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DETERMINATION OF GROSS ALPHA AND GROSS BETA RADIOACTIVITY LEVELS OF BOTTLED MINERAL WATER SAMPLES IN TURKEY

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TÜRKİYE'DE ŞİŞELENMİŞ MİNERALLİ SULARIN TOPLAM ALFA VE TOPLAM BETA RADYOAKTİVİTE SEVİYELERİNİN BELİRLENMESİ

Abstract

In this study, 32 different commercially bottled mineral waters were collected from various regions of Turkey. Gross alpha and gross beta activities of these bottled mineral water samples were measured by using proportional counters. The gross alpha activity concentrations of 32 samples were determined between Minimum Detectable Activity (*MDA*) (0.011-0.681 Bq/L) and 1.86 Bq/L with an average value of 0.541 Bq/L. Similarly, the gross beta activity concentrations were determined between MDA (0.010-0.245 Bq/L) and 1.85 Bq/L with an average value of 0.813 Bq/L. The gross alpha and gross beta activities of the mineral waters in Turkey determined in this study were compared with the corresponding values of the different countries.

The results of the study showed that the gross alpha and gross beta activities in all collected commercially bottled mineral water samples except one are within the limits of regulations published by Turkish Ministry of Health. Nevertheless, periodic monitoring of the mineral waters is required to ensure the safe level in terms of radioactivity.

Özet

Bu çalışmada Türkiye'nin çeşitli bölgelerinden ticari olarak şişelenmiş 32 farklı mineralli su numuneleri toplanmıştır. Bu mineralli su numunelerinin toplam alfa ve toplam beta radyoaktiviteleri orantılı sayaçlar kullanılarak ölçülmüştür. 32 farklı numunenin toplam alfa radyoaktivite konsantrasyonu *Ölçülebilir Minimum Değer (ÖMD)* (0.011-0.681 Bq/L) ile 1.86 Bq/L arasında değişen değerlerde ortalama 0.541 Bq/L olarak tespit edilmiştir. Toplam beta radyoaktivite konsantrasyonu ise *ÖMD* (0.010-0.245 Bq/L) ile 1.85 Bq/L arasında değişen değerlerde ortalama 0.813 Bq/L olarak tespit edilmiştir. Türkiye'de bulunan mineralli sularda tespit edilen toplam alfa ve toplam beta aktivite değerleri farklı ülkeler için yapılan benzer çalışmalarda bulunan değerler ile karşılaştırılmıştır.

Bu çalışmada elde edilen sonuçlara göre toplanan mineralli su numunelerinden bir tanesi hariç diğer tüm numunelerde toplam alfa ve toplam beta aktivite konsantrasyonları Türkiye Sağlık Bakanlığı tarafından yayınlanmış olan yönetmelikteki limitlerin altında kalmıştır. Fakat yine de radyoaktivite açısından güvenli seviyelerin devam ettiğinden emin olabilmek için mineralli suların periyodik olarak izlenmesi gerekmektedir.

Anahtar kelimeler: Mineralli sular, Toplam alfa aktivitesi, Toplam beta aktivitesi, Orantılı sayaç.
Key words: Mineral Waters, Gross Alpha Activity, Gross Beta Activity, Proportional counter.

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1. Introduction

Naturally occurring or anthropogenic radiation sources constitute environmental radioactivity. The studies of Scientific Committee of United Nations show that 98% of the environmental radiation is caused by natural radiation sources. The exposed radiation dose of people differs from region to region depending on some factors such as sea level, radioactivity in soil, food and water consumption and inhaled air. Since the concerns about the radiologic quality of the drinking waters increase, the necessity of the data showing the radiologic quality has also increased. The radiologic control of the waters consumed by people has become an obligation. This control involves the determination of radioactivity levels of natural and anthropogenic radionuclides in the water sample (United States Environmental Protection Agency, 2002).

Water present or passing in earth crust contains radioactivity. Natural radionuclides in earth crust, accordingly in water are ^{238}U, ^{232}Th and their decay products. Alpha radioactivity mainly originates from ^{226}Ra and ^{222}Rn radioisotopes. In some cases ^{40}K may increase gross beta activity, depending on dissolved potassium in the water (Sidhu et al., 1998).

The consumption of mineral water increases each passing year in our country like in the world, due to being healthful drink. By considering this increase in the consumption, determination of radioactivity concentrations in mineral water has an importance for the future.

Water used directly or indirectly by people may have various effects on human health depending on its chemical and radioactive properties. For that reason, properties and characteristics of waters consumed by people should be identified [United States Environmental Protection Agency, 2002; Gans, 1985 and Hem, 1985].

It is recommended in regulations of World Health Organization that, if a water source exposes 0.1 mSv or more radiation dose to human, it should be radiologically examined in detail (ICRP, 1991; WHO, 2006).

For tritium and total indication dose, the limit values are given as 100 Bq/L and 0.1 mSv/year respectively in the “Parameters and Parametric Values” part of the Euopean Union regulation, published in 2005, named “European Communities (Natural Mineral waters, Spring waters, and other waters in bottles or containers) Regulations” with S.I. No: 79. In the same regulation, it is stated that if the water in interest is not surface water or does not affect surface water, there is no need to measure the above parameters (tritium and total indication dose) (EC, 2007). And it is also stated that in case of exceeding the limiting values, the reason of the matter and effects to the human health should be determined and reported.

Since the analyses of the radioisotopes of natural radionuclides such as U, Th, Ra require different radiochemical separation and distillation processes, the analysis of each radioisotope is time consuming and expensive. Therefore, firstly the gross alpha and gross beta analyses are performed and then if necessary, detailed radionuclide analyses are done.

Turkey appears in first ten countries in the point of having mineral water reserves. While the yearly consumption of mineral water in European Union is about 150 L, this amount is about 6.5 L in Turkey (Baba et al., 2008).

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In the clause 6 of 01.12.2004 dated regulation of Turkish Health Ministry, the limit values for gross alpha and gross beta activities in mineral waters are defined as 1.5 Bq/L and 2.0 Bq/L respectively (Turkish Ministry of Health, 2007).

In this study, 32 different commercially bottled mineral waters are analyzed and then examined according to the current regulations of Health Ministry of Turkey and WHO (WHO, 2006; Turkish Ministry of Health, 2007).

2. Material and Method

2.1. Sampling

The bottled water samples are collected from 32 different mineral water bottling facilities in Turkey. The samples are taken in plastic bottles of 5 L and delivered to laboratory in 2 days. The bottles were cleaned before filling with water sample. The collected water samples were acidified with concentrated nitric acid of pH ≤ 2, trying to avoid precipitations, polymerizations and colloidal formations. There are many companies producing natural bottled mineral water from different depth aquifers. Fig. 1 shows the sampling points of the bottled mineral waters investigated radiologically.

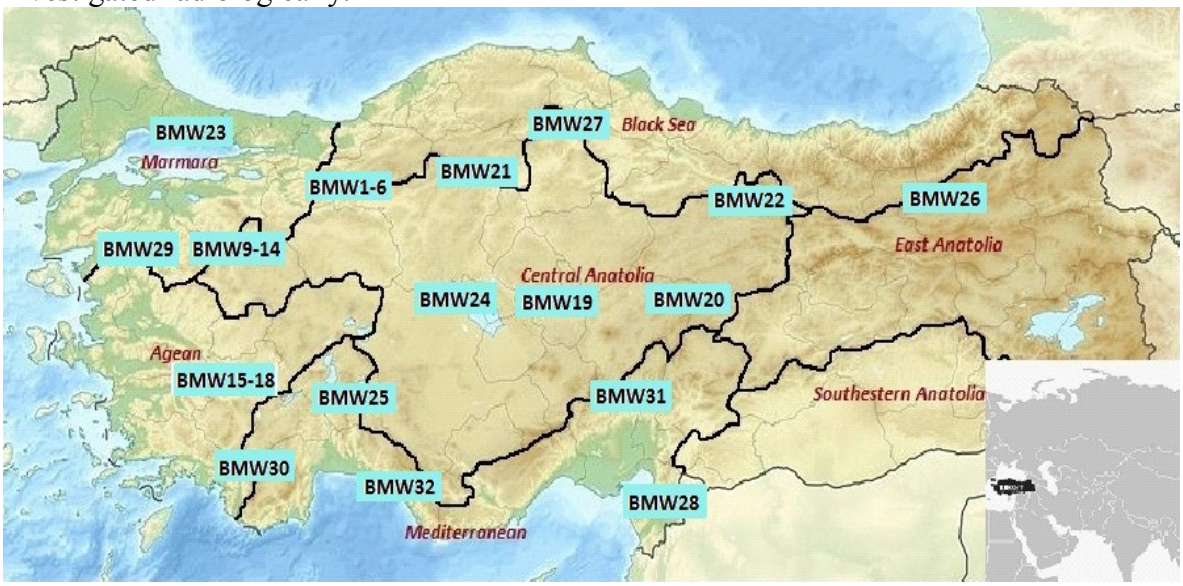

Figure 1. Sampling locations of bottling facilities for mineral waters in Turkey (BMW: bottled mineral water) (Şahin, 2017).

In this study, the most of the bottled natural mineral waters samples were collected from Marmara regions of Turkey. Fourteen mineral waters; BMW1-6, BMW 9-14, BMW 23 and BMW 29 were collected from Marmara region. Six mineral waters; BMW15-18, BMW 25 and, BMW 30 were taken from Aegean region. Four mineral waters; BMW21-22, BMW 26-27 were collected from Black Sea region. Four mineral waters; BMW19-20, BMW 24 and BMW 31 were obtained from Central Anatolia region. Two mineral waters; BMW28 and BMW 32 were collected from Mediterranean region of Turkey.

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2.2. Sample preparation and Measurements

After acidification of collected bottled mineral water samples, they were waited for at least 16 hours prior to starting sample preparation. Sample preparation is as follows; firstly, some water sample was taken from the plastic bottle and put into a clean beaker. Secondly, water in the beaker was evaporated under infrared lamp until 20-30 mL sample was left in the beaker. After that, remaining sample was put on a planchet and all samples were dried under infrared lamp. Lastly, gained residue was kept in drying oven at 105 ^{o}C for about 2 hours to get constant weight. In this step, the important point was to get the residue on planchet with minimum self-absorption. The planchet was kept in a desiccator until the measurement to prevent the sample from absorbing moisture. The amount of residue differs from sample to sample depending on water's type and quality. For that reason, at the beginning, 100 mL sample was put into a beaker and residue amount analysis was done. According to this residue amount, volume of the sample was redefined and the above procedure was applied starting from first step.

In this study, two different ultra-low background alpha/beta counting systems, PIC WPC 9550 automatic counter and PIC MPC 9604 multi-detector system, were used for the measurements.

The counting systems have been calibrated by the calibration sets prepared by using standard reference materials containing Am-241 (Isotope Products 1263-71-3) and Sr-90 (Isotope Products 1217-1-1). The efficiency calibration curves of the system for alpha and beta particles are shown in Fig. 2 and Fig. 3, respectively. The counting efficiency of the detector is determined according to the amount of the residue of the sample. The crosstalk effect of the system between the alpha and beta particles are taken into account in the activity calculations.

Before measurement of samples, alpha and beta background count rates were determined. For background counting, empty planchet was placed in to counting system and counted for same counting time as sample (900 min). The background count rates of our systems varied between 0.04 cpm and 0.08 cpm for alpha, and between 0.4 cpm and 0.9 cpm for beta.

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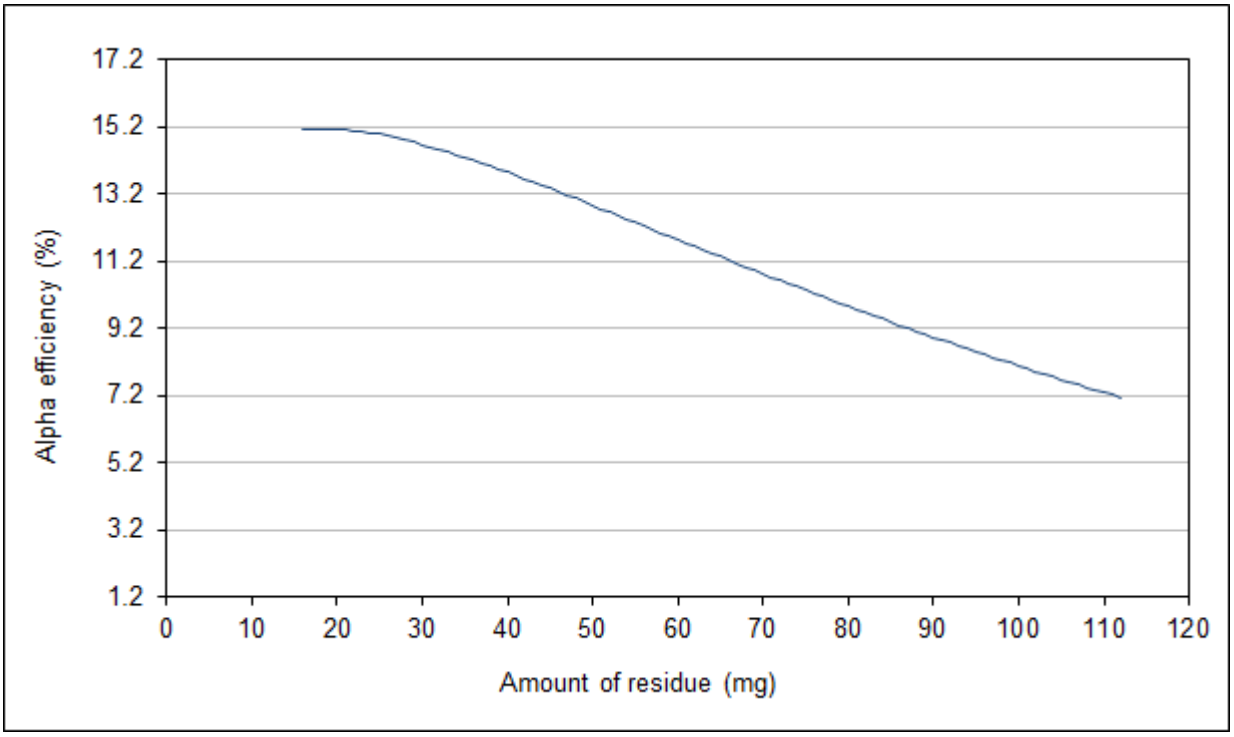

Figure 2. Alfa particle counting efficiency of the detector depending on the amount of the residue of the sample.

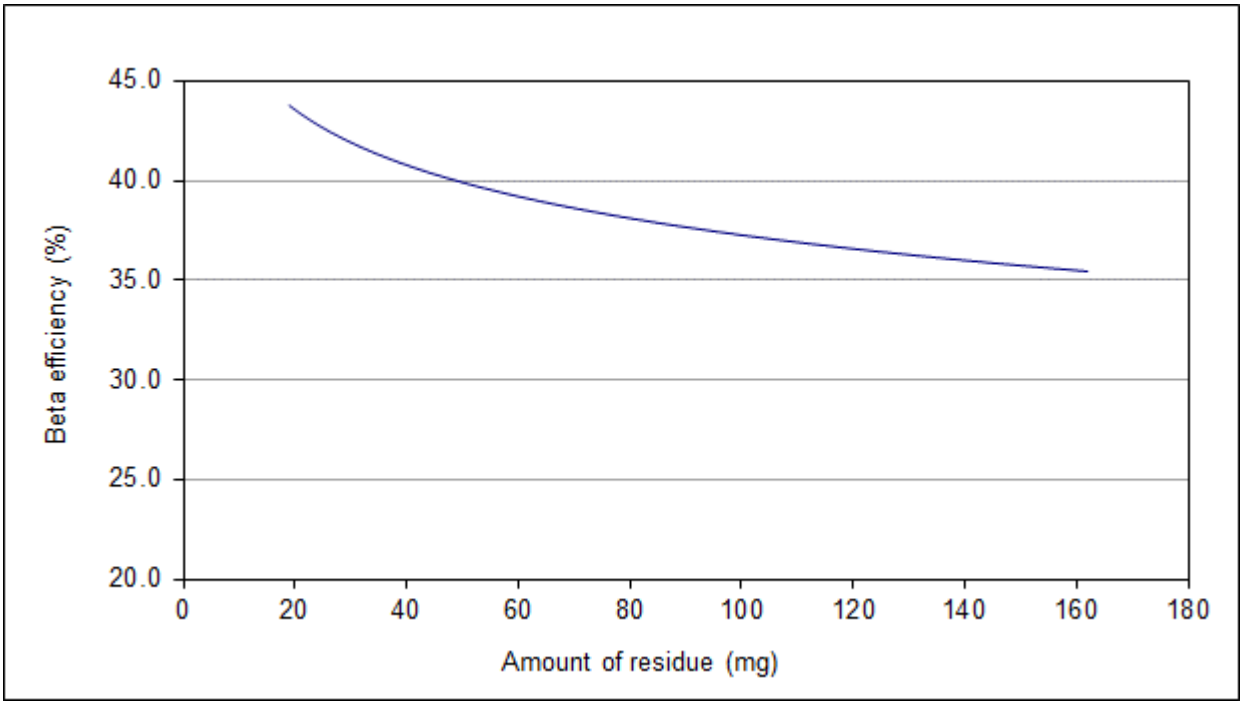

Figure 3. Beta particle counting efficiency of the detector depending on the amount of the residue of the sample.

The minimum detectable activities of gross alpha and gross beta measurements were calculated by using the following well-known and widely used formulas (Currie, 1968):

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$$MDA_{\alpha} = \frac{2.71 + 3.29\sqrt{cpm_{\alpha-bgk}t_s(1+\frac{t_s}{t_b})}}{t_s \;\; 60 \;\; V_s \;\; eff_{\alpha}} \quad (1)$$

$$MDA_{\beta} = \frac{2.71 + 3.29\sqrt{cpm_{\beta-bgk}t_s(1+\frac{t_s}{t_b})}}{t_s \;\; 60 \;\; V_s \;\; eff_{\beta}} \quad (2)$$

Where; MDA_{α} is Minimum Detectable Activity for alpha (Bq/L), MDA_{β} is Minimum Detectable Activity for beta (Bq/L), $cpm_{\alpha\text{-}bkg}$ is alpha background count Rate (count/minute), $cpm_{\beta\text{-}bkg}$ is beta background count rate (count/minute), ts eff_{α} is efficiency of alpha, eff_{β} is efficiency of beta, V_s is sample volume (L) and 60 is conversion factor from minute to second.

The *MDA* values varied between 0.011 – 0.681 Bq/L for the gross alpha measurements and between 0.010 – 0.245 Bq/L for the gross beta measurements, which can be seen in Table 2. Since the MDA strongly depends on sample volume, efficiency (amount of residue) and sample counting time knowing that the other factors are constant, it varied extremely from sample to sample. Background count rates were obtained 0.072 cpm for alpha and 0.634 cpm for beta. The gross alpha and gross beta activity concentrations were calculated by using calibration data of the system. Then results were reported in Bq/L. Gross alpha and gross beta activity concentrations and corresponding uncertainties were calculated by using the activity calculation formula in Eq. 3.

$$A_{\alpha,\beta}\left({}^{Bq}/_{L}\right) = \frac{N_{\alpha,\beta}}{60 \;\; eff_{\alpha,\beta} \;\; V_s} \quad (3)$$

In this equation; $A_{\alpha,\beta}$ is the gross alpha or gross beta activity, $N_{\alpha,\beta}$ is net gross alpha count rate or net gross beta count rate (in cpm), $eff_{\alpha,\beta}$ is gross alpha or gross beta counting efficiency (%), V_s is volume of sample aliquot in liter and 60 is conversion factor from minute to second.

Inter-comparison water samples from International Atomic Energy Agency (IAEA-2010-03), National Physical Laboratory (NPL-2010 and NPL-2011) were used for quality control (Table 1).

Table1. Intercomparison test results for measured gross alpha and gross beta activities and certified values.

Test	Gross Alpha		Gross Beta	
	Measured value	**Certified Value**	**Measured value**	**Certified Value**
IAEA-2010-03	4.80±0.17 Bq/kg	4.80±0.10 Bq/kg	9.97±0.21 Bq/kg	9.10±0.20 Bq/kg
NPL-2010	80.0±2.45 Bq/g	80.57±0.21 Bq/g	3.54±0.12 Bq/g	3.80±0.07 Bq/g
NPL-2011	0.072±0.004 Bq/g	0.065±0.005 Bq/g	3.26±0.051 Bq/g	3.226±0.007 Bq/g

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3. Results and Discussion

3.1. Gross alpha and gross beta activity concentrations

The measured activity concentrations of gross alpha and gross beta in bottled mineral water samples of different commercial products in Turkey are given in Table 2. The gross alpha activities in bottled mineral water samples are determined between *MDA* value and 1.86 ± 0.30 Bq/L, where the *MDA* values varies between 0.011 – 0.681 Bq/L. The Fig. 4 was created by assuming the *MDA* value of a sample as its activity concentration where the sample had no calculated activity concentration but only *MDA* value. Average gross alpha activity concentration of bottled mineral water was determined as 0.541 Bq/L. The highest gross alpha concentration is reported for the sample BMW 4. The recommended level of gross alpha activity concentration is indicated as 0.5 Bq/L in the third edition of WHO guidelines for drinking water quality (WHO, 2006). If the gross alpha activity does not exceed 0.5 Bq/L, it can be assumed that the annual total indicative dose is less than 0.1 mSv per year (ICRP, 1991; WHO, 2006). According to the mineral water directive of Turkish Ministry of Health, the recommended level is applied as 1.5 Bq/L in Turkey (Turkish Ministry of Health, 2004). Only one sample has a gross alpha concentration greater than the limit value prescribed by Turkish Ministry of Health for the mineral waters and three samples have gross alpha concentrations greater than the limit value prescribed by WHO for the drinking waters (Fig. 4).

Table 2. Results of bottled mineral waters (BMW) screening in Turkey.

Sample code	Gross Alfa (Bq/L)	Gross Beta (Bq/L)
BMW-1	< MDA=0.012	< MDA=0.011
BMW-2	< MDA=0.011	< MDA=0.011
BMW-3	< MDA=0.012	0.021 ± 0.007
BMW-4	1.86± 0.30	1.59 ± 0.17
BMW-5	< MDA=0.014	< MDA=0.010
BMW-6	< MDA=0.012	< MDA=0.011
BMW-7	0.292 ± 0.055	0.526 ± 0.052
BMW-8	0.448 ± 0.130	1.70 ± 0.14
BMW-9	< MDA=0.255	0.820 ± 0.090
BMW-10	0.297 ± 0.102	1.19 ± 0.11
BMW-11	< MDA=0.363	1.17 ± 0.16
BMW-12	< MDA=0.336	1.21 ± 0.19
BMW-13	0.418 ± 0.155	1.85 ± 0.17
BMW-14	0.292 ± 0.059	0.370 ± 0.055
BMW-15	< MDA=0.016	0.034 ± 0.006
BMW-16	< MDA=0.055	0.20 ± 0.03
BMW-17	< MDA=0.158	0.570 ± 0.080
BMW-18	< MDA=0.320	1.46 ± 0.15
BMW-19	< MDA=0.138	0.702 ± 0.076
BMW-20	< MDA=0.230	0.966 ± 0.102
BMW-21	< MDA=0.254	< MDA=0.245
BMW-22	< MDA=0.208	< MDA=0.211
BMW-23	< MDA=0.681	1.31 ± 0.24

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Sample code	Gross Alfa (Bq/L)	Gross Beta (Bq/L)
BMW-24	< MDA=0.031	0.070 ± 0.012
BMW-25	0.286 ± 0.057	0.223 ± 0.037
BMW-26	0.373 ± 0.114	< MDA=0.144
BMW-27	< MDA=0.062	< MDA=0.049
BMW-28	< MDA=0.039	< MDA=0.045
BMW-29	< MDA=0.197	0.595 ± 0.106
BMW-30	0.604 ± 0.055	0.745 ± 0.045
BMW-31	< MDA=0.274	0.565 ± 0.114
BMW-32	< MDA=0.032	< MDA=0.029

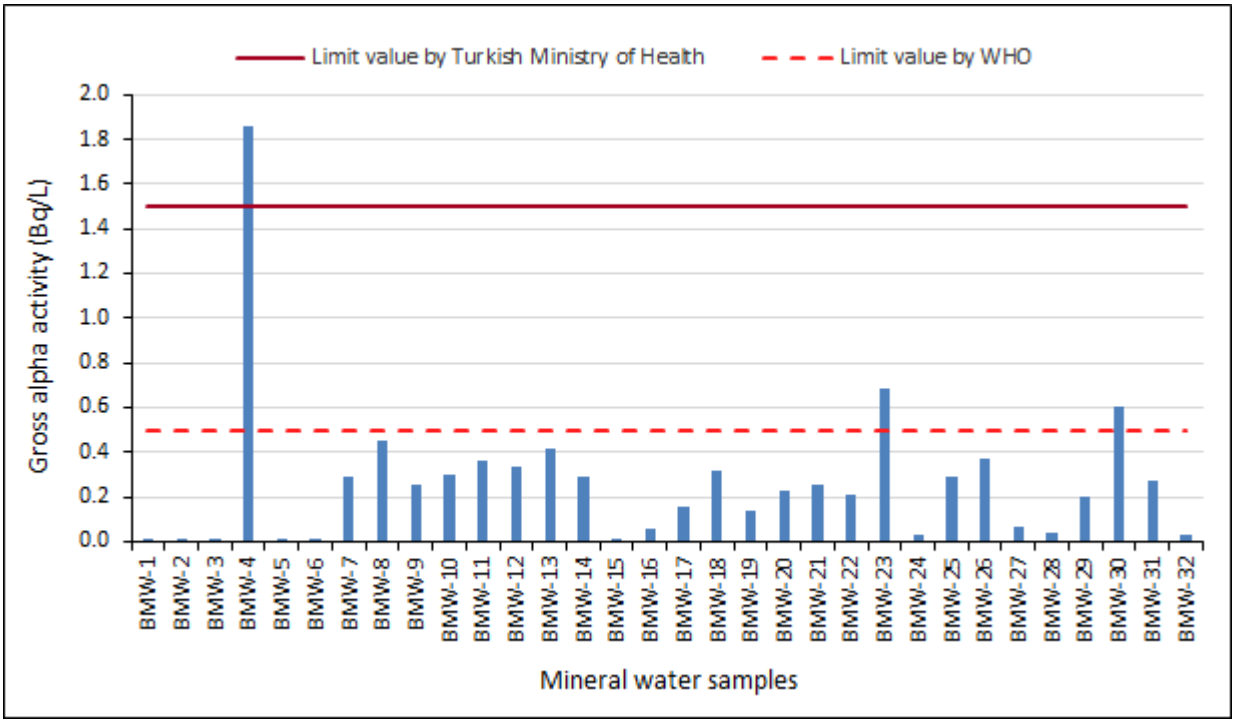

Figure 4. Gross alpha activity of bottled mineral waters samples (BMW). The limit values recommended by for WHO and Turkish Ministry of Health are 0.5 Bq/L and 1.5 Bq/L, respectively.

The measured activity concentrations of gross beta in bottled mineral water samples are given in Table 2. The observed gross beta activities found in bottled mineral water samples vary from *MDA* to 1.85 ± 0.17 Bq/L (Şahin, 2017) where the *MDA* values varies between 0.010 – 0245 Bq/L. The Fig. 5 was created by assuming the *MDA* value of a sample as its activity concentration where the sample had no calculated activity concentration but only *MDA* value. *Average gross beta activity concentration of bottled mineral water samples was determined as 0.813 Bq/L. The highest gross beta concentration is reported for the sample BMW 13. The* recommended level of gross beta activity concentration is defined as 1.0 Bq/L in the third edition of WHO guidelines for drinking water quality (EC, 2007). According to this guideline, if gross beta activity does not exceed 1.0 Bq/L, it can be assumed that the annual total indicative dose of adults is less than 0.1

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mSv per year (ICRP, 1991; WHO, 2006). The results of this study (Fig. 5) show that the measured activity concentrations of gross beta in bottled mineral water samples are less than 2.0 Bq/L which is the recommended limit by Turkish Ministry of Health (Turkish Ministry of Health, 2004).

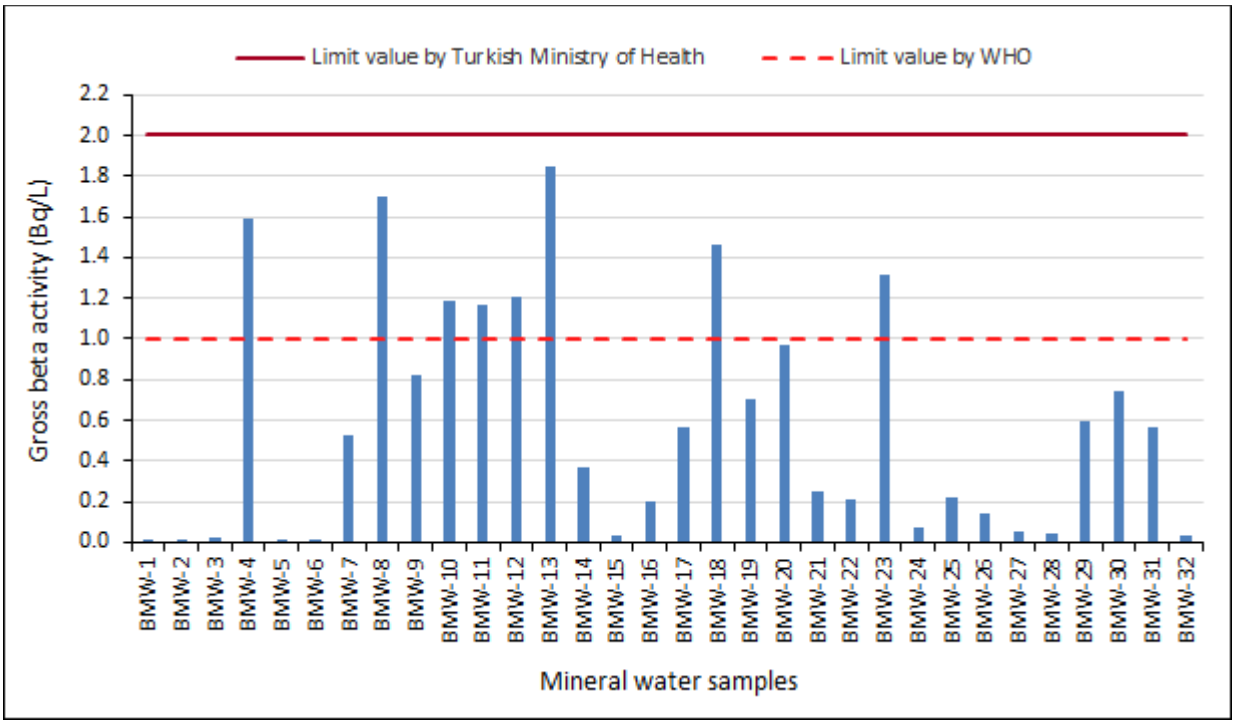

Figure 5. Gross beta activity of bottled mineral water samples (BMW). The limit values recommended by for WHO and Turkish Ministry of Health are 1 Bq/L and 2 Bq/L, respectively.

The gross alpha and gross beta concentrations determined in mineral waters of Turkey were compared with others countries of the world (Table 3). The gross alpha and the gross beta activity concentrations of the mineral waters in Turkey are almost in the same range as the results determined in Hungary (Jobbágy et al., 2011 and Kovács et al., 2004), a bit higher from the results determined in Italy (Desideri et al., 2007; Sgorbati et al., 1997) but much higher than the results determined in Bangkok (Srisuksawad et al., 2007). Eventually, it can be said that the results obtained in this study are generally comparable with the results of the other studies performed in several countries.

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Table 3. Overview of the gross alpha and gross beta activities in bottled mineral water samples around the world.

Region/Country	Type of water	Gross alpha (mBq/L)	Gross beta (mBq/L)
Central Italy (Desideri et al., 2007)	Spring water	4.02 - 277.50	24.91 - 930.00
Balaton Upland /Hungary (Jobbágy et al., 2004)	Spring water	26 - 1749	33 - 2015
Hungary (Kovács et al., 2011)	Mineral water	35 - 2600	≤ 8 - 3340
Bangkok Metropolitan Area (Srisuksawad et al., 2007)	Mineral water	90 - 580	190 - 450
Lombardia / Northern Italy (Sgorbati et al., 1997)	Mineral water	≤ 3 - 550	27 - 1108
This Study	Mineral water	≤ 11 - 1860	≤ 10 - 1860

4. Conclusion

In this work, the gross alpha and gross beta radioactivity levels are examined in the bottled water samples collected from 32 different mineral water bottling facilities in Turkey. Gross alpha values range from MDA (min. value of 11 mBq/L) to 1.86 ± 0.30 Bq/L and gross beta values range from MDA (min. value of 10 mBq/L) to 1.85 ± 0.17 Bq/L. These activity concentration values are comparable with the values found in literature. Recommended gross alpha limit values indicated in the guideline of Turkish Health Service about activity concentrations for mineral water samples was exceeded in only one case. The gross beta activities of all bottled mineral water are less than the limit value recommended by directives of Turkish Health Service.

Due to the importance of consuming bottled mineral water, the radioactivity level must be controlled strictly. The results of these study will contribute to the radioactivity level database of Turkey and provide a reference point for the future changes.

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