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Monitoring the levels of radon and toxic elements pollutants in bottled drinking water

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▶ ***Original article***

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ABSTRACT

Background: The existence of radioactive isotopes and toxic elements in water poses a potential threat to public health. Due to the high consumption of bottled water, the focus of this study is on measuring the concentration of radioactive isotopes (^{222}Rn and ^{226}Ra) and toxic elements (Cr, Mn, Co, Ni, Cu, Zn, Cd, Pb) in different brands of bottled drinking water. ***Materials and Methods:*** Therefore, twenty-four samples of bottled water have been carefully selected from local markets in Saudi Arabia. Nuclear track detector of type CR-39 and Inductive Coupled Plasma-Mass Spectrometer were used to measure radioactivity concentrations and toxic elements, respectively. ***Results:*** It is observed that the activity concentration of ^{222}Rn and ^{226}Ra in all samples was lower than the recommended values set by different agencies such as WHO, and EPA. The annual effective dose for three age groups (infants, children, and adults) was calculated from the concentration of ^{222}Rn. These measurements provide basic information for consumers who could be at risk of exposure through bottled water consumption. ***Conclusion:*** then all types of bottled drinking water are suitable and safe for daily population ingestion.

Keywords: *Bottled drinking water, radon, toxic elements, CR-39, inductive coupled plasma-mass spectrometer.*

INTRODUCTION

Water has many different and varied benefits to humans. The average amount that a person needs to drink is eight and ten cups per day. Therefore, its quality is of great importance to human health and it must be free from any toxic and natural radioactive elements. Toxic elements such as chromium (Cr), cobalt (Co), nickel (Ni), copper (Cu), zinc (Zn), arsenic (As), manganese (Mn), cadmium (Cd), lead (Pb), and mercury (Hg), and radioactive materials such as uranium, thorium and their decay products (radium, and radon) have several impacts on human (1, 2). The radiation released from radium could affect tissues in the bone marrow which could cause a bone tumor. During the decay of radium, radon gas is formed. Long exposure time to radon and its daughters will cause health changes in respiratory functions which may lead to lung cancer (3). In addition, a large amount of radon in drinking water may cause a substantial risk such as stomach and gastrointestinal cancer (4). Therefore, measurement of radium, and radon concentration in water is important from the radiological perspective. The Environmental Protection Agency (EPA) and World Health Organization (WHO) have developed a standard level of 11 Bq/L and 100 Bq/L for drinking water, respectively (5,6). Furthermore, it has been given that the annual effective dose produced by radioisotopes of radon in drinking water is 0.1 mSv/y (6, 7). CR-39 is one of the most reliable detectors that are widely used in ^{222}Rn measurement and belongs to etched nuclear track detectors (ENTDs). CR-39 polymer detectors are made from allyl diglycol carbonate monomer and its chemical formula $C_{12}H_{18}O_7$ [8]. The pollution of drinking water with toxic metals is a substantial threat to society and the environment. The amount of the toxic metals that are absorbed by the digestive system varies greatly depending on the chemical structure of the metals, the age of the individual and the nutritional status. Once a toxic metal is absorbed, it spreads itself in organs and tissues. The excretion process occurs mainly through the kidney and digestive system. However, metals tend to stay in some storage areas, such as the kidney, liver, and bones for years [9]. The kidneys are the first target organs affected by metal toxicity, in addition, other infections may occur [10]. Therefore, a high level of toxic metals in bottled drinking water may have detrimental side effects. Many studies have been submitted on natural radioactivity and toxic elements concentration in water in several countries [11-18]. Due to the large consumption of bottled drinking water this research focuses on determining the natural radioactive and toxic elements concentration in different brands of bottled drinking water currently consumed in Saudi Arabia, using CR-39 nuclear track detector and Inductive Coupled Plasma-Mass Spectrometer, respectively. Also, estimating the radiological risk resulting in the human using the water in this region.

MATERIALS AND METHODS

Different types of bottled drinking water samples were selected from the local markets in Saudi Arabia. Twenty-four samples of bottled drinking water were collected from (Nova, Hana, Safa Makah, Mawared, Berain, Aquafina, Nestle, and Al-Qassim) types. According to table 1, three samples of each type of bottled drinking water were analyzed by placed it in a plastic flask with 5 cm radius and 15 cm height as shown in figure 1. A Sheet of CR-39 detector was cut into pieces of 1 cm^2 and pasted at the bottom of the plastic lid per flask. The lid was closed tightly to ensure that there are no outlets for alpha particles emitted from ^{222}Rn. One flask without water sample was used as a reference to calculate the background radiation. The flasks were left for six months to ensure that the equilibrium was reached between radioactive isotopes [19]. After exposure time all detectors were collected together and etched by NaOH solution with normality of 6.25 N at 70 °C for 7h using water bath technique [8]. The number of alpha tracks can be observed after the etching process using an optical microscope with magnification 400X. Toxic elements namely Cr, Mn, Co, Ni, Cu, Zn, Cd and Pb in bottled drinking water samples were measured with Inductive Coupled Plasma-Mass Spectrometer, (ICP-MS) Thermo Fisher scientific, Instrument at Central Laboratory College of Science (CLCS) King Saud University, Riyadh, KSA. The external calibration was carried out using multi-element standard of the ultra-scientific analytical solution. Each sample was analyzed in three replicates. In this study all data were analyzed using statistical program "SPSS program" with using appropriate statistical tests described in results section. P value <0.05 was considered as significant level.

RESULTS AND DISCUSSION

Radioactivity analysis

Concentration levels of ^{222}Rn and ^{226}Ra were assessed for twenty-four samples from different local bottled drinking water brands commercially consumed in Saudi Arabia, which are recorded in table 1. Table 1 shows that the activity concentration range of ^{222}Rn in Nova, Hana, Safa Makah, Mawared, Berain, quafina, Nestle and Al-Qassim bottled drinking waters. This study shows that the concentration of ^{222}Rn are lower than WHO action level of 100 Bql^{-1} in all water brands [20], and also lower than EPA action level of 11 Bql^{-1} [21]. The results show variation in radioactivity concentration of ^{222}Rn and ^{226}Ra in all the bottled water brands, this may be due to radioactivity in bottled water is based on various parameters such the

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geochemistry of origin isotopes, the interaction among water and rigid juncture with which it reaches into friction through its period during the terrestrial crust (22, 23). According to WHO guidelines, the activity concentration of ^{226}Ra in all bottled drinking water are within the WHO action level 1 Bql^{-1} (7). Figure 2 displays the comparison between the activity concentration for both ^{222}Rn and ^{226}Ra. It is observed that in all samples the concentration of ^{222}Rn was lower than those from ^{226}Ra. ^{226}Ra appears with a high concentration in all samples due to the location of the main source of the water. The common main source of bottled water is deep aquifers that have been in longer contact with the host rock of the aquifer than shallow ground water or spring water (24, 25). By comparing our results for radon concentrations in this study with the data of other studies for different sites in the world. It's clear that, the range of radon concentration in bottled water in this study was lower when compared to the values reported in Austria, Kuwait, Egypt, and Serbia (26, 27, 32, 33), and was higher than the values reported in Iran, Brazil, Thailand, and Iraq (28-30, 31) as shown in table (2).

Table 1. Activity concentrations of ^{222}Rn, and ^{226}Ra in bottled drinking water samples.

Sample No	Sample source	Sample Code	Rn-222 (Bq/L)	Ra-226 (Bq/L)10^{-3}
1	Nova1	N1	1.15±0.002	1.45±0.002
2	Nova2	N2	1.23±0.014	1.56±0.020
3	Nova3	N3	1.45±0.059	1.83±0.076
4	Hana1	H1	1.49±0.067	1.88±0.086
5	Hana2	H2	1.31±0.031	1.65±0.039
6	Hana3	H3	1.89±0.149	2.39±0.190
7	SafaMakah 1	S1	0.67±0.100	0.85±0.125
8	Safa Makah2	S2	0.96±0.041	1.22±0.049
9	Safa Makah 3	S3	1.10±0.012	1.39±0.014
10	Mawared1	M1	1.03±0.027	1.30±0.033
11	Mawared2	M2	0.93±0.047	1.18±0.057
12	Mawared3	M3	0.89±0.055	1.12±0.069
13	Berain1	B1	0.46±0.143	0.58±0.180
14	Berain2	B2	0.80±0.073	1.01±0.092
15	Berain3	B3	0.62±0.110	0.79±0.137
16	Aquafina1	A1	0.41±0.153	0.52±0.192
17	Aquafina2	A2	0.48±0.139	0.61±0.174
18	Aquafina3	A3	0.57±0.120	0.72±0.151
19	Nestle1	E1	1.85±0.141	2.34±0.180
20	Nestle2	E2	2.11±0.194	2.67±0.247
21	Nestle3	E3	2.46±0.265	3.11±0.337
22	Al-Qassim1	Q1	1.60±0.090	2.02±0.114
23	Al-Qassim2	Q2	1.18±0.004	1.49±0.006
24	Al-Qassim3	Q3	1.13±0.006	1.43±0.006

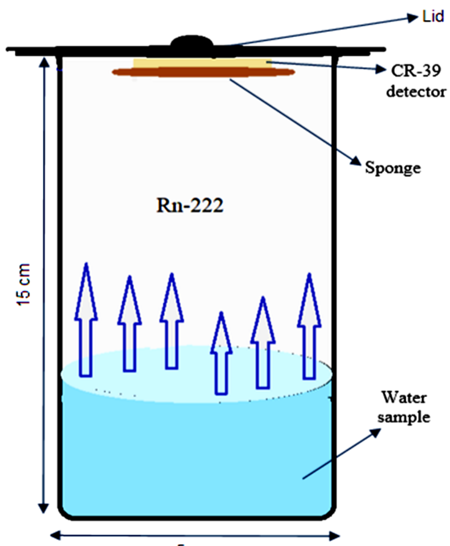

Figure 1. Closed-can technique of radon measurements.

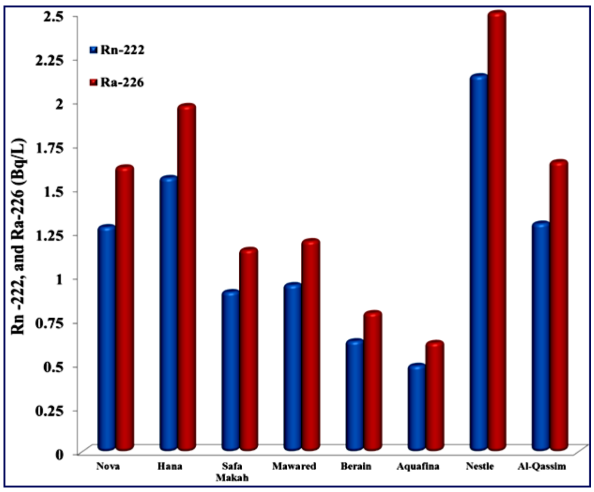

Figure 2. Distribution of ^{222}Rn, and ^{226}Ra in bottled drinking water.

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Table 2. Comparison concentration of Rn-222 in bottled waters with different sites in the world.

Country	Source of water	Rn-222 (Bq/L)	References
Saudi Arabia	Bottled drinking water	0.41- 2.46	Present work
Austria	Bottled drinking water	0.12–18	(26)
Serbia	Bottled drinking water	0.91-1463	(27)
Thailand	Bottled drinking water	0.2– 0.3	(28)
Brazil	Bottled drinking water	0.1	(29)
Iran	Bottled drinking water	0 – 0.90	(30)
Iraqi	Bottled drinking water	0.0354 - 0.248	(31)
Kuwait	Bottled drinking water	1.02 - 6.05	(32)
Egypt	Bottled drinking water	0.93 - 6.89	(33)

Statistical analysis

Table 3 displays the descriptive statistics of activity concentrations for ^{222}Rn, and ^{226}Ra in (Bq/L) for twenty-four samples of bottled drinking water. Descriptive statistics have elementary statistics (mean, median, variance, std. deviation, skewness, kurtosis, etc.) of a group of data. The mean, median, std. deviation, variance, skewness, kurtosis of activity concentrations of ^{222}Rn are 1.1571 Bql^{-1}, 1.1150 Bql^{-1}, 0.54082 Bql^{-1}, 0.292 Bql^{-1}, 0.699, and 0.080 respectively. In addition, for ^{226}Ra they are 2.9575 Bql^{-1}, 2.8500 Bql^{-1}, 1.3811 Bql^{-1}, 1.907 Bql $^{-1}$, 0.706, 0.092, respectively. Figures 3 and 4 display normal distributions for histogram of activity concentrations for ^{222}Rn, and ^{226}Ra, this normality of frequency distributions for both radioactive isotopes ^{222}Rn and ^{226}Ra are indicated by two parameters; One-Sample Kolmogorov-Smirnov test, and the values of the kurtosis. The first one has many applications such as commerce processes, environmental analysis, and production suppose suitable normality of data is ordinarily established using tests of normality (37, 35). Kolmogorov Smirnov test was applied on the current data of the activity concentrations of ^{222}Rn, and ^{226}Ra using SPSS statistical program. Asymp. Sig (p-values) are 0.919, and 0.960 for ^{222}Rn, and ^{226}Ra respectively, both p-values > 0.05 which means the data of the concentration of ^{222}Rn, and ^{226}Ra follows the Poisson distribution as shown in figures 3 and 4 (35,36). The second parameter, which indicates the Gaussian distribution is the value of the kurtosis. The q-kurtosis of activity concentrations of ^{222}Rn, and ^{226}Ra are 0.080, 0.092 respectively, since q-kurtosis is finite in the range of $0 \le q < 3$, this range of q-kurtosis is sufficient to refer for q-Gaussian distributed data sets as one does in the standard definition of the kurtosis (37,38).

Table 3. Descriptive Statistics of ^{222}Rn, and ^{226}Ra in (Bq/L).

Descriptive Statistics	Rn-222 (Bql^{-1})	Ra-226 (Bql^{-1})
Mean	1.1571	2.9575
Median	1.1150	2.8500
Std. Deviation	0.54082	1.3811
Variance	0.292	1.907
Skewness	0.699	0.706
Std. Error of Skewness	0.472	0.472
Kurtosis	0.080	0.092
Std. Error of Kurtosis	0.918	0.918
Range	2.05	5.23
Minimum	0.41	1.06
Maximum	2.46	6.29
One-Sample Kolmogorov-Smirnov Test		
Kolmogorov-Smirnov Z	0.554	0.506
Asymp. Sig. (2-tailed)	0.919	0.960

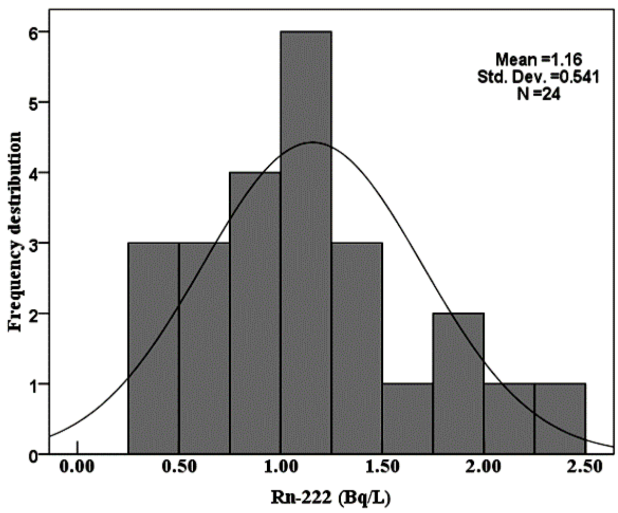

Figure 3. Frequency distribution of activity concentration of ^{222}Rn (Bq/L).

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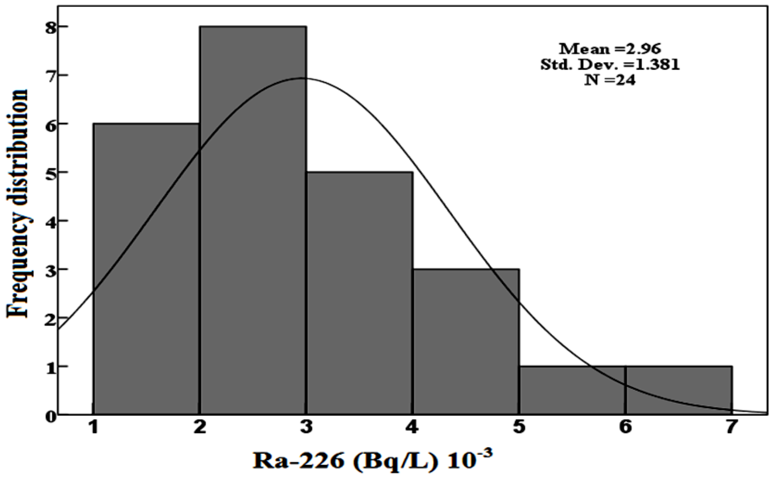

Figure 4. Frequency distribution of activity concentration of ^{226}Ra (Bq/L).

Contribution of ingestion annual effective dose

The ingestion annual effective doses for the individuals resulting from the ^{222}Rn concentration in different drinking water samples among different types of age groups (infants (1–2 y), children (7–12 y) and adults (>17 y) are summarized in table 4. It is safe to assume that Nova, Hana, Safa Makah, Mawared, Berain, Aquafina, Nestle, and Al-Qassim were the main sources of bottled drinking water of types for daily water consumption in Saudi Arabia. Table 4 gives the mean annual effective dose due to ingestion of ^{222}Rn in drinking water for infants, children and, adults respectively. Figure 5 illustrates contribution of each type of bottled drinking water to the total ingestion annual effective dose due to ^{222}Rn among different types of age groups. The percent contribution dose due to ^{222}Rn for the infant age group can be ordered as follows; Nestle > Hana >Nova, and Al-Qassim > Safa Makah, and Mawared > Berain > Aquafina. The Nestle bottled water has an ingestion effective dose of 23% from the total dose percentage while the Aquafina bottled water has a lower contribution of 5% from the total dose percent. The contribution dose for children age group can be ordered as follows; Nestle > Hana, Nova, and Al-Qassim > Safa Makah, and Mawared > Berain, and Aquafina. For children age group the Nestle water has an ingestion effective dose of 25% from the total dose percent while two other bottled drinking water brands, Berain and Aquafina, have a lower contribution of 5% from the total dose percent. The contribution dose for adult's age group can be ordered as follows; Nestle > Hana >Al-Qassim > Nova > Mawared and Aquafina > Safa Makah, and Berain. For adult age group, the Nestle water has the ingestion effective dose of 25% from the total dose percent, while three other bottled drinking water brands, Safa Makah, and Berain, have a lower contribution are 8% from the total dose percentage. From the current analysis of data of ingestion dose, all values of annual effective dose for all types of bottled drinking water due to ingestion of ^{222}Rn for three different age groups (infants, children and adults) are lower than the recommended value of 0.1 mSvy^{-1} [7, 39].

Toxic elements analysis

The results of toxic elements analysis using inductive Coupled Plasma-Mass Spectrometer for bottled drinking water are concluded in table 6, along with the worldwide standard levels. Figure 6 shows the average concentration of toxic elements is ordered as Zn > Cr > Pb > Ni > Cu > Cd > Co > Mn, and all toxic elements concentrations are lower than the Action levels of WHO organization [7] as seen in table 5. The relation between radon concentrations and toxic elements were produced in figure 7a to 7g. All relations have poor negative correlations between ^{222}Rn and all toxic elements in all types of bottled drinking water, these poor correlations may be due to various geochemical behavior for radon and toxic elements [40, 41].

Physicochemical parameters

Table 7 shows the physicochemical parameters in different brands of bottled drinking waters. All physicochemical parameters such as TDS, PH, K, Na, Mg, Ca, Fe, Nitrates, fluorides, Chlorides, SO_4, and HCO_3 were consistent with the permissible levels according to different agencies like WHO, EU [42], and FDA [43] in these bottled water samples. Hence, all bottled drinking water brands which were analyzed in this work are safe, and proper for human intake.

Table 4. Annual effective dose (mSv/y) due ^{222}Rn concentration in different brands of bottled water.

Sample No	Sample source	Sample code	Annual effective dose (mSv/y) 10^{-3}		
			Infant	Children	Adult
1	Nova1	N1	6.07 ± 0.010	2.23 ± 0.004	2.93 ± 0.006
2	Nova2	N2	6.53 ± 0.083	2.40 ± 0.031	3.15 ± 0.039
3	Nova3	N3	7.67 ± 0.316	2.82 ± 0.116	3.71 ± 0.153
4	Hana1	H1	7.86 ± 0.355	2.89 ± 0.131	3.80 ± 0.171
5	Hana2	H2	6.92 ± 0.163	2.55 ± 0.061	3.34 ± 0.078
6	Hana3	H3	9.98 ± 0.788	3.67 ± 0.290	4.82 ± 0.380
7	Safa Meka1	S1	3.55 ± 0.525	1.31 ± 0.192	1.72 ± 0.253
8	Safa Meka2	S2	5.09 ± 0.210	1.87 ± 0.078	2.46 ± 0.102
9	Safa Meka3	S3	5.81 ± 0.063	2.14 ± 0.022	2.81 ± 0.031
10	Mawared1	M1	5.45 ±0.137	2.01 ± 0.049	2.63 ± 0.067
11	Mawared2	M2	4.92 ± 0.245	1.81 ± 0.090	2.38 ± 0.118
12	Mawared3	M3	4.70 ± 0.290	1.73 ± 0.106	2.27 ± 0.141
13	Berain1	B1	2.44 ± 0.751	0.90 ± 0.276	1.18 ± 0.363
14	Berain2	B2	4.22 ± 0.388	1.55 ± 0.143	2.04 ± 0.188
15	Berain3	B3	3.30 ± 0.576	1.21 ± 0.212	1.59 ± 0.280
16	Aquafina1	A1	2.19 ± 0.802	0.80 ± 0.296	1.06 ± 0.388
17	Aquafina2	A2	2.56 ± 0.727	0.94 ± 0.267	1.24 ± 0.351
18	Aquafina3	A3	2.99 ± 0.639	1.10 ± 0.235	1.44 ± 0.310
19	Nestla1	E1	9.79 ± 0.749	3.60 ± 0.276	4.73 ± 0.361
20	Nestla2	E2	11.17 ± 1.031	4.11 ± 0.380	5.40 ± 0.498
21	Nestla3	E3	13.02 ± 1.408	4.79 ± 0.518	6.29 ± 0.680
22	Al-Qassim1	Q1	8.46 ± 0.477	3.11 ± 0.176	4.09 ± 0.231
23	Al-Qassim2	Q2	6.24 ± 0.024	2.30 ± 0.010	3.01 ± 0.010
24	Al-Qassim3	Q3	5.98 ± 0.029	2.20 ± 0.010	2.89 ± 0.014

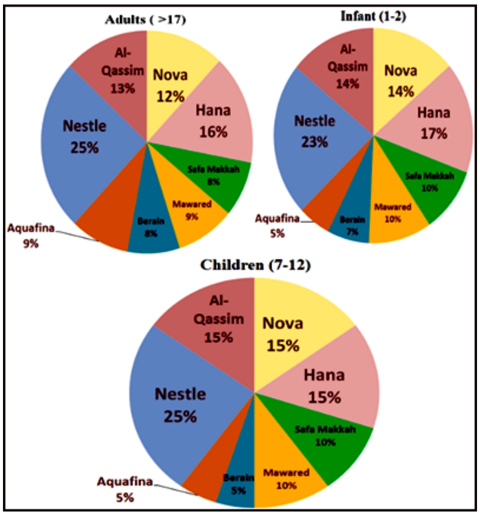

Figure 5. Contribution of each type of bottled water to the total ingestion annual effective dose due to ^{222}Rn among different types of age groups.

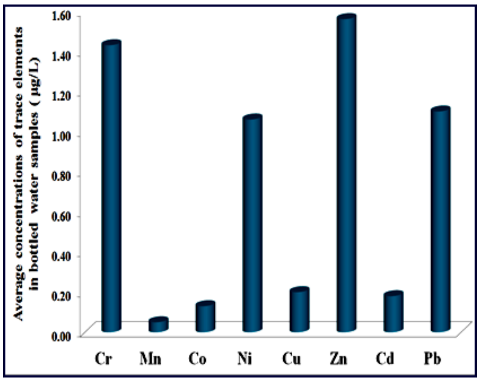

Figure 6. Average concentration of trace element ($\mu g/L$) in different brands of bottled drinking water.

Table 5. Mean of annual effective dose (mSv/y) due to ^{222}Rn ingestion for three groups of age (Adult, children, and infant) in different types of bottled drinking water.

Water type	Mean of annual effective dose (mSv/y) 10^{-3}		
	Adult	Children	Infant
Nova	6.75 ± 0.14	2.49 ± 0.05	3.26 ± 0.07
Hana	8.25 ± 0.44	3.04 ± 0.16	3.99 ± 0.21
Safa Makah	4.82 ± 0.27	1.77 ± 0.10	2.33 ± 0.13
Mawared	5.02 ± 0.22	1.85 ± 0.08	2.43 ± 0.11
Berain	3.32 ± 0.57	1.22 ± 0.21	1.60 ± 0.28
Aquafina	2.58 ± 0.72	0.95 ± 0.27	1.25 ± 0.35
Nestle	11.33 ± 1.06	4.17 ± 0.39	5.47 ± 0.51
Al-Qassim	6.89 ± 0.18	2.54 ± 0.07	3.33 ± 0.09

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Table 6. Concentrations of trace element (µg/L) in different brands of bottled drinking waters, and worldwide standard levels.

No	Water type	Cr	Mn	Co	Ni	Cu	Zn	Cd	Pb
1	Nova	2.34 ± 0.08	0.23 ± 0.05	0.41± 0.05	1.51 ± 0.05	0.35±0.04	1.61 ± 0.18	0.76 ± 0.05	8.50±0.26
2	Hana	1.21 ± 0.30	0.00	0.05 ± 0.08	0.84 ± 0.06	0.16±0.10	1.10 ± 0.30	0.08 ± 0.01	0.00
3	Safa Makah	1.34 ± 0.087	0.04 ± 0.01	0.12 ± 0.02	1.14 ± 0.08	0.10 ±0.02	2.29 ± 0.11	0.10 ± 0.01	0.00
4	Mawared	1.87 ± 0.05	0.05 ± 0.02	0.08 ± 0.003	1.23 ± 0.13	0.16±0.04	2.05 ± 0.20	0.08 ±0.006	0.00
5	Berain	1.20 ± 0.11	0.00	0.06 ± 0.004	0.86 ± 0.02	0.34±0.11	0.99 ± 0.24	0.10 ± 0.01	0.30±0.01
6	Aquafina	1.02 ± 0.03	0.07 ± 0.02	0.07 ± 0.01	0.80 ± 0.05	0.00	1.07 ± 0.04	0.08 ± 0.01	0.00
7	Nestle	1.25 ± 0.20	0.00	0.10 ± 0.03	0.91 ± 0.13	0.24±0.10	1.67 ± 0.25	0.10 ± 0.02	0.00
8	Al-Qassim	1.21 ± 0.40	0.03 ± 0.01	0.14 ± 0.04	1.15 ± 0.26	0.06±0.10	1.73 ± 0.46	0.10 ± 0.02	0.00
9	Average	1.43	0.05	0.13	1.06	0.20	1.56	0.18	1.10
10	Action level WHO	50	500	50	70	2000	3000	3	10

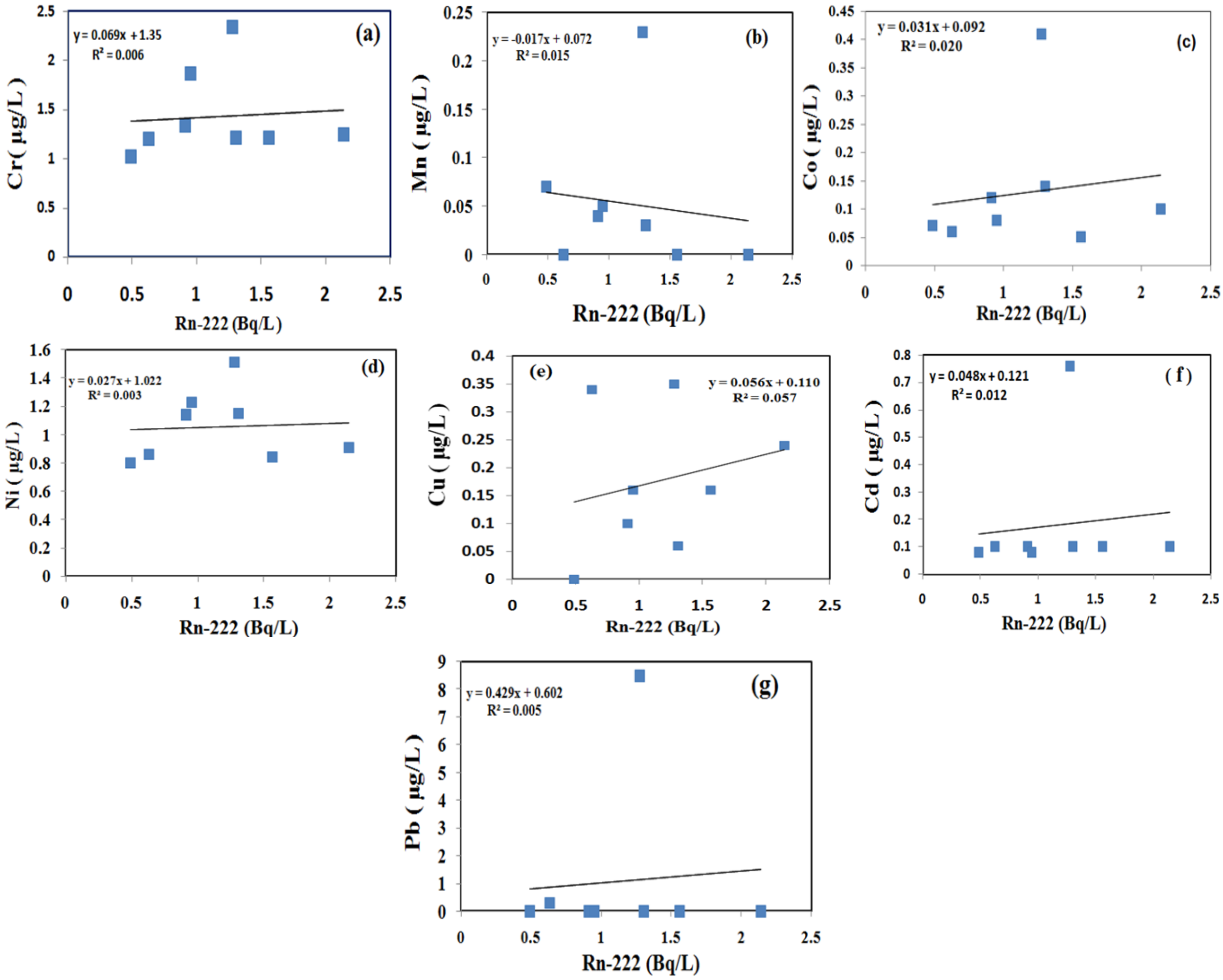

Figure 7. Relation between concentration of 222Rn and toxic elements in bottled drinking water: **(a)** Cr vs. Rn, **(b)** Mn vs. Rn, **(c)** Co vs. Rn, **(d)** Ni vs. Rn, **(e)** Cu vs. Rn, **(f)** Cd vs. Rn, **(g)** Pb vs. Rn.

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Table 7. Physicochemical parameters in different brands of bottled drinking water in mg/L.

Water Type	TDS	PH	K	Na	Mg	Ca	Fe	Nitrates	fluorides	Chlorides	So_4	$HCO3^-$
Nova	120	7.4	1.2	17	3.4	11	0	3	1	19	26	26
Hana	120	7.8	8	3	9	21	0	0.2	1	32	28	18
Safa Makah	110	7.2	0.20	13	1.5	12	0	0.04	1	26	7	28
Mawared	120	7.2	1.5	12.3	3	14.4	0	2	0.9	17.5	28	24
Berain	155	8	0.9	25	2	15	0.01	0.1	1.1	32	7	70
Aquafina	110	7	1	16	13	Less than 5	0.01	Less than 0.1	1	27.5	51	1.3
Nestle	120	7	0.2	9.5	2.3	27	Less than 0.02	Less than 1	0.8	50	10	22
Al- Qassim	140-180	6.85-7.4	1	25	2	12	0	5	0.95	35	25	10
Action level	500	7.0–9.2	——	200 mg/l	50	75–200 mg/l	0.2^b	$10\text{-}40^c$	1.5	300	250	50

a (WHO., 2011), b (EU., 2005), c (FDA., 1998)

CONCLUSION

The following bottled drinking water; Nova, Hana, Safa, Makah, Mawared, Berain, Aquafina, Nestle, and Al-Qassim are widely consumed in Saudi Arabia. Radioactivity analysis was done for all bottled drinking water samples providing additional evidence about the health risks. The activity concentration of ^{222}Rn, and ^{226}Ra was below the safe limits recommended by WHO organization in all the bottled drinking water tested. The annual effective dose of total ingestion for three different age groups (infants, children and adults) was assessed and the value for each age group was lower than the recommended value of 0.1mSv/y by WHO. The concentration of toxic elements and physicochemical parameters were within the global levels according to different agents, and no relations were found between radon concentrations and toxic elements, then all types of bottled drinking water are suitable and safe for daily population ingestion.

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