

STUDY ON GROSS ALPHA AND BETA RADIOACTIVITY OF SAMPLES OF BOTTLED MINERAL WATERS IN ROMANIA

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Abstract

Measurement of gross alpha and beta radioactivity in drinking water is an important screening method for assessing radiological water quality. In recent years, the trend of using bottled water has expanded significantly, therefore monitorization of the level of radioactivity has become essential. The present study aimed to evaluate the total radioactivity of bottled mineral water samples from different regions of Transylvania, Romania over a 6 year-period (2017-2022). The obtained results indicate concentrations of gross alpha and beta activities ranging from 0.0057 to 0.0801 Bq/L with a mean value of 0.0383 Bq/L for gross alpha activity, and between 0.0929 Bq/L and 0.7953 Bq/L with a mean value of 0.4415 Bq/L for gross beta activity. The radioactivity values of bottled mineral waters were situated within the permitted levels regulated by WHO, with reference values of 0.5 Bq/L for gross alpha activity and 1 Bq/L for gross beta activity, respectively. The obtained values were also within the limits recommended by the Council Directive 2013/51/ EURATOM, of 0.1 Bq/L for gross alpha activity and 1 Bq/l for gross beta activity, respectively. According to the results of this study, the investigated bottled mineral waters can be considered radiologically safe for consumption.

Keywords: gross alpha activity, gross beta activity, mineral waters, permissible limits, radiological water quality.

1. INTRODUCTION

The World Health Organization (WHO), the International Commission on Radiation Protection (ICRP) and the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) recommend that adults drink at least 1-2 L of water per day to demonstrate the positive effects on human health (Grande et al., 2015; Piñero García et al., 2021). Therefore, the supply of clean and safe drinking water (tap water, spring water, mineral water, purified water, distilled water, etc.) is very important.

Currently, due to natural events and human activities, water resources (streams, lakes, groundwater, aquifers, springs, etc.) are easily contaminated by various toxic organic and inorganic compounds and radionuclides (Khandaker et al., 2015; Yu et al., 2020). It is estimated that about one third of the world's population uses groundwater as the main source of drinking water (Calin et al., 2015). Groundwater contains dissolved mineral substances as well as natural radionuclides in the decay series ^{238}U , ^{232}Th and ^{40}K , found at different concentrations (Alseroury et al., 2018). The presence and content of these radionuclides in water are related to seasonal changes in precipitation,

infiltration time, mineralogy and geochemical composition of rocks and soils, redox conditions and weathering (Turhan et al., 2013; Chmielewska et al., 2020). High concentrations of radionuclides determine the emission of ionizing radiation (alpha, beta and gamma radiation), which, once ingested and/or inhaled, can have serious effects on human health (Khandaker et al., 2017). Therefore, the radioactive quality of drinking water must be strictly monitored on a regular basis to ensure the protection of human health.

Bottled drinking water is one of the main routes of drinking water distribution worldwide and represents a purer, safer and tastier alternative (Pourfadakari et al., 2022). Recently, the trend of replacing tap water with bottled mineral water is increasing due to consumer preferences for therapeutic and nutritional practices (Seid et al., 2020). The radiological quality of commercial bottled mineral water must be carefully and systematically controlled, due to the potential natural radioactivity, a great number of studies being carried out worldwide on this topic (Kasić et al., 2015; Labidi et al., 2018; Catani et al., 2020; Geleva et al., 2020; Kinahan et al., 2020).

The purpose of this study was to assess the levels of gross alpha and beta radioactivity in nine samples of bottled mineral water originating from Arad, Hunedoara and Harghita counties, Romania, over a 6 year-period (2017-2022), monitoring which becomes important in the context of avoiding consumption risks for population.

2. MATERIALS AND METHODS

Sampling

A total of nine water samples were collected from commonly used mineral water springs in West- and Eastern Transylvania, Romania, as follows: 4 samples from Hunedoara County ("Cezara" carbonated mineral water, "Perla Apusenilor" carbonated mineral water, "Vreau din România" carbonated mineral water and "Aqua Sara" carbonated mineral water), 4 samples from Arad County ("Lipova" carbonated mineral water, "Baile Lipova" carbonated mineral water, "Lipova" partially decarbonated water and "Baile Lipova" partially decarbonated water) and 1 sample from Harghita County ("Borsec" carbonated mineral water). Samples were collected using polyethylene bottles. Analysis was performed within one day after sampling, according to SR ISO 5667-3 recommendations.

Determination of gross alpha and beta radioactivity

Gross alpha and beta radioactivity were determined according to SR ISO 9696/2018 and SR ISO 9697/2019 standards, respectively. The sample stabilized by acidulation was evaporated to almost total dryness to give a volume of approximately 50 ml. After cooling, 1 ml of concentrated sulfuric acid was added and carefully evaporated to dryness, then calcinated using a calcination furnace (Nabertherm) at 350°C for 1 h. From the obtained residue, gross alpha radioactivity was determined using alpha TENNELEC TC 256 equipment calibrated with a standard source of Americium-241 (^{241}Am) according to SR-ISO 9696, 2018. Gross beta radioactivity was measured using the ROBOTRON 20050 equipment, previously calibrated with a standard source of (Sr-Y)-90, according to SR-ISO 9697, 2019.

The gross alpha volume activity of the water sample, c_A , expressed in Bq/L, was determined according to equation (1):

$$c_A = \frac{r_g - r_0}{V\varepsilon} = [r_g - r_0]w, \quad (1)$$

where,

c_A = alpha volume activity (Bq/L)

r_0 = counting speed of the radiation background

r_g = overall sample counting speed

V = volume of test sample equivalent to the mass of residue on the counting tray, (l).

ε = counting efficiency for the specified radioactive standard, ^{241}Am , (efficiency)

The volume of test sample (V), expressed in L, equivalent to the mass of residue deposited on the counting tray, was calculated according to formula (2):

$$V = \frac{V_t}{m} m_r \quad (2)$$

where,

m = mass expressed as calcined residue in volume V (mg);

m_r = mass of the sample residue deposited on the counting tray, (mg);

V_t = volume of water sample (L);

$$w = \frac{1}{V\varepsilon}$$

The counting efficiency, ε , efficiency, is obtained according to equation (3):

$$\varepsilon = \frac{r_s - r_0}{A} \quad (3)$$

where:

r_0 = counting speed of the radiation background;

r_s = calibration counting speed;

A = alpha activity of the calibration source; (Bq).

The gross beta volume activity of the water sample, c_A , expressed in Bq/L, is obtained by equation (4):

$$c_A = \frac{r_g - r_0 - \chi(r_{g\alpha} - r_{0\alpha})}{V\varepsilon} = \left[r_g - r_0 - \chi(r_{g\alpha} - r_{0\alpha}) \right] w \quad (4)$$

where,

c_A = beta volume activity (Bq/L);

r_0 = counting speed of the radiation background;

$r_{0\alpha}$ = rate of counting of the radiation background;

r_g = overall sample counting speed;

$r_{g\alpha}$ = overall sample counting rate from alpha calibration source to beta meter;

V = volume of test sample equivalent to the mass of residue on the counting tray (L);

ε = counting efficiency for specified radioactive standard, $^{90}\text{Sr-Y}$

χ = alpha-beta intermodulation, percentage of number of alpha particles entering through the beta window of the alpha calibration source.

The volume V of the test sample (L), equivalent to the mass of residue deposited on the counting tray, was calculated according to formula (5):

$$V = \frac{V_t}{m} m_r \quad (5)$$

where:

V_t = volume of water sample (L);

m = mass of calcined residue (mg);

m_r = mass of the sample residue deposited on the counting tray, (mg);

The counting efficiency, ε , is obtained by calculation using the equation (6):

$$\varepsilon = \frac{r_s - r_0}{A} \quad (6)$$

where:

r_s = the calibration counting speed;

r_0 = the radiation background counting speed;

A = beta activity of calibration source, (Bq).

If necessary, the alpha-beta intermodular correction factor, χ , can be calculated using equation (7):

$$\chi = \frac{r_{s\alpha \rightarrow \beta}}{r_{s\alpha}} \quad (7)$$

where:

$r_{s\alpha \rightarrow \beta}$ represents the counting speed in the beta window when measured with the standard alpha source.

If equipment other than the gas proportional meter is used, then alpha-beta intermodulation can be neglected $\chi=0$.

3. RESULTS AND DISCUSSIONS

The results on gross alpha and gross beta radioactivity of the investigated samples of bottled mineral water samples are presented in Table 1.

As shown in Table 1, the lowest value of gross alpha radioactivity (0.0057 Bq/, 2018) was recorded for the sample “Lipova” mineral water from Arad County, while the highest gross alpha activity (0.0801 Bq/L, 2017) was recorded in “Aqua Sara” carbonated mineral water from Hunedoara County, followed by the “Perla Apusenilor” carbonated mineral water from Hunedoara County (0.0723 Bq/L, 2019). The lowest value of gross beta activity (0.0929 Bq/L, 2018) was found in the partially decarbonated mineral water sample “Lipova”, while the highest gross beta activity (0.7953 Bq/L, 2017) was recorded for the “Aqua Sara” carbonated mineral water, followed by “Borsec” carbonated mineral water from Harghita County with a gross beta activity of 0.7356 Bq/L (2017).

Table 1. Gross alpha and beta radioactivity of mineral water samples, from different regions of Transylvania, Romania (2017-2022). The values are presented as mean \pm standard deviation

No.	SAMPLE	SOURCE	RADIOACTIVITY (Bq/ L)	YEAR					
				2017	2018	2019	2020	2021	2022
1.	Mineral sparkling water “Cezara”-	Hunedoara	Gross Alpha	0.0250± 0.0007	0.0473± 0.0008	0.0105± 0.0001	0.0169± 0.0001	0.0286± 0.0005	0.0222± 0.0001
			Gross Beta	0.2521± 0.0008	0.5691± 0.0584	0.2440± 0.0183	0.3469± 0.0257	0.3592± 0.0216	0.2827± 0.0182
2.	Mineral sparkling water “Perla Apusenilor”		Gross Alpha	0.0602± 0.0002	0.0538± 0.0003	0.0723± 0.0008	0.0674± 0.0005	0.0471± 0.0003	0.0627± 0.0005
			Gross Beta	0.4146± 0.0322	0.3239± 0.0287	0.5023± 0.0376	0.4588± 0.0339	0.3615± 0.0218	0.5622± 0.0335
3.	Mineral sparkling water “Vreau din România”		Gross Alpha	0.0181± 0.0003	0.0112± 0.0002	0.0229± 0.0003	0.0420± 0.0004	0.0413± 0.0003	0.0418± 0.0004
			Gross Beta	0.2960± 0.0261	0.1439± 0.0129	0.6362± 0.0475	0.5679± 0.0417	0.4687± 0.0290	0.5141± 0.0318
4.	Mineral sparkling water “Aqua Sara”		Gross Alpha	0.0801± 0.0010	0.0648± 0.0003	0.0448± 0.0006	0.0595± 0.0005	0.0356± 0.0002	0.0656± 0.0006
			Gross Beta	0.7953± 0.0621	0.6936± 0.0615	0.4663± 0.0348	0.5359± 0.0393	0.4148± 0.0249	0.6717± 0.0416
5.	Mineral sparkling water “Lipova”	Arad	Gross Alpha	0.0163± 0.0003	0.0060 ±0.0001	0.0112± 0.0002	0.0227± 0.0004	0.0334± 0.0002	0.0311± 0.0004
			Gross Beta	0.3446± 0.0311	0.1456± 0.0130	0.1570± 0.0118	0.2703± 0.0200	0.3211± 0.0194	0.2670± 0.0188
6.	Mineral sparkling water “Băile Lipova”		Gross Alpha	0.0355± 0.0001	0.0283± 0.0006	0.0146± 0.0002	0.0306± 0.0001	0.0368± 0.0003	0.0256± 0.0002
			Gross beta	0.4676± 0.0433	0.1896± 0.0168	0.4064± 0.0307	0.4577± 0.0342	0.4562± 0.0275	0.3451± 0.0202
7.	Partially decarbonization water “Lipova”		Gross Alpha	0.0263± 0.0005	0.0057± 0.0001	0.0192± 0.0001	0.0119± 0.0001	0.0239± 0.0002	0.0135± 0.0002
			Gross Beta	0.3639± 0.0332	0.0929± 0.0082	0.2123± 0.0133	0.2024± 0.0150	0.2555± 0.0154	0.2245± 0.0144
8.	Partially decarbonization water “Băile Lipova”		Gross Alpha	0.0220± 0.0004	0.0120± 0.0001	0.0155± 0.0002	0.0289± 0.0006	0.0141± 0.0001	0.0281± 0.0003
			Gross Beta	0.3043± 0.0284	0.2112± 0.0143	0.2973± 0.0165	0.3191± 0.0238	0.2247± 0.0135	0.3368± 0.0197
9.	Mineral sparkling water Borsec	Harghita	Gross Alpha	0.0620± 0.0004	0.0681± 0.0005	0.0590± 0.0003	0.0524± 0.0003	0.0610± 0.0004	0.0533± 0.0003
			Gross Beta	0.7356± 0.0633	0.5215± 0.0455	0.5131± 0.0384	0.7156± 0.0542	0.6639± 0.0411	0.5210± 0.0343

The measurement of gross alpha and gross beta radioactivities are usually conducted for a primary screening of water radiological quality. The WHO has recommended guidelines on gross alpha and gross beta activity in drinking water, setting maximum permissible limits of 0.5 Bq/L and 1.0 Bq/L respectively (WHO, 2022).

The average value of gross alpha for the year 2017 was 0.0383 Bq/L and of gross beta of 0.4415 Bq/L, respectively. For the year 2018 there was an average value of 0.0330 Bq/L for gross alpha and 0.3212 Bq/L for gross beta. In 2019, there was an average value for gross alpha of 0.0300 Bq/L, gross beta of 0.3816 Bq/L, while in 2020, the average gross alpha activity was 0.0369 Bq/L, and gross beta activity was 0.4305 Bq/L. For 2021, the average value of gross alpha activity was 0.0357 Bq/L and gross beta activity of 0.3917 Bq/L, while in 2022 average gross alpha values were 0.0382 Bq/L and gross beta 0.4139 Bq/L. Within the studied period, the highest values of gross alpha and beta activity were registered in 2017.

All the determined radioactivity values for both gross alpha and gross beta of bottled mineral water collected during 2017-2022 were situated within the limits allowed by the WHO (WHO, 2022) and the European Directive (EURATOM 2013/51).

A comparative investigation on mean values of gross alpha and gross beta radioactivity obtained in this study and other reported values, is presented in Table 2.

Table 2. Gross alpha and gross beta radioactivity of bottled mineral water samples of different origin.

Sample	Region/Country	Gross alpha activity (mean) (Bq/L)	Gross beta activity (mean) (Bq/L)	Ref.
Mineral water	West Region/ Romania Eastern Transylvania/Romania	0.0057-0.0801 (0.0383)	0.0929-0.7953 (0.4415)	Present study
	Bucovina/ Romania	0.0003-0.0550	0.0012-0.0288	Calin et al., 2015
	Southwest region/ Bulgaria	0.003-0.671 (0.107)	0.054-0.375 (0.164)	Geleva et al., 2023
	Hungary	< 0.008-3.34	0.035-2.6	Kovacs et al., 2004
	Italy	< 0.004-0.28	< 0.025-0.93	Desideri et al., 2007
	Türkiye	0.008-0.101 (0.021)	0.017-0.177 (0.059)	Turhan et al., 2019
	Sao Paulo/Brazil	< 0.01-0.11	< 0.05-1.60	Nisti et al., 2022
	City Urumqi /China	0.0044-0.1306 (0.0348)	0.0173-0.3203 (0.0834)	Kobyas et al., 2011
	Krakow / Poland	0.0022-0.938 (0.162)	0.053-10.537 (0.845)	Chau et al., 2009

As noticed in Table 2, the mean values of gross alpha and gross beta radioactivity obtained for mineral water samples from West and East regions of Romania, investigated in this study over a 6 year-period were similar or lower than most of those reported in other studies, but higher than those reported in the Turkish study (0.021 Bq/L for gross alpha activity, 0.059 Bq/L for gross beta activity, respectively). Other studies, such as those conducted in Poland and Hungary, showed samples with gross alpha and beta radioactivity values exceeding the permitted limits set by WHO.

4. CONCLUSIONS

Screening drinking water samples for gross alpha and gross beta radioactivity is important in assessing the radiological water quality.

In the present study, gross alpha and gross beta radioactivity of nine samples of bottled mineral water samples, of different Romania origin, indicates an average value for the investigated period, 2017-2022, of 0.0383 Bq/L and 0.4415 Bq/L, respectively. The lowest values of gross alpha and gross beta activity were recorded in “Lipova” partially decarbonated water, collected in 2018, as being 0.0057 Bq/L for gross alpha and 0.0929 Bq/L for gross beta. The highest value of gross alpha and gross beta activity was identified in “Aqua Sara” carbonated mineral water collected in 2017, as being 0.0801 Bq/L for gross alpha activity and 0.7953 Bq/L for gross beta activity. These values are significantly lower than the recommended limit values of 0.5 Bq/L for gross alpha activity and 1 Bq/L for gross beta activity (WHO), and of 0.1 Bq/L for gross alpha activity and 1 Bq/L for gross beta activity (Council Directive 2013/51/EURATOM).

The obtained results are significant in the context of assessment of the risk to public health caused by the consumption of radiologically contaminated water.

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