

Radioactivity Survey and Risk Assessment Study for Drinking Water in the Artvin Province, Turkey

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Abstract In this study, 117 spring water samples, collected across the Artvin province, were analyzed for gross α and gross β radioactivity. The values of the activity concentrations of the gross α and gross β measured in the natural spring water samples ranged from 5 to 771 mBqL⁻¹ with the mean of 46 mBqL⁻¹ and from 13 to 808 mBqL⁻¹ with the mean of 91 mBqL⁻¹. All values of measured water samples except one were within the limits, as recommended by the World Health Organization (WHO). Gross alpha- and beta-interpolated values of the region were determined and mapped by using the Kriging method. In addition to the measured gross alpha and beta activity result of natural spring water samples, for each sample, annual effective dose equivalent was calculated and lifetime cancer risks were estimated. Our study showed that two values of water samples exceeded WHO limits and lifetime

cancer risk due to the water radioactivity ranges between 1.3×10^{-5} and 20.6×10^{-4} with the mean of 10.1×10^{-5} .

Keywords Gross α –gross β · Natural spring water · Geostatistical analysis · Effective dose equivalent · Lifetime risk · Artvin

1 Introduction

We live in an environment which contains radiation originating from man-made and natural resources that may pose a threat for human health in case they exceed certain values. In our environment, air, water, soil, and foodstuff contain some radioactive isotopes such as uranium, radium, thorium, and radon that emit alpha, beta, and gamma radiation to the environment. Water quality has an importance in environmental studies because of its daily human consumption and its ability to transport pollutants in nature (Amrani, 2002). Radionuclides in drinking water cause internal exposure in humans, which occurs when the decayed radionuclides are taken into the body through ingestion and indirect inhalation when they are incorporated as part of the human food chain (Değerlier and Karahan, 2010). Since radionuclides (²²⁶Ra, ²²⁸Ra, ²¹⁰Po, ⁴⁰K) present similarities in structural characteristics to Ca and its precipitates, their accumulation in the body occurs especially in bones and teeth (Dewit et al. 2001; Wrenn et al., 1985; Whicker and Schultz, 1982).

There are several standards limiting the amount of radionuclide concentration in drinking water. The

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practical approach is to use a screening procedure with no regard to the identity of specific radionuclides (WHO, 2011). Measurement of the activity concentrations of the gross α and gross β is suitable for a preliminary screening procedure to determine whether further analysis related to a specific radionuclide is necessary (Turhan, 2013). Also, the gross α and gross β activity can be used for detecting changes in the radiological characteristics of the drinking water sources (WHO, 2011). Generally, limit values of gross α and gross β have been recommended as 500 and 1000 mBqL⁻¹, respectively. Below these limit values of gross α and gross β , drinking water is acceptable for human consumption and no action to reduce radioactivity is required.

There are some studies about varied water (tap, surface, ground, natural spring, thermal, and mineral) radioactivity concentration in Turkey and several other countries (Karahan et al., 2000; Bozkurt et al., 2007; Forte et al., 2007; Bonotto et al., 2009; Değerlier and Karahan, 2010; Kobya et al., 2010; Görür et al., 2011; Jobbagy et al., 2011; Taskin et al., 2012; Turhan et al., 2013; Görür and Camgöz, 2014). However, particular systematic data on the radioactivity of natural spring water sources in the Artvin province is not available in the literature.

The province of Artvin is situated in the most northeast of Turkey between the latitudes of 40° 36' and 41° 31' N and the longitudes of 41° 09' and 42° 35' E. The Artvin province has an area of 7436 km² and nearly 1 % of Turkey's total geographical area. It has a high hydroelectric energy potential and rich spring water sources. The spring water of this region is not only consumed by the 200,000 people living in the Artvin province but also bottled and distributed for consumption across the country. Due to this fact, it is important to measure the radioactivity level for the spring water of the region in order to determine the lifetime cancer risk for consumers. Some countries have established their own water quality standards to meet their national priorities, taking into account their economic, technical, social, cultural, and political requirements (Fatima, 2007). Turkey did not have guidelines for gross α and gross β activity concentrations in drinking water until the first guidelines were put into effect by the Institute of Turkish Standards in 1997 (Görür and Camgöz, 2014).

The aim of this study is to determine the gross α and gross β concentrations in natural spring water, to evaluate the effective dose equivalent due to water

radioactivity concentration and excess lifetime cancer risk assessment, and to determine the distribution of lifetime risk in terms of water radioactivity by calculating the lifetime cancer risk for the Artvin province.

2 Material and Methods

2.1 Sample Collection

In order to measure the gross α and gross β activities in water, natural spring water samples were collected from 117 different sampling stations in the Artvin Province. Sampling sites are listed in Table 1 and shown in Fig. 1. The natural spring water samples were collected in 5000 mL capacity polystyrene bottles. Then, water samples were acidified with HNO₃ to pH 2 to prevent any loss by sorption of the radionuclides around the container walls and to reduce the growth of microorganisms.

2.2 Experimental Procedure

The activity concentrations of gross α and gross β in the natural spring water samples were measured using a gas proportional alpha/beta counter of low background multiple detector type (Berthold LB 770). LB 770 10-channel α - β low-level counter is capable of measuring α - β of 10 planchets simultaneously. For each sample, there are two separate measuring channels for alpha and beta activities. Slider and counter tubes are surrounded by a 100-mm-thick shielding made of machined lead bricks to reduce ambient radiation. The background of each detector was determined by counting an empty planchet for 100 min. The calibration of the system was performed by using one planchet containing a certified solution of ²⁴¹Am and another planchet containing certified solution of ⁹⁰Sr/⁹⁰Y (Turhan et al., 2013).

The minimum detectable activity (MDA) was calculated using (Görür et al., 2011):

$$\text{MDA} \left(\text{Bq/L} \right) = \frac{2.71 + 4.65 \sqrt{\text{CR}_B T}}{V T \varepsilon 60} \quad (1)$$

where CR_B is the background count rate (counts per minute), *V* is the volume sample (L), *T* is the measurement time (min), and ε is efficiency. The MDA for gross α and gross β was calculated as 5 and 7 mBqL⁻¹, respectively.

Table 1 Activity concentrations of gross alpha and gross beta in natural spring water samples

Places	Sample number	Gross α (mBqL ⁻¹)				Gross β (mBqL ⁻¹)			
		Min.	Max.	Mean	SD	Min.	Max.	Mean	SD
Artvin Centrum	20	7±3	286±33	50±9	62.71	41±17	407±33	141±17	77.89
Arhavi	15	5±3	39±18	15±6	8.91	17±8	94±10	42±9	25.15
Hopa	15	6±4	86±23	40±8	24.27	23±7	212±40	85±13	51.5
Yusufeli	18	27±7	771±58	96±13	171.70	63±10	808±46	171±21	89.87
Ardanuç	16	10±5	141±16	59±11	41.61	23±6	383±22	108±16	61.45
Şavşat	16	6±3	74±11	22±7	17.47	13±8	167±12	61±14	47.85
Murgul	8	14±6	42±8	27±8	9.77	26±13	87±10	48±12	19.74
Borçka	9	21±6	50±7	30±7	8.71	22±8	198±15	76±11	43.17
Provincial wide	117	5±3	771±58	46±9	77.56	13±8	808±46	91±15	97.03

SD standard deviation

2.3 Measurement of the Gross α in the Natural Spring Water Samples

The activity concentrations of gross α of the ground water samples were measured using method SM 7110 precipitation. Two hundred fifty-milliliter aliquot of each sample was transferred to a beaker. Drops of 2–3 dilute detergent was added to the prepared aliquot. The

beaker placed on hot-plate magnetic stirrer was mixed by adding 20 mL of 2 N H₂SO₄. The stirring process was continued for 10 min after boiling to ensure more mixing of the solution. Then, 0.5 mL of barium carrier was added to the solution, and the stirring process was continued for 30 min. Bromocresol purple indicator (0.5 mL), 1 mL of iron carrier, and 5 mL of paper pulp/water mixture were added. Drops of 6 NH₄OH

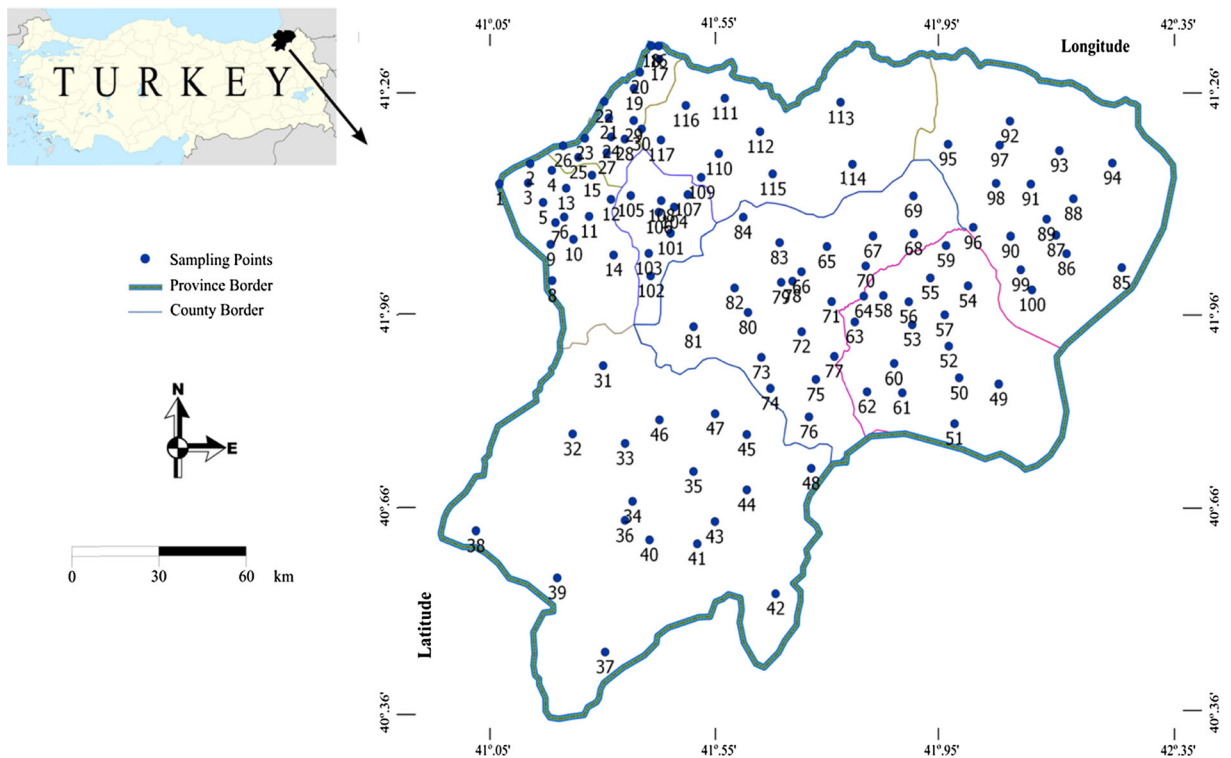


Fig. 1 Study area and samples collection points in Artvin Province, Turkey

M were added until the color of the solution turned from yellow to purple, and the stirring process was continued for 30 min more. Then, the solution was allowed to stand for precipitation. The precipitate was then filtered through a filter paper with a vacuum pump. Finally, the precipitate was dried under an infrared lamp. The residue was counted twice for a period of 100 min. The gross α activity concentration was determined by taking the average result of the two counts. The abovementioned procedures were repeated for each water sample (Turhan et al., 2013).

2.4 Measurement of Gross β in the Natural Spring Water Samples

Measurements of the activity concentrations of gross β in the natural spring water samples were performed using the EPA 900 evaporation method (Krieger and Whittaker, 1980). Two hundred fifty-milliliter aliquot of each sample prepared in a beaker was first acidified with HNO_3 of 2–3 mL, and then the solution was evaporated to a volume of 15–20 mL on a hot plate. The solution was transferred on a tared 6-cm diameter steel planchet and dried in an oven at 105 °C for at least 2 h. The sample residue was cooled in a desiccator for about 30 min, and then it was weighed. The residue was counted twice for a period of 500 min. The gross β activity concentration was determined by taking the average result of two counts. The abovementioned procedures were repeated for each water sample (Turhan et al., 2013).

2.5 Determination of the Effective Dose Equivalent

To calculate the doses, we used (Sajo-Bohus et al., 1996; USA-EPA, 1998):

$$DR_W = A_W \times IR_W \times ID_F \times 2 \quad (\text{for both } \alpha \text{ and } \beta) \quad (2)$$

where DR_W is the dose equivalent effective (Sv/year), A_W is activity (Bq/L), IR_W is the intake of water for one person in 1 year (730 L), and ID_F is the ingestion effective dose equivalent factor for 3.58×10^{-7} Sv/Bq for alpha. In the calculation following the procedures of Fernandez et al. (1992), it is considered that more than 50 % of the annual dose corresponds to radium (gross alpha radium).

2.6 Lifetime Risk Assessment

Lifetime cancer risk assessment was calculated using:

$$\text{Lifetime Risk} = DR_W \times DL \times RF \quad (3)$$

where DR_W is the annual effective dose equivalent (Sv/year), DL is duration of life (70 years), and RF is risk factor (Sv^{-1}). For risk assessment, the nominal probability coefficient of $7.3 \times 10^{-2} \text{ Sv}^{-1}$ recommended by ICRP (1991) was adopted (Görür and Camgöz, 2014).

2.7 Geostatistical Analyses

Geostatistics is a calculation method which is different from other conventional statistics methods based on stationary random function, which is the correlation between sample values with their sampling coordinates (Clark, 1979; Pebesma and Wesseling, 1998). In this method, spatial variable depends on distance value and function of variogram or semivariogram. This function is expressed as variance of two variables which are at a distance of h to each other (Diggle and Riberio, 2007). The semivariance containing all spatial variables is expressed through:

$$\gamma(h) = \frac{1}{2N(h)} \sum_{i=1}^{N(h)} (Z(x_i) - Z(x_i + h))^2 \quad (4)$$

where $\gamma(h)$ is the value of semivariance, h is the distance between measured samples of pairs, $N(h)$ is the number of pairs of sample points separated by the lag distance h , $Z(x_i)$ is the measured value of the variable in i point, and $Z(x_i + h)$ is the measured value of the variable in $x_i + h$ point. This experimental semivariogram function is calculated for each distance, and then it is fitted with a suitable parametric function (spherical, exponential, Gaussian, linear, periodic, etc.). The fitted model provides necessary input parameters for the Kriging interpolation (the weight coefficients). In the Kriging method, which is known to be mathematically the "best linear unbiased estimator" (BLUE) in literature, there should be a normal distribution for the accurate calculations of weighted values (Boogaart and Schaeben 2002). Otherwise, the shape of variograms can be shifted causing miscalculations of Kriging weights. For this reason, data conversion is applied in order to achieve normal distribution out of a data that

Table 2 Comparison of gross α and gross β concentrations in waters with literature

Place	Water type	Average gross α (mBqL ⁻¹)	Average gross β (mBqL ⁻¹)	References
Adana	Ground	9.6	86	Değerlier and Karahan, 2010
Nevşehir	Ground	192	579	Turhan et al., 2013
Bursa	Drinking	68.5	67.1	Taşkın et al., 2012
Bolu	Drinking	87.6	127.6	Görür and Camgöz, 2014
İstanbul	Drinking	23	70	Karahan et al., 2000
Tekirdağ	Drinking	44	100	Kam et al., 2010
Ş.Urfa	Drinking	38	132.4	Bozkurt et al., 2007
Italy	Drinking	8–349	25–273	Forte et al., 2007
Brazil (Sao Paulo)	Drinking	1–400	120–860	Bonotto et al., 2009
Spain	Drinking	30–880	40–2280	Palomo et al., 2007
Hungary	Natural spring	35–1749	33–2015	Jobbygy et al., 2011
Trabzon	Natural spring	8	25	Kobyta et al., 2010
Rize	Natural spring	21	25	Kobyta et al., 2010
Artvin	Natural spring	45.9	91.2	This study

shows lognormal distribution (Krige, 1966; McGrath et al., 2004).

All analyses are carried out in the R environment for statistical computing and visualization (Ihaka and Gentleman, 1996) and the gstat (Pebesma and Wesseling, 1998) and sp R packages (R Development Core Team, 2005).

3 Results and Discussion

3.1 The Activity Concentrations of the Gross α and Gross β

According to the catchment area, results of gross alpha and beta radioactivity for spring water samples, collected from 117 sampling stations, are shown in Table 1. Gross alpha activity for water samples ranged from 5 to 771 mBqL⁻¹ with a mean of 46 mBqL⁻¹. The highest gross alpha radioactivity concentration was 771 mBqL⁻¹ for the samples which were collected from Yusufeli, and this exceeded the limited value of gross alpha (500 mBqL⁻¹) for drinking water, as recommended by WHO. Also, the highest gross beta radioactivity concentration was 808 mBqL⁻¹ for the sample which was collected from Yusufeli and was within the limited value of gross beta (1000 mBqL⁻¹) for drinking water recommended by WHO. In addition, our study showed

that gross beta mean values are two times higher than gross alpha values for the Artvin Province. The results for the gross alpha and beta radioactivity concentrations of spring water samples which were collected from 117 sampling stations showed that all values of measured water samples except one were within the limits which were recommended by WHO (2011). For this reason, further analysis related to specific radionuclide is not necessary.

Comparison of the activity concentrations of gross α and gross β measured in the natural spring water samples in the present study with doses reported for different regions is shown in Table 2. It is shown that the gross alpha and beta activity concentration results of Artvin are essentially lower than those of the Rize and Trabzon Provinces which are in same geographic region.

In addition to the study results of the Nevşehir and Bolu Provinces in Turkey, study results in Italy, Brazil, Spain, and Hungary were relatively higher than our study results, and other study (such as Adana, İstanbul, Trabzon, Rize, etc.) results were similar to ours.

3.2 Geostatistic Analyses and Interpolated Estimated Maps

Estimation of ordinary Kriging is created by using an isotopic experimental semivariogram which is fitted by a suitable model. We have determined that the most

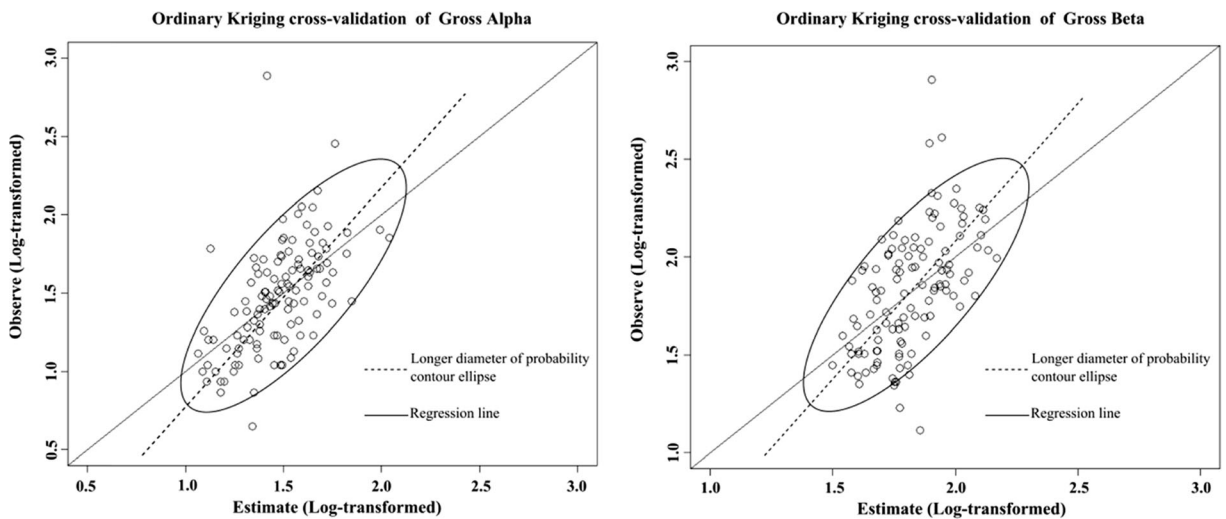


Fig. 2 The graph of scatter plot and cross-validation for gross alpha and beta activity (this figure plot using Log by the R code)

suitable model for the estimation of interpolated map for gross alpha activity concentrations was the spherical model while for gross beta activity concentrations, it was the exponential model.

According to the determined models for gross alpha and beta activity concentrations of estimated ordinary Kriging, cross-validity graphs for each measurement points is shown in Fig. 2. We have found out that there

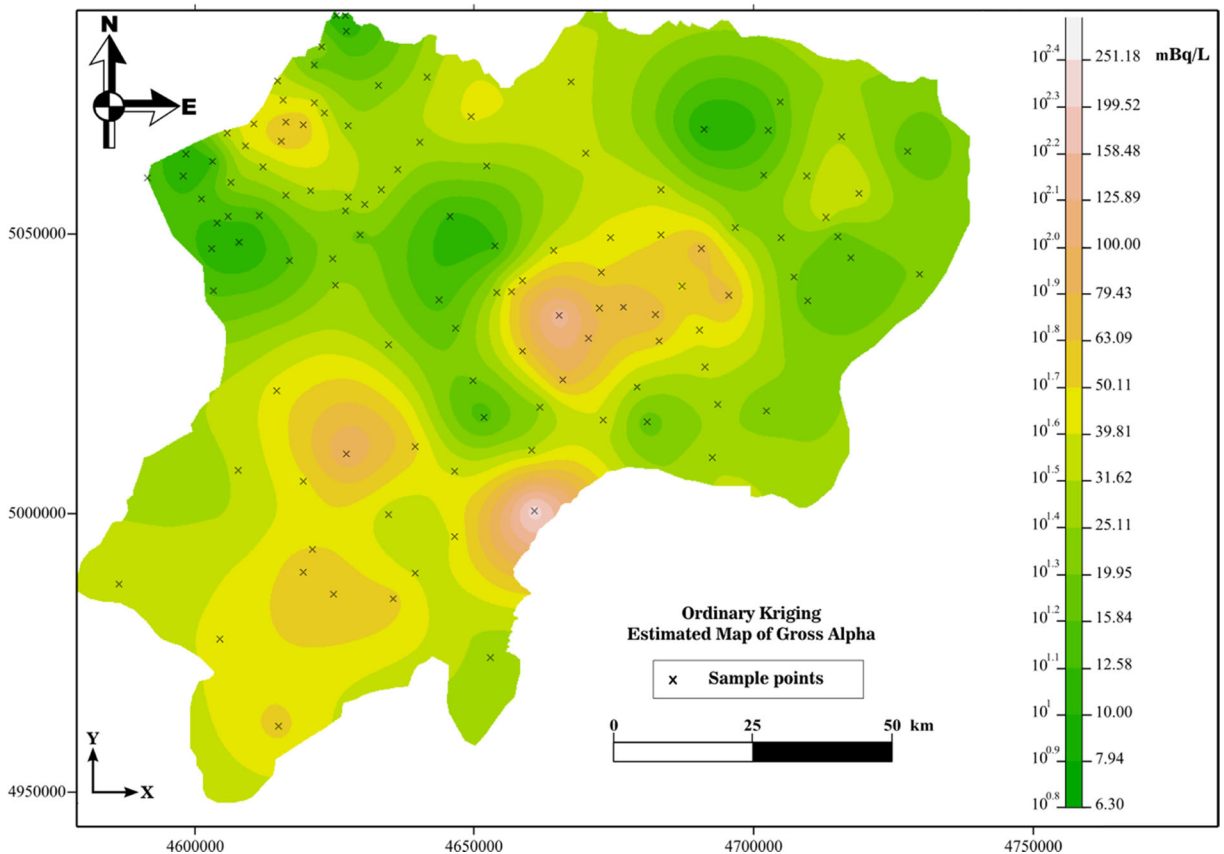


Fig. 3 Ordinary Kriging interpolated maps for gross alpha activity concentration (this figure plot using by the R code)

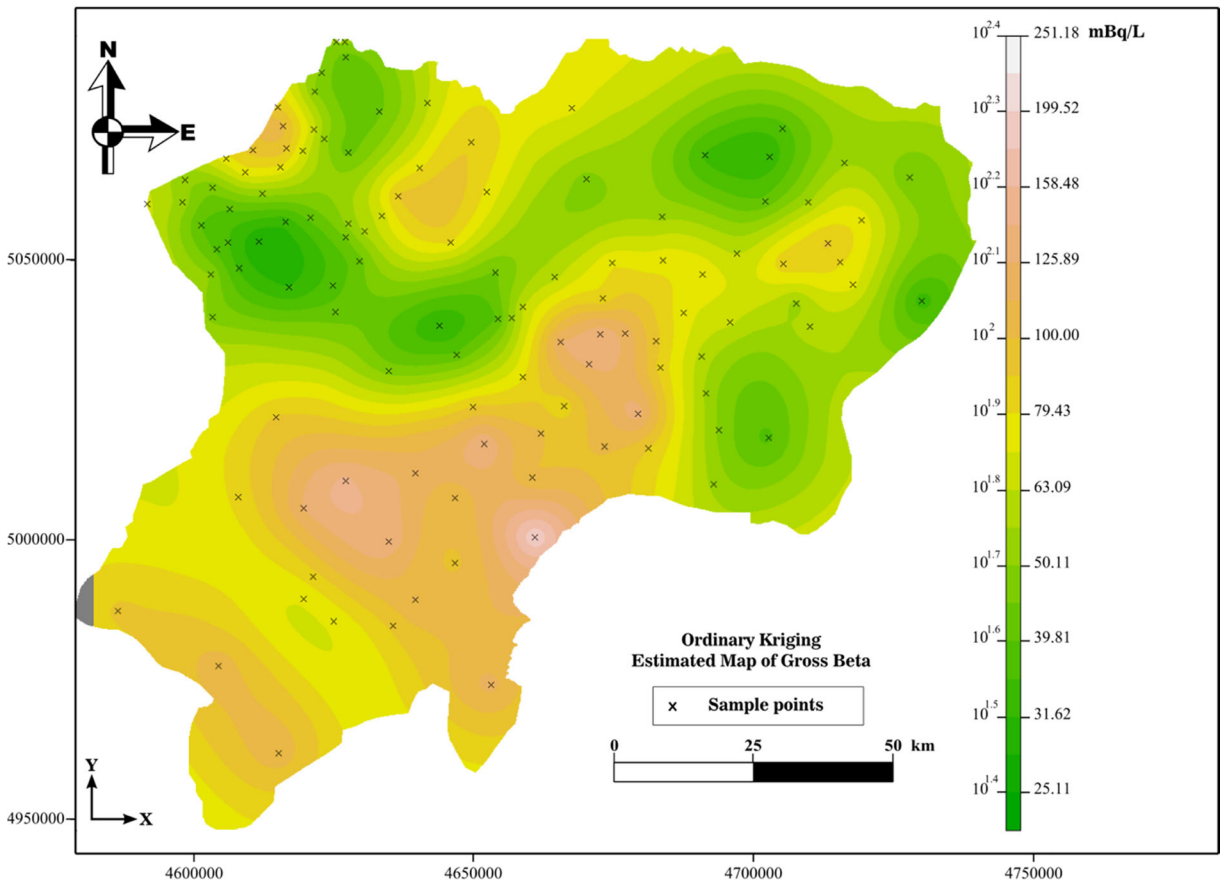


Fig. 4 Ordinary Kriging interpolated maps for gross beta activity concentration (this figure plot using by the R code)

was a strong linear correlation between certain values and estimated values for both graphs. A number of estimated points which are in the study area were in the probability contour ellipse. In addition, the fact that there is only a small angle deviation from the long part

of the regression line of the probability counter ellipse shows that both estimated values of standard error were at a very low level.

According to the determined models, ordinary Kriging-interpolated estimated maps of study areas for

Table 3 Annual effective dose equivalent and excess lifetime cancer risk for the spring water samples

Places	Annual effective doses mean ($\mu\text{Sv year}^{-1}$)			Estimated lifetime risk		
	Min.	Max.	Mean	Min.	Max.	Mean
Artvin Centrum	4.70	149.49	27.00	2.40×10^{-5}	7.63×10^{-4}	12.25×10^{-5}
Arhavi	2.61	20.38	7.87	1.34×10^{-5}	1.04×10^{-4}	3.99×10^{-5}
Hopa	3.14	44.95	21.08	1.60×10^{-5}	2.30×10^{-4}	8.60×10^{-5}
Yusufeli	14.11	402.99	50.50	7.21×10^{-5}	20.59×10^{-4}	22.68×10^{-5}
Ardanuç	5.23	73.70	31.00	2.67×10^{-5}	3.77×10^{-4}	13.84×10^{-5}
Şavşat	3.14	21.95	10.62	1.60×10^{-5}	1.12×10^{-4}	5.95×10^{-5}
Murgul	8.89	26.13	17.60	4.54×10^{-5}	1.33×10^{-4}	8.58×10^{-5}
Borçka	3.66	14.11	9.40	1.87×10^{-5}	7.21×10^{-5}	5.2×10^{-5}
Provincial wide	2.61	402.99	21.08	1.34×10^{-5}	20.59×10^{-4}	10.12×10^{-5}

both gross alpha and beta were shown in Figs. 3 and 4. Especially in the north part of the study region, gross alpha and beta activity concentration levels were low, whereas in south and central parts of the study region, activity concentration levels were relatively high.

3.3 The Annual Effective Dose Equivalent and Lifetime Risk Assessment

Annual effective dose equivalents and excess lifetime cancer risk derived from gross alpha and beta radioactivity concentrations of spring waters in the Artvin province are shown in Table 3. Annual effective dose equivalent values were between 2.61 and 402.99 μSv with the mean of 21.08 μSv in Artvin. These results were lower than the WHO limit value which is 0.1 mSv. However, two values of water samples exceeded the WHO limits, one of them was 0.4 mSv, collected from Yusufeli, and the other was 0.15 mSv, collected from centre of Artvin.

When lifetime cancer risk due to the water radioactivity concentration was investigated, risk values ranged between 1.34×10^{-5} and 20.59×10^{-4} with the mean of 10.12×10^{-5} . The results show that there is no risk for consumption of spring water in the study area and it can be consumed safely.

In our study, we found that the spring water of the Artvin Province contains low-level radioactive isotopes in general and it is acceptable as high-quality drinking water in terms of radioactivity.

4 Conclusions

Gross alpha and beta activity analysis was performed for 117 natural spring water samples which were collected from the Artvin Province. This is the first detailed study of radioactivity concentrations in natural spring water samples in the Artvin Province. When the gross alpha and beta radioactivity levels of the water samples were investigated, all water samples' radioactivity levels were found out to be within limit values. However, the gross alpha activity concentration value of one sample which was collected in Yusufeli exceeded the limit value recommended by the WHO. Moreover, results of this study were compared with similar studies in literature. In addition, radiological estimated interpolation maps were drawn using the ordinary Kriging method for the Artvin Province. In this way, the findings obtained with the

results of gross alpha and beta activity values, which were collected from the study area, were used for the estimation of radiological interpolated values for other parts of the region, where water samples were not collected. Gross alpha and beta measurement results were used to calculate the annual effective dose equivalent for each water sample. Just two of all samples exceeded the limit values. One of them was collected from Yusufeli which were 4 times higher than the limit value of 500 mBqL^{-1} . The other sample was collected from central Artvin, which was 1.5 times higher than the limit values.

Finally, excess lifetime cancer risks were calculated by using values of the annual effective dose equivalent for each water sample, and estimation of lifetime cancer risk tables was done for the general population of the Artvin Province. The data obtained in this study is a baseline which can be used to evaluate possible future changes. It should provide a good baseline for setting standards for water quality in Turkey.

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