

Assessment of natural radioactivity and consequent radiological hazard in different brands of commercialized bottled mineral water produced in China

Lang Yu, Guangwen Feng, Qian Liu, Chao Tang, Baoshan Wu, Peihong Mao and Changlong Cai

ABSTRACT

As one of the drinking water quality parameters, natural radioactivity parameters are recommended to prevent a potential health threat to the public. In this study, the gross- α and gross- β activity concentrations in 15 different brands of commercial bottled mineral water consumed in China were analyzed to evaluate the quality and corresponding health impact on the population. The activity concentrations of gross- α and gross- β in different samples varied from 4.4 to 130.6 and 17.3–320.3 mBq L⁻¹, respectively. The values of the annual effective dose equivalent rate (AED) for infants, children and adults ranged from 1.3 to 21.6, 2.9–52.5 and 5.5–97.8 μ Sv y⁻¹, respectively. The average excess lifetime cancer risk caused by the consumption of bottled mineral water samples was estimated as 6.0×10^{-5} . These results show that all the measured gross- α and gross- β are found to be obviously less than the guidance level by WHO and the domestic standard. The values of AED are below the World Health Organization (WHO) recommended limit of 0.1 mSv y⁻¹. Combined with the lifetime cancer risk assessment, it is concluded that there is no significant risk for consumption of the observed brands of bottled mineral water and it can be consumed safely.

Key words | bottled mineral water, gross- α and gross- β , natural radioactivity, radiological hazard

HIGHLIGHTS

- Natural radioactivity parameters are recommended to prevent a potential health threat to the public.
- The gross- α and gross- β activity concentrations in different brands commercial bottled mineral water were analyzed to evaluate the quality and corresponding health impact on the population.
- The results obtained show that all the measured values of gross- α and gross- β in bottled mineral water samples are found to be obviously less than the guidance level by WHO and the domestic standard.
- The values of AED due to α - and β -emitters estimated for infants, children and adults are below the WHO recommended a limit of 0.1 mSv y⁻¹.
- Combined with the lifetime cancer risk assessment, it is concluded that there is no significant risk for consumption of the observed brands of bottled mineral water and it can be consumed safely.

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INTRODUCTION

Nowadays, the safety of drinking water is becoming one of the most social concerns, because the water supply source is vulnerable to contamination from toxic substances, microorganisms, radionuclides etc. (Varga 2011; Carvalho & Fajgelj 2013; Bodini *et al.* 2018). In addition, the relation between water quality problems and the risk of acute or chronic health has been shown by epidemiologic evidence, especially in diarrheal diseases, pulmonary diseases, gastric diseases, bone and sinus diseases and so on (Steynberg 2002; Hunter 2003; Isaji 2003; Ahmed 2004; Bonotto 2004; Cevik *et al.* 2006; Fatima *et al.* 2007; Ajayi & Achuka 2009; WHO 2009, 2011; Vogeltanz-holm & Schwartz 2018). Therefore, drinking water quality has become the focus of our attention (Isaji 2003; Varga 2011; Carvalho & Fajgelj 2013; He *et al.* 2014; Al-Omran *et al.* 2015).

As one of the drinking water parameters, radioactivity parameters are recommended by World Health Organization (WHO) to evaluate drinking water quality (Fatima *et al.* 2007; Trabidou & Florou 2010). Radionuclides, including uranium series, thorium series and potassium, are generally released in trace quantities to the environmental aquatic system (Dueñas *et al.* 1999; UNSCEAR 2000). The concentrations of these radionuclides in water depend on the geological characteristics of the soil and rocks or human activities. The occurrence of radionuclides in the body, ingested from drinking water and the food-chain, gives rise to internal exposure to humans, which may produce a damaging effect and a serious health risk (Dueñas *et al.* 1999; UNSCEAR 2000; Fatima *et al.* 2007; Khandaker *et al.* 2017). Therefore, radiation released from ingested drinking water theoretically has potential radiological hazards to the public exposed to significant levels. It is important to obtain the levels of the natural radioactivity as gross- α and gross- β in drinking water for providing useful information in the monitoring of drinking water quality from the perspective of public health protection (WHO 2011; Asaduzzaman *et al.* 2015; Turhan 2019).

In recent years bottled mineral water for consumption and therapeutic purposes has widely increased all over the world. Bottled mineral water contains different levels of beneficial elements, such as magnesium, zinc, iron, calcium, potassium, etc. (Chau & Michalec 2009; Taskin *et al.* 2013). However, it

should be noted that some of these minerals are usually accompanied by a higher content of natural radionuclides (Chau & Michalec 2009). In 2013, China became the world's largest consumer market of bottled water, and this has greatly increased their awareness of natural radionuclides as a potential source of ionized radiation (Rodwan 2016; GB 8537 2018). Therefore, it is important to determine the activity concentrations of natural radioactivity as gross- α and gross- β in bottled mineral water samples collected from the different brands produced in China, and to assess the radiological hazard to the human population. The results obtained in this study are presented, discussed and compared with worldwide safety limits and the corresponding results for bottled mineral water of different national origins in the literature.

MATERIALS AND METHODS

Sample collection and preparation

In this study, 45 samples of bottled drinking mineral water from 15 different brands were purchased from local supermarkets in Urumqi, China. The bottled drinking mineral water was chosen in such a way that it represented the best-selling brands, produced from different water source locations according to the production address marked on the bottle body, to ensure the variation of measurement values. Three samples of each brand of bottled mineral water were purchased from the same retailer at the same time with the same production date. The distribution of water sources is shown in Figure 1. Three samples for each brand were prepared randomly from the local markets of Urumqi city. The methods described in the relevant domestic standards were used in the pretreatment of bottled mineral water samples (HJ 898 2017; HJ 899 2017). Each water sample was prepared by using the following processes:

1. Evaporation concentration. First, a volume of approximately 2,600–12,100 mL of each bottled mineral water sample was transferred into a beaker, where a small amount of concentrated nitric acid was added to avoid precipitation of radionuclides onto the beaker wall. Second, each water sample was heated on an electric furnace at

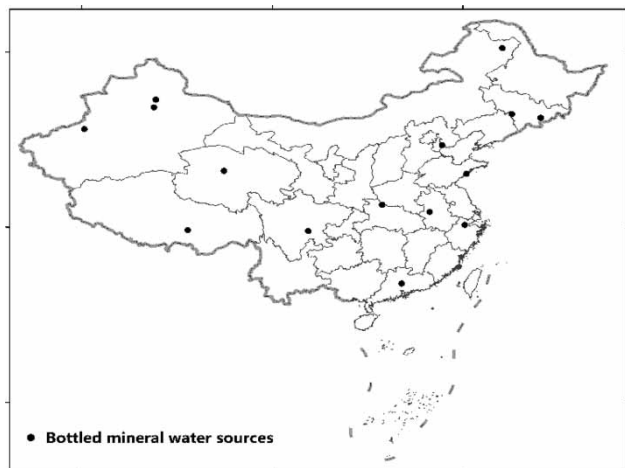


Figure 1 | The distribution of water source of nine different brands of commercial bottled mineral water.

80 °C (in order to avoid boiling) and enriched to 50 ± 5 mL. Finally, the evaporated sample was transferred into evaporating utensils, and a small amount of deionized water above 80 °C was used to wash the beaker to prevent the precipitation of salt crystal onto the beaker wall.

2. Sulfation. A volume of approximately 1 mL of sulfuric acid ($\rho = 1.84$ g/mL) was slowly added into evaporating utensils along the wall of the utensils. To prevent splashing, the evaporating utensils were heated on the infrared lamp until fumes of sulfuric acid evolved, then the evaporating utensils were continually heated on an electric furnace (below 350 °C) until the fumes were exhausted.
3. Dryness. The evaporating utensils with residue were heated for 1 hour to dryness in a muffle furnace at 350 °C, and they were then placed in a desiccator to cool, weigh and determine the mass of the dry residue.
4. Sample preparation. The residues were transferred to mortar, ground into a fine powder, and then transferred quantitatively to a stainless steel planchet (approximately 5 cm diameter) and evenly spread by organic solvent as ethanol or acetone. The stainless steel planchet with residues was kept in the desiccator to avoid moisture until counting was performed in the gross α/β counters (Ferdous et al. 2016). Before using the stainless steel planchets, the planchets were cleaned using detergent and rinsed using distilled water, then left to be dried in a drying oven at 105 °C to avoid sample contamination.

Analysis of the gross- α and gross- β activity concentration

In this work, the activity concentrations of the gross- α and gross- β in the bottled mineral water samples were measured using a gas-flow proportional α/β counter (ORTEC, MPC 9604 model), which is a low background 4-channel multiple detector type and could offer simultaneous α/β measurement of four planchets. The detectors were gas-flow window-type counters with an ultra-thin window. The counting gas was P-10 gas that contained a mixture of argon and methane in the ratio of 90:10 respectively. In order to decrease the influence of background radiation, the slider and counter tubes were shielded by the machined lead bricks of 10 cm thickness. Before the sample measurement, the background measurement and detector efficiency calibration must be performed. The environmental background of each detector was determined using an empty planchet (blank sample) under identical measured conditions. The detector efficiency calibration was performed by using a radioactivity reference material of ^{241}Am (13.6 Bq g^{-1}) for α activity and a radioactivity reference material of KCl (16.1 Bq g^{-1}) for β activity respectively, which was manufactured by the National Institute of Metrology, China. The samples (including a blank sample) were placed a certain distance below the detector under the shield, and the accumulating counting time was greater than 1,000 min.

Measurements were carried out at the laboratory of Radiation Environment Supervision Station of the Department of Ecology and Environment, Xinjiang. Periodical efficiency calibration, background and sample retesting were conducted for quality assurance.

The activity concentrations of gross- α or gross- β ($A_{\alpha,\beta}$ in Bq L^{-1}) of the bottled mineral water samples are calculated by Equation (1) (Avwiri & Agbalagba 2007; HJ 898 2017; HJ 899 2017):

$$A_{\alpha,\beta} = \frac{(R_{s(\alpha,\beta)} - R_{0(\alpha,\beta)})}{(R_{r(\alpha,\beta)} - R_{0(\alpha,\beta)})} \cdot C_{\alpha,\beta} \cdot \frac{m}{1,000} \cdot \frac{1.02}{V} \quad (1)$$

where $R_{s(\alpha,\beta)}$ is the gross- α or gross- β count rate of sample, in s^{-1} ; $R_{0(\alpha,\beta)}$ is the gross- α or gross- β count rate of background, in s^{-1} ; $R_{r(\alpha,\beta)}$ is the gross- α or gross- β count rate of reference material, in s^{-1} ; $C_{\alpha,\beta}$ is the gross- α or gross- β

activity concentration of reference material, in Bq g^{-1} ; V is the volume of the sample evaporated in liters and m is the mass in mg of the residue from volume V and 1.02 is included to correct for 20 mL of nitric acid added per liter as a stabilizer.

The minimum detectable activity (MDA in mBq L^{-1}) is calculated by Equation (2) (Turhan *et al.* 2013):

$$\text{MDA} = \frac{2.71 + 4.65 \sqrt{R_{0(\alpha,\beta)} \cdot t}}{V \cdot t \cdot \varepsilon \cdot 60} \quad (2)$$

where t is time, in seconds. The ε is detector efficiency. Other terms have been described above.

RESULTS AND DISCUSSION

Activity concentration of gross- α and gross- β in bottled mineral water

The activity concentrations of gross- α and gross- β in bottled mineral water samples, together with their respective standard deviations, are presented in Table 1. As can be seen in Table 1, the activity concentrations of gross- α and gross- β in bottled mineral water samples are in the range of 4.4–130.6 and 17.3–320.3 mBq L^{-1} , with mean values of 34.8 and 83.4 mBq L^{-1} , respectively. The results obtained show that all the measured values of gross- α and gross- β in bottled mineral water samples are found to be obviously less than the screening limit of 0.5 and 1.00 Bq L^{-1} respectively (WHO 2011), so the activity concentration analysis of individual radionuclides is not required. This also means that all the concentration values of individual radionuclides in bottled mineral water samples are unlikely to exceed the guidance levels of natural and artificial radionuclides, as recommended by WHO and the domestic standards (GB 5749 2006; WHO 2011). As known from the above analysis, the distribution of the activity concentration in the samples is not uniform, which may be caused by different geology or different processes of drinking water treatment (IAEA 2003). The activity concentrations of B_12 and B_14 samples are higher than that of other brands, the main reason may be that the water sources were derived from groundwater sources and springs which have high carbonate alkalinity (WHO 2018).

Table 1 | Activity concentration of gross- α and gross- β in bottled mineral water samples produced in China

Brands No.	Activity concentration (mBq L^{-1})	
	Gross- α	Gross- β
B_1 ^a (3 ^b)	11.6 ± 1.4	21.7 ± 1.0
B_2 (3)	7.1 ± 0.5	17.3 ± 0.9
B_3(3)	11.7 ± 0.5	88.4 ± 1.4
B_4(3)	52.2 ± 3.3	115.2 ± 1.1
B_5(3)	5.0 ± 0.2	25.6 ± 0.4
B_6(3)	4.4 ± 1.2	49.0 ± 1.6
B_7(3)	8.3 ± 0.5	159.6 ± 4.1
B_8(3)	37.1 ± 7.4	94.6 ± 7.2
B_9(3)	8.2 ± 0.9	58.6 ± 2.9
B_10(3)	15.8 ± 0.5	48.0 ± 1.1
B_11(3)	43.8 ± 1.4	40.2 ± 0.7
B_12(3)	109.0 ± 3.7	320.3 ± 1.1
B_13(3)	20.4 ± 3.0	49.7 ± 2.9
B_14(3)	130.6 ± 4.7	117.0 ± 6.2
B_15(3)	56.5 ± 3.5	46.3 ± 3.3
Range	4.4–130.6	17.3–320.3
Mean ± SD	34.8 ± 38.1	83.4 ± 75.3

^aB_1–B_15 represent brands 1–15, respectively.

^bNumber of samples.

On the basis of obtaining data, the correlations between the gross- α and gross- β activity concentrations in the analyzed bottled mineral water samples and the dry residues were investigated, see Figures 2 and 3. As can be seen from Figures 2 and 3, although the natural radioactivity levels (gross- α and gross- β activity concentrations) generally increase with increasing dry residue, their correlation in the present study is generally poor. Moderate linear correlation ($R = 0.4592$) can be found between gross- α activity concentrations and dry residue, while a weak linear correlation ($R = 0.2182$) can be found between gross- β activity concentrations and dry residue. Similar findings have been reported previously, so all investigated brands of bottled mineral water samples are classified as the low-mineral water class (residue $< 500 \text{ mg L}^{-1}$) or the oligo-mineral water class (residue $< 50 \text{ mg L}^{-1}$) (Rusconi *et al.* 2004).

As shown in Table 2, comparing the measured values of gross- α and gross- β of the analysis with other countries, it can be concluded that the activity concentrations of

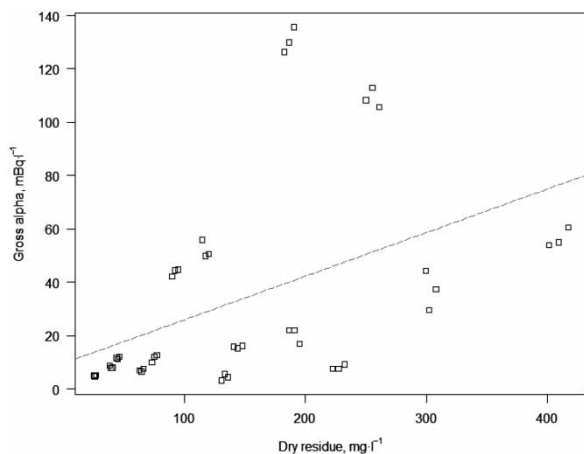


Figure 2 | Correlation between the activity concentration of gross- α and the amount of dry residue.

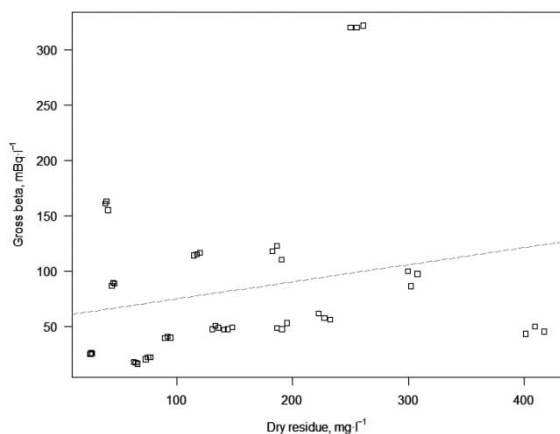


Figure 3 | Correlation between the activity concentration of gross- β and the amount of dry residue.

gross- α and gross- β in the investigated bottled mineral water samples from other studies are significantly different. It can be also observed that the values of gross- α and gross- β in this study are in the range of reported international values (Sánchez *et al.* 1999; Kovács *et al.* 2004; Rusconi *et al.* 2004; Desideri *et al.* 2007; Chau & Michalec 2009; Kobya *et al.* 2011; Taskin *et al.* 2013).

Radiological assessment

Annual effective dose rate

The annual effective dose equivalent rate (*AED*) from the ingestion of natural radionuclides in bottled mineral water is estimated to assess the radiological hazard for infants,

Table 2 | Comparison of activity concentrations (mBq L^{-1}) with published results in bottled mineral water samples from different countries

Countries	Gross- α	Gross- β	References
Poland (23 ^a)	2.2–938	53–10,537	Chau & Michalec (2009)
Turkey (40)	7–3,042	21–4,845	Taskin <i>et al.</i> (2013)
Italy (21)	<3–550	27–1,108	Rusconi <i>et al.</i> (2004)
Spain (28)	21–7,800	<20–5,800	Sánchez <i>et al.</i> (1999)
France (2)	<100	<120	Sánchez <i>et al.</i> (1999)
Portugal (13)	<7–8,400	<9–4,800	Sánchez <i>et al.</i> (1999)
Hungary (18)	<8–3,340	35–2,600	Kovács <i>et al.</i> (2004)
Italy (51)	<4.02–277.05	<24.91–930.00	Desideri <i>et al.</i> (2007)
Turkey (13)	7–898	13–854	Kobya <i>et al.</i> (2011)
China (45)	4.4–130.6	17.3–320.3	Present study

^aNumber of samples.

children and adults using Equation (3) (Alomari *et al.* 2019; Bello *et al.* 2020):

$$\text{AED}(\mu\text{Sv} \cdot \text{y}^{-1}) = A_{\alpha,\beta} \cdot CF \cdot W_c \quad (3)$$

where *CF* is the effective dose conversion factor for main α - or β -emitting radionuclides and *W_c* is the annual water consumption, taken as 120, 292 and 543 L y^{-1} for infants, children and adults, respectively (Chinese Society of Nutrition 2014). In the estimation of the *AED*, the average value of *CF* is taken as 0.30 $\mu\text{Sv Bq}^{-1}$ for α and 0.46 $\mu\text{Sv Bq}^{-1}$ for β (IAEA 1996; WHO 2011).

The calculated values of the *AED* for the bottled mineral water samples are presented in Table 3. The results show that the values of the *AED* for infants, children and adults range of 1.3–21.6 $\mu\text{Sv y}^{-1}$ with a mean of 5.9 $\mu\text{Sv y}^{-1}$, 2.9–52.5 $\mu\text{Sv y}^{-1}$ with a mean of 14.2 $\mu\text{Sv y}^{-1}$ and 5.5–97.8 $\mu\text{Sv y}^{-1}$ with a mean of 26.5 $\mu\text{Sv y}^{-1}$, respectively. The values of *AED* of all surveyed samples are much less than the WHO recommended reference value of 0.1 mSv y^{-1} . As can be seen in Table 3, the contribution of *AED* from α -emitting radionuclides to total *AED* is approximately 21.5%, while the contribution of *AED* from β -emitting radionuclides to total *AED* is approximately 78.5%.

Table 3 | The annual effective dose equivalent rate (AED) estimated for infants, children and adults

Brands No.	Annual effective dose equivalent rate ($\mu\text{Sv y}^{-1}$)								
	Infants			Children			Adults		
	α -emitters	β -emitters	Total	α -emitters	β -emitters	Total	α -emitters	β -emitters	Total
B_1	0.4	1.2	1.6	1.0	2.9	3.9	1.9	5.4	7.3
B_2	0.3	1.0	1.3	0.6	2.3	2.9	1.2	4.3	5.5
B_3	0.4	4.9	5.3	1.0	11.9	12.9	1.9	22.1	24.0
B_4	1.9	6.4	8.2	4.6	15.5	20.1	8.5	28.8	37.3
B_5	0.2	1.4	1.6	0.4	3.4	3.8	0.8	6.4	7.2
B_6	0.2	2.7	2.9	0.4	6.6	7.0	0.7	12.2	12.9
B_7	0.3	8.8	9.1	0.7	21.4	22.2	1.4	39.9	41.3
B_8	1.3	5.2	6.5	3.2	12.7	15.9	6.0	23.6	29.6
B_9	0.3	3.2	3.5	0.7	7.9	8.6	1.3	14.6	15.9
B_10	0.6	2.6	3.2	1.4	6.4	7.8	2.6	12.0	14.6
B_11	1.6	2.2	3.8	3.8	5.4	9.2	7.1	10.0	17.1
B_12	3.9	17.7	21.6	9.5	43.0	52.5	17.8	80.0	97.8
B_13	0.7	2.7	3.4	1.8	6.7	8.5	3.3	12.4	15.7
B_14	4.7	6.5	11.2	11.4	15.7	27.1	21.3	29.2	50.5
B_15	2.0	2.6	4.6	4.9	6.2	11.1	9.2	11.6	20.8
Range	0.2–4.7	1.0–17.7	1.3–21.6	0.4–11.4	2.3–43.0	2.9–52.5	0.7–21.3	4.3–80.0	5.5–97.8
Mean	1.3	4.6	5.9	3.0	11.2	14.2	5.7	20.8	26.5
SD	1.4	4.3	5.2	3.4	10.4	12.7	6.3	19.3	23.7

The lifetime cancer risk assessment

The estimated lifetime cancer risk is calculated using Equation (4) (Gorur & Camgoz 2014):

$$\text{Lifetime risk} = AED \cdot DL \cdot RF \quad (4)$$

where AED is the annual effective dose equivalent rate (Sv y^{-1}), DL is duration of life (70 years) and RF is the risk factor (Sv^{-1}). In the estimation of the lifetime cancer risk, the nominal probability coefficient of $5.5 \times 10^{-2} \text{Sv}^{-1}$ is adopted (WHO 2009, 2011, 2018).

The excess lifetime cancer risk levels from the direct ingestion of the natural radionuclides in bottled mineral water are estimated as shown in Table 4. The excess lifetime cancer risk values in bottled mineral water samples range from 1.3×10^{-5} to 2.2×10^{-4} with a mean of 6.0×10^{-5} . It is observed that the obtained results are lower than the estimated value of 3.85×10^{-4} recommended by WHO (2009, 2011, 2018). The results also show that there is no significant

risk for consumption of the observed brands of bottled mineral water and it can be consumed safely.

CONCLUSIONS

In this study, the gross- α and gross- β activity concentrations in 15 different brands of commercial bottled mineral water produced in China were analyzed to evaluate the quality and corresponding health impact on the population. According to the experimental results, the activity concentrations of gross- α and gross- β in bottled mineral water samples are in the range of 4.4–130.6 and 17.3–320.3 mBq L^{-1} , with mean values of 34.8 and 83.4 mBq L^{-1} , respectively. The values of the AED for infants, children and adults are in the range of 1.3–21.6 $\mu\text{Sv y}^{-1}$ with a mean of 5.9 $\mu\text{Sv y}^{-1}$, 2.9–52.5 $\mu\text{Sv y}^{-1}$ with a mean of 14.2 $\mu\text{Sv y}^{-1}$ and 5.5–97.8 $\mu\text{Sv y}^{-1}$ with a mean of 26.5 $\mu\text{Sv y}^{-1}$, respectively. The excess lifetime cancer risk values in bottled mineral water samples range from 1.3×10^{-5} to 2.2×10^{-4} with a mean of 6.0×10^{-5} .

Table 4 | The estimated lifetime risk of natural radioactivity for infants, children and adults (70 years) in bottled mineral water

Brand no.	Lifetime cancer risk (α and β)			
	Infants (2 years)	Children (15 years)	Adults (53 years)	Total (70 years)
B_1	1.8×10^{-7}	3.2×10^{-6}	1.3×10^{-5}	1.7×10^{-5}
B_2	1.4×10^{-7}	2.4×10^{-6}	1.0×10^{-5}	1.3×10^{-5}
B_3	5.8×10^{-7}	1.1×10^{-5}	4.4×10^{-5}	5.5×10^{-5}
B_4	9.0×10^{-7}	1.7×10^{-5}	6.8×10^{-5}	8.5×10^{-5}
B_5	1.8×10^{-7}	3.1×10^{-6}	1.3×10^{-5}	1.6×10^{-5}
B_6	3.2×10^{-7}	5.8×10^{-6}	2.3×10^{-5}	3.0×10^{-5}
B_7	1.0×10^{-6}	1.8×10^{-5}	7.5×10^{-5}	9.4×10^{-5}
B_8	7.2×10^{-7}	1.3×10^{-5}	5.4×10^{-5}	6.8×10^{-5}
B_9	3.9×10^{-7}	7.1×10^{-6}	2.9×10^{-5}	3.6×10^{-5}
B_10	3.5×10^{-7}	6.4×10^{-6}	2.6×10^{-5}	3.3×10^{-5}
B_11	4.2×10^{-7}	7.6×10^{-6}	3.1×10^{-5}	3.9×10^{-5}
B_12	2.4×10^{-6}	4.3×10^{-5}	1.8×10^{-4}	2.2×10^{-4}
B_13	3.7×10^{-7}	7.0×10^{-6}	2.8×10^{-5}	3.9×10^{-5}
B_14	1.2×10^{-6}	2.2×10^{-5}	9.2×10^{-5}	1.2×10^{-4}
B_15	5.1×10^{-7}	9.2×10^{-6}	3.8×10^{-5}	4.7×10^{-5}
Range	1.4×10^{-7} – 2.4×10^{-6}	2.4×10^{-6} – 4.3×10^{-5}	1.0×10^{-5} – 1.8×10^{-4}	1.3×10^{-5} – 2.2×10^{-4}
Mean	6.4×10^{-7}	1.2×10^{-5}	4.8×10^{-5}	6.0×10^{-5}

These results show that all the measured values of gross- α and gross- β in bottled mineral water samples are found to be obviously less than the guidance level by WHO and domestic standards. A poorly linear correlation is found between gross- α/β activity concentrations and dry residues. The values of AED due to α - and β -emitters estimated for infants, children and adults are below the WHO recommended limit of 0.1 mSv y^{-1} . Combined with the lifetime cancer risk assessment, it is concluded that there is no significant risk for consumption of the observed brands of bottled mineral water and it can be consumed safely.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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