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Radioactivity assessment and lifetime risk survey from bottled water in Iran

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Abstract

In the last few years, the tendency to utilization of bottled water has significantly expanded. Therefore, its radioactivity level must be strictly evaluated. This work aims to measure the activity concentrations of gross beta and alpha in bottled drinking water to evaluate its quality and annual effective dose as well as the lifetime risk. The radioactivity of 30 bottled mineral water samples from several brands was analyzed with Wallac Quantulus 1220 LSC. The measurement results showed that the gross beta and alpha activity concentrations in bottled mineral water samples ranged from 29 to 49 $mBqL^{-1}$ with an average of 38.7 $mBqL^{-1}$ and 48 to 76 $mBqL^{-1}$ with an average of 60.8 $mBqL^{-1}$ respectively. Furthermore, Annual effective doses ranged from 30.11 to 48.3 $\mu Sv\ y^{-1}$ with an average value of 38.54 $\mu Sv\ y^{-1}$, which is below the 0.1 $mSv\ y^{-1}$ as reference dose limit. The variation of lifetime risk is from 1.70×10^{-4} to 2.60×10^{-4} with an average of 2.14×10^{-4}. According to this study's results, radiologically, these bottled mineral waters can be considered safe drinking water in Iran.

Keywords: annual effective dose, bottled water, gross alpha and beta, lifetime risk

1. Introduction

Without a doubt, water is vitally important for human health and life. 1.8–2.0 liter of water per day is requisite for humans to be healthy. Hence drinking water must be safe and high quality for human consumption [1]. Nevertheless, urbanization and industrial development pollute the drinking water supplies due to the fast growth of the human populace [2].

The main sources of water contamination are household and industrial waste release, heavy metals disposal, marine debris and radioactive contaminant wastes.

Using water with contagious agents, chemical components and radionuclides can cause risks to health [3]. 80% of global sicknesses are due to waterborne diseases which is a significant reason for death in numerous areas of the world, particularly in kids. In addition, the rates of naturally occurring radioactivity in drinking water can be high, because it is mainly derived from groundwater that comes in contact with bedrock and soil-containing elements such as elements in the uranium and thorium decay series [4].

Furthermore, renovating the old drinking water systems is time consuming and costly. Besides, water leaking increases water to 30% in the water supply network [5]. Such problematic shortcomings make bottled water a better replacement instead of tap water. Also, bottled water provides a handy choice for many people to prevent dehydration.

For this reason, the use of bottled water is notably growing and expanding worldwide due to economic and safety matters [6]. In 2011, worldwide bottled water consumption reached 232 billion liters, a 31 percent rise from 178 billion liters used 5 years earlier [7].

Regarding quantity, China used the maximum amount of the world's bottled water in 2015, and by 2020 is expected to account for 20 percent of global bottled water utilization. Per capita, bottled water consumption in the European Union differs greatly from country to country, with an average intake of 104 liters per year [8].

The growing use of bottled water requires determining its competency for public use. Due to the low concentrations of radioactive materials in potable water, the procedure of detecting radionuclides distinctly one by one and assessing their concentration is time-consuming and high-priced [1].

The valuable parameters for basic water screening are gross beta and alpha activities; by evaluating these two parameters, important details concerning the natural radionuclides in water and their related safety risks correlated with water usage can be obtained [9].

According to the Institute of Standards and Industrial Research of Iran, the suggested guideline for activity concentration in potable water is 1 Bq/L for gross beta activity and 0.5 Bq/L for gross alpha activity. The acceptable levels for human potable water consumption are below those screening levels of gross activities. When measuring amounts surpassing the screening level, additional researches are needed to determine the radionuclides that lead to the elevated activity to measure the resulting dose of radiation to consumers [1,9].

According to existing researches, several radioactivity experiments have been done in different water samples like tap water, surface water and hot spring water, obtained from several cities in Iran and other countries [10-16]. However, gross beta and alpha radioactivity concentrations evaluations in bottled mineral water and the resulting excess lifetime cancer risk haven't been carried out in Iran yet.

Hence this research aims to evaluate the activity concentration of gross beta and alpha in bottled mineral water to analyze bottled water contamination in Iran and identify the possible safety risks for the population. The research also plans to quantify the associated annual effective dose as well as the lifetime hazards because of water use.

2. Materials and methods

2. 1. Sample collection

The 30 bottled mineral water samples from different brands that are most widely used in Iran were bought from randomly selected markets in 1.5 L size plastic bottles (made from PET). The bottled drinking water samples were taken to the laboratory, and for commercial concerns and observance of research ethics, the bottled potable water samples were labeled as BDW. The sample collection process was repeated three times (with an interval of 4 months).

To prevent the sorption of radionuclides around the walls of the container, the HNO_3 was used to acidify the BDW samples to pH < 2. The samples were stored in a cold room at temperatures set to 4 °C before the experiment.

2. 2. Experimental setup

Wallac Quantulus 1220 LSC was used to simultaneous measurement of the gross alpha and gross beta activities. The Advantages of LSC are listed below:

- 4π counting geometry (it's equal to a geometrical factor = 1)
- Without self-absorption of the samples (like about planchet counting)
- Counting efficiency equivalent to almost 100% (in planchet counting and solid state spectrometry, less than 50%)
- Easy preparation of sample by combining it with a commercial cocktail
- Concurrent alpha/beta counting (with the splitting of alpha/beta spectrum by pulse shape analysis)

The liquid scintillation counting (LSC) is a significant method not only for the evaluation of pure beta-emitting radionuclides but also for investigating of radionuclides that decay through alpha emission and electron capture [17].

2. 3. Minimum detectable activity

The least radioactivity that can be obtained when assessing a sample using a detection system is called Minimum Detectable Activity (MDA). The multiple factors on which MDA is dependent on them are counting time, sample size, counting efficiency and background. Increasing the time to count or the sample size can increase MDA. For the LSC detection system, the below equation can give MDA [18,19]:

$$\mathrm{MDA} = \mathrm{L_d} / (\mathrm{eff} \times \mathrm{V} \times \mathrm{T} \times 60), \quad (1)$$

where the sample volume is shown with *V*, the parameter *T* is indicated to the duration of the measurements (in min) and the counting efficiency is expressed by eff. *The* L_d was specified as:

$$\mathrm{L_d} = 2.71 + 4.65\sqrt{(\mathrm{B_g} \times \mathrm{T})}, \quad (2)$$

which *Bg* is the background radiation in counts per minute.

2. 4. Sample preparation and measurement of gross alpha and beta

First, 250 mL of each prepared sample was moved to a glass container. To prevent any accumulation of the samples on the walls and gathering of organic materials and also to avoid variations in the ions state in the samples, the dilute HNO_3 was utilized to acidify the samples to be pH 2.5. Next, to dry the glass was put on the hot plate. The heating temperature was controlled so as not to reach 80º C, to avoid evaporation of alpha and beta emitters which led to inaccurate readings.

Each sample was stirred through a heating procedure by utilizing a stirring magnetic capsule to preserve the homogeneousness of the samples and convey heat efficiently. Then, the residue of the compound changed to a solution by adding HNO_3 0.1M and was fully dissolved.

To prepare appropriate volume of the sample, the dilution process was done by double distilled water. After that, the samples were moved into a 20 mL vial. To create a final sample, 10 mL of Ultima Gold™ scintillation cocktail (from PerkinElmer Inc) was added to the vial until the total volume reached 20 mL. Finally, the ready sample was moved to a Liquid Scintillation Counting (LSC) for counting and analysis.

The standard solution comprising pure beta and alpha emitting materials with various pre-identified activities, measured individually, was used to determine the efficacy calibration for the gross beta and alpha counts.

The relevant standard solution of pure beta and alpha emitting materials was utilized to measure the gross alpha/beta detector efficiency under optimal PSA

Table 1. The activity concentration of gross beta and alpha in the bottled mineral water samples (n=3)

Sampling code	pH	Gross alpha (mBq L^{-1})	Gross beta (mBq L^{-1})
BDW 1	7.4	32 ± 0.83	61 ± 1.51
BDW 2	7.1	42 ± 1.01	54 ± 1.24
BDW 3	7.3	38 ± 0.95	57 ± 1.35
BDW 4	7.2	41 ± 0.98	62 ± 1.41
BDW 5	7.8	37 ± 0.93	49 ± 1.16
BDW 6	7.5	36 ± 0.9	51 ± 1.21
BDW 7	7.6	44 ± 1.06	54 ± 1.23
BDW 8	7.8	29 ± 0.75	51 ± 1.26
BDW 9	7.2	32 ± 0.84	48 ± 1.19
BDW 10	7.4	43 ± 1.03	58 ± 1.33
BDW 11	7.2	35 ± 0.91	73 ± 1.8
BDW 12	7.5	34 ± 0.88	67 ± 1.65
BDW 13	7.4	42 ± 1.02	57 ± 1.3
BDW 14	7.3	45 ± 1.08	65 ± 1.48
BDW 15	7.5	35 ± 0.91	53 ± 1.31
BDW 16	7.6	49 ± 1.18	69 ± 1.57
BDW 17	7.7	42 ± 1	74 ± 1.69
BDW 18	7.2	33 ± 0.86	62 ± 1.53
BDW 19	7.6	38 ± 0.95	69 ± 1.64
BDW 20	7.5	40 ± 1	66 ± 1.59
BDW 21	7.3	45 ± 1.08	58 ± 1.32
BDW 22	7.3	39 ± 0.98	68 ± 1.62
BDW 23	7.8	42 ± 1.02	76 ± 1.73
BDW 24	7.1	34 ± 0.89	63 ± 1.56
BDW 25	7.5	37 ± 0.93	60 ± 1.43
BDW 26	7.4	37 ± 0.92	54 ± 1.28
BDW 27	7.6	41 ± 0.97	59 ± 1.36
BDW 28	7.4	32 ± 0.84	71 ± 1.75
BDW 29	7.3	48 ± 1.15	49 ± 1.12
BDW 30	7.5	39 ± 0.96	66 ± 1.57

conditions:

$$\text{eff} = \left(N_S - B_g\right) / \left(A_{\text{Standard}} \times V \times T \times 60\right), \quad (3)$$

where, gross efficiency was shown by eff, the count of alpha or beta was displayed by N_S, the background count was indicated by Bg and also A_{Standard} indicated the pre-identified activity of standards and V corresponds to the standard solution volume.

The radioactivity concentrations of gross beta or alpha in a given quantity is then determined by the equation

$$A = N / \left(\text{eff} \times V \times 60\right), \quad (4)$$

where N is the actual counting rate, eff is the detector efficiency and V is the volume of the sample in L. To convert decay per minute (dpm) to decay per second (dps), a coefficient of 60 was used in the above equation.

2. 5. Assessment of effective dose

The effective dose for adults due to swallowing both beta and alpha emitting materials in potable water was determined using the equation [20]:

$$\text{effective dose}(\text{mSvy}^{-1}) = A \times W \times CF, \quad (5)$$

where A is the gross beta or alpha activity concentration, W is the water intake per each person annually (Ly^{-1}) and CF is the conversion factor of ingestion dose for gross alpha or beta (mSv Bq^{-1}).

The annual water consumption can alter depending on some parameters, like the outside temperature. In this study, the annual water consumption adopted is 730 L for adults [1].

3. Results and discussion

The important thing in measuring low radioactive concentrations is to determine the minimum radioactivity that can be accurately identified. In other words, one of the most significant indicators of a method's success is its detection limit. MDA values were calculated according to equation 3 for the gross alpha and beta. The MDA values for blank sample measurement for a duration of 150 min on PSA= 110 are 0.025 Bq L^{-1} and 0.035 Bq L^{-1}, for alpha and beta, respectively.

The values of the gross beta and alpha activity concentration calculated in the bottled mineral water samples are shown in table 1. The measured gross beta and alpha activity concentrations in bottled mineral water samples vary from 29 to 49 mBq L^{-1} with an average of 38.7 mBq L^{-1} and 48 to 76 mBq L^{-1} with an average of 60.8 mBq L^{-1} , respectively.

As can be seen from table 1, the gross beta activities are greater than the related gross alpha activities. It is clear to see from table 1 that all gross alpha concentration values are lower than the suggested upper limit value, that is, 0.5 Bq L^{-1}. Also, gross beta concentration values are lower than the recommended upper limit value, which means 1 Bq L^{-1}.

Basic descriptive statistics such as standard deviation, standard error, mean, maximum, minimum, kurtosis and skewness for gross beta and alpha radioactivity concentrations in the 30 mineral water samples are presented in table 2.

Table 2. Statistical data for gross beta and alpha activity measured in the 30 mineral water samples.

	Gross alpha (mBq L^{-1})	Gross beta (mBq L^{-1})
Mean	38.7	60.8
Standard Error	0.91	1.46
Standard Deviation	4.99	7.97
Kurtosis	-0.55	-0.96
Skewness	0.13	0.14
Minimum	29	48
Maximum	49	76

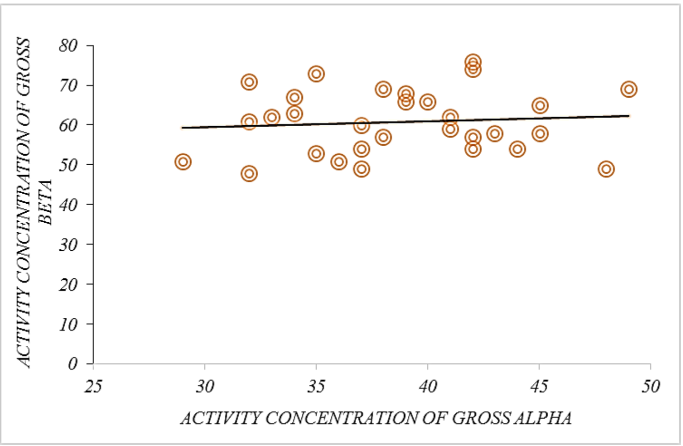

Figure 1. The weak correlation between gross beta and alpha radioactivity concentrations (mBq L^{-1}).

Table 3. Comparison of the gross beta and alpha radioactivity concentrations results of this work and other study.

Origin	Gross alpha activity concentration (mBq L^{-1})	Gross beta activity concentration (mBq L^{-1})	References
Albania	39	220	Cfarku *et al.*, 2014 [21]
Spain	< 30 – 860	< 40 – 2280	Palomo *et al.*, 2007 [22]
Turkey 1	8 – 101	17 – 177	Turhan *et al.*, 2019 [23]
Turkey 2 (mineral)	164	555	Taskin *et al.*, 2013 [24]
Serbia	1 – 13	44 – 173	Janković *et al.*, 2012 [25]
Greece	8 – 94	71 – 350	Karamanis *et al.*, 2007 [26]
Bangladesh	0.73 – 0.96	65.5 – 77.3	Ferdous *et al.*, 2016 [27]
Mexico (Purified)	< 11 – 415	< 26 – 695	Rangel *et al.*, 2002 [28]
Mexico (mineral)	< 11 – 601	211	Rangel *et al.*, 2001 [29]
Catalonia	20 – 1700	40 – 2900	Ortega *et al.*, 1996 [30]
Iran (bottled mineral water)	29 – 49	48 – 76	This work

The arithmetic mean is 39 and 61, the standard deviation is 5 and 8, the standard error is 0.17 and 0.27, the coefficient of skewness is 0.13 and 0.14, coefficient of kurtosis is -0.55 and -0.96 for alpha and beta, respectively. Skewness coefficients have positive values, and kurtosis coefficients have negative values. This circumstance recommends that the distributions appeared in table 1 are relatively asymmetric.

Figure 1 shows the correlation between gross beta and alpha activity concentrations of the bottled mineral water samples. The correlation coefficient is one of the criteria used to determine the association (or dependence) of two variables. The correlation coefficient indicates how much the change in one variable is consistent with the change in the other variable. The weak correlation (0.1) between the results of gross beta and alpha means that it is not possible to predict the gross beta concentrations from the gross alpha concentration for other water samples.

Table 3 reports the measured gross beta and alpha radioactivity concentrations in water samples in other countries. The reported values for gross alpha in different countries are in an extensive range from 1 in Serbia to 1700 in Catalonia. The reported gross beta radioactivity concentrations range from 17 in Turkey to 2900 in Catalonia.

The annual effective dose correlated with radiation exposure by drinking the bottled mineral water sample

Table 4. Estimated effective dose and lifetime risk from ingestion of gross beta and alpha activities in bottled water.

Sampling code	Effective dose (alpha) (μSv y^{-1})	Effective dose (beta) (μSv y^{-1})	Lifetime risk
BDW 1	6.54	30.73	2.07E-04
BDW 2	8.58	27.20	1.99E-04
BDW 3	7.77	28.71	2.02E-04
BDW 4	8.38	31.23	2.20E-04
BDW 5	7.56	24.68	1.79E-04
BDW 6	7.36	25.69	1.83E-04
BDW 7	8.99	27.20	2.01E-04
BDW 8	5.93	25.69	1.75E-04
BDW 9	6.54	24.18	1.70E-04
BDW 10	8.79	29.21	2.11E-04
BDW 11	7.15	36.77	2.44E-04
BDW 12	6.95	33.75	2.26E-04
BDW 13	8.58	28.71	2.07E-04
BDW 14	9.20	32.74	2.33E-04
BDW 15	7.15	26.70	1.88E-04
BDW 16	10.02	34.76	2.48E-04
BDW 17	8.58	37.27	2.54E-04
BDW 18	6.75	31.23	2.11E-04
BDW 19	7.77	34.76	2.36E-04
BDW 20	8.18	33.24	2.30E-04
BDW 21	9.20	29.21	2.13E-04
BDW 22	7.97	34.25	2.34E-04
BDW 23	8.58	38.28	2.60E-04
BDW 24	6.95	31.73	2.15E-04
BDW 25	7.56	30.22	2.10E-04
BDW 26	7.56	27.20	1.93E-04
BDW 27	8.38	29.72	2.11E-04
BDW 28	6.54	35.76	2.35E-04
BDW 29	9.81	24.68	1.91E-04
BDW 30	7.97	33.24	2.29E-04

was determined to evaluate the health risk of adult persons in the society. The effective dose based on mSv was calculated utilizing the gross beta and alpha activity concentrations, the dosage coefficient and the water intake annually according to equation 7. Table 4 displays the computed effective dose values for the analyzed water samples.

The annual effective dose values varied from 30.11 to 48.3 µSv with an average value of 38.54 µSv, which is below the 0.1 mSv y^{-1} as reference dose limit.

To comply with international safety requirements, it is necessary to examine all potential indicators related to water consumption, including radiation health danger. Lifetime risk as a result of the water intake was also assessed in the following equation:

$$\text{Lifetime risk} = \text{effective dose} \times \text{LE} \times \text{RF}, \qquad (6)$$

Where "effective dose" is the lifetime-mean dose taken annually, considering 76 years as life expectancy, LE, for Iran and the risk factor was shown by RF risk factor for radiation-induced stochastic health effects is declared 7.3×10^{-2} [20,31].

The variation of risk is from 1.70E-04 to 2.60E-04 with an average of 2.14E-04 that is slightly more than the acceptable limit of 10^{-4} [32]. In other words, according to these results, we can expect about 2 cases of the disease for 10 thousand exposed people.

4. Conclusion

Available evidences suggest that this research is the first comprehensive one on the radioactivity concentration in the bottled mineral water of Iran and the associated health hazards. Measurements of gross beta and alpha radioactivity are used as the first phase in determining water radioactivity. It is because of the simpleness of the technique, since it offers a general evaluation of both beta and alpha radioactivity levels for a cost-effective procedure during a comparatively short time. Of the entire samples, the findings have been considerably smaller than the WHO-suggested reference level.

In addition, the average value of lifetime risk is nearly equal to the appropriate 10^{-4} limit. This research findings reveal that bottled mineral waters are radiologically healthy and do not pose a significate public radiation threat. The radiometric baseline values of bottled mineral water for this area were yielded from data obtained in this study. This can help establish a national guideline for existing natural radioactive materials in potable water.

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