorbed dose of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  and a comparison of age-dependent cumulative annual effective doses for children and adults in this study were illustrated in Fig. 1 & Fig 2.



**Fig. 2. A comparison of age-dependent cumulative annual effective doses**

milligrams per liter. In Table 4, the concentrations of Zn of the water samples under investigation ranged between 0.04 to 0.26 mg l<sup>-1</sup>, these concentrations of Zn, were lower than the guidelines of WHO and EPA (2003). The concentrations of Mn in all water samples were found to be less than WHO standard and ranged between 0.02 mg l<sup>-1</sup> and 0.24 mg l<sup>-1</sup>. Cu and Ni concentrations in the water samples ranged between 0.06, 0.12 mg l<sup>-1</sup>, 0.016, and 0.26 mg l<sup>-1</sup> respectively.

The highest concentrations of Pb were 0.02 mg l<sup>-1</sup> and lower than the detection limit. The concentrations of Cu, Ni and Pb were lower than the WHO and EPA and, standards. Fe concentrations in the water samples were between 0.09 and 0.51 mg l<sup>-1</sup>, only four samples (w6, w11, w13 and w14) concentrations exceeded the recommended values given by WHO guideline. The mean elemental contractions in the water samples were 0.25, 0.15, 0.08, 0.07, 0.03 and 0.02 mg l<sup>-1</sup> for Fe, Zn, Cu, Mn, Ni and Pb, respectively and decreased as Fe >Zn > Cu> Mn > Ni >Pb. These results are below the guideline values as presented in Table 4. Therefore, the present water samples are safe for drinking purposes.

**Table 4. Concentrations of heavy metals in drinking water samples together with WHO and EPA limits**

Sample Code	Heavy metals concentration (mg l <sup>-1</sup> )					
	Zn	Fe	Mn	Cu	Ni	Pb
W1	0.22	0.23	0.02	0.06	0.050	<D.L
W2	0.07	0.15	0.04	0.07	0.025	0.01
W3	0.14	0.11	0.03	0.06	0.016	0.01
W4	0.16	0.26	0.24	0.11	0.031	0.01
W5	0.11	0.09	0.08	0.07	0.032	0.02
W6	0.19	0.47	0.07	0.06	0.043	0.02
W7	0.26	0.27	0.08	0.08	0.037	<D.L
W8	0.06	0.13	0.14	0.06	0.035	<D.L
W9	0.19	0.30	0.02	0.08	0.026	0.02
W10	0.04	0.09	0.12	0.07	0.013	0.02
W11	0.26	0.31	0.04	0.10	0.009	<D.L
W12	0.15	0.13	0.05	0.12	0.022	<D.L
W13	0.06	0.51	0.08	0.06	0.027	0.01
W14	0.16	0.45	0.07	0.11	0.022	<D.L
W15	0.20	0.16	0.04	0.11	0.043	0.01
Average	0.15	0.25	0.07	0.08	0.03	0.014
WHO(2006)	-	-	0.4	2	0.07	0.01
EPA(2002)	5	0.3	0.05	1.3	0.05	0.015

**Table 5. Mean concentrations, ADDs, and HQs of heavy metals in drinking water samples**

Heavy metals	Mean concentration	RfD (mg/kg body weight/day)	ADD (mg/kg body weight/day)	HQ ((Hazard Quotient))
Zn	0.15	$3.0 \times 10^{-1}$	0.0032	0.011
Fe	0.25	$7.0 \times 10^{-1}$	0.0054	0.008
Mn	0.07	$2.4 \times 10^{-2}$	0.0015	0.063
Cu	0.08	$4.0 \times 10^{-2}$	0.0017	0.043
Ni	0.03	$2.0 \times 10^{-2}$	0.0006	0.030
Pb	0.014	$3.5 \times 10^{-3}$	0.0003	0.086

### Heavy metals analysis

Table 4, contains the heavy metal's concentration results and the comparison with the recommended values of the World Health Organization (*WHO 2006*) and United States Environmental Protection Agency (*EPA 2002*). In the present investigation, the elemental concentrations are expressed in

### Hazard quotient

"The Risk of the heavy metals through ingestion may be characterized using a hazard quotient (HQ). This as the ratio of the average daily dose (ADD; milligrams per kilogram of body weight per day) and the reference dose (RfD, milligrams

per kilogram per day). The HQ was calculated by using the following equation" (Wang *et al.*, 2005, Akoto *et al*, 2014):

$$\text{HQ} = \text{ADD} / \text{RfD} \quad \dots \dots \dots \quad (3)$$

If  $\text{HQ} > 1.00$ , representing that there is a potential risk associated with that metal. The ADD of metals was determined using the following equation (Zhuang *et al.*, 2009):

$$\text{ADD} = C_{\text{metal}} \times W / m \quad \dots \dots \dots \quad (4)$$

Where  $C_{\text{metal}}$  is the concentration of heavy metals in water sample; W represents the average daily consumption of water, which is 1.5 liter for adults (UNSCEAR2000). ; m is the body weight of 70 kg for adults. The RfD values of all the considered heavy metals were taken from US-EPA(1993). Estimated exposure and hazard quotient due to the intake of drinking water under investigation were summarized in Table 5. The HQ ranges from 0.011 (Cu) to 0.086 (Pb). All the HQ values were below the critical value of 1 as reported by US-EPA. Consequently, these drinking waters are suitable for the human use with no harmful health effect.

## Conclusion

$^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  activity concentrations in the drinking water samples collected from various locations in Makkah region, Saudi Arabia, were measured using gamma-ray spectrometry. The average value's results of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  concentrations in the water samples were 0.32, 0.12 and 10.96 Bq  $^{-1}$ , respectively, these concentrations were quite low. The total annual effective dose was found to be less than the reported value in WHO 2006. Thus, these radionuclides in the studied drinking water samples do not create any significant health risk to the public. The obtained results of heavy elements as Zn, Fe, Mn, Cu, Ni and Pb showed that the investigated elemental concentrations in the water samples did not exceed the limit values reported by WHO, EPA and TSE-266 guidelines. The hazard quotients of all the measured heavy metals in drinking water samples were below 1, posing no risk to the individuals and there is no special radiological significance of the studied waters. The obtained data from this work of heavy metals and natural radionuclides are necessary for establishing a guidelines and safety standards for drinking water in Saudi Arabia and can be used to evaluate possible future changes.

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