Journal of Water & Health

Journal of Water and Health Vol 21 No 12, 1847 doi: 10.2166/wh.2023.205

Tritium contents in drinking and surface seawaters before the nuclear power plant planned in Sinop (Türkiye) and their radiological risks on human population

Serdar Dizman [a,*], Feyza Zeynep Aşık[b], Ali Erdem Özçelik[c], Recep Keser[d] and Filiz Korkmaz Görür[e]

[a] Department of Physics, Faculty of Arts and Sciences, Recep Tayyip Erdogan University, Rize 53100, Türkiye
[b] Institute of Graduate Studies, Recep Tayyip Erdogan University, Rize 53100, Türkiye
[c] Department of Landscape and Architecture (Geomatics Engineer), Faculty of Engineering and Architecture, Recep Tayyip Erdogan University, Rize 53100, Türkiye
[d] Department of Basic Sciences, Faculty of Engineering, Samsun University, Samsun 55100, Türkiye
[e] Department of Physics, Faculty of Arts and Science, Bolu Abant İzzet Baysal University, Bolu 14100, Türkiye
*Corresponding author. E-mail: serdar.dizman@erdogan.edu.tr

SD, 0000-0002-6511-9526

ABSTRACT

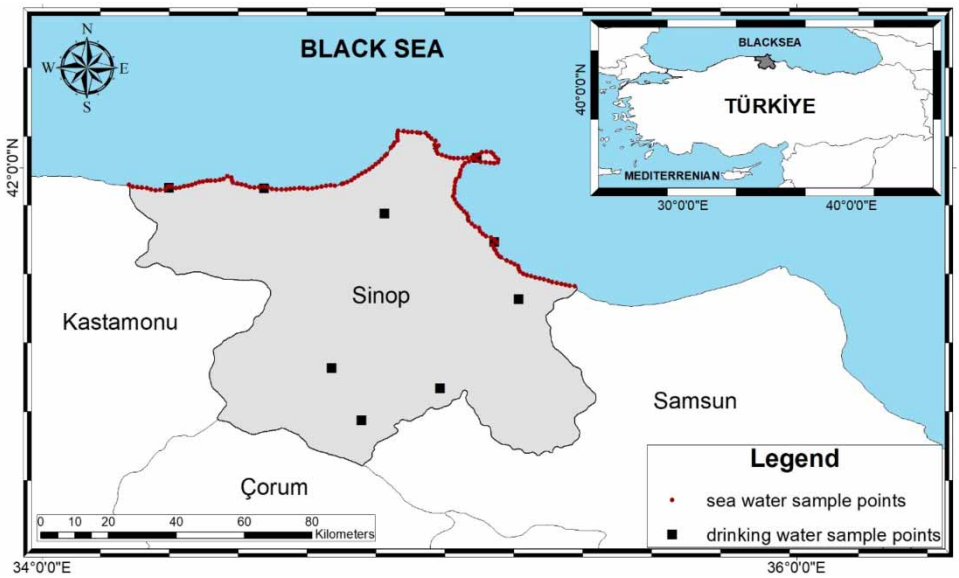
This study aims to determine the background levels of tritium radioisotope in drinking and seawater samples of Sinop province before the nuclear power plant was established in Sinop. In this context, a total of 174 water samples were collected, these are as follows: nine drinking water samples from the Sinop center and districts and 165 seawater samples from the seacoast from Samsun to Kastamonu. Tritium concentrations in the collected water samples were measured by the liquid scintillation counter. The minimum detectable activity for the method used was found to be 1.48 Bq/L. The tritium concentrations of the seawater and drinking water samples were found in the range of <MDA (minimum detectable activity)-6.29 Bq/L and 2.45–3.17 Bq/L, respectively. In addition, the annual effective dose rates of the people consuming these drinking waters due to the tritium radioisotope were calculated separately for infants, children and adults and found to be 12.12, 16.96 and 35.38 nSv, respectively.

Key words: health risk, nuclear power plant, population dose, tritium, Türkiye

HIGHLIGHTS

- Tritium concentrations in the seawater and drinking water samples were determined.
- A radiological map of the study area was created.
- Annual effective dose rates for infants, children and adults were calculated.
- Lifetime cancer risk values for females and males were calculated.
- All of the tritium concentrations were found to be lower than the limit values.

GRAPHICAL ABSTRACT



INTRODUCTION

Tritium, a radioactive isotope of hydrogen, emits a beta particle with a maximum energy of 18.6 keV as it decays to helium and its half-life is 12.3 years (Lucas & Unterweger 2000). Tritium isotopes are both naturally occurred and artificially produced. High-energy cosmic rays reacting with nuclei of oxygen and nitrogen in the upper layers of the atmosphere continually produce natural tritium (Masarik & Beer 2009). Many anthropogenic nuclear operations, such as the testing of nuclear bombs and the operation of nuclear reactors, create artificial tritium. Furthermore, ^{3}H can be artificially created via neutron capture in reactors (Okada & Momoshima 1993). The atmosphere's main sources of tritium come from atmospheric thermonuclear tests conducted between 1953 and 1963 (Harms *et al.* 2016). The atmosphere's fast oxidation of formed tritium results in the formation of element tritium (HT), tritiated water (HTO) and tritiated methane (CH_3T).

At present, the Akkuyu nuclear power plant with a power of 4,800 MW is under construction in Mersin province of Türkiye and the first unit of this power plant is planned to be put into operation in 2023 (DNEPI 2012; Dizman *et al.* 2015). Tritium's chemical forms are closely related to types of the nuclear power plant. In pressurized water (PWR) and boiling water (BWR) type reactors, tritium releases mostly occur as HTO and only 1% is in gaseous HT form (Hou 2018). Tritium's health hazard depends on its chemical form and HTO is one of the tritium types that are dangerous for people to consume. HTO can interact directly with the metabolic process after entering the body through the skin, water and food. This can have negative health implications such as an increased risk of cancer and mutation (Matsumoto *et al.* 2021). Because of this, determining the tritium activity levels in waters meant for human consumption is crucial. Many researches on determining the tritium contents in water samples from diverse sources, including seawater samples, have been conducted in various nations (Damianova *et al.* 2016; Osman *et al.* 2016; Deng *et al.* 2017; Ansari *et al.* 2018; Dizman *et al.* 2018; Jean-Baptiste *et al.* 2018; Dizman & Mukhtarli 2021). Although there are many studies on determining tritium activity in drinking water in Türkiye, no research has been conducted on seawater in recent years (Görür & Genç 2012; Gören *et al.* 2014; Karataşlı *et al.* 2017; Dizman & Korkmaz Görür 2019).

The Black Sea receives large amounts of water via the discharges of the central- and eastern-European rivers. There are nuclear power plants on some of these rivers. Therefore, the surface tritium concentration is higher than those of other seas at comparable latitude band. A systematic monitoring of tritium in the Black Sea is not available. However, some studies have been carried out on the determination of tritium in the Black Sea (Nikitin & Vakulovsky 1996; Rank *et al.* 1999; Top 1999).

The aim of this work is to determine both tritium concentrations and radiological risks in surface seawaters and drinking water before the nuclear power plant to be established in Sinop province starts to operate. In order to achieve this, the annual

effective dose rates in three different groups (infants, children and adults) of the Sinop population were assessed, along with the lifelong cancer risks associated with tritium exposure by gender. In addition, determined radioactivity values have been compared with the limit values suggested by the international organizations (Turkish Standards Institution, European Commission and World Health Organization). The results of this study can contribute to the monitoring of tritium radioisotope in the Black Sea and can be utilized as a guide for future research. Moreover, the results will be very valuable, especially for studies to be carried out after the nuclear power plant comes into operation.

MATERIALS AND METHODS

Study area

The study area is the Sinop province (Figure 1). Sinop province is located on the Boztepe Cape and Peninsula lying on the Black Sea coastline and is located between 41° 12′ and 42° 06′ north latitudes and 34° 14′ and 35° 26′ east longitudes (SV 2023). Its area is 5,862 km^2. The central population of Sinop province is 65,489 and the total population is 216,460 (TUIK 2020). There are many beaches in Sinop province and sea fishing is done intensively.

An intergovernmental agreement was signed in 2013 with France and Japan for Türkiye's second nuclear power plant in Sinop. In accordance with the signed agreement, it is planned to establish a French–Japanese joint design ATMEA-1 type nuclear power plant with a total power of 4,480 MW, consisting of four units of 1,120 MW each (NADA 2015). It is stated that the nuclear power plant will be established in İnceburun locality (NADA 2015).

Sample collection and preparation

Firstly, a total of 165 seawater samples were taken at 1 km intervals along the coastline of Sinop province (from the Samsun provincial border to the Kastamonu provincial border). Later, a total of nine drinking water samples were taken, one from each of Sinop center and districts. Both seawater and drinking water samples were taken with 500 mL plastic bottles. The drinking water samples were taken from the city water supply consumed by the people in the relevant district. The coordinates of each sampled point were recorded with the GPS device (Magellan Explorist 510). Then, the water samples were transferred to the laboratory for the tritium measurement. The water samples were first filtered to remove physical particles. The samples were then distilled. The distillation procedure described in detail by Tjahaja and Sukmabuana was carried out by adding 6 M NaOH and 0.05 N $KMnO_4$ into a 250 mL water sample (Tjahaja & Sukmabuana 2012). The plastic vials used in measurements (Zinsser Analytics, 20 mL) were first filled with 10 mL of the distilled samples, and the remaining vials were filled with Ultima Gold LLT scintillation (Perkin Elmer Inc.). The background sample (BKG) was created using groundwater that had undergone two distillations and had little tritium present.

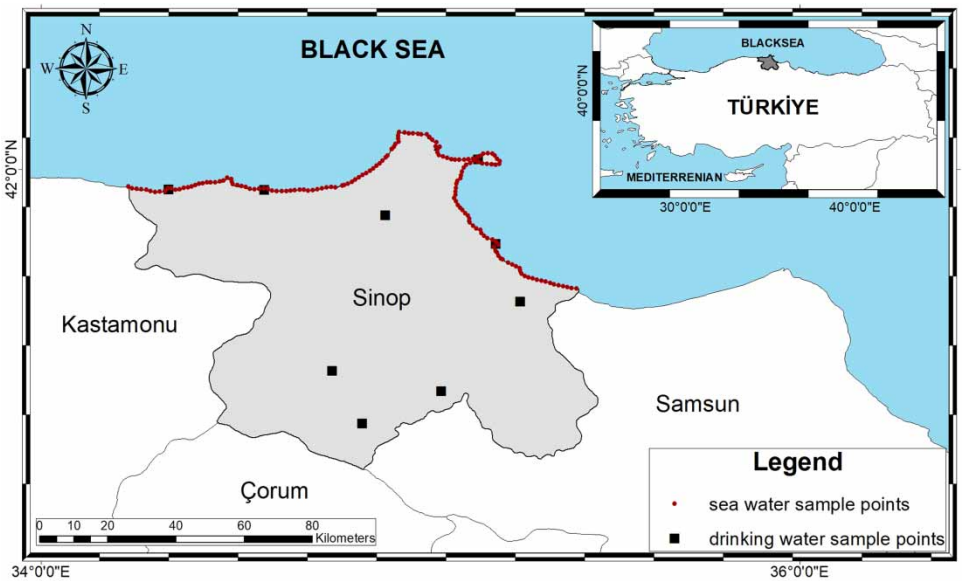


Figure 1 | Study area and sampling points.

Radioactivity analysis

Liquid scintillation counter (LSC) (Perkin Elmer, LSC Tricarb 2910 TR) was used to measure the tritium in the water samples. LSC is a method that is frequently used for the determination of tritium in environmental samples. To calculate the detection efficiency of the LSC, a laboratory (DWS) standard was prepared using a certified liquid tritium standard (Eckert & Ziegler, P.O. No.: P700723, Source No.: 1676–44). The prepared standards and samples were measured in the LCS for 1,500 min (150 min × 10 rounds). The detection efficiency of the LSC was calculated using the following equation:

$$E = \frac{a_{st} - \text{bkg}}{\text{dpm}_{st}} \tag{1}$$

where E is the detection efficiency, a_{st} is the mean of the count rate of the DWS standard (cpm), bkg is the mean of the count rate of the background sample (cpm) and dpm_{st} is the activity of the DWS standard at the measurement date (dpm).

Equation (2) was used to determine the tritium concentrations in the water samples:

$$A\ (\text{Bq/L}) = \frac{(\text{ac} - \text{bkg})}{60 \times E \times V} \tag{2}$$

where ac is the sample count rate (cpm), E is the efficiency and V is the sample volume (L). Equation (3) was used to determine this method's minimum detectable activity (MDA) (Currie 1968):

$$\text{MDA (Bq/L)} = \frac{3.29\sqrt{\frac{\text{bkg}}{t_s} + \frac{\text{bkg}}{t_b}} + \frac{2.71}{t_s}}{60 \times E \times V} \tag{3}$$

where t_s is the counting time of the samples (minute) and t_b is the counting time of the background sample (minute). The annual effective dose rates (AED) linked to radiation exposure from ingesting water samples are used to evaluate the individuals' health risks and are calculated using the following equation:

$$\text{AED (nSv)} = A \times \text{CW} \times \text{DCT} \times 10^9 \tag{4}$$

where A is the concentration of tritium in water (Bq/L), CW is the people's amount of water consumption (the water consumption amounts for adults, children and infants are 730, 350 and 250 L, respectively) (WHO 2011) and DCT is the tritium's dose coefficient (1.8×10^{-11} Sv/Bq).

Equation (5) is used to compute the lifetime cancer risk (LCR) values for females and males owing to the drinking of the water samples:

$$\text{LCR} = A \times \text{CW} \times \text{ALT} \times \text{TCRC} \tag{5}$$

where ALT is the mean lifetime (year) for individuals and TCRC is tritium's cancer risk coefficient (9.44×10^{-13} Bq^{-1}) (EPA 1999). The mean lifetimes for males and females in Türkiye are 75.3 and 80.7 years, respectively (TUIK 2017).

A reference sample with known activity was prepared from the certified tritium source (Eckert & Ziegler, Source No.: 1676–44, P.O. No.: P700723) and tested alongside the samples in order to ascertain the measuring system's accuracy. The reference sample's spiked activity is prepared as 715.60 ± 17.86 Bq/L. Equation (6) is used to determine the Z-score value used by ISO 13528 (ISO 2015) to assess the quality of analytical techniques:

$$Z = \frac{A_s - A_m}{\sqrt{U_s^2 + U_m^2}} \tag{6}$$

where A_s is the reference sample's spiked activity (Bq/L), A_m is the reference sample's measurement activity (Bq/L), at a 95% level of confidence ($k = 2$), U_s and U_m are spiked and measurement uncertainties (Bq/L) of the reference sample, respectively. Z-score must have a value between −1 and 1. The SPSS (Statistical Package for the Social Sciences) computer program (version 21.0) was used to conduct statistical analyses.

RESULTS AND DISCUSSION

Radioactivity in the waters

A common unit of measurement used to express tritium concentrations in measurements of environmental samples is the tritium unit (TU), which is equal to one tritium atom per 1×10^{18} hydrogen atoms (1 TU = 0.118 Bq/L). The results obtained in this study are given in both Bq/L and TU units.

Average cpm (count per minute) values of DWS and background sample in LSC were found as 2,377.67 cpm for DWS and 2.44 cpm for BKG. Using these values, the efficiency and MDA values were calculated as 26% and 1.48 Bq/L (12.54 TU), respectively.

The tritium concentrations of 62 seawater samples (37.6% of the samples) were below the MDA. Descriptive statistics of determined tritium concentrations in the seawater samples taken from Sinop coast of the Black Sea are given in Table 1. The tritium concentration distributions of seawater samples according to the sample code are shown in Figure 2. According to Figure 2, it can be said that the tritium concentration values of the seawater samples are homogeneous. Besides, the interpolation map created for the seawater samples taken along the Sinop coastline is shown in Figure 3.

Table 1 | Descriptive statistics of tritium concentrations in the seawater samples from Sinop coast

Statistics	Value
Number of samples	165
Minimum[a]	1.47
Maximum[a]	6.29
Mean[a]	2.57
Standard deviation[a]	1.11
Median[a]	2.28
Skewness	2.06
Kurtosis	3.84

[a]In Bq/L.

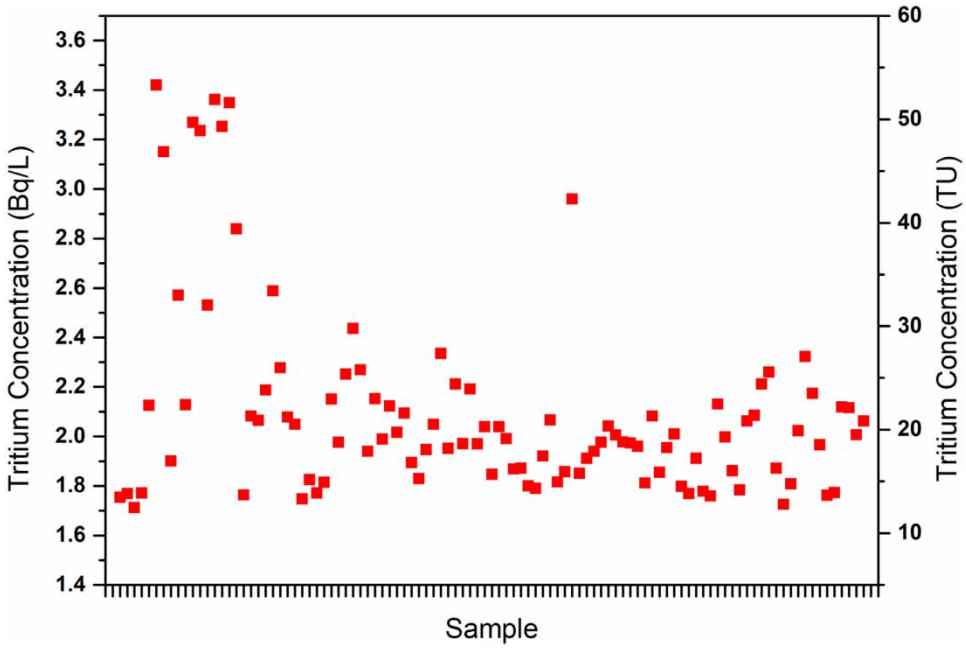


Figure 2 | The tritium concentration (>MDA) distributions of examined seawater samples.

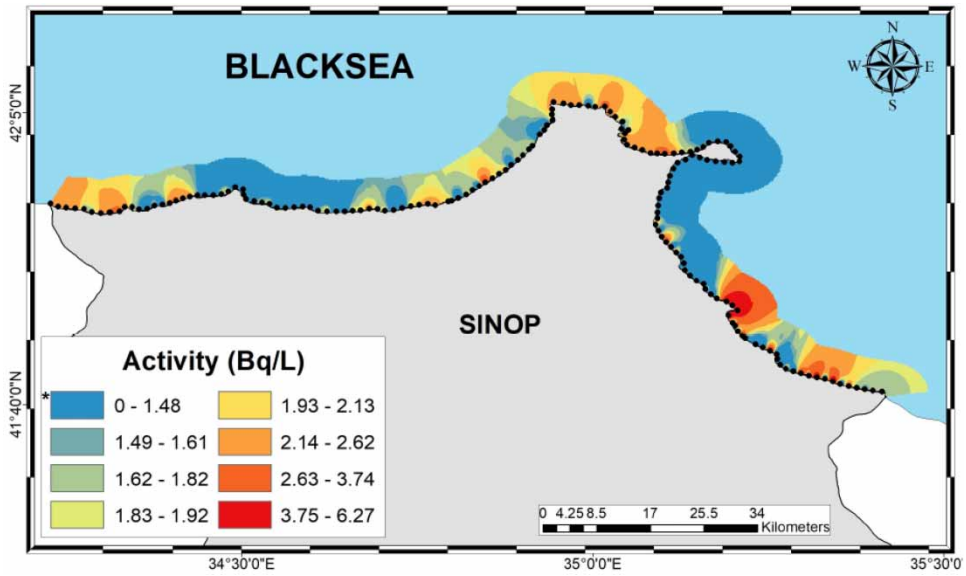


Figure 3 | Interpolation map (*: <MDA) created for the seawater samples taken along the Sinop coastline.

Tritium concentrations of 57% of seawater samples (94 samples) are in the range >MDA-4 Bq/L. The categorical (<MDA, >MDA-2, >2–4, >4–6 and >6) variation frequency of tritium concentration values of seawater samples is shown in Figure 4.

The coordinates and tritium concentration values of drinking water samples taken from the center and districts of Sinop province are given in Table 2. In addition, the interpolation maps created for the drinking water samples according to the sampling region are shown in Figure 5. The mean tritium concentrations of analyzed seawater and drinking water samples were calculated to be 2.57 $\pm$ 0.47 Bq/L (21.78 TU) and 2.69 $\pm$ 0.43 Bq/L (22.81 TU), respectively. The maximum tritium concentration values are found as 6.29 Bq/L (53.29 TU) for the seawater samples ('S7' coded sample) and 3.17 Bq/L (26.86 TU) for the drinking water samples ('Center' coded sample), respectively. The mean tritium content of the samples of drinking

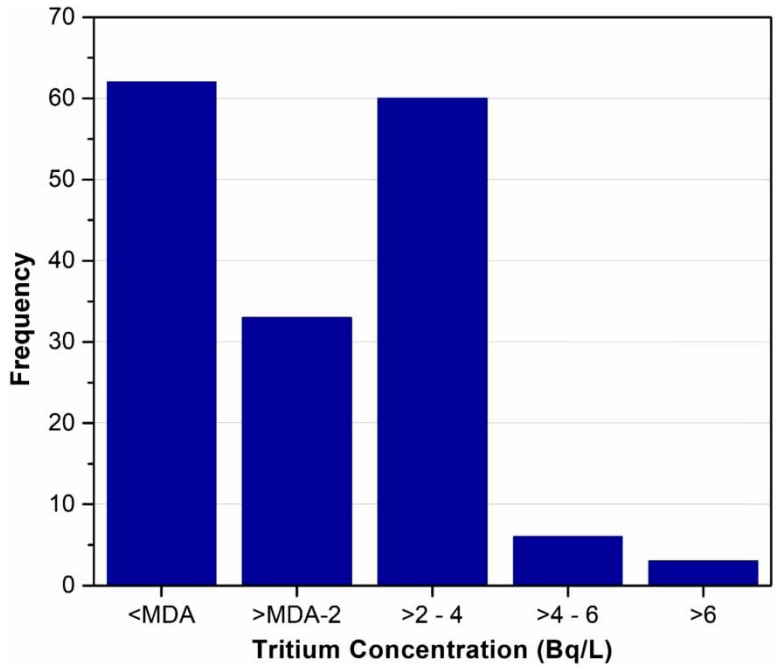


Figure 4 | The frequency distributions of tritium concentration values of seawater samples.

Table 2 | The coordinates and tritium concentrations from which drinking water samples were taken

	GPS coordinates		Tritium concentration	
District	Latitude (N)	Longitude (E)	Bq/L	TU
Ayancık	41.946160	34.588380	2.69 ± 0.33	22.84 ± 2.83
Boyabat	41.468670	34.766650	2.46 ± 0.40	20.83 ± 3.36
Center	42.026310	35.151600	3.17 ± 0.47	26.86 ± 4.01
Dikmen	41.651191	35.263431	2.95 ± 0.47	24.96 ± 4.00
Durağan	41.415410	35.054680	2.84 ± 0.39	24.03 ± 3.29
Erfelek	41.879766	34.906961	2.45 ± 0.48	20.77 ± 4.09
Gerze	41.803305	35.198520	2.57 ± 0.36	21.80 ± 3.01
Saraydüzü	41.329750	34.846080	2.57 ± 0.42	21.80 ± 3.57
Türkeli	41.947175	34.335724	2.53 ± 0.55	21.42 ± 4.67

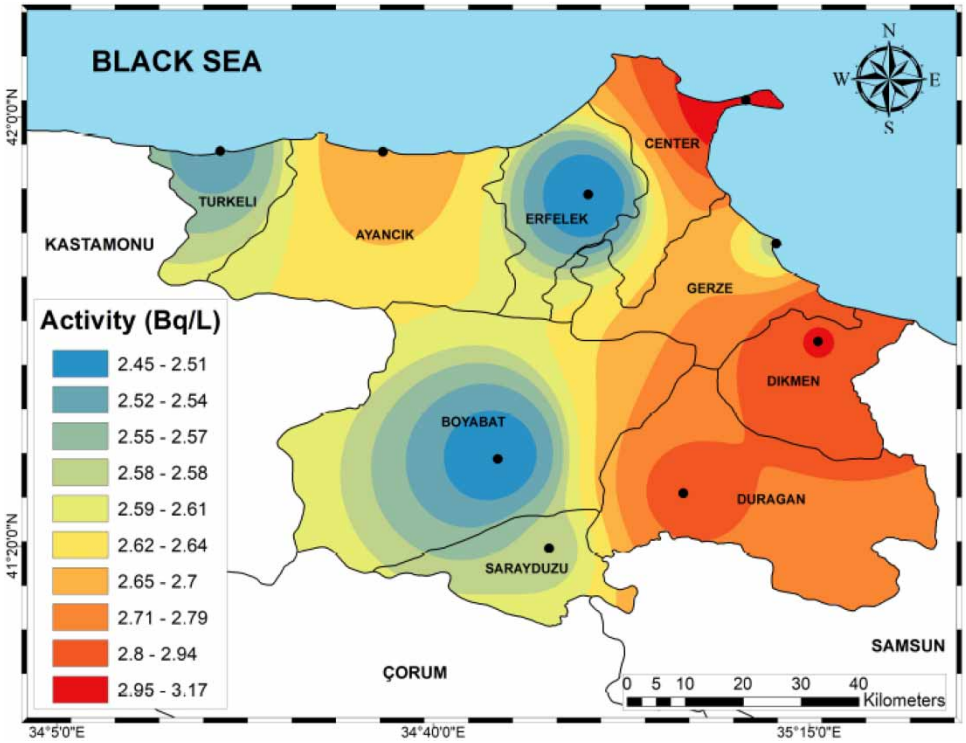


Figure 5 | Interpolation map created for drinking water samples.

water is higher than that of the samples of seawater. Nevertheless, it should be noted that the tritium levels in the water samples analyzed were far lower than the standard for drinking water, which is 100 Bq/L (EC 2003; TSI 2005). The World Health Organization (WHO), an international organization, also recommends the limit value for tritium concentration in drinking water as 10,000 Bq/L (WHO 2011). Therefore, it can be said that there is no radiological risk in terms of tritium radioisotope for the examined sea and drinking water samples.

As a result of this work, the tritium concentration values obtained in seawater samples could only be compared with the study conducted by Top in 1999 in Türkiye. The tritium concentrations found in this study were lower than the determined values in the study done by Top (1999). On the other hand, there are studies on the determination of tritium activity concentrations in seawater samples in different countries of the world. The study in which tritium concentrations were determined in seawater samples and the average tritium concentration values found in studies conducted in different countries are given in Table S1 of the Supplementary Materials.

The average tritium concentration of seawater samples taken along the coast of Sinop province located on the Black Sea coast is lower than from the tritium concentration of the seawater samples by Russian side of the Black Sea, from the tritium concentration in the seawater samples from different points of the Turkish side of the Black Sea, from the tritium concentration of the Pacific ocean water samples from the Misava port in Japan, from the tritium concentration of the water sample from the Bohai sea in China and from the tritium concentration of the Mediterranean water sample from the French coast. However, it was found to be higher than the tritium level of water samples from various locations in the Black Sea and the average tritium level of Irish Seawater samples. As seen in Table S1, it can be said that this tritium concentration difference between seas depends on the mobility of the water in the sea (current, circulation, etc.), whether it is in the gulf or whether there are nuclear power plants on the sea coast. In addition, tritium can exhibit considerable environmental variability and behavior in different ecosystems.

In our country and around the world, investigations have been done to determine the tritium activity concentrations in drinking water. This study in which tritium concentrations were determined in drinking water samples and the average tritium concentration values found in studies conducted in different countries are given in Table S2 of the Supplementary Material. As seen in Table S2, the average tritium concentration determined in the examined drinking waters was found lower than the tritium concentrations in the drinking waters of Italy, Portugal and Türkiye (Mersin), but higher than the tritium concentrations in the drinking waters of Spain, Bulgaria and Türkiye (Rize). It can be said that these concentration differences between and within countries may be due to the fact that tritium varies greatly in the environment.

Evaluation of radiation risks

The AED values of individuals (infant, child and adult) who consume drinking water samples taken from the center and districts of Sinop province were calculated and found values are given in Table S3 of the Supplementary Material. It has been determined that these calculated annual effective dose rates vary between 11.03 and 14.26 nSv (Erfelek – Center) in infants, between 15.44 and 19.97 nSv (Erfelek – Center) in children and between 32.19 and 41.65 nSv (Erfelek – Center) in adults. Average annual effective dose rates were calculated as 12.12 nSv for infants, 16.96 nSv for children and 35.38 nSv for adults. In a study conducted in Japan in 1993, it was shown that 52% of the tritium taken into the body by individuals originates from drinking water (Okada & Momoshima 1993). The distribution of the annual effective dose values calculated for infants, children and adults according to the sampling region is shown in Figure 6. When the results of this study are compared with a previous study conducted by Sudprasert *et al.* (2022), which reported that the average annual effective dose values due to the intake of the tritium in tap water samples in Bangkok were 10 nSv for infants, 14 nSv for children and 30 nSv for adults, it appears that our values are slightly higher. Gören *et al.* (2014) have reported average annual effective dose rates in some drinking water samples in Adana (Türkiye) as 24 nSv for infants, 34 nSv for children and 70 nSv for adults. The results of this study are lower than the values found by Gören. The annual effective dose rate allowed for the public by the IAEA and ICRP is 1 mSv (ICRP 2007; IAEA 2018). Also, it is recommended by WHO and EC that the annual effective dose to be taken by individuals with the consumption of drinking water for tritium radioisotope should not exceed 0.1 mSv (EC 2003; WHO 2011). In this study, the AED values found for infants, children and adults are considerably lower than these allowable dose limits. Therefore, it can be claimed that there is no risk associated with people consuming the drinking water that was the subject of this investigation in terms of tritium radioisotope.

Table 3 lists the estimated lifetime cancer risk values for people (males and females) who drank water samples collected from Sinop Province's center and districts. The lifetime cancer risk values determined for males and females varied from 1.27×10^{-7} to 1.64×10^{-7} and 1.36×10^{-7} to 1.76×10^{-7}, respectively. Whole LCR values of adults are found to be well below the permissible value (1.6×10^{-3}) for cancer risk as radiological (UNSCEAR 2000; Patra *et al.* 2013). Hereof, these findings demonstrate that there are no radiological risks associated with consuming the investigated waters. In addition, the findings of this study will be probably used as a reference in further studies.

Validation of measurement

The measurement outcomes of the reference sample are found as 710.81 ± 18.62 Bq/L. Experimentally determined tritium activity agrees with the spiked activity. Additionally, given that the Z value (0.19) ranges from -1 to 1, these results demonstrate that the current analytical procedure produced satisfactory results.

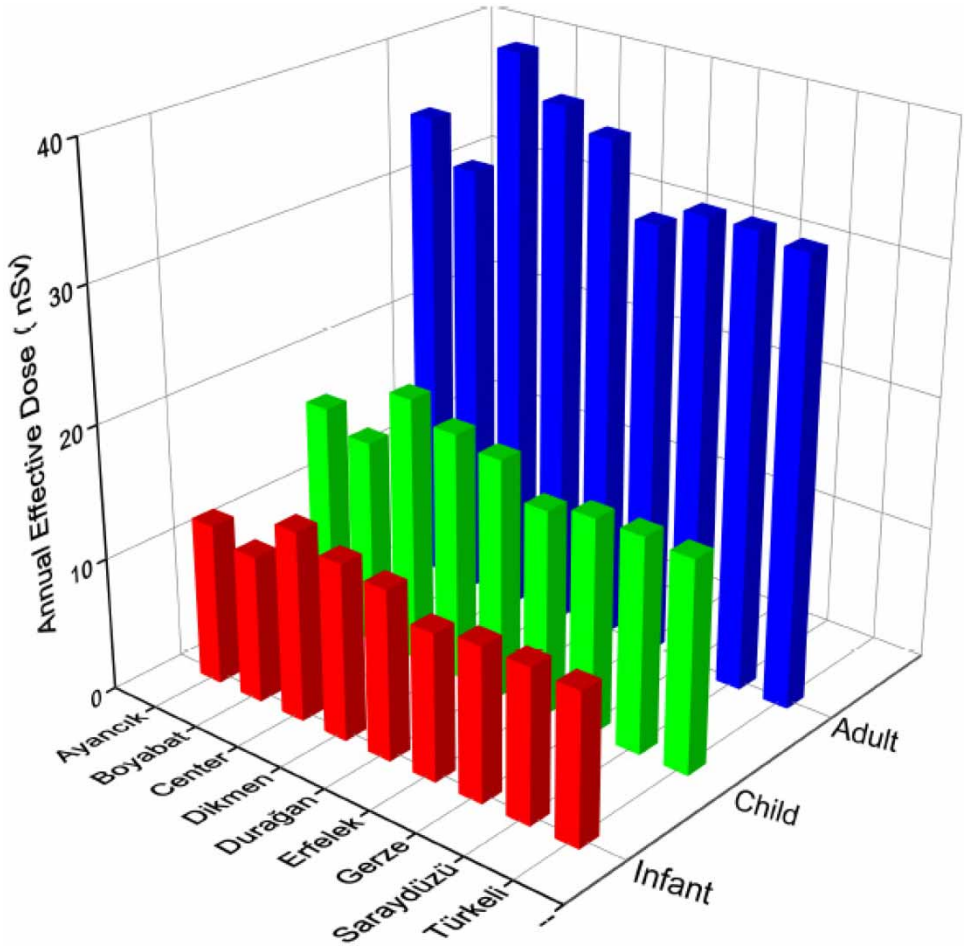


Figure 6 | The distribution of the annual effective dose values calculated for individuals.

Table 3 | Lifetime cancer risk values calculated for females and males consuming examined drinking waters

	Lifetime cancer risk ($\times 10^{-7}$)	
District	**Female**	**Male**
Ayancık	1.50	1.40
Boyabat	1.37	1.28
Center	1.76	1.64
Dikmen	1.64	1.53
Durağan	1.58	1.47
Erfelek	1.36	1.27
Gerze	1.43	1.33
Saraydüzü	1.43	1.33
Türkeli	1.41	1.31

CONCLUSIONS

In this study, the tritium contents of drinking and sea water samples taken from Sinop province were examined before Turkey's second nuclear power plant, which is planned to be established in Sinop province, went into operation. The tritium

contents in drinking and seawater samples were measured using Liquid Scintillation Counter (LSC). The average tritium contents of examined drinking and sea water samples were determined to be 22.81 and 21.78 TU, respectively. In addition, the annual effective dose rates for people consuming the examined drinking waters were estimated and these values were found to be 12.12, 16.96 and 35.38 nSv for infants, children and adults, respectively. The Turkish Standards Institution (TSI), the World Health Organization (WHO) and the European Commission (EC) control the maximum allowable level of tritium and radioactivity levels in drinking water. This work confirmed that the tritium levels in the drinking and seawater samples analyzed were low and much below the tritium exposure limits advised by international organizations. As a result, there is no tritium radiation risk associated with the tested water samples. In addition, the findings of this study can serve as a guide for future investigation.

ACKNOWLEDGEMENTS

This research was done as part of a master's thesis that was written in 2020 at Recep Tayyip Erdogan University.

AUTHOR CONTRIBUTIONS

S. D. conceptualized the whole article, conducted the formal analysis, developed the methodology, wrote the original draft, wrote the review and edited the article. F. Z. A. conducted the formal analysis and investigated the work, A. E. O. conducted the formal analysis, investigated the work, and visualized the project. R. K. conducted the formal analysis and investigated the work. F. K. G. conducted the formal analysis and investigated the work and rendered support in funding acquisition.

FUNDING

The Scientific and Technological Research Council of Türkiye (TUBITAK) funded this work under the project number '117Y285'.

DATA AVAILABILITY STATEMENT

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

CONFLICT OF INTEREST

The authors declare there is no conflict.

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First received 12 July 2023; accepted in revised form 24 November 2023. Available online 8 December 2023