

A Study on Determination of Natural Radionuclide Concentrations in Tap Water Samples Collected from the Districts of Sinop Province, Türkiye

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Abstract

Since Turkey's second nuclear power plant is planned to be built in Sinop province, it is very important to determine the background levels of radioactivity in this region. In the present study, activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K natural radionuclides in nine tap water samples collected from the districts of Sinop province located in the north of Turkey were determined using a high purity germanium detector (HPGe). The coordinates of sampling areas were also recorded by GPS device (Magellan Explorist 510). The mean activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K radionuclides in the studied waters were calculated to be 1.74 ± 0.49 Bq/L, 1.39 ± 0.35 Bq/L and 5.32 ± 1.41 Bq/L. In addition, annual effective dose equivalents from the intake of these radionuclides in the tap water samples were calculated for infants, children and adults and the found results were compared with the values recommended by international organizations. As a result, there is no risk in terms of natural radioactivity in the consumption of studied waters by humans.

Keywords: Natural radioactivity, Radionuclide, Tap water, Nuclear power plant, Sinop

Sinop il ve İlçelerinden Alınan Musluk Suyu Örneklerinde Doğal Radyonüklit Konsantrasyonlarının Belirlenmesi Üzerine Bir Çalışma

Öz

Türkiye'nin ikinci nükleer santralının Sinop ilinde kurulması planlandığından bu bölgedeki radyoaktivite arka plan düzeylerinin belirlenmesi çok önemlidir. Bu çalışmada, Türkiye'nin kuzeyinde yer alan Sinop il merkezi ve ilçelerinden toplanan dokuz musluk suyu örneğinde ²²⁶Ra, ²³²Th ve ⁴⁰K doğal radyonüklitlerinin aktivite konsantrasyonları yüksek saflıkta bir germanyum dedektörü (HPGe) kullanılarak belirlenmiştir. Örnekleme noktalarının koordinatları ayrıca GPS cihazı (Magellan Explorist 510) ile kaydedilmiştir. İncelenen sulardaki ²²⁶Ra, ²³²Th ve ⁴⁰K radyonüklitlerinin ortalama aktivite konsantrasyonları $1,74 \pm 0,49$ Bq/L, $1,39 \pm 0,35$ Bq/L ve $5,32 \pm 1,41$ Bq/L olarak hesaplanmıştır. Ayrıca bebekler, çocuklar ve yetişkinler için su örneklerinin tüketilmesi sonucunda bu radyonüklitlerin alınmasıyla elde edilen yıllık etkin doz eşdeğerleri hesaplanmış ve bulunan sonuçlar uluslararası kuruluşlar tarafından önerilen değerlerle karşılaştırılmıştır. Sonuç olarak incelenen suların insanlar tarafından tüketilmesinde doğal radyoaktivite açısından herhangi bir risk bulunmadığı belirlenmiştir.

Anahtar Kelimeler: Doğal radyoaktivite, Radyonüklit, Musluk suyu, Nükleer güç santrali, Sinop

1. Introduction

Environmental radiation in the any region originates from a number of naturally occurring and man-made sources. The measurement of concentrations of naturally occurring radioactive materials (NORM) in waters used by people is useful for determining human population exposure to ionizing radiation by ingestion and domestic uses because the radiation doses from these pathways are strongly related to the amount of radionuclides present [1,2]. NORM consist of potassium, uranium, thorium and any of their decay products such as radium, radon and thoron [3].

Water constitutes about three quarters of our planet and human body. Due to the importance of water for human life, its quality must be strictly controlled. For this reason, studies of drinking water for human consumption must be performed in order to guarantee that they have a low level of radioactivity [4].

Recently, studies related to the determination of the natural radioactivity (^{226}Ra , ^{232}Th and ^{40}K) in drinking water from different sources were performed worldwide [5-12]. However, concentration levels of ^{226}Ra , ^{232}Th and ^{40}K in drinking water used in Sinop city and its districts and the radiological impacts of the consumption of drinking water have not been reported in the literature previously.

The objective of this study is to obtain the concentrations of natural radionuclides in tap water samples collected from Sinop province in Turkey. The reason why Sinop province was chosen in this study is that Turkey's second nuclear power plant will be built to this province. In addition, annual effective doses taken by individuals through consumption of the studied waters were calculated and compared with the value recommended by World Health Organization (WHO). The results obtained from this study will be a reference for the studies carried out after the establishment of the nuclear power plant.

2. Materials and Method

A. Study Area

The Sinop was established on the Boztepe Cape and Peninsula, which extends to the north of the Black Sea coastline. It is located between the latitudes of $41^{\circ} 12'$ and $42^{\circ} 06' \text{ N}$ and the longitudes of $34^{\circ} 14'$ and $35^{\circ} 26' \text{ E}$ (Figure 1). Sinop is a province which is famous in summer tourism, where the history and culture embrace all together with natural beauties. The population of Sinop province is 218,408 people [13].

B. Collection and Preparation of Samples

Nine tap water samples were collected with 1000 mL capacity polypropylene bottles from centre and districts of Sinop. The samples were acidified with hydrochloric acid (Merc, fuming 37%) soon after sampling to prevent adsorption of radionuclides onto the walls of the containing bottle [14]. The coordinates of sampling points were also recorded by a GPS device (Magellan Explorist 510). Then, the water samples were transported to laboratory (Nuclear Physics Research) for the natural radioactivity measurement. In the laboratory, the water samples were first filtered to remove undesired particles. All water samples were evaporated in a furnace to

reduce their volume to approximately 100 mL. Then, the water samples were weighed and transferred into uncontaminated empty Marinelli beakers of uniform size [15]. The water samples were stored four week to allow daughter products to come into radioactive secular equilibrium with their parents ^{226}Ra and ^{222}Rn before radiometric analysis.

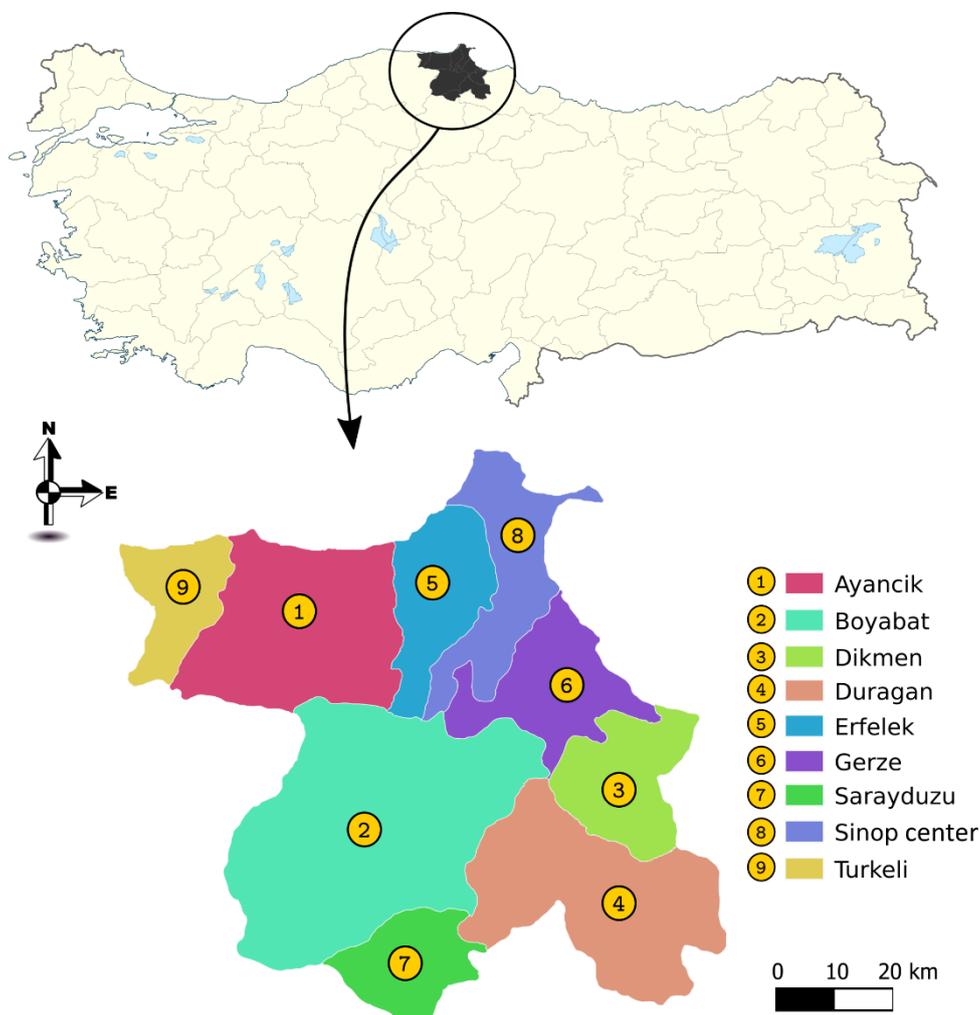


Figure 1. Map of Sinop province

C. Determination of Radioactivity

Natural radioactivity concentration measurements (^{226}Ra , ^{232}Th and ^{40}K radioisotopes) in the samples were carried out using ORTEC brand (Model: GEM55P4-95) High Purity Germanium Detector (HPGe), which has a separation power (resolution) of 1.9 keV at 1332.5 keV and relative efficiency of 55%. Separation power can literarily be referred to as the resolution of a detector. High resolution enables the separation of two gamma lines which are close to each other thereby eliminating spectral interferences. The energy resolution of a detector is typically discussed in terms of the full width half maximum (FWHM) of the peak in related gamma-ray energy. The gamma spectrometer system consists of a detector, a spectroscopy amplifier, a preamplifier, an ADC system that converts analogue counts into electronic signals and a multichannel analyzer (MCA). The experimental scheme of the gamma spectrometer system used in the measurements is given in Figure 2. The detector (HPGe) was enclosed in a 100 mm

thick lead shield to reduce the natural external background radiation. The water samples were measured 80000 second at the detector and the image of one from the obtained spectra is presented in Figure 3. Gamma Vision, a data analysis program, was used to evaluate the spectra obtained from counting. ^{226}Ra concentration was estimated from the gamma-ray peaks at 351.92 keV of ^{214}Pb and 609.32 keV of ^{214}Bi . ^{232}Th concentration was estimated from the gamma-ray peaks at 583.19 keV of ^{208}Tl and 911.16 keV of ^{228}Ac . ^{40}K concentrations were estimated using its 1460 keV gamma ray peak. The natural radioactivity concentrations in the water samples after determining the peak areas at these energies were determined using equation 1.

$$A(\text{Bq/L}) = \frac{C}{\varepsilon \cdot I \cdot V \cdot t} \quad (1)$$

where C is the net counts rate under the full energy peaks, ε is detector efficiency in specific energy, I is the possibility of absolute transition, V is volume of the sample (L) and t is the counting time in seconds.

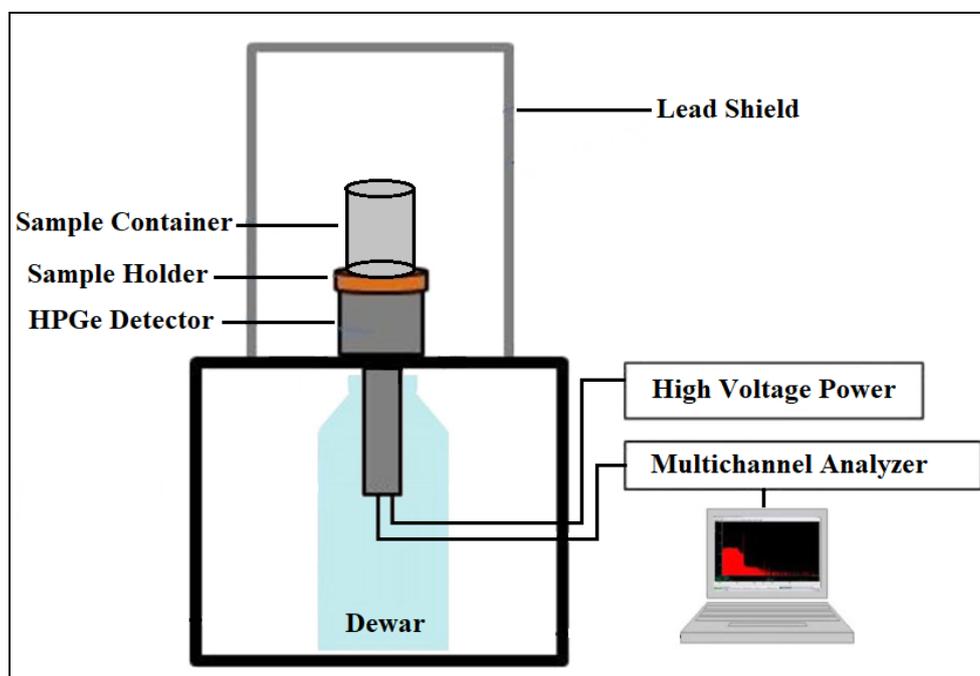


Figure 2. The experimental scheme of the gamma spectrometer system

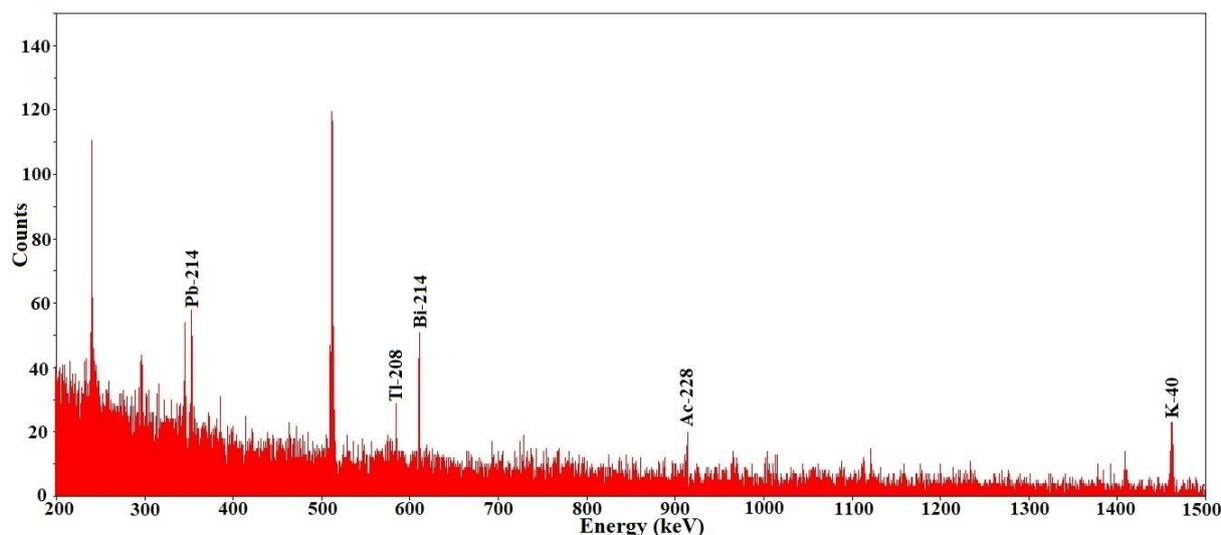


Figure 3. A typical spectrum image obtained from measuring samples at the detector

Energy calibration and efficiency of the detector have been carried out with a ^{152}Eu calibration source (Amersham Company UK) [16]. For this, the ^{152}Eu standard source was placed in front of the detector and counted for 15 minutes. Then the count rate values of the spectra formed at 121.73, 244.54, 344.05, 410.57, 443.73, 778.77, 866.94, 963.78, 1085.44, 1111.47, 1212.01, 1298.54 and 1407.13 keV energies of the ^{152}Eu standard source were obtained. Using these values, detector efficiencies at energies of interest were calculated using equation 2. The efficiency graph obtained is shown in Figure 4.

$$\varepsilon = \frac{N}{A \cdot I_{\gamma} \cdot t} \quad (2)$$

where, ε is the efficiency at the gamma energy of interest, N is the net count rate at the energy of interest, A is the current activity (Bq) of the standard source used for the efficiency calibration, I_{γ} is the abundance of gamma rays at the energy of interest and t is the counting time in seconds.

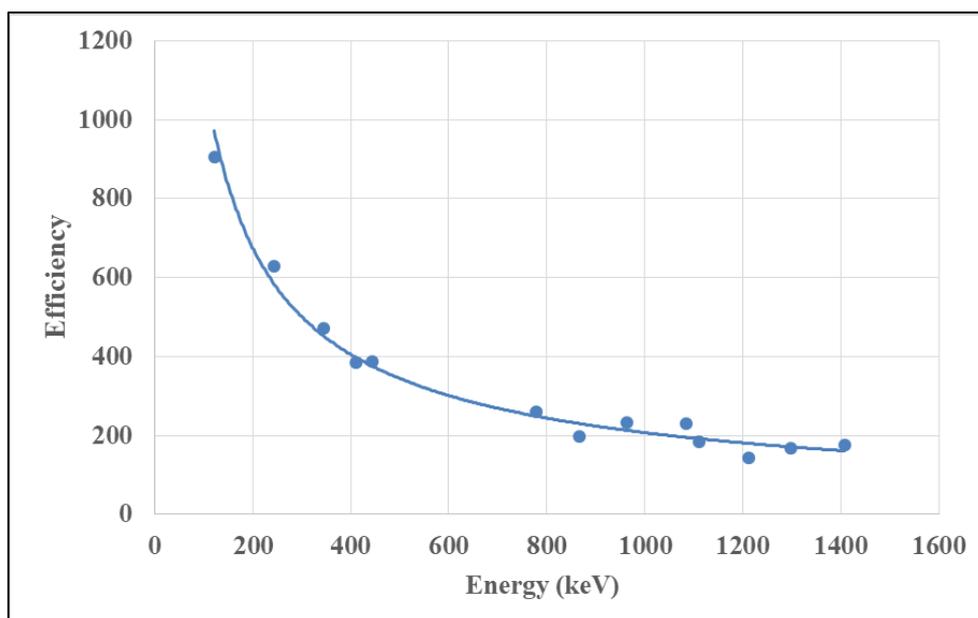


Figure 4. The efficiency graph obtained from Eu-152 standard source.

Annual effective dose (AED) taken through consumption of the studied water samples was calculated for infants, children and adults using the equation 3 [17].

$$\text{AED (mSv/y)} = C \times \text{CR} \times \text{DC} \quad (3)$$

where C is radionuclide concentration in water (Bq/L), CR is the consumption rate (L/y) of drinking water (250 for infants, 350 for children and 730 L for adults) [18], DC is dose coefficients for radionuclides (2.8×10^{-4} for ^{226}Ra , 6.9×10^{-4} for ^{232}Th and 6.2×10^{-6} mSv/Bq for ^{40}K) [19].

D. Geostatistics

Geostatistics is a spatial analysis method that takes into account the relationship between samples and the coordinates from which the samples are taken [20]. In this context, it differs from traditional statistical methods. This method is based on the theory of stationary randomness [21]. In this method, an estimate of the distribution can be calculated over the average value [22]. The relationships depending on the distances between spatial variables in geostatistical calculations are determined by the semi-variogram function. In present study, the Ordinary Kriging method that a geostatistical method that is easy to calculate and can predict the distribution at an appropriate mean value was used [23]. This method is mathematically defined as the best known linear and unbiased estimator [24].

3. Results and Discussion

The GPS coordinates taken and natural radioactivity concentrations (^{226}Ra , ^{232}Th and ^{40}K) of the 9 tap water samples collected from centre and districts of Sinop are given in Table 1. The natural radioactivity levels found for the studied water samples are also shown in Figure 5. The concentrations found in the present study ranged from 1.33 ± 0.65 (Dikmen district) to 2.09 ± 0.41

(Boyabat district) Bq/L for ^{226}Ra , from 0.95 ± 0.29 (Türkeli district) to 1.73 ± 0.30 (Saraydüzü district) Bq/L for ^{232}Th , from 3.91 ± 1.03 (Centre) to 6.59 ± 1.98 (Gerze district) Bq/L for ^{40}K .

Table 1. The taken GPS coordinates and natural radionuclide concentrations of the water samples

Sample Code	GPS Coordinates		Radionuclide Concentration (Bq/L)		
	Latitude	Longitude	^{226}Ra	^{232}Th	^{40}K
Ayancık	41.946160	34.588380	1.88 ± 0.56	1.46 ± 0.39	5.18 ± 1.22
Boyabat	41.468670	34.766650	2.09 ± 0.41	1.61 ± 0.27	4.39 ± 1.65
Dikmen	41.651191	35.263431	1.33 ± 0.65	1.47 ± 0.41	5.87 ± 1.58
Durağan	41.415410	35.054680	1.34 ± 0.53	1.60 ± 0.33	4.16 ± 1.08
Erfelek	41.879766	34.906961	1.73 ± 0.57	1.26 ± 0.28	5.90 ± 1.18
Gerze	41.803305	35.198520	1.78 ± 0.44	1.30 ± 0.44	6.59 ± 1.98
Center	42.026310	35.151600	1.69 ± 0.37	1.17 ± 0.43	3.91 ± 1.03
Saraydüzü	41.329750	34.846080	1.99 ± 0.47	1.73 ± 0.30	5.38 ± 1.18
Türkeli	41.947175	34.335724	1.82 ± 0.42	0.95 ± 0.29	6.54 ± 1.83

The average activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in the water samples were found to be 1.74 ± 0.49 , 1.39 ± 0.35 and 5.32 ± 1.41 Bq/L, respectively. The limit values of ^{226}Ra , ^{232}Th and ^{40}K radioisotopes in drinking waters is stated by World Health Organization (WHO) to be 1, 0.1 and 10 Bq/L, respectively [18]. The natural radioactivity values obtained in this study were slightly higher than the recommended these values except for K-40 radioisotope.

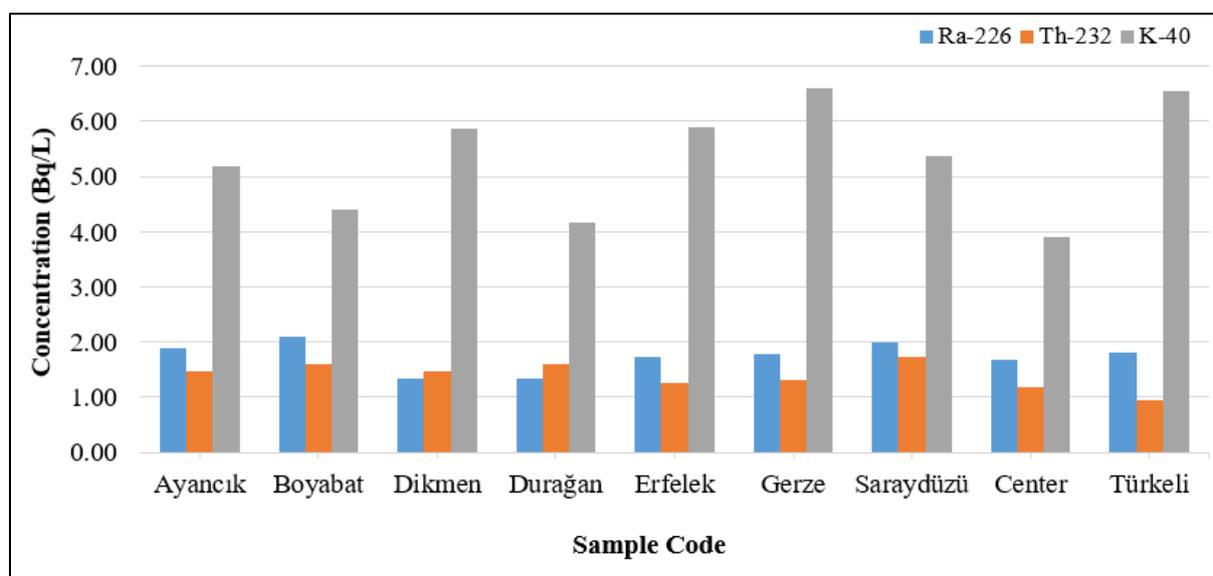


Figure 5. ^{226}Ra , ^{232}Th and ^{40}K concentrations in the water samples

The interpolation maps created for the water samples taken from districts and centre of Sinop province are given in Figure 6. These interpolation maps reveal the general pattern of radionuclide activity distributions and show estimate of an average activity concentration for

unmeasured areas. According to the maps given in figure 3, it is observed that Ra-226 and Th-232 radionuclide activities are at high levels, especially in the inner parts of the Sinop, while the concentration of radiation activity towards the shores generally decreases. While the K-40 activity concentrations in the center and districts of Sinop province are higher in the coastal areas, they are lower in the interior areas.

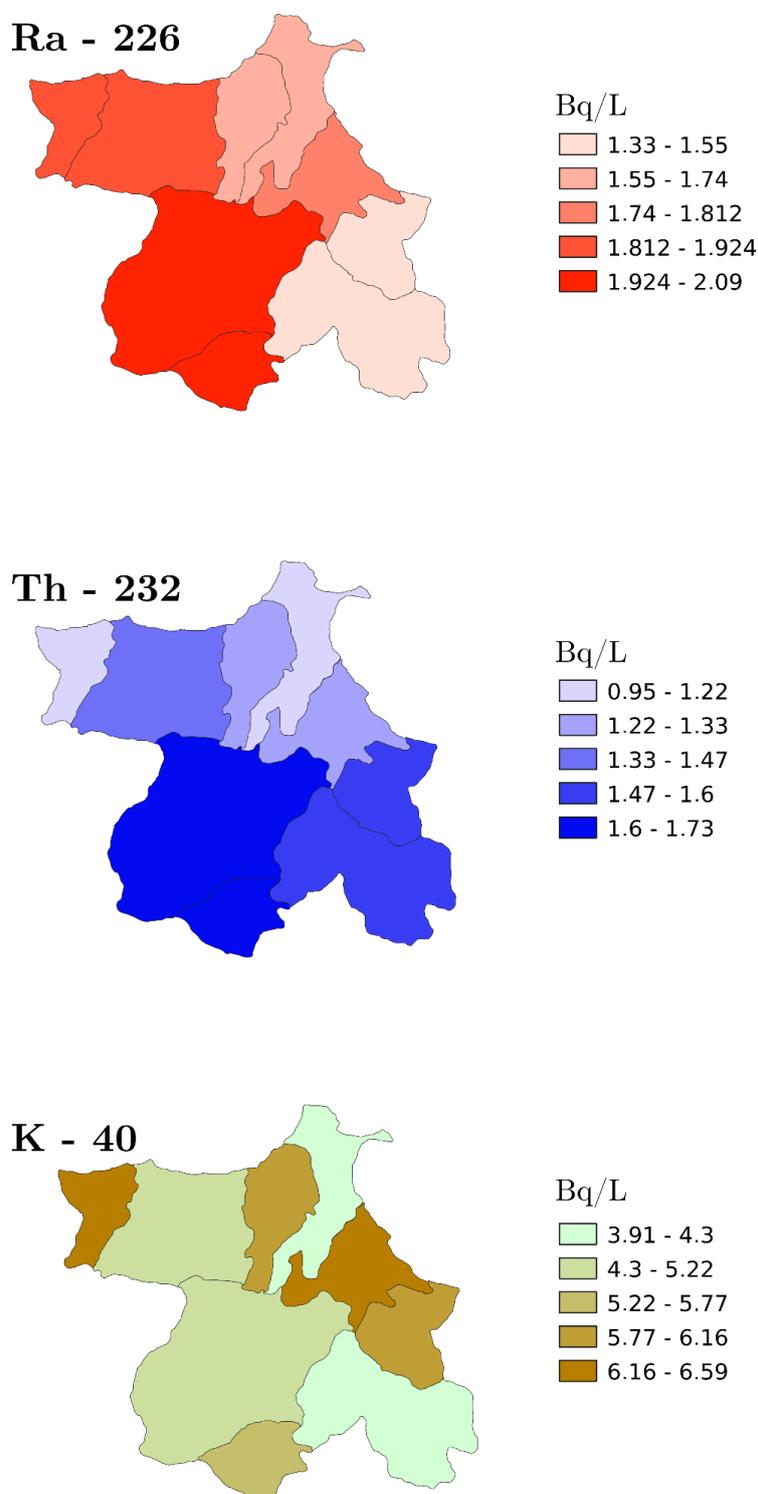


Figure 6. Geo-spatial distribution maps of the activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K for the water samples

The annual effective dose values taken by individuals through consumption of the tap water samples was are presented in Table 2. The values of total annual effective dose found for infant, child and adult varied from 0.30 to 0.45 mSv/y with an average of 0.37 mSv/y, 0.42 to 0.62 mSv/y with an average of 0.52 mSv/y, and 0.88 to 1.30 mSv/y with an average of 1.08 mSv/y, respectively. The values of total annual effective dose calculated for infant, child and adult were found below the individual dose criterion of 1 mSv/y recommended by WHO [18] except for the value found for adults. The total annual effective dose values taken by individuals through consumption of the studied waters are also shown in Figure 7.

Table 2. Annual effective dose values estimated for infant, child and adult

Sample Code	Total Annual Effective Dose (mSv/y)		
	Infant	Child	Adult
Ayancık	0.391	0.548	1.143
Boyabat	0.431	0.603	1.258
Dikmen	0.356	0.498	1.039
Durağan	0.376	0.527	1.099
Erfelek	0.348	0.487	1.015
Gerze	0.359	0.503	1.048
Saraydüzü	0.446	0.624	1.303
Centre	0.326	0.457	0.952
Türkeli	0.301	0.422	0.880

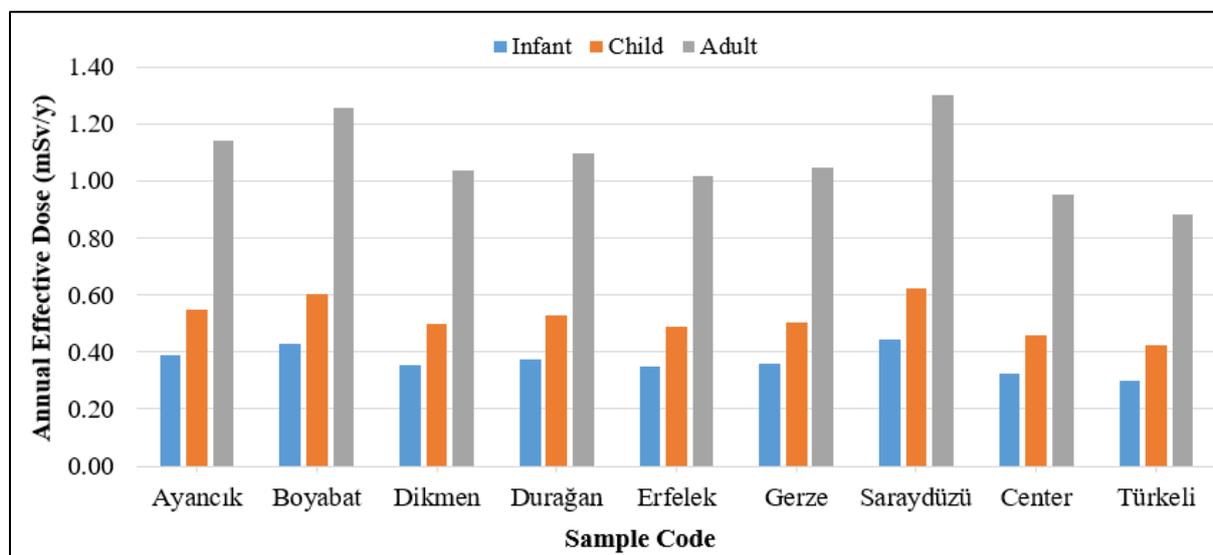


Figure 7. The annual effective dose values taken by individuals (infant, child and adult)

4. Conclusion

^{226}Ra , ^{232}Th and ^{40}K concentrations of nine tap water samples collected from centre and districts of Sinop province were determined using high purity germanium detector (ORTEC) with a 55% relative efficiency. The annual effective dose values taken by individuals through consumption

of the tap water samples were also calculated. The results of this study show that the annual effective doses calculated for the infants, children and adults were below the individual dose criterion of 1 mSv/y recommended by WHO except for the value found for adults. In addition, the annual effective dose value calculated for adults were slightly higher than the recommended this value. As a result, it can be said that the consumption of studied water will not pose any radiological risk in terms of human health. Also, the obtained results can be contributed to the baseline data of radionuclide concentrations in this area.

Ethics in Publishing

There are no ethical issues regarding the publication of this study.

Acknowledgment

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