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# Spatial distributions of natural radionuclide concentrations of bottled mineral water: doses estimation and health risk assessment

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**Abstract**

**Background:** Evaluation of the various types of water reveal that groundwater comprises 99% of the earth's available fresh water. Many factors affect the type and degree of mineralization, as well as the natural radionuclides content in these types of water. The consumption of bottled natural mineral water, which comes from groundwater, among Iranians is gradually increasing. Therefore, the detection of high concentrations of radionuclides, associated with consumption of groundwater, is proposed as a public health problem in several areas.

**Methods:** In this study, the activity concentration of natural radionuclides such as $^{226}$Ra, $^{228}$Ra, $^{210}$Pb, and $^{40}$K, annual effective dose for three age groups (<1 year, 7-12 years, and >17 years), and excess lifetime cancer risk due to the ingestion of natural radionuclides present in 70 different commercial bottled mineral waters from most provinces of Iran, were evaluated. Activity concentrations were measured using gamma spectrometry and a high purity germanium detector (HPGe).

**Results:** The results showed that the activity concentrations of natural radionuclides were higher than those reported in the same studies in other countries. Also, the annul effective dose for the three age groups was much higher than the recommended value (0.1 mSvyr$^{-1}$), as reported by the WHO. The excess lifetime cancer risk for three radionuclides, $^{226}$Ra, $^{228}$Ra, and $^{40}$K, were less than the acceptable value of $10^{-3}$ for radiological risk, while the risk for $^{210}$Pb was higher than the recommended value.

**Conclusion:** According to the results of this study, the frequent use of bottled waters produced in different provinces of Iran possess health hazards to consumers.

**Keywords:** Mineral waters, Spectrometry, Gamma, Radium-226, Potassium-40, Iran

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## Introduction

Fresh water comprises a very small fraction of all Earth's water, only 2.5% of all the water supply on earth is fresh water, while groundwater encompasses approximately 99% of earth's available fresh water. There are three classes of rocks including igneous, sedimentary, and metamorphic rocks. They contain certain amounts of natural radioactivity attributable to the elements of uranium (U) and thorium (Th) decay series (1). Different concentrations of natural radionuclides, including $^{40}$K, as well as the Th and U decay series especially $^{226}$Ra,$^{228}$Ra, $^{234}$U, $^{238}$U, and $^{210}$Pb are widely distributed in nature. Many factors affect the type and degree of mineralization, as well as the natural radionuclides content in these types of water. The most important factors include: (1) the type of rock forming the substrate of aquifer, because by their interaction, water can dissolve some of the salts and natural radionuclides present in the substrate composition, (2) the contact time of water-substrate, which depends on the type of substrate and the water filtration velocity (longer water residence time implies higher solubilization of different components of the substrate rock) (2). Furthermore, it depends on the local geological conditions and interactions between the water

and solid phases through which it comes into contact during its circulation across the terrestrial crust. These factors cause different types and degree of mineralization (3-5). Radium is considered as a highly toxic element and requires attention to human health. Two natural radium isotopes ($^{226}$Ra and $^{228}$Ra) create concern in water supplies. $^{226}$Ra and $^{228}$Ra are the results of $^{238}$U and $^{232}$Th decay, respectively. Radium enters groundwater through aquifers soil disintegration. The behavior of radium in body is similar to that of calcium, it deposits in bone, which in turn can cause bone and head-sinus cancer (6). Natural bottled mineral waters come from groundwater sources all around the world. The consumption of bottled mineral water by Iranians is gradually increasing. According to this trend, the consumption of mineral water has increased rapidly and has become popular among the public (7). According to the reports of the United Nations Scientific Committee on the effects of atomic radiation (UNSCEAR) in 2000, an average radiation dose of 0.29 mSvyr$^{-1}$ is received via ingestion of natural radionuclides of $^{238}$U and $^{232}$Th series and $^{40}$K during habitual consumption of food and water worldwide (8). Therefore, it is of utmost importance to strictly control bottled water quality from the radiological aspect and ensure compliance with the national and international standards for radionuclides. In fact, measurement of natural radioactivity in drinking water is necessary for public health studies, which allows the evaluation of population exposure to radiation due to the consumption of bottled mineral water. Consequently, several studies have been conducted in recent years to investigate the natural radioactivity level, annual effective dose, and excess life time cancer risk for public in various types of water such as bottled, groundwater, spring, and tap waters in South Korea (9), United Arab Emirates (10), Algeria (11), Guilan (12), Malysia (13), Vietnam (14), Ghana (15), and Ramsar (16). Several predominant dissolved natural radionuclides in water, which cause health hazards, are members of the U and Th decay series, allowing the assessment of population exposure to radiation through water consumption (17,18). This study aimed to investigate the activity concentrations and annual effective doses of $^{40}$K, $^{228}$Ra, $^{226}$Ra, and $^{210}$Pb in bottled mineral waters produced in Iran.

## Materials and Methods

### Sampling locations and chemical characteristics

From June to September 2014, 70 available bottles of mineral water were recognized as those produced in Iran. The main sources of bottled mineral water are from the North and West regions of Iran, because the major sources of groundwater and springs originate from the Zagros and Alborz mountain ranges. Figure 1 shows the locations of sampling sites.

The samples were not filtered prior to analyses for various parameters. Moreover, they were initially analyzed for non-radioactive contaminants. The pH of the samples

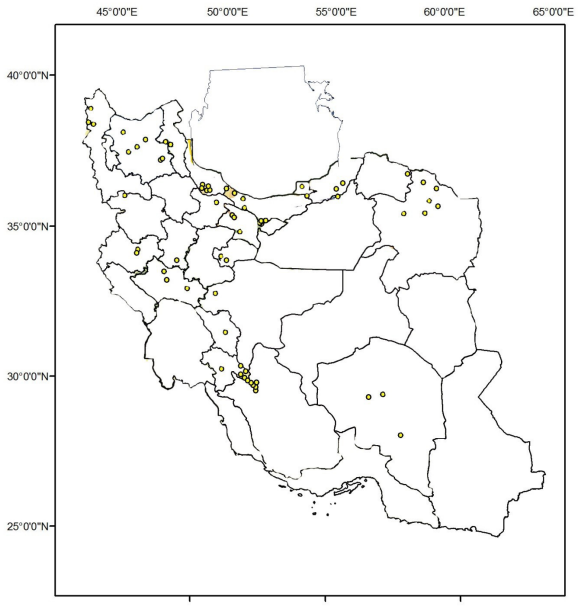


**Figure 1.** The origin points of bottled mineral water in Iran.

was measured using a pH meter (Corning 120 pH/MV meter). The turbidity was determined using a turbidity meter (HACH model 2100A). The conductivity and TDS were measured using an EC/CDS meter (HACH model 44600). The concentrations of sodium ($Na^+$), magnesium ($Mg^{2+}$), calcium ($Ca^{2+}$), chloride ($Cl^-$), fluoride ($F^-$), bicarbonate ($CaCO_3^-$), nitrate ($NO_3^-$), and sulfate ($SO_4^{2-}$) were measured according to the standard methods for the examination of water and wastewater (22nd Edition) (19). All measurements were carried out at room temperature (25°C). The mean, maximum, and minimum values of non-radioactive parameters for all 70 samples of bottled mineral waters are given in Table 1.

### Analytical methods of radioactivity

The activity concentrations of $^{210}$Pb, $^{40}$K, $^{228}$Ra, and $^{226}$Ra in the bottled mineral water samples were determined using gamma-ray spectrometry with a high resolution provided by a high purity germanium (HPGe) detector (DSG, Germany, Model NCG4020). Each water sample was placed on the top of the HPGe detector and counted at least 7200 s. The activity concentration of $^{226}$Ra was estimated from its gamma-ray peak at 186 keV. The activity concentration of $^{228}$Ra was determined using the energy peak of 911 keV of $^{228}$Ac. The activity concentrations of $^{210}$Pb and $^{40}$K were estimated from the gamma-ray peak of 46 keV and 1462 kev, respectively. Gamma-ray spectra were analyzed using InterWinner 7.0. All samples were tightly sealed and left for 3 weeks or more to make sure that equilibrium between the radionucleotides was achieved.

### Quantification of $^{226}$Ra

$^{226}$Ra was directly quantified by measuring the 186 keV energy line. The 186 keV energy line especially in gamma

**Table 1.** Physicochemical properties of 70 bottled mineral water samples (mg $L^{-1}$)

| Chemical Parameter | Mean | Max | Min | SD* |
|---|---|---|---|---|
| pH | 7.71 | 8.32 | 7 | 0.24 |
| Turbidity | 0.15 | 1.09 | 0.019 | 0.17 |
| Hardness | 148.17 | 448 | 0 | 76.50 |
| Ca | 40.89 | 91.38 | 0 | 20.36 |
| Mg | 11.32 | 74.17 | 0 | 10.96 |
| $Cl^-$ | 10.61 | 75.93 | 0.99 | 11.95 |
| Na | 106.31 | 887.31 | 2.9 | 138.80 |
| K | 6.22 | 37.65 | 0,023 | 7.78 |
| $SO_4^{2-}$ | 27.83 | 140.81 | 1.65 | 31.74 |
| $HCO_3^-$ | 131.87 | 278 | 30 | 54.46 |
| $NO_3^-$ | 12.61 | 49.51 | 0.76 | 10.18 |

spectrometry of naturally occurring radioactive material (NORM) was subjected to significant interference from $^{235}U$, which has an energy line at 185.71 keV. During each spectrum analysis, the 185.71 keV peak should be removed. It is assumed that the entire 186 keV peak is due to $^{226}Ra$. Therefore, the following correction factors were used to correct the $^{226}Ra$ value.

*Corrected Ra-226=0.5709 × apparent Ra - 226*

The correction factor of 0.5709 was calculated by Giles. Giles shows that in the Uranium decay series, three out of every seven gamma-rays at 186 keV are due to $^{235}U$. Thus, the real activity of $^{226}Ra$ acquired from the measurement of the gamma ray at 186.21 keV was corrected by multiplying by 0.5709 (4/7) (20). The corrected $^{26}Ra$ values are presented in Table 2.

## Shielding and background spectrum

HPGe gamma-ray detectors are very sensitive and powerful tools for estimating gamma-ray radiation. High-resolution gamma-ray spectrometry with the HPGe detector with a relative efficiency of 40% was used to

**Table 2.** Activity concentrations of $^{226}Ra$, $^{228}Ra$, $^{210}Pb$, $^{40}K$ (Bq $L^{-1}$) determined in bottled mineral waters

| Sample | Locality | $^{226}R$ | $^{228}Ra$ | $^{210}Pb$ | $^{40}K$ |
|---|---|---|---|---|---|
| MW1 | Yasuj | <0.017 | 0.59 | 9.51 | 4.40 |
| MW2 | Saqez | 0.265 | <0.013 | 16.37 | <1.29 |
| MW3 | Mazandaran | <0.017 | <0.013 | 2.04 | <1.29 |
| MW4 | Shiraz | <0.017 | <0.013 | 16.36 | <1.29 |
| MW5 | Tonekabon | <0.017 | <0.013 | 25.88 | 28.58 |
| MW6 | Khorramabad | <0.017 | 1.79 | 9.87 | 22.27 |
| MW7 | Shiraz | <0.017 | 2.03 | 2.63 | 17.99 |
| MW8 | Ramsar | <0.017 | <0.013 | 10.19 | 13.76 |
| MW9 | Mashhad | <0.017 | 1.06 | 5.19 | 24.04 |
| MW10 | Yasuj | <0.017 | 1.18 | 12.48 | 18.21 |
| MW11 | Shiraz | <0.017 | 1.32 | 2.89 | 26.90 |
| MW12 | Kerman | <0.017 | <0.013 | 10.57 | 18.49 |
| MW13 | Shiraz | <0.017 | <0.013 | 8.19 | 36.21 |
| MW14 | Mashhad | <0.017 | 1.64 | 23.40 | 29.59 |
| MW15 | Shiraz | 0.027 | <0.013 | <0.15 | <1.29 |
| MW16 | Shiraz | <0.017 | <0.013 | 19.58 | <1.29 |
| MW17 | Rudbar | 0.243 | <0.013 | 8.26 | <1.29 |
| MW18 | Yasuj | <0.017 | 2.26 | 12.23 | 2.48 |
| MW19 | Tabriz | <0.017 | 4.37 | 23.40 | <1.29 |
| MW20 | Qom | <0.017 | <0.013 | 12.16 | 10.48 |
| MW21 | Yasuj | <0.017 | <0.013 | 19.76 | <1.29 |
| MW22 | Mashhad | 0.484 | 0.014 | 24.94 | <1.29 |
| MW23 | Qazvin | 0.755 | 0.70 | 29.12 | 22.97 |
| MW24 | Tehran | 0.545 | <0.013 | 21.10 | <1.29 |
| MW25 | Rasht | <0.017 | 1.08 | 2.11 | 46.92 |
| MW26 | Arak | <0.017 | 0.97 | 27.60 | 10.40 |
| MW27 | Shahrekord | <0.017 | 3.48 | 12.21 | 38.65 |
| MW28 | Tehran | 0.214 | <0.013 | 1.92 | 25.95 |
| MW29 | Khorramabad | 0.503 | 1.84 | 8.13 | 9.10 |
| MW30 | Koohsar | 0.452 | <0.013 | 10.03 | 1.54 |
| MW31 | Bandar Gaz | <0.017 | <0.013 | 25.43 | 64.60 |
| MW32 | Mashhad | <0.017 | 0.44 | 5.86 | 44.65 |
| MW33 | Rudbar | <0.017 | <0.013 | 17.09 | 12.56 |
| MW34 | Karaj | <0.017 | <0.013 | 9.07 | 22.67 |
| MW35 | Bandar Gaz | 2.22 | <0.013 | 28.44 | 389.17 |
| MW36 | Amol | 0.571 | 0.98 | 16.17 | <1.29 |
| MW37 | Tehran | 0.344 | 1.05 | <0.15 | 54.26 |
| MW38 | Rudbar | 0.351 | <0.013 | 28.38 | 5.44 |
| MW39 | Dorud | <0.017 | <0.013 | 24.67 | 25.36 |
| MW40 | Qazvin | <0.017 | 0.15 | 7.03 | <1.29 |
| MW41 | Miyaneh | 0.643 | 0.38 | 12.46 | 28.13 |
| MW42 | Chaldoran | 0.281 | <0.013 | 12.80 | <1.29 |
| MW43 | Abali | <0.017 | 0.61 | 11.65 | 73.88 |
| MW44 | Shirvan | 0.240 | <0.013 | 11.85 | <1.29 |
| MW45 | Esfahan | <0.017 | 1.42 | 21.37 | <1.29 |
| MW46 | Marand | <0.017 | <0.013 | <0.15 | 2.75 |
| MW47 | Hamadan | 0.018 | <0.013 | 4.08 | <1.29 |
| MW48 | Neyshabur | <0.017 | 1.47 | <0.15 | <1.29 |
| MW49 | Mashhad | 0.329 | <0.013 | 27.15 | 46.57 |
| MW50 | Tabriz | <0.017 | 0.83 | 10.76 | <1.29 |
| MW51 | Khoy | 0.350 | 0.71 | 23.44 | <1.29 |
| MW52 | Ardebil | 0.068 | 1.72 | 6.66 | 20.54 |
| MW53 | Khoy | 0.614 | 1.71 | <0.15 | 19.86 |
| MW54 | Shahrekord | <0.017 | 13.75 | 4.82 | 143.36 |
| MW55 | Chalus | <0.017 | 2.33 | <0.15 | 17.28 |
| MW56 | Kermanshah | 0.356 | 1.20 | <0.15 | 6.00 |
| MW57 | Kandovan | <0.017 | 2.98 | 7.24 | 105.91 |
| MW58 | Ramiyan | <0.017 | 1.37 | 10.86 | 32.89 |
| MW59 | Kermanshah | <0.017 | 12.29 | 11.90 | 17.788 |
| MW60 | Khorramabad | <0.017 | 1.50 | 12.13 | 21.20 |
| MW61 | Mino Dasht | 2.037 | 2.49 | 7.34 | 1.30 |
| MW62 | Ardebil | <0.117 | <0.013 | 13.11 | 13.47 |
| MW63 | Polur | <0.017 | <0.013 | <0.15 | <1.29 |
| MW64 | Miyaneh | <0.071 | 1.58 | 9.15 | <1.29 |
| MW65 | Tabriz | 0.121 | <0.013 | 9.15 | <1.29 |
| MW66 | Shahrekord | <0.017 | 1.65 | 0.87 | 13.66 |
| MW67 | Yasuj | <0.017 | 1.54 | 21.69 | 26.63 |
| MW68 | Yasuj | 0.282 | 4.06 | 20.22 | 145.57 |
| MW69 | Kerman | 0.246 | 3.24 | 10.12 | 322.00 |
| MW70 | Kerman | 0.907 | 4.07 | 41.02 | 194.59 |

*Standard deviation.

count gamma-ray in the bottled water samples. The HPGe detector contains a 70 mm lead shield. The shielding reduces the gamma-ray background from surrounding area. Because of the uranium, thorium and potassium are in building material of counting room, furniture, and even people, they may be radioactive sources. Therefore, detecting the gamma-ray background spectra is essential, especially if the activity of naturally occurred radioactivity materials (NORM) are measured. In order to determine the gamma-ray background due to natural radionuclides in the surrounding area, an empty bottle of water under the same measuring conditions was used (20). The background spectrum of gamma radiation in the HPGe gamma-ray spectrometer is shown in Figure 2.

### Quality control

Appropriate quality-assurance processes were performed and precautions were taken to ensure the reliability of the results. Three types of calibrations (energy, resolution, and efficiency calibration) were fulfilled for the gamma spectrometer. Efficiency calibration was performed using the IAEA standard source with physical dimensions and density similar to those of the samples. The efficiency was measured on an HPGe using $^{152}$Eu efficiency at 344.286 keV.

### Spatial distributions

The most straightforward way to establish the relationship between measured radionuclides and color palette indices in map data is via ArcGIS software (21). Inverse distance weighted (IDW) interpolation determines cell values using a linearly weighted combination of a set of sample points. The weight is a function of the inverse distance. The surface being interpolated should be that of a locationally dependent variable. This method assumes that the variable being mapped decreases in influence with distance from its sampled location (22). In the present study, IDW analysis was used in GIS to measure and calculate the interpolation error for each radionuclide. The critical step in creating a topographic map from the survey data is interpolation. The data collected from sampling sites requires interpolation in order to fill the spaces where point data are not estimating the values of the assumed sites within the area covered by the point of observations. The assumptions regarding the spatial variation of elevation and the location of data collection points, are important as they can greatly affect the results (23).

## Results

### Activity concentrations of radionuclides in bottled mineral waters

The activity concentrations of bottled mineral water for each sample are shown in Table 2. Radioactivity in mineral water samples depends on several factors, such as the interaction between water and the solid phases. Concentrations of radionuclides in the various types of water samples vary over a large range. The activity concentrations of $^{226}$Ra are found to be in the range of <0.03 to 3.88 Becquerel per liter (BqL$^{-1}$). The elevated concentrations of $^{226}$Ra were restricted to natural mineral water in the studied samples. This is because water originating from deeper aquifers were in longer contact with the host rock of the aquifers. The concentrations of $^{226}$Ra were below the detection limit (<0.017 Bq L$^{-1}$) in 57.14% of the samples. The activity concentrations of $^{228}$Ra are within the range of <0.013 to 13.75 BqL$^{-1}$. While the concentrations of $^{228}$Ra were below the detection limit (<0.013 BqL$^{-1}$) in 41.42% of the samples. The concentration of $^{210}$Pb in the samples was analyzed. The concentrations ranged from <0.15 to 41.02 BqL$^{-1}$, with an average concentration of 13.62 BqL$^{-1}$. The concentration of $^{210}$Pb was below the detection limit (<0.15 BqL$^{-1}$) in 11.42% of the samples. The activity concentration of $^{40}$K was in the range of <1.29 to 389.17 BqL$^{-1}$ with an average concentration of 47.52 BqL$^{-1}$. Because of the poor shielding of the gamma-ray detector and the existence of natural potassium in environment, the activity concentration of $^{40}$K was high. The error analysis for $^{40}$K was estimated 27%. The concentration of $^{40}$K was below the detection

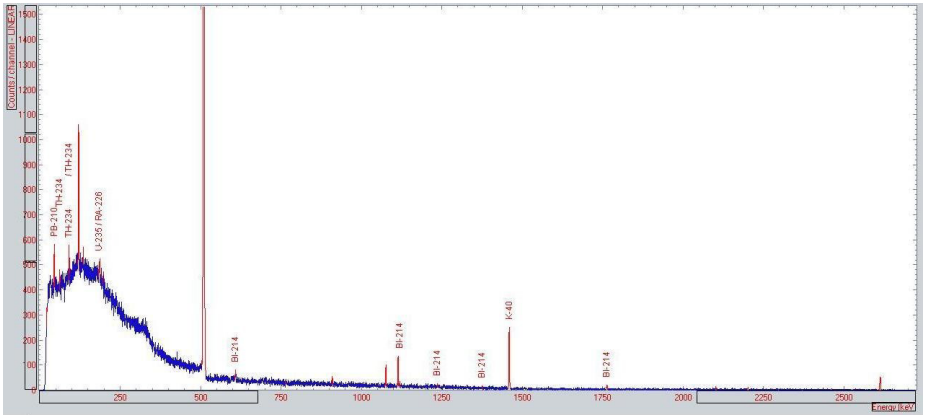


**Figure 2.** Gamma-ray spectrum of detected background.

limit (<1.29 $BqL^{-1}$) in 31.62% of the samples. A summary of the $^{226}Ra$, $^{228}Ra$, $^{210}Pb$, and $^{40}K$ activity concentrations in other countries and those observed in this study is shown in Table 3. The concentrations of all radionuclides obtained in the present study are different from those reported in other countries, such as Turkey, Spain and etc (Table 3) (2,17). The mean, maximum, minimum, and standard deviation of natural radionuclides in all 70 samples of bottled mineral water are presented in Table 4. According to the results of statistical analyses, there was no relationship between the chemical parameters and concentrations of natural radionuclides. A positive relationship was found between concentrations of $^{228}Ra$ and $^{40}K$ ($P=0.02$). Moreover, there was a relationship between the concentrations of $^{210}Pb$ and $^{228}Ra$ ($P=0.012$). No other relationships were found between the other radionuclides.

## An estimation of the annual effective dose

The annual effective dose ($mSv/yr^{-1}$) based on the intake of radionuclides through ingestion of bottled mineral waters for different age groups was estimated using the dose conversion factors recommended by the International Atomic Energy Agency (IAEA) (Table 5).

The WHO estimated the water intake rate (5,24). The age-dependent ingestion dose of E (Sv) comes from the consumption of bottled mineral waters produced in Iran, was estimated using Eq. (1) (25):

*Ingestion Dose (Sv/yr) = CONC (Bq/l) × Intake (l/y) × CDF (Sv/Bq)*

Where *CDF* is the ingestion dose conversion factor for specific radionuclides ($SvBq^{-1}$), which is shown in Table 6. The water consumption in three age groups of <1 year, 2-7

**Table 3.** Comparative evaluation of $^{226}Ra$, $^{228}Ra$, $^{210}Pb$ and $^{40}K$ concentrations in this study (Iran) and other countries (Bq $L^{-1}$)

| Country | | $^{226}Ra$ | $^{228}Ra$ | $^{210}Pb$ | $^{40}K$ |
|---|---|---|---|---|---|
| Iran (this work) | Max<br>Min | 2.22<br><0.017 | 13.75<br><0.013 | 41.02<br><0.15 | 389.17<br><1.29 |
| Slovenia (34) | Max<br>Min | 0.032<br>0.00014 | 0.0053<br>0.00005 | 0.0132<br>0.0006 | -<br>- |
| Nigeria (32) | Max<br>Min | 15.50<br>2.22 | 7.04<br>0.04 | -<br>- | 34.08<br>0.57 |
| Turkey (17) | Max<br>Min | -<br>- | - | 2.57<br>0.496 | -<br>- |
| Algeria (35) | Max<br>Min | 0.143<br>0.0134 | 0.054<br>0.0075 | -<br>- | -<br>- |
| Brazil (36) | Max<br>Min | 0.647<br>0.0033 | 0.471<br>0.012 | -<br>- | -<br>- |
| Egypt (37) | Max<br>Min | 0.92<br>0.44 | 0.87<br>0.3 | -<br>- | -<br>- |
| Italy (38) | Max<br>Min | 0.103<br>0.0002 | 0.025<br>0.0001 | -<br>- | -<br>- |
| Morocco (33) | Max<br>Min | 3.696<br>0.0091 | 0.620<br>0.0024 | -<br>- | -<br>- |
| Pakistan (18) | Max<br>Min | 0.015<br>0.008 | -<br>- | -<br>- | 0.015<br>0.008 |
| Tunisia (3) | Max<br>Min | 1.77<br>0.088 | -<br>- | -<br>- | -<br>- |
| Austria (39) | Max<br>Min | 0.225<br>0.02 | -<br>- | -<br>- | -<br>- |
| Hungary (31) | Max<br>Min | 2.94<br>0.0077 | -<br>- | -<br>- | -<br>- |
| Poland (40) | Max<br>Min | 0.355<br>0.029 | 0.32<br>0.268 | -<br>- | -<br>- |

**Table 4.** Mean isotopic activity concentrations with their max, min, and standard deviation (Bq $L^{-1}$)

| Radionuclide | Mean | Max | Min | Standard Deviation | Error Analysis |
|---|---|---|---|---|---|
| $^{226}Ra$ | 0.207 | 2.22 | <0.017 | 0.39 | 10% |
| $^{228}Ra$ | 2.19 | 13.75 | <0.013 | 2.32 | 10% |
| $^{210}Pb$ | 13.62 | 41.02 | <0.15 | 9.21 | 67% |
| $^{40}K$ | 47.52 | 389.17 | <1.29 | 66.37 | 27% |

**Table 5**. Total annual effective dose for three age groups (mSvyr$^{-1}$)

| Sample | Locality | Total Effective Dose | | |
|---|---|---|---|---|
| | | <1 yr | 7-12 yr | >17 yr |
| MW1 | Yasuj | 5.67 | 7.68 | 5.46 |
| MW2 | Saqez | 8.29 | 11.19 | 8.64 |
| MW3 | Mazandaran | 1.16 | 1.54 | 1.14 |
| MW4 | Shiraz | 8.25 | 11.06 | 8.55 |
| MW5 | Tonekabon | 15.74 | 20.64 | 14.89 |
| MW6 | Khorramabad | 8.70 | 11.68 | 7.20 |
| MW7 | Shiraz | 4.88 | 6.69 | 3.37 |
| MW8 | Ramsar | 6.46 | 8.44 | 6.00 |
| MW9 | Mashhad | 5.93 | 7.79 | 4.51 |
| MW10 | Yasuj | 9.06 | 12.10 | 8.02 |
| MW11 | Shiraz | 5.31 | 6.95 | 3.59 |
| MW12 | Kerman | 7.13 | 9.26 | 6.44 |
| MW13 | Shiraz | 7.76 | 9.79 | 6.14 |
| MW14 | Mashhad | 16.02 | 21.36 | 14.52 |
| MW15 | Shiraz | 0.22 | 0.28 | 0.16 |
| MW16 | Shiraz | 9.84 | 13.20 | 10.22 |
| MW17 | Rudbar | 4.27 | 5.79 | 4.43 |
| MW18 | Yasuj | 8.24 | 11.53 | 7.63 |
| MW19 | Tabriz | 15.46 | 21.72 | 14.47 |
| MW20 | Qom | 7.10 | 9.36 | 6.85 |
| MW21 | Yasuj | 9.93 | 13.32 | 10.31 |
| MW22 | Mashhad | 12.56 | 16.99 | 13.15 |
| MW23 | Qazvin | 17.47 | 23.43 | 16.88 |
| MW24 | Tehran | 10.67 | 14.48 | 11.18 |
| MW25 | Rasht | 6.76 | 8.48 | 4.09 |
| MW26 | Arak | 15.57 | 20.94 | 15.34 |
| MW27 | Shahrkord | 12.97 | 17.50 | 10.16 |
| MW28 | Tehran | 3.64 | 4.49 | 2.42 |
| MW29 | Khorramabad | 6.61 | 9.26 | 5.81 |
| MW30 | Koohsar | 5.20 | 7.10 | 5.44 |
| MW31 | Bandar Gaz | 19.19 | 24.63 | 16.52 |
| MW32 | Mashhad | 7.84 | 9.85 | 5.62 |
| MW33 | Rudbar | 9.75 | 12.89 | 9.51 |
| MW34 | Karaj | 6.82 | 8.76 | 5.88 |
| MW35 | Bandar Gaz | 16.22 | 22.14 | 16.42 |
| MW36 | Amol | 9.06 | 12.54 | 9.14 |
| MW37 | Tehran | 6.56 | 8.17 | 3.55 |
| MW38 | Rudbar | 14.67 | 19.72 | 15.10 |
| MW39 | Dorud | 14.81 | 19.45 | 14.09 |
| MW40 | Qazvin | 3.75 | 5.05 | 3.79 |
| MW41 | Miyaneh | 9.46 | 12.47 | 8.32 |
| MW42 | Chaldoran | 6.52 | 8.82 | 6.80 |
| MW43 | Abali | 13.84 | 17.42 | 10.22 |
| MW44 | Shirvan | 6.05 | 8.17 | 6.29 |
| MW45 | Esfahan | 11.94 | 16.38 | 11.94 |
| MW46 | Marand | 0.37 | 0.47 | 0.26 |
| MW47 | Hamadan | 2.17 | 2.90 | 2.19 |
| MW48 | Neyshabur | 1.47 | 2.28 | 0.92 |
| MW49 | Mashhad | 18.25 | 23.78 | 16.58 |
| MW50 | Tabriz | 6.17 | 8.46 | 6.08 |
| MW51 | Khoy | 12.39 | 16.95 | 12.77 |

**Table 5**. Continued

| Sample | Locality | Total Effective Dose | | |
|---|---|---|---|---|
| | | <1 yr | 7-12 yr | >17 yr |
| MW52 | Ardebil | 6.88 | 9.32 | 5.52 |
| MW53 | Khoy | 3.66 | 5.12 | 2.20 |
| MW54 | Shahrekord | 46.99 | 60.60 | 26.70 |
| MW55 | Chalus | 3.83 | 5.35 | 2.22 |
| MW56 | Kermanshah | 1.83 | 2.66 | 1.17 |
| MW57 | Kandovan | 16.95 | 21.60 | 10.92 |
| MW58 | Ramiyan | 9.91 | 13.03 | 8.04 |
| MW59 | Kermanshah | 56.1 | 71.26 | 33.01 |
| MW60 | Khorramabad | 9.48 | 12.75 | 8.24 |
| MW61 | Mino Dasht | 6.2 | 9.49 | 5.92 |
| MW62 | Ardebil | 7.89 | 10.44 | 7.58 |
| MW63 | Polur | 0.22 | 0.30 | 0.18 |
| MW64 | Miyaneh | 6.03 | 8.423 | 5.68 |
| MW65 | Tabriz | 4.69 | 6.32 | 4.86 |
| MW66 | Shahrekord | 3.24 | 4.49 | 2.05 |
| MW67 | Yasuj | 14.78 | 19.73 | 13.44 |
| MW68 | Yasuj | 28.38 | 51.83 | 20.20 |
| MW69 | Kerman | 22.45 | 36.04 | 14.42 |
| MW70 | Kerman | 43.76 | 87.62 | 33.72 |

**Table 6.** Dose conversion factors for three age groups (μSv Bq$^{-1}$)

| Radionuclide | Effective Dose Coefficient (μSv Bq$^{-1}$) | | |
|---|---|---|---|
| | Adults (>17 yr) | Children (7-12 yr) | Infants (≤ 1 yr) |
| Ra-228 | 0.69 | 3.9 | 5.7 |
| Ra-226 | 0.28 | 0.8 | 0.96 |
| Pb-210 | 0.69 | 1.9 | 3.6 |
| k-40 | 0.069 | 0.34 | 0.68 |

years, and >17 years, was assumed to be 150, 350, and 730 L, respectively.

The total annual effective doses for infants (<1 year), children (7-12 years), and adults (>17 years) are listed in Table 5. The high contribution of dose values is due to the injection of $^{210}$Pb in the bottled mineral waters. The dose ranges for infants, children, and adults were 0.22 to 56.29, 0.28 to 87.62, and 0.16 to 31.72 mSvyr$^{-1}$, respectively. As shown in Figure 3, the dose received by children (7-12 years) was higher than that received by the other two age groups (>17 years and <1 year), and the dose received by infants was higher than that received by adults (<17 years). This can be due to both dose conversion factors for this age group, as well as the usage of mineral water for infant formula production. The reference levels of effective doses from one-year consumption of drinking water recommended by the WHO and IAEA for infants, children, and adults are 0.26, 0.2, and 0.1 mSvyr$^{-1}$, respectively (26). The doses estimated in this study are much higher than the recommended reference levels for all three age groups.

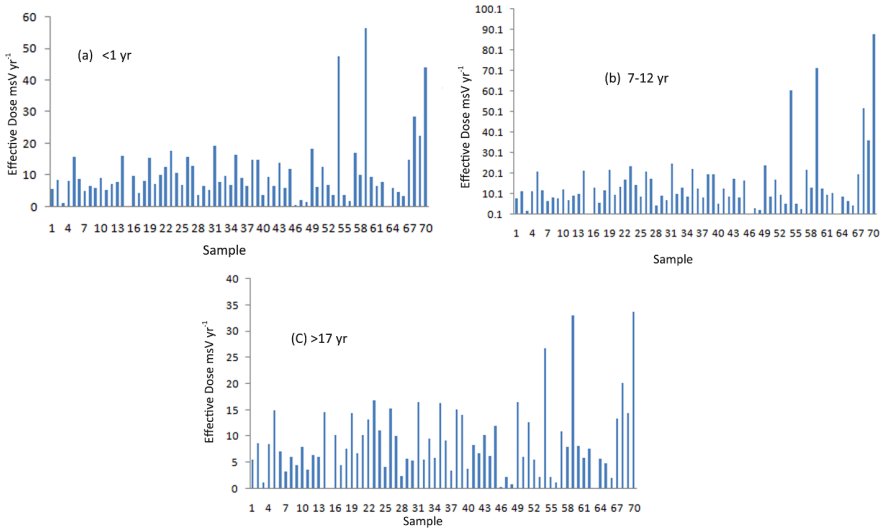


**Figure 3.** Total effective doses for three age groups of (a) <1 y, (b) 7-12 y, and (c) >17 y for mineral bottled water samples.

## Risk assessment

Radiation protection is based on the assumption that any exposure to radiation involves some risk. For prolonged exposure, as is the case for ingestion of drinking water containing radionuclides over an extended period of time, there is evidence of increased cancer risk in humans, at doses above 100 mSv. There is no evidence of increased cancer risk at doses below 100 mSv. The risk of developing cancer, as a result of radionuclide intake through ingestion of bottled mineral water, was assumed in procedures proposed by the USEPA, listed in Table 7 (27).

The coefficients used to assess the potential risk of cancer are from the long-term exposure to radionuclides in environmental media (25). The cancer risk due to intake of radionuclides in bottled mineral water is estimated according to the EPA formula:

*Risk = Radionuclide Intake (Bq) × Cancer Risk Coefficient (1/Bq)*

Intake of any radionuclide can be calculated using the following USEPA equation:

$$Intake = CF \times IR \times FI \times EF \times ED$$

Where *CF* is radionuclide concentration ($BqL^{-1}$), *IR* is the ingestion rate ($Ld^{-1}$), *FI* is the fraction ingested from contaminated source (unitless), *EF* is the exposure frequency (day $yr^{-1}$), and *ED* is the exposure duration (yr). The ingestion rate of water consumption in humans is 2 $Ld^{-1}$. Furthermore, the FI and EF are 100% or 1 and 365 d $y^{-1}$, and the ED (life expectancy at birth) is 74 years, according to the WHO recommendation reported in 2013 (22). The estimated risk of cancer due to the intake of radionuclides ($^{226}Ra$, $^{228}Ra$, $^{210}Pb$, and $^{40}k$) is reported in Table 6. The excess lifetime cancer risk for $^{226}Ra$, $^{228}Ra$, $^{210}Pb$, and $^{40}K$ was estimated to be $8.04\times10^{-5}$, $1.3\times10^{-3}$, $1.1\times10^{-2}$, and $7.6\times10^{-6}$, respectively. For all radionuclides except $^{210}Pb$, the results are lower than the acceptable level ($10^{-3}$) (25). In a similar study conducted in Yemen, the excess lifetime cancer risk for $^{226}Ra$ and $^{228}Ra$ were reported as $5\times10^{-5}$ and $3.2\times10^{-4}$, respectively, which are less than the WHO acceptable level and the results of the present study.

**Table 7.** Excess lifetime cancer risk (USEPA)

| Radionuclide | Intake ($Bq^{-1}$) | Mortality Risk ($Bq^{-1}$) | Excess Lifetime Cancer Risk |
|---|---|---|---|
| Ra-226 | 11215.2 | 7.17E-09 | 0.0000804 |
| Ra-228 | 69794.39 | 2.00E-08 | 0.001396 |
| Pb-210 | 674179.1 | 1.75E-08 | 0.011798 |
| K-40 | 1782358 | 4.30E-10 | 0.000766 |

## The spatial distribution of radionuclides found in bottled mineral waters

The use of geographic information system (GIS) can provide more accurate results when assessing environmental characteristics. In this study, GIS was used to create graphics to display the spatial distribution of complex environmental radiation of natural radionuclides in bottled mineral water samples (28). The distribution of $^{210}Pb$ in bottled mineral waters and its sources was performed by GIS, and the results are shown in Figure 4. As shown in this figure, in many provinces of Iran, especially in Khorasan and Kerman provinces, high concentrations of lead in bottled mineral waters were reported. Also, high activity concentrations of $^{226}Ra$ were reported in bottled mineral waters produced in Kerman, Tehran, and Lorestan provinces. Furthermore, the activity concentrations of $^{228}Ra$ and $^{40}K$ in West Azerbaijan, Golestan, and Kerman were higher than those reported in other provinces of Iran. The interpolation errors for $^{226}Ra$,

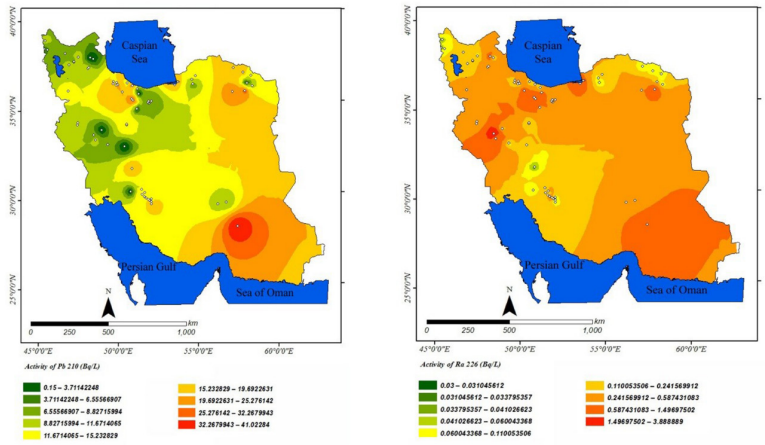


**Figure 4.** Spatial distribution of $^{210}$Pb and $^{226}$Ra in bottled mineral water as indicated by the median value at the locality

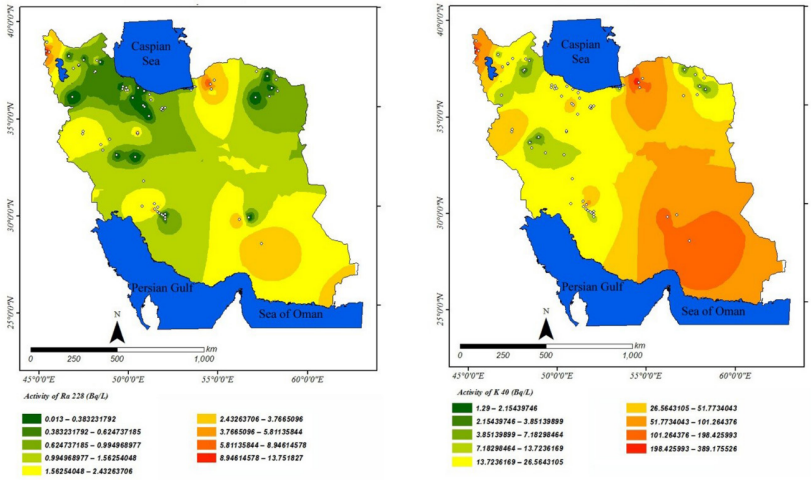


**Figure 5.** Spatial distribution of $^{40}$K and $^{228}$Ra in bottled mineral water as indicated by the median value at the locality.

$^{228}$Ra, $^{210}$Pb, and $^{210}$K were 0.011, 0.128, 0.354, and 3.249, respectively.

## Discussion

The consumption of bottled mineral water by Iranian is increasing day by day. The enormous increase of mineral bottled water brands and their consumption is a crucial issue because they have direct impacts on the public health. These types of water have variable chemical content, depending on the characteristics and peculiarities of their original aquifers, and also, contain trace amount of natural radionuclides mainly belonging to uranium and thorium series (29). Several studies have been conducted to evaluate the risk of cancer associated with high concentrations of natural radionuclides in water (25,30). To evaluate the gamma radiation dose and risk assessment due to the NORMs in different brands of bottled mineral water, 70 samples from west of Iran were examined. The difference between the number of samples in west and northwest of Iran and other parts of Iran is due to variable climate conditions and situation of aquifers in Iran.

In this study, the World Health Organization (WHO) guideline for drinking water quality was used to evaluate the activity concentrations obtained for each of the radionuclides (5). According to this guideline, the activity concentration for $^{226}$Ra is 1 BqL$^{-1}$, but in 8.57% of the bottled mineral water samples, it was higher than the recommended level. The permitted activity concentration for $^{228}$Ra is 0.1 BqL$^{-1}$, but it was higher in 56% of the samples. The recommended concentration for $^{210}$Pb is 0.1 BqL$^{-1}$, which was higher in 90% of the samples. Also, the WHO recommended the standard value of 10 BqL$^{-1}$ for $^{40}$K, which was higher in 57.14% of the samples. Also, Figures 4 and 5 show the distribution of these radionuclides in Iran. The activity concentrations of $^{226}$Ra in bottled mineral water samples ranged from 0.017 to 2.22 Bq L$^{-1}$. According to Table 3, the concentrations of $^{226}$Ra reported in the present study are very similar to the results of a study conducted in Hungary (31), but lower than those reported in bottled water and natural groundwater

in Nigeria and Morocco (32,33). The concentrations of $^{226}$Ra reported in this study are higher than the results of some studies (3,18,34-40). The WHO established a guideline for radioactivity level of some radionuclides in drinking water. The WHO recommended value for $^{226}$Ra concentration in drinking water is 1 Bq $L^{-1}$ (41), which was higher in the present study. The activity concentrations of $^{228}$Ra in this study ranged from 0.013 to 13.75 Bq $L^{-1}$, which is higher than the concentrations of $^{226}$Ra reported in all studies (0.225-0.02 and 2.94-0.0077 Bq $L^{-1}$) (31,39). The activity concentrations of $^{210}$Pb in the present study ranged from 0.15 to 41.2, which can be considered as the highest activity concentration of all the analyzed radionuclides; for instance, activity concentrations of $^{210}$Pb reported in a study in Turkey was 0.496-2.57 Bq $L^{-1}$ while it was reported 0.0006-0.0132 Bq $L^{-1}$ in another study in Slovenia (17,34). According to Table 4, the highest analysis error belonged to $^{210}$Pb isotope, which is due to poor lead shielding of the HPGe detector. The WHO recommended value for $^{210}$Pb activity concentration in drinking water is 0.1 Bq $L^{-1}$, which was below the limited level in only 8 samples studied in the present study. Thus, according to the high consumption of bottled water in Iran, the consequences of consumption of these types of water should be expected. In the present study, in 70 water samples of bottled mineral water in Iran, the activity concentration of $^{40}$K was between 1.29 and 389.1 Bq $L^{-1}$. In comparison, $^{40}$K activity concentration in bottled water was reported 0.008-0.015 Bq $L^{-1}$ in Pakistan (18) and 0.57-34.08 Bq $L^{-1}$ in Nigeria (32), which are lower than that reported in this study.

As the annual consumption of bottled mineral water is increasing day by day, and due to the fact that more than 98% of the total population are exposed to radiation comes from natural sources (42), it is necessary to estimate the total annual effective dose received from consumption of bottled mineral water. According to Table 2 and by applying equation 1, the effective dose received through consumption of bottled mineral water for three age groups (infants <1 year, children 7-12 years, and adults >17 years) was estimated, which are shown in Table 5. Several studies estimated the annual effective doses for different age groups. These studies have provided wide data on the assessment of the total annual effective dose for the consumption of different types of water such as mineral water, tap water, and spring water in different age groups. Also, the variability of different radionuclides in studies affects the variability of annual effective dose. According to the obtained results, the effective doses estimated for infants, children, and adults were higher than the maximum recommended reference levels of 0.26, 0.2, and 0.1 mSv $y^{-1}$ for infants, children, and adults, respectively (26). In a study on mineral waters in Croatia, the mean annual effective doses for $^{228}$Ra, $^{226}$Ra, and $^{210}$Pb for three age groups (1-2, 7-12 and >17 years) were estimated. In Croatia, the total annual effective dose for infants, children, and adults for $^{228}$Ra was 18.8 ± 2.1, 29.8 ± 3.3, and 7.5 ± 0.8 mSv $y^{-1}$, respectively, and for $^{226}$Ra, it was reported 6.1 ± 0.4, 11.8 ± 0.7, and 5.9 ± 0.3 mSv $y^{-1}$, respectively. In this study, the average of the total annual effective dose for three age groups, <1, 7-12, and >17 years, was 10.67 ± 9.88, 14.88 ± 15.14, and 33.72 ± 6.76 mSv $y^{-1}$, respectively (1). Another study in Slovenia estimated the annual effective doses for mineral water and tap water for three age groups of infants, children, and adults (34). The mean annual effective doses for infants, children, and adults for mineral water was 6.6, 10.1, and 6.9 mSv $y^{-1}$, respectively, and for tap water, it was reported 5.6, 8.1, and 4 mSv $y^{-1}$, respectively. A number of studies have investigated the risk of cancer associated with high concentrations of natural radionuclides in water. In the present study, the concentrations of $^{226}$Ra, $^{228}$Ra, $^{210}$Pb, and $^{40}$K in the bottled mineral waters were evaluated and the excess lifetime cancer risk was calculated. Over the lifetime consumption of bottled mineral waters at an average of 2 L $d^{-1}$, the cancer risk was estimated $8.04\times10^{-5}$, $1.3\times10^{-3}$, $1.1\times10^{-2}$, and $7.6\times10^{-6}$ for $^{226}$Ra, $^{228}$Ra, $^{210}$Pb, and $^{40}$K, respectively. For all radionuclides except $^{210}$Pb, the results are lower than the acceptable level ($10^{-3}$) (43). In a similar study conducted in Yemen, the results estimated for $^{226}$Ra and $^{228}$Ra were $5\times10^{-5}$ and $3.2\times10^{-4}$, respectively, which are less than the acceptable level for cancer risk and the results of the present study (30). In another study conducted in India, the excess lifetime cancer risk for uranium in drinking water was reported $4.3\times10^{-8}$ to $1.7\times10^{-5}$ (25). To measure radioactivity level, some studies have been conducted on drinking water in north and east of Iran. The results of this study can be compared with those of a study conducted on drinking water of 11 cities of Kermanshah province to evaluate the concentration of natural radionuclides $^{226}$Ra, $^{232}$Th, $^{228}$Ra, and $^{40}$K (44). The results obtained for $^{226}$Ra in Kermanshah province (0.57 ± 0.22) are consistent with the results of this study. Also, a study was conducted in Guilan province to determine the risk of $^{226}$Ra in drinking water samples (12). The average concentration of $^{226}$Ra (7.6 ± 0.6 mBq$L^{-1}$) and cancer mortality risk due to $^{226}$Ra ($3.28\times10^{-6}$) in drinking water in Guilan province were lower than those reported in the present study.

## Conclusion

The annual effective dose for three age groups (infants, children, and adults) and the lifetime cancer risk due to natural radioactivity in bottled mineral waters in Iran was estimated using gamma spectrometry. The activity concentration of radionuclides studied was compared to the WHO recommended level in drinking water, which in 5.87% of the samples, the concentration of $^{226}$Ra was above the recommended level (1 Bq$L^{-1}$). Also, the concentrations of $^{210}$Pb and $^{228}$Ra in 90% and 56% of the samples were above the WHO recommended level (0.1 Bq$L^{-1}$). The annual effective dose calculated for three age groups (1-2, 7-12, and >17 years) due to one-year consumption of bottled mineral waters revealed that infants (<1 year)

received the highest doses, followed by children and adults, respectively. In most provinces of Iran, high activity concentrations of $^{210}$Pb was reported. The results of estimation of lifetime cancer risk showed that the excess lifetime cancer risk for $^{210}$Pb ($1.1\times10^{-2}$) was higher than the recommended value ($10^{-3}$). Therefore, it can be concluded that some bottled mineral waters in Iran are not safe for radiological aspect of natural radionuclides $^{40}$K, $^{210}$Pb, $^{228}$Ra, and $^{226}$Ra, and frequent use of bottled waters produced in different provinces of Iran may pose health hazards to consumers in future.

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## Ethical issues

All ethical considerations have been considered in this study. The authors hereby certify that all data collected during the study are as stated in the manuscript, and no data from the study has been or will be published separated elsewhere.

## Competing interests

The authors declare that they have no conflict of interests.

## Authors' contributions

The manuscript was read and approved by all authors. All authors approved the order of authors listed in the manuscript.

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