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ORIGINAL ARTICLE

Gross α and β activity concentrations in various water from Karaman, Turkey

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Abstract Natural activity concentrations in water sources are necessary to assess the effects of exposure to environmental radiation. The purpose of this study is to determine the activity concentrations of gross α and β in various water samples collected from 30 different locations in Karaman province, Turkey. The estimated values of activities of gross α and β obtained from water samples vary from 0.006 to 0.125 Bq $L^{-1}$and from 0.001 to 0.667 Bq L^{-1}, respectively. The gross β activities have been found always higher than the corresponding gross α activities for all samples. The obtained values indicated that concentration levels of α and β emitting radionuclides in samples have not exceeded WHO recommendations. The results represented here that the AED values are below of recommended reference level (0.1 mSv $year^{-1}$) by the WHO for all water samples in this study.

Keywords Water · Gross α · Gross β · Activity · Karaman · Effective dose equivalent

Introduction

Monitoring of any release of radioactivity in water is very important for biological effects of radiation on humans and environmental protection. Uranium (^{234,238}U), radium (^{226,228}Ra), potassium (^{40}K) and radon (^{222}Rn) are all soluble in surface waters such as wells, lakes and rivers. Radon present in water sources is readily released into outdoor air as it passes over rocks and soils. Naturally occurring radionuclides in drinking-water usually give radiation doses higher than those provided by artificial radionuclides. The process of identifying individual radionuclides and determining their concentration is time-consuming and expensive. So concentrations in drinking-water are low. Although the contribution of drinking-water to total exposure to radionuclides is very small, the health risks associated with the presence of naturally occurring radionuclides in drinking-water should also be taken into consideration. Gross α and β activities are very useful parameters for the preliminary screening of waters. For these reasons, firstly, gross α and β activities in water resources need to determine without regard to the identity of specific radionuclides (WHO 2011). According to the recommended guideline, activity concentrations should be 0.5 Bq L^{-1} for gross α and 1.0 Bq L^{-1} for gross β activity concentrations in drinking water (WHO 2011; Murad et al. 2014).

Karaman, located among the cities of Konya-Mersin-Antalya in the south of central Anatolia, is an important commercial, agricultural and industry area. The city of Karaman is located between the latitudes of 37°36′–36°24′N and 32°24′–34°24′E. The total urban area of Karaman province is about 9163 km^2 and the urban population is approximately 234,000 people live (Agar et al. 2014). This studied region is quite close to Akkuyu Nuclear Power Plant (NPP) which will be operated in Turkey, at Mersin on the Mediterranean coast. Since there is no available information about activity concentrations reported in water samples in the Karaman province so far, this paper will be an important contribution to the field.

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This study aims to determine the environmental radioactivity level of the Karaman province, Turkey based on measurements of gross α and β activities in various water sources. In the next section, we present the materials and method of the present study. In "Results and discussion", we show the results obtained by using a gas-flow proportional counter and "Conclusions" is devoted to our summary and discussion.

Materials and methods

Sample collection and preparation

US Environmental Protection Agency (EPA) (Krieger 1976) has established drinking water standards to protect public health. In the study, EPA-900 method has been used for the determination of gross α and gross β in investigated drinking water samples. This method covers the measurement of gross α and gross β particle activities in drinking water. In order to assess the levels of gross α and β activity in waters, thirty water samples were collected from Kazımkarabekir, Sarıveliler, Ayrancı, Başyayla and Ermenek districts in Karaman province. The locations of sampling site are indicated in Fig. 1. All the water samples were collected in 5000 mL capacity linear polypropylene bottles and immediately taken to the laboratory for analysis. Then, these samples were acidified with HNO_3 to be pH 2 to prevent precipitation and adsorption of contents of water sample on container walls and to avoid any biological activities. After acidification of collected bottled mineral water samples, they waited at least 16 h prior to start sample preparation. Gross α and β activity was determined as follows; first some water sample is taken from the plastic bottle and put into a clean beaker. Second, water in the beaker is evaporated under infrared lamp until 20–30 mL sample is left in the beaker. Then, remaining sample is put on a planchet and all samples are dried under the infrared lamp. Third, gained residue is kept in a drying oven at 105 °C for about 2 h to get constant weight. In this step, the important point is to get the residue on planchet with minimum self absorption. The amount of residue is defined as 5 mg cm^{-1} for calibration. The amount of residue differs from sample to sample depending on water's type and quality. For that reason, 100 mL sample is put into a beaker at the beginning and residue amount

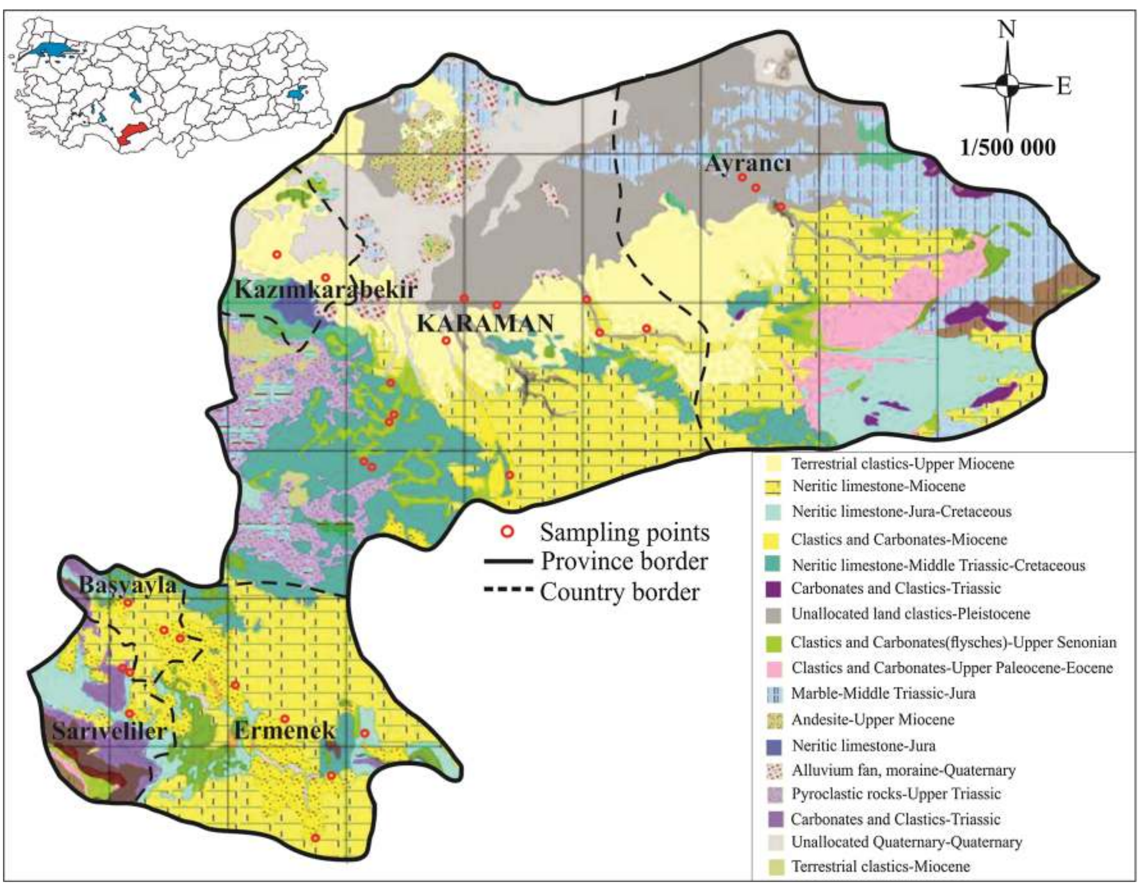


Fig. 1 A geological map of the studied area (Akbaş et al. 2002)

analysis is done. According to this residue amount, volume of the sample is redefined and the above procedure is applied starting from first step.

Analytical methods

Gross α and β activity concentrations in water samples were determined by a gas-flow proportional counter (PIC-MPC 9604–α/β counter). The sample time was set as 900 min for all samples. The counting gas (P-10) was a mixture of 90 % argon and 10 % methane. Lead shielding was used to attenuate external radiation. The operating voltage on the detector was selected as 1515 V.

For background counting, empty planchet is placed into counting system and counted for same counting time as sample (900 min). The background count rates of our systems vary between 0.04 and 0.08 cpm for α, and between 0.4 and 0.9 cpm for β.

The minimum detectable activities of gross α and gross β measurements were calculated by using following equations:

$$\mathrm{MDA}_{\alpha} = \frac{\frac{2.71}{t_{(\mathrm{Samplecounttime})i}} + 3.29\sqrt{\frac{\mathrm{cpm}_{\alpha\mathrm{BKG}}}{t_{(\mathrm{SampleCountTime})}} + \frac{\mathrm{cpm}_{\alpha\mathrm{BKG}}}{t_{(\mathrm{BackgroundCountTime})}}}}{\%\,\mathrm{Eff}_{\alpha}} \times \frac{1}{60 \times V_{(\mathrm{SampleofVolume})}} \quad (1)$$

and

$$\mathrm{MDA}_{\beta} = \frac{\frac{2.71}{t_{(\mathrm{Samplecounttime})i}} + 3.29\sqrt{\frac{\mathrm{cpm}_{\beta\mathrm{BKG}}}{t_{(\mathrm{SampleCountTime})}} + \frac{\mathrm{cpm}_{\beta\mathrm{BKG}}}{t_{(\mathrm{BackgroundCountTime})}}}}{\%\,\mathrm{Eff}_{\beta}} \times \frac{1}{60 \times V_{(\mathrm{SampleofVolume})}} \quad (2)$$

where MDA_{α} and MDA_{β} is minimum detectable activity for α and β (Bq L^{-1}), respectively, $\mathrm{cpm}_{\alpha\mathrm{BKG}}$ is α background count rate (count per minute), $\mathrm{cpm}_{\beta\mathrm{BKG}}$ is β background count rate (count per minute), % Eff_{α} is percent efficiency of α, % Eff_{β} is percent efficiency of β, V is sample of volume (L) and 60 is conversion factor from minute to second.

The MDA values of gross α and gross β have been calculated as 0.037 and 0.045 Bq L^{-1} respectively. Background count rates have been obtained 0.072 cpm for gross α and 0.634 cpm for gross β. In order to calibrate the low level counting system, standard solutions that contained known activities of ^{241}Am (450 Bq) for αs and ^{90}Sr (600 Bq) for βs which are similar to the sample geometry have been used. The gross α and β activity concentrations were calculated by using calibration data of our system. The calibration of the low level counting system used in the measurements was carried out. Then results were reported in Bq L^{-1}. Gross α and gross β activity concentrations and corresponding uncertainties were calculated by using Eq. 3 (Saleh and Abu Shayeb 2014):

$$A\alpha, \beta\ (BqL^{-1}) = \frac{N}{60 \times \mathrm{Eff} \times V} \quad (3)$$

where N is net gross α count rate or net gross β count rate (cpm), Eff is gross α or gross β counting efficiency (%), V is volume of sample aliquot in liter and 60 is conversion factor from minute to second in this equation.

The doses was calculated by the following equation (USA-EPA 1988; Sajo-Bohus et al. 1997) :

$$\mathrm{DR}_W = A_W \cdot \mathrm{IR}_W \cdot \mathrm{ID}_F$$

where DR_W is annual effective dose (AED) equivalent (µSv $year^{-1}$), A_W is gross α and gross β activity concentration (mBq L^{-1}), IR_W is intake of water for person in 1 year. The AED equivalents were determined for adults (17 years $<$ age), children (2–17 years) and lactation age (1 years $>$ babies) that drink 730, 350 and 250 L of water per year, respectively. Finally, the individual effective dose equivalents were assessed for adults who drink 2 L of water per day.

The total indicative dose (TID) was calculated for three classes of ages using the following approach. ID_F the annual effective dose conversion factors (mSv Bq^{-1}). The gross α activities were assumed to be gained from ^{238}U, ^{234}U, ^{230}Th, ^{226}Ra, ^{210}Po, ^{232}Th, respectively. The gross β activities were assumed to be gained from ^{210}Pb and ^{228}Ra. For our calculations, we used the following dose conversion factors proposed by the WHO (WHO 2004; Damla et al. 2009; Görür et al. 2011; Al-Amir et al. 2012; Gorur and Camgoz 2014; Akbulut and Taskin, 2015): 4.5×10^{-5} mSv Bq^{-1} for ^{238}U, 4.9×10^{-5} mSv Bq^{-1} for ^{234}U, 2.1×10^{-4} mSv Bq^{-1} for ^{230}Th, 2.8×10^{-4} mSv Bq^{-1} for ^{226}Ra, 1.2×10^{-3} mSv Bq^{-1} for ^{210}Po, 2.3×10^{-4} mSv Bq^{-1} for ^{232}Th, 6.9×10^{-4} mSv Bq^{-1} for ^{210}Pb, and 6.9×10^{-4} mSv Bq^{-1} for ^{228}Ra.

Mapping

In this study, the locations of the sampling site were labeled for mapping of the region. Also, spatial distributions of gross α and β concentrations in the study area were displayed on contour maps by using Surfer 8.0 for Windows software, a contouring and 3D surface mapping program. This version provides over twelve interpolation methods, each having specific functions and related parameters. Kriging, is one of these interpolation methods that can be used for mapping environmental data, was used in contour maps. This method is a geostatistical gridding method which has proven useful and popular in many fields and produces visually appealing maps from irregularly spaced data (Surfer 2002; Yılmaz 2007). It depends on mathematical and statistical models for optimal spatial prediction. Information on the exact spatial locations allows distances between observation to be calculated and autocorrelation to be modelled as a function of inverse distance in geostatistical methods (Burrough and McDonell 1988). In order to quantify the spatial autocorrelation in the data, Kriging uses the semivariogram which is a function of the distance and direction separating two locations. This function is used to measures average degree of dissimilarity among unmeasured and nearby values and to describe the weighted sum of data that derive the contribution of the surrounding measured points to the estimation of new values at the unmeasured sites within the area (Krivoruchko and Gotway 2004; Erdogan 2009).

Results and discussion

The values of gross α and β activity measurements for water samples together with pH and residue collected from tap, river and lake in Karaman province are demonstrated in Table 1. The specific activity is expressed in Bq L^{-1}. According to the results, concentrations varying from 0.006 Bq L^{-1} (Basyayla) to 0.125 Bq L^{-1} (Karaman) and from 0.005 Bq L^{-1} (Sarıveliler) to 0.667 Bq L^{-1} (Kazımkarabekir) were observed for the gross α and β activities in water samples under investigation, respectively. The average activity concentrations in all water samples are 0.0325 and 0.0681 Bq $L^{-1}$for gross α and gross β, respectively. It is clearly seen in Table 1 that all values of the gross β activity are higher than the corresponding gross α activity except for Sariveliler tap sample (S4), Karaman tap sample (S15) and Karaman river (S23) sample. It was assessed that concentrations of gross α and β

Table 1 The concentrations of gross α and β activity (Bq L^{-1}) for various water samples

District (users)	Sample	Water type	Residue (mg)	pH	Gross α	Gross β
Kazımkarabekir (4324)	S1	Tap	82.9	7.8	0.009 ± 0.001	0.018 ± 0.005
	S2	Tap	105.2	7.6	0.076 ± 0.029	0.667 ± 0.020
	S3	River	79.3	8.5	0.018 ± 0.005	0.025 ± 0.005
Sarıveliler (12,783)	S4	Tap	55.8	7.6	0.010 ± 0.007	0.005 ± 0.006
	S5	Tap	54.3	7.6	0.032 ± 0.009	0.040 ± 0.008
	S6	River	48.8	8.2	0.010 ± 0.005	0.023 ± 0.007
Başyayla (4497)	S7	Tap	75.7	7.3	0.023 ± 0.013	0.026 ± 0.007
	S8	River	88.3	8.4	0.006 ± 0.004	0.012 ± 0.006
	S9	Tap	123.9	7.4	0.033 ± 0.017	0.066 ± 0.007
Ermenek (30,361)	S10	Tap	99.9	8.1	0.010 ± 0.004	0.015 ± 0.005
	S11	Lake	111.4	8.2	0.030 ± 0.009	0.054 ± 0.005
	S12	Tap	124	8	0.041 ± 0.012	0.051 ± 0.006
	S13	River	106.7	8.1	0.028 ± 0.010	0.079 ± 0.015
	S14	Lake	122.3	8.1	0.015 ± 0.012	0.031 ± 0.006
Karaman Center (172,854)	S15	Tap	106.9	7.1	0.115 ± 0.025	0.058 ± 0.008
	S16	Tap	163.2	7.2	0.021 ± 0.007	0.038 ± 0.006
	S17	Tap	76.4	7.4	0.023 ± 0.008	0.066 ± 0.006
	S18	River	102.1	7.8	0.012 ± 0.007	0.022 ± 0.008
	S19	Lake	96.5	8.0	0.027 ± 0.012	0.058 ± 0.006
	S20	Tap	102.9	7.3	0.020 ± 0.006	0.034 ± 0.005
	S21	Tap	115.3	7.5	0.031 ± 0.009	0.060 ± 0.006
	S22	Tap	100.1	7.5	0.007 ± 0.001	0.017 ± 0.005
	S23	River	107.8	7.4	0.125 ± 0.030	0.102 ± 0.012
	S24	Tap	125	7.5	0.012 ± 0.005	0.035 ± 0.010
	S25	Tap	109.3	7.3	0.038 ± 0.009	0.040 ± 0.007
	S26	Lake	123.5	9.1	0.049 ± 0.018	0.152 ± 0.008
Ayrancı (9186)	S27	Tap	62	7.5	0.015 ± 0.008	0.064 ± 0.011
	S28	Tap	66.1	7.4	0.042 ± 0.012	0.084 ± 0.010
	S29	Tap	51.8	7.8	0.032 ± 0.009	0.048 ± 0.012
	S30	Lake	26.3	8.1	0.011 ± 0.011	0.054 ± 0.013

Table 2 Comparisons of average activities for gross α and β (Bq L^{-1}) in waters with literature

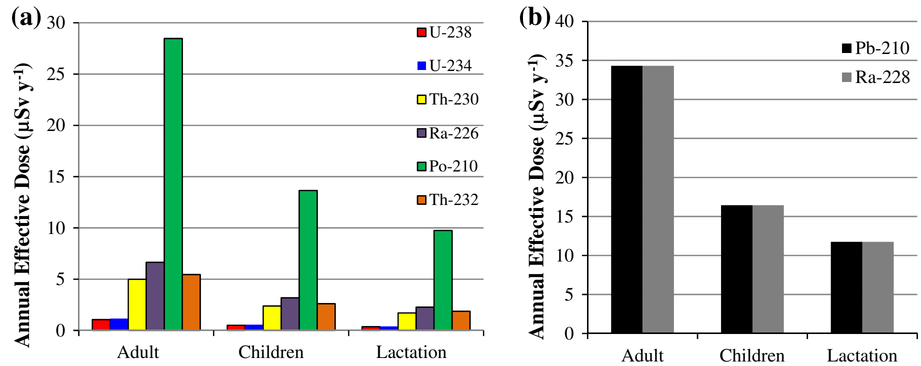
Water type	Region	Gross α	Gross β	References
Tap	Istanbul	0.0228	0.0664	Karahan et al. (2000)
	Tekirdag	0.044	0.1	Kam et al. (2010a)
	Trabzon	0.0065	0.1008	Damla et al. (2006)
	Sanliurfa	0.038	0.1324	Bozkurt et al. (2007)
	Adana	0.0096	0.086	Degerliler and Karahan (2010)
	Batman	0.0338	0.0803	Damla et al. (2009)
	Gaziantep	0.0498	0.1284	Osmanlioglu et al. (2007)
	Samsun	0.0519	0.0778	Görür et al. (2011)
	Canakkale	0.0599	0.0841	Kam et al. (2010b)
	Bolu	0.06811	0.16944	Gorur and Camgoz (2014)
	Erzincan	0.0477	0.104	Yalcin et al. 2012
	Rize	0.022	0.085	Akbulut and Taskin (2015)
	Italy	0.008–0.349	0.025–0.273	Forte et al. 2007
	Brasil	0.001–0.400	0.120–0.860	Bonotto et al. (2009)
	Ghana	0.0423	0.1732	Darko et al. (2015)
	Spain	0.02–2.42	0.05–5.8	Duenas et al. (1998)
	Serbia	0.029–0.21	0.4	Todorovic et al. (2012)
	Malesia	0.004–0.02	0.082–0.35	Saleh et al. (2015)
	Karaman	0.031	0.0753	Present work
River	Adana	0.005	0.2453	Degerliler and Karahan (2010)
	Batman	0.0468	0.0779	Damla et al. (2009)
	Samsun	0.142	0.180	Görür et al. (2011)
	Bolu	0.0876	0.1276	Gorur and Camgoz (2014)
	Fırtına	0.033	0.070	Küçükömeroğlu et al. (2008)
	Karaman	0.0421	0.0438	Present work
Lake	Bolu	0.04123	0.1276	Gorur and Camgoz (2014)
	Seyhan	0.012	0.0426	Degerliler and Karahan (2010)
	Karagol	0.03	2.62	Akyil et al. (2009)
	Golcuk	0.75	2.35	Akyil et al. (2009)
	Catalbogaz	0.03	1.77	Akyil et al. (2009)
	Karaman	0.0264	0.0698	Present work

activity for drinking, lake and river water samples are lower than the maximum permissible values (0.5 Bq L^{-1} for α activity and 1 Bq $L^{-1}$for β activity) of the WHO guidelines (WHO 2011) for drinking water quality.

The average values of gross α and β activity concentrations are comparable to the data from other studies in different parts of Turkey and the world as seen in Table 2. The average concentrations values obtained for the gross α in tap water samples are lower than observed concentrations for Tekirdag, Batman, Gaziantep, Sanliurfa, Samsun, Canakkale, Bolu, Erzincan, Spain, Serbia, Malesia and Ghana but higher than those of Adana, Istanbul, Rize, Trabzon, Brasil, and Italy. The average concentrations values obtained for gross β in tap water samples higher than Istanbul but lower than those of Trabzon, Tekirdag, Batman, Sanliurfa, Samsun, Adana, Canakkale, Gaziantep, Bolu, Erzincan, Rize, Brasil, Italy, Spain, Serbia, Malesia and Ghana.

The calculated AED values of α and β emitters in investigated water samples are given in Fig. 2a, b, respectively. The water supplied from different sources such as rivers and lakes is rarely used as drinking water although tap waters are generally used for drinking. The gross α and β activities are assumed to be from α and β emitting radionuclides ^{238}U, ^{234}U, ^{230}Th, ^{226}Ra, ^{210}Po, ^{232}Th and ^{228}Ra, respectively. Contributions of the tap, river and lake waters to total annual effective dose equivalent from ^{238}U, ^{234}U, ^{230}Th, ^{226}Ra, ^{210}Po, ^{232}Th and ^{228}Ra are 1.07, 1.16, 4.98, 6.64, 28.47, 5.46 and 34.32 µSv year^{-1} for adults, 0.51, 0.56, 2.39, 3.19, 13.65, 2.62 and 16.45 µSv year^{-1} for children, 0.37, 0.40, 1.71, 2.28 9.75 1.87 and 11.75 µSv year^{-1} for lactation age, respectively. It can be observed that the estimated annual effective doses for adult, children and lactation members in study area are below the recommended reference level of 0.1 mSv year^{-1} by WHO for water samples. Therefore, a

Fig. 2 The calculated annual effective doses of (**a**) α and (**b**) β emitters in waters (μSv year^{-1})



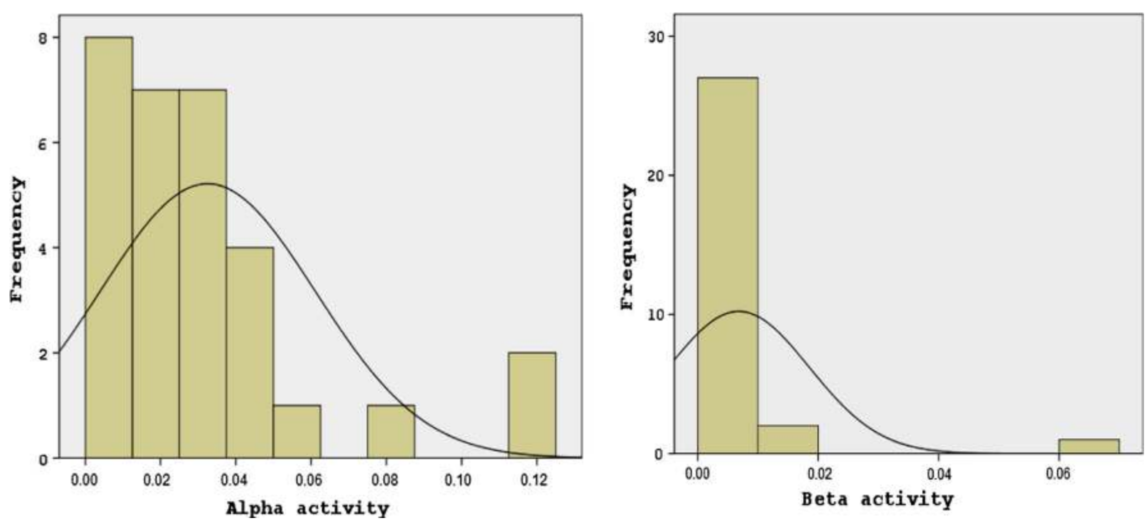


Fig. 3 The frequency distribution of gross α (*left*) and β (*right*) activities of Karaman

variety of water sources such as tap, river and lake can be used as drinking water without any treatment of the supply to decrease the concentrations of radioactive contaminants.

After gross α and β activity measurements, the obtained data are statistically analyzed by using SPSS computer software. Figure 3 shows the corresponding frequency distribution of activities detected for gross α and β in water samples. The gross α activity concentrations obtained in this work are lower than 0.06 Bq L^{-1} in 90 % of water samples while the gross β activity concentrations are lower than 0.1 Bq L^{-1} in 96 % of the water samples according to Fig. 3. Also, the average values are generally comparable with the reported data from different regions of Turkey, as well as some international data as seen in Table 2. Table 3 indicates the statistical data such as the arithmetic and geometric mean values, standard deviation, skewness, kurtosis coefficient and the type of theoretical frequency distribution that best fits each empirical distribution corresponding to the activities measured in water samples. It can be easily seen from Table 3 that the positive values of kurtosis coefficient of gross α and β concentrations (4.572 and 25.714, respectively) indicates a higher and narrower distribution than normal. Moreover, the positive values of skewness calculated for activity concentrations of gross α and β (2.118 and 4.925, respectively) represents the asymmetric distribution with the right tail being longer than the left as can be viewed in Fig. 3.

Concentrations of natural radionuclides that have been found to depend on the local geological and geographical conditions differ from area to area, although these

Table 3 Statistical values of gross α and β activity concentrations (in Bq L^{-1}) in water samples

Statistic data	Gross α	Gross β
Arithmetic mean	0.0325	0.0681
Geometric mean	0.0245	0.0422
Arithmetic standard deviation	0.02868	0.11709
Skewness	2.118	4.925
Kurtosis	4.572	25.714
Frequency distribution	Log-normal	Log-normal

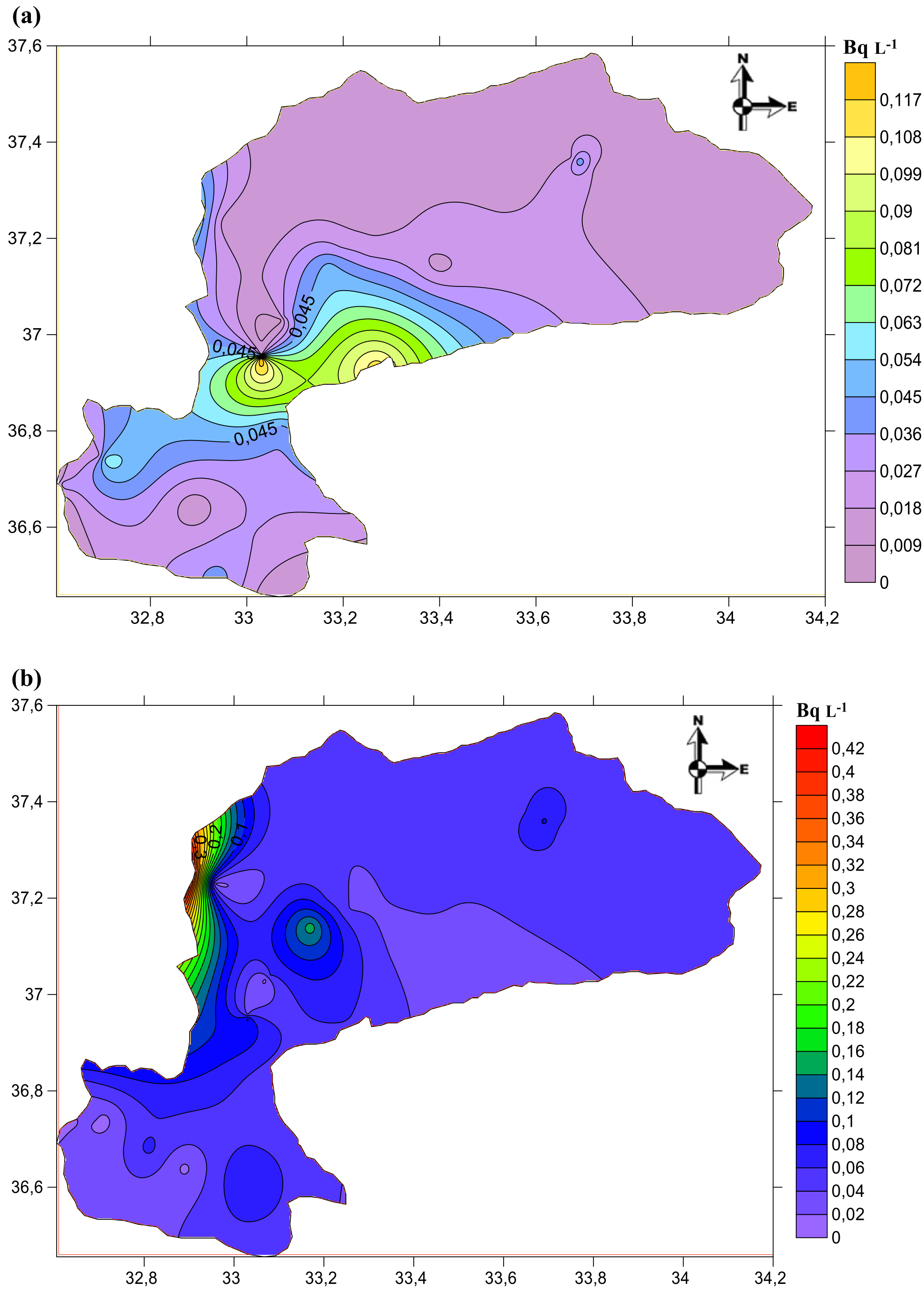


Fig. 4 Contour maps of (**a**) gross α and (**b**) β activities ($Bq\ L^{-1}$) of Karaman

radionuclides are widely distributed. Figure 4a, b represent spatial distributions of gross α and β concentrations in study area, respectively. The contour maps are drawn based on the radionuclide activity concentrations measured in water samples. As shown in contour maps (Fig. 4a), it is clear that the gross α concentrations distributed approximately in thecenter part of Karaman. Besides, the largest contribution of radioactivity for gross β concentration in waters is generally distributed near Kazımkarabekir which is the west part of study area. The high gross α and β activity concentrations in water samples may be due to the different geological origin and chemical composition of spring waters. Natural radioactivity is directly related to the kind of geological layers crossed by waters. Karaman geological formation is generally consisted by limestone, marble and terrestrial clastics. It is demonstrated that the gross α and gross β radioactivity concentrations are greatly affected by Karaman geological structure.

Conclusions

In this study, the concentrations of gross α and β activity in various water samples collected from Karaman province, Turkey were determined by using a gas-flow proportional counter. Also, to the best of our knowledge, the present work is one of the first studies on the radioactivity measurements in some water samples around Karaman–Turkey, contributing useful baseline data.

The WHO advises 0.5 Bq L^{-1} for gross α and 1.0 Bq L^{-1} for gross β activity as limit values for drinking water. Gross β activities of all water samples are seriously below the reference value of 1.0 Bq L^{-1}. Most of the gross α activity in waters is attributed to decay of uranium and thorium isotopes. Also, main sources of the gross β activity are arisen from radioactive potassium (^{40}K) isotope. It can be determined that the annual effective doses received by adult, children and lactation members are lower than the WHO in this area. The waters under investigation is almost neutral or weakly alkaline at pH 7.1–9.1.

The estimated gross α and β radioactivity concentrations in these water samples will contribute to a radioactivity database in the future. The results may also be used as reference data for monitoring possible radioactivity pollutions in the future since Turkey will be operating a nuclear power reactor in this area.

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