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Article *in* Desalination and Water Treatment · June 2012

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Filiz Korkmaz Görür [a] & Erdem Genç [b]

[a] Abant Izzet Baysal University, Faculty of Sciences and Arts, Department of Physics, Bolu, 14280, Turkey Phone: Tel. +90 374 253 43 24 Fax: Tel. +90 374 253 43 24

[b] Rize University, Faculty of Sciences and Arts, Department of Physics, Rize, 53100, Turkey

Available online: 15 May 2012

To cite this article: Filiz Korkmaz Görür & Erdem Genç (2012): The tritium, deuterium and oxygen-18 isotope levels determination in various waters in Rize and Trabzon, Desalination and Water Treatment, 44:1-3, 215-222

To link to this article: http://dx.doi.org/10.1080/19443994.2012.691734

Desalination and Water Treatment
www.deswater.com
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doi: 10/5004/dwt.2012.3096

Taylor & Francis
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44 (2012) 215–222
June

The tritium, deuterium and oxygen-18 isotope levels determination in various waters in Rize and Trabzon

Filiz Korkmaz Görür[a,*], Erdem Genç[b]

[a]*Abant Izzet Baysal University, Faculty of Sciences and Arts, Department of Physics, Bolu 14280, Turkey*
Tel. +90 374 253 43 24; Fax: +90 374 253 46 42; email: filizkorkmaz@yahoo.com
[b]*Rize University, Faculty of Sciences and Arts, Department of Physics, Rize 53100, Turkey*

Received 08 July 2011; Accepted 06 November 2011

ABSTRACT

Rize and Trabzon are provinces at north-east site of Turkey, on the eastern Black Sea coast. There has been no information about isotopic $\delta^{18}O$, δD and tritium determination reported in water samples in the Rize and Trabzon provinces up to now. This study includes research about oxygen-18 ($\delta^{18}O$), deuterium (δD) and tritium (T) compositions of different water samples collected from tap, spring and mineral waters in Rize and Trabzon. The obtained results illustrated that activity concentrations of tritium in tap water samples did not exceed limits and recommendations of World Health Organization (WHO), United States Environmental Protection Agency (US EPA), Turkish Standards Institution (TSI) and Turkey Ministry of Health (MOH). The results of isotopic measurements show a little variation. Tritium value decreases from Rize to Trabzon, $\delta^{18}O$, δD and d-excess values increase from Rize to Trabzon. It shows Rize waters are younger than Trabzon waters and have shallow circulations. When δD and $\delta^{18}O$ values are examined, it's observed that tap, mineral and spring water samples of Rize and Trabzon are showed tendency to Mediterranean origin precipitations.

Keywords: $\delta^{18}O$; δD; Tritium; Rize; Trabzon

1. Introduction

There are lots of applications in hydrology and meteorology because of the importance of deuterium, tritium and oxygen-18.

Environmental isotope hydrologic techniques have proved to be effective tools for solving many hydrological problems [1] and in many cases, provide information that could not be obtained by any other conventional means [2–4]. They are very useful tracers to study the present and ancient hydrological processes and to understand surface and groundwater interconnections, the source and mechanism of recharge [5,6], groundwater circulation and its renewability [7,8], recharge areas and transit times of the aquifer [9,10], hydraulic inter-relationships [11,12] and source and mechanism of groundwater contamination [13,14]. Environmental isotopes now routinely contribute to more productive water investigations, complementing geochemistry and physical hydrogeology.

Tritium is a radioactive isotope that emits low-energy beta particles, with a maximum energy of 18.6 keV, and with a half-life of 12.6 y [15]. It is found in nature and it can also be produced by human activities. The natural origin is associated to the interaction of cosmic rays with the atmosphere. However, the environmental levels of this radionuclide were enhanced between 1945 and 1963 during nuclear weapon tests. Nowadays tritium is also produced in nuclear reactors that provide electricity, mainly of the pressurized water reactors type (PWR),

*Corresponding author.

as a by-product of the fission of uranium, which is a significant source of tritium in the environment [16]. As part of the water molecule ($^3HHO^1/^1H^3HO$), tritium perfectly follows water in atmospheric, oceanic, and hydrological transport and mixing processes [17].

Tritium concentration culminated in the atmospheric moisture of the Northern Hemisphere in 1963 [18]. It has also been released in large quantities from nuclear reprocessing facilities [19].

The activity of ^3H is expressed in tritium units (TU); one TU equals to one ^3H atom in 1018 ^1H atoms. The tritium activity in precipitation before the tests was of about 5 TU in central Europe. This number reached an average of about 5000 TU in northern Europe in 1963 [20].

Application of tritium to hydrologic problems was first proposed by Libby [21] and Bregmann and Libby [22]. Tritium analysis has been useful in many areas such as hydrogeology [23–25], nuclear industry monitoring [26,27], and for estimating the groundwater residence time [28,29] as it is directly incorporated into water molecule. Since the mid-eighties the tritium values in rain water have been 10 TU in the northern hemisphere [30], except for some local anthropogenic releases of tritium from the nuclear industry and other uses of tritiated materials.

Stable isotopes have been routinely used as valuable tools for estimating hydrogeological processes and characteristics. Especially, the stable oxygen and hydrogen isotopes have been used as ideal conservative tracers for identifying water sources because they constitute water molecules [2]. Therefore, the stable oxygen and hydrogen isotopes have been widely used to constrain hydrological cycles and to establish water budgets in various terrestrial environments. The analysis of stable isotopic composition of water has become an effective means for investigating the complex hydrologic system on a range of spatial and temporal scales. Because of the ubiquity of oxygen in the Earth system, the $\delta^{18}O$ value within each subsystem is controlled by a multitude of factors, with the end result of a final $\delta^{18}O$ value measured in the speleothem. As a result, the controls on $\delta^{18}O$ of the water before reaching the drip and cave are numerous and complex.

The main aim of this study is to determine the levels of tritium, deuterium and oxygen-18 isotopes in tap, spring and mineral waters. It was also aimed to obtain the information about their origin, circulation and nutrition and also to compare the results with other studies and standard limit values.

2. Experimental procedure

2.1. Sample collection and preparation

In order to measure the tritium, δD, $\delta^{18}O$ values in tap, mineral and spring waters samples were collected from 20 different sampling stations in Rize and Trabzon provinces (Fig. 1) by polyethylene bottles of 1 l capacity, preserved airtight using inner sealing cap in order to avoid evaporation and stored in a refrigerator at about 4°C until measurement. The bottles were cleaned up by using the modified procedure as given by Laxen and Harrison [31]. The samples for environmental ^3H were collected while the bore is still pumping according to the procedure given by Rosen et al. [32].

The tritium measurements ware carried out according to IAEA method by a means of Perkin Elmer QUANTULUS 1220 Ultra Low Level Scintillation Counting System (LSC). This spectrometer is specifically designed for determining very low activities. The detector has a multi-channel analyzer that separates tritium signals in the sample. The applied method consists of two major parts including: pre-concentration of tritium in the analyzed water sample using electrolytic enrichment and detection of tritium activity in the concentrated sample using liquid scintillation spectrometry.

To remove any impurity, to reduce quenching and to prevent the interference of other radionuclides which might adulterate the results, samples were distilled before they were measured by LSC. Distillation was performed by adding 0.05 N KMnO4 and 6 M NaOH to a volume of 500 ml of the water samples and spikes.

In second step, 250 ml of distilled water samples in each cell are electrolytically reduced to approximately 18 ml in about a week. A 1 g Na_2O_2 is added to each samples and spikes to provide electric conduction in water. Hence water samples and spikes turn into alkaline forms ($Na_2O_2 + H_2O \rightarrow 2\ NaOH + 1/2\ O_2$). To bring the weight of the sample from 250 g to about 18 g a total charge of 690 Ah is passed through each cell. After electrolysis, the cells were removed from the refrigerator unit, allowed to attain room temperature and then condensed water on the outer surfaces of the cells was removed.

Thirdly, the content of each cell was neutralized by adding 3 g PbCl2 to each of the 20 ml samples.

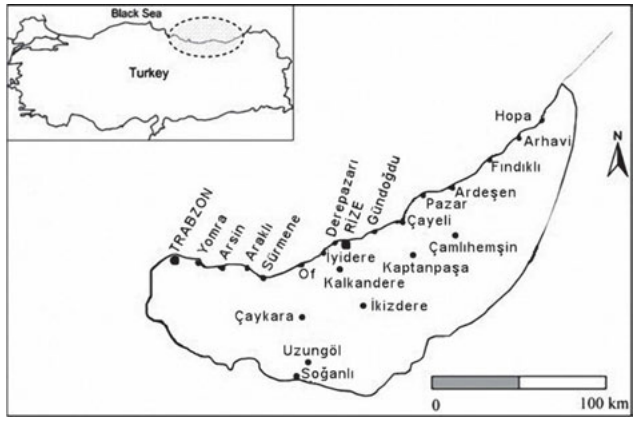

Fig. 1. Location of sample collection sites.

The sample was subsequently subjected to a final distillation to prevent salinity, which may produce heterogeneity in the sample-scintillator mix [33]. An aliquot of 8 ml from the second 18 ml fraction of the distillate was placed in a 20 ml polyethylene scintillation vial, and mixed with 12 ml of the liquid scintillation cocktail (Optiphase Hifase™ 3) supplied by Wallac from Turku, Finland. The first parameter evaluated was the scintillation cocktail. The influence of the cocktail on the efficiency of detection was investigated. To do so, we used a standard sample of tritium of known activity and a scintillator. To achieve maximum counting efficiency, it is vital importance that the mixture between the scintillation cocktail and sample is stable and homogeneous.

To prevent luminescence in the vial that contains the sample, it is recommended to store the samples for a minimum of 24 h in the dark before it is measured by LSC. Finally, dead water, laboratory standard and spike standard also were counted with the samples for about 600 min. Tritium activity concentration in the samples were obtained by using Eq. (1).

$$A_{TU} = \frac{N_{(SA)} \times A_{(Std)}}{N_{(Std)} \times Z} \times D \quad (1)$$

where A_{TU} is tritium activity of sample (TU), N_{SA} is net count rate of the sample (count per minute (cpm)), N_{Std} is net count rate of the standard (cpm), A_{Std} is activity concentration of the standard (TU), Z is tritium enrichment factor for the given sample and D is the factor taking into account decay of tritium in the sample from the date of measurement to the date of the sampling.

The oxygen isotopic composition was measured by using the Epstein and Mayeda [34] water – CO_2 equilibration technique with IRMS Dual Inlet. The carbon dioxide gas was extracted and cryogenically purified. Hydrogen isotope ratios were measured by using the zinc reduction method [35]. The samples collected from October 2010 to January 2011 underwent manual preparation. The analytical reproducibility is ±0.1‰ for $\delta^{18}O$ and ±1‰ for δD. The obtained isotopic values are reported usually in delta δ units (permil deviation of the isotope ratio from the international standard Vienna-standard Mean Ocean Water (VSMOW)), δ being defined by the following relationship:

$$\delta = \left[\left(R - R_{VSMOW} \right) / R_{VSMOW} \right] \times 10^3 \quad (2)$$

where R = $^{18}O/^{16}O$ or D/H. We also report the values of the deuterium excess that can be calculated as suggested by Craig [36] d-excess = $\delta D - 8\delta^{18}O$ on the basis of the world meteorological water line. The d-excess gives the measure of deviation of isotopic ratios of the samples from the international standard V-SMOW in per mil (‰) units.

3. Results and discussion

3.1. Tritium in water

Tritium levels in various water samples in Rize and Trabzon region are shown in Table 1.

The tritium concentrations in 17 tap water samples collected in Rize and Trabzon varied from 6.64 TU (Güneysu) to 9.70 TU (Çayeli) and the average tritium concentrations are 7.56 TU and 7.31 TU in Rize and Trabzon provinces, respectively. The tritium concentrations for spring and mineral waters have an average of 5.68 TU and 0.45 TU, respectively. These results are much lower than the upper levels determined by WHO (10,000 Bq/l) [37], US EPA (740 Bq/l) [38], TSI (100 Bq/l) [39] and MOH (100 Bq/l) [40]. Thus, the results indicate that all tap, spring and mineral water samples taken in Rize and Trabzon provinces are suitable for drinking.

The tritium level in young waters is in same level as in precipitation; as the water moves downward and laterally, the tritium concentration decreases with time. Therefore for Andon, Ridos and Güneysu waters it can be said that they have deep circulation.

The long turnover time of waters points to deep circulation of the recharging water through possibly a large geothermal reservoir. Tritium concentrations less than the detection limit of 0.3 TU [41] is interpreted as that water recharged before 1952. Tritium concentrations below 1 TU (negligible tritium content) were considered to indicate that water is at least 50 y old (recharged prior to the production of tritium from hydrogen bombs, which started in the early 1950s = pre-modern) and tritium values that equal to or were greater than 1 TU, were considered as modern water. Values of tritium of about 3 TU indicate a residence time of the water of about 30–40 y. The tritium values ranging from 1 to 8 TU could be attributed as an admixture of recent water with old water and water having been subjected to radioactive decay. Tritium values ranging from 9 to 18 TU could be attributed as recent water and tritium values ranging from 19 to 28 TU also could be attributed as thermonuclear water [42]. Taking this information into consideration collected water samples are divided into the four groups. Andon water was considered to indicate at least 50 y age (pre modern) and has a deeper circulation, Ridos water was considered to be a modern water and with a residence time about 30–40 y. Çayeli water was considered as recent water. Other groups of waters were considered as an admixture of recent water with old water and have shallow circulations.

Table 1
^{3}H, δD and δ^{18}O values in tap, mineral and spring waters of Rize and Trabzon

Water type	Locations	^{3}H (TU)	δ^{18}O	δD	d-excess
Tap water	Fındıklı	7.38 ± 0.14	–11.21 ± 0.32	–74.07 ± 0.55	15.61
	Ardeşen	7.47 ± 0.15	–11.40 ± 0.15	–73.70 ± 0.12	17.50
	Çamlıhemşin	7.35 ± 1.10	–11.03 ± 0.17	–70.74 ± 0.71	17.50
	Pazar	7.29 ± 0.14	–10.21 ± 0.13	–62.66 ± 0.72	19.02
	Hemşin	7.60 ± 1.10	–10.30 ± 0.30	–64.95 ± 0.74	17.45
	Çayeli	9.70 ± 1.25	–11.08 ± 0.18	–70.50 ± 1.09	18.14
	Rize	8.14 ± 0.15	–11.23 ± 0.16	–70.63 ± 1.09	19.21
	Derepazarı	6.66 ± 0.13	–11.53 ± 0.17	–70.71 ± 1.02	21.53
	İyidere	7.85 ± 1.10	–11.57 ± 0.01	–69.92 ± 0.15	22.64
	Kalkandere	7.45 ± 1.10	–10.74 ± 0.21	–70.10 ± 0.81	15.82
	İkizdere	7.20 ± 1.10	–12.47 ± 0.16	–82.21 ± 0.42	17.55
	Of	7.45 ± 1.10	–10.44 ± 0.07	–65.32 ± 0.66	18.20
	Sürmene	8.05 ± 1.15	–9.74 ± 0.30	–62.41 ± 0.55	15.51
	Araklı	6.65 ± 1.05	–10.05 ± 0.04	–65.32 ± 0.67	18.68
	Arsin	6.95 ± 1.05	–10.20 ± 0.13	–63.05 ± 0.61	18.55
	Yomra	7.45 ± 1.10	–10.46 ± 0.36	–63.35 ± 0.70	20.33
	Güneysu	6.64 ± 0.70	–11.04 ± 0.26	–72.96 ± 0.96	15.36
Mineral water	Andon	0.45 ± 0.65	–11.05 ± 0.28	–71.84 ± 0.58	16.56
Termal spring	Ayder	8.10 ± 1.15	–14.09 ± 0.23	–91.99 ± 0.33	20.73
	Ridos	3.25 ± 0.80	–12.37 ± 0.29	–98.50 ± 0.49	0.46

As shown in Table 2; tritium concentrations were comparable to other studies in various regions. The average activities obtained for tritium in tap waters are higher than observed activities for Spain country, in mineral waters lower than observed activities for Spain (Girona) country, in thermal spring waters higher than observed activities for West Syria, Morocco and Eastern India.

Table 2
Average ^{3}H activity values in different locations

Water type	Location	Average ^{3}H(TU)	References
Tap water	Spain	2.46	[50]
	Rize	7.56	Present study
	Trabzon	7.31	Present study
Mineral water	Spain (Girona)	<0.68	[51]
	Rize	0.45	Present study
Termal spring	West Syria (Figeh)	4.26	[52]
	Morocco	5.50	[53]
	Eastern India	1.23	[54]
	West Syria (Al-sin)	2.98	[52]
	Rize	5.68	Present study

3.2. Relationship between $\delta^{18}O$ and δD

δ^{18}O and δD compositions in various water samples in Rize and Trabzon regions are presented in Table 1.

The δ^{18}O and δD compositions of tap water samples collected in Rize and Trabzon varied from –12.47 (İkizdere) to –9.74 (Sürmene) and from –82.21 (İkizdere) to –62.41 (Sürmene). Average δ^{18}O and δD compositions in tap water samples are –11.15 and –10.18 and –71.10 and –63.89 and spring and mineral water samples –13.23 and –11.05, –95.25 and –71.84, respectively.

The isotopic composition of hydrogen and oxygen in water is expressed in units of δD and δ^{18}O, respectively. In precipitation these quantities follow in general the so called global meteoric water line equation: $\delta D = 8\delta^{18}O + 10$ [36].

The meteoric water line equation for the Eastern Mediterranean is estimated to be on average $\delta D = 8\delta^{18}O + 22‰$ [43]. The area of the Mediterranean basin is sensitive to climatic change [44].

Fig. 2 shows Global Meteoric Water Line (GMWL) and Mediterranean precipitation line (MMWL); δ^{18}O and δD values of the water samples between the lines or

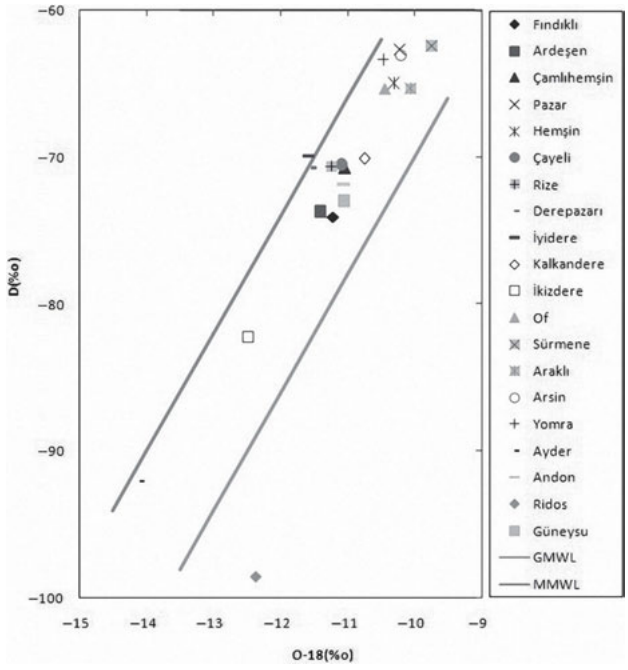

Fig. 2. Relationship between $\delta^{18}O$ and δD values of the water samples.

to Mediterranean precipitation line (especially Ayder, İyidere, Derepazarı and Yomra) can indicate the almost Mediterranean precipitation origin; Ridos sample outside of this range seems to be affected by evaporation.

Ayder, Ridos and İkizdere are high-altitude continental regions and their waters have lower isotopic compositions. Sürmene, Araklı, Arsin and Pazar waters collected along the seashore and have higher isotopic compositions. Generally, the stable isotopic compositions of precipitation decrease with decreasing temperature (temperature effect) and with increasing rainfall amount (amount effect). Yurtsever and Gat [45] pointed out that the temperature effect is generally pronounced in high-latitude continental regions, whereas the amount effect is pronounced in tropical regions. The average temperature is low at higher altitudes; the altitude effect, as a tendency for a systematic decrease of stable isotope composition with increasing altitude, is theoretically associated with the concept that this parameter is a function of cooling of air masses as they rise to higher elevation [45].

As shown in Table 3, $\delta^{18}O$ and δD values were comparable with other studies in various regions. The average values obtained for $\delta^{18}O$ and δD in tap waters are lower than United States; in thermal spring waters lower than West Syria, Morocco, Korea and Eastern India.

3.3. Deuterium excess

Deuterium excess values in various water samples in Rize and Trabzon region are shown in Table 1.

The d-excess values for tap water samples collected in Rize and Trabzon varied from 15.36 (Güneysu) to 22.64 (İyidere). The average d-excess values for tap water samples are 18.11 and 18.25 and for spring and mineral water samples 10.60 and 16.56.

On the global scale, the d value is close to 10 for meteoric waters [46] and it may vary from region to region. Vapor and condensation, produced under very low relative humidity and high evaporation will cause high d-values and similar low evaporation with low d values is likely to occur at high humidity [46]. Due to these reasons, for water samples of İyidere, Derepazarı and Ayder it can be said that there was high evaporation and for Ridos, Güneysu and Sürmene water samples low evaporation.

The d-excess values may be used for fingerprinting to particular sources, like high deuterium excess values correspond to the Mediterranean region and low values

Table 3
Average $\delta^{18}O$ and δD values in different locations

Water type	Location	Average $\delta^{18}O$	Average δD	Average d-excess	Reference
Tap water	United State	–8.16	–59.36	–	[55]
	Rize	–11.15	–71.10	18.11	Present study
	Trabzon	–10.18	–63.89	18.25	Present study
Mineral	Rize	–11.05	–71.84	16.56	Present study
Termal spring	Morocco	–6.86	–42.05	–	[53]
	Eastern India	–4.08	–28.04	–	[54]
	West Syria (Figeh)	–8.91	–50.90	20.10	[52]
	West Syria (Al-sin)	–6.49	–31.80	20.10	[52]
	Korea	–11.67	–78.00	12.11	[56]
	Rize	–13.23	–95.25	10.60	Present study

to high-latitude regions [47,48]. Therefore, it can be said that Ayder, İyidere, Derepazarı and Yomra waters are Mediterranean origin.

Andon and Ridos water has low tritium and d-excess results are consistent. The d-excess is inversely correlated with the electrical conductivity, indicating that the water with more negative value of d-excess tends to be saltier. Therefore, results reveal that these waters have more electrical conductivity and tend to be saltier.

Consistence of d-excess values indicates that the groundwater storage is significantly large and well mixed and the groundwater age is relatively old; therefore it can concluded that Andon and Ridos water have deep circulations.

As shown in Table 3, d-excess values were comparable with other studies for various regions. The average values obtained for d-excess in thermal spring waters are lower than West Syria (Figeh), West Syria (Al-sin) and Korea.

3.4. Relationship between $\delta^{18}O - {}^{3}H$ and ${}^{3}H - \delta D$

Generally, precipitation at higher latitudes would have more negative $\delta^{18}O$ and δD values [49]. As the water circulates long time, tritium concentration decreases in time.

High ${}^{3}H - \delta D$ values show that waters have shallow circulations and low ${}^{3}H - \delta D$ values show that waters have deep circulations.

According to relationship between the $\delta^{18}O - {}^{3}H$, ${}^{3}H - \delta D$ as shown in Fig. 3 and Fig. 4; Ayder, İkizdere and Ridos are fed on areas that are higher than the others.

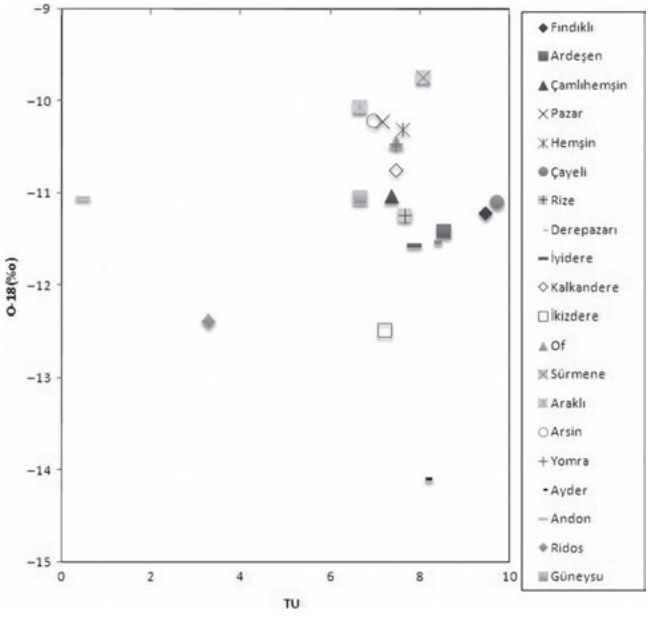

Fig. 3. Relationship between $\delta^{18}O$ and ${}^{3}H$ values of the water samples.

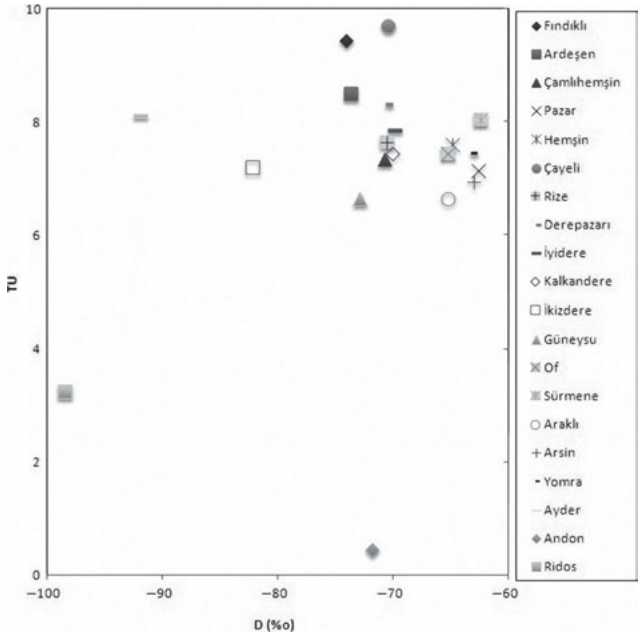

Fig. 4. Relationship between ${}^{3}H$ and δD values in the water samples.

The results of isotopic measurements show a little variation. Tritium value decreases from Rize to Trabzon, $\delta^{18}O$, δD and d-excess values increase from Rize to Trabzon. It shows Rize waters are younger than Trabzon waters and have shallow circulations.

4. Conclusions

This study has identified that tritium concentration in tap, spring and mineral waters in Rize and Trabzon provinces satisfying the WHO, USEPA, TSI and MOH regulations for drinking water. The present study, however, has been the first isotopic research in tap water in Rize and Trabzon and will make several noteworthy contributions to further researches to be done in this area.

Tendency to the Mediterranean origin precipitation is important topic and must be discussed. Because of quite limited research in isotopic field at East Black Sea Region, the more research about water isotopes will give the more information about waters origin, nutrition, circulations, ages and climates that clarify these comments.

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