ELSEVIER

Journal of
Environmental Radioactivity 43 (1999) 231–245

JOURNAL OF
ENVIRONMENTAL
RADIOACTIVITY

Oxygen-18, deuterium and tritium in the Black Sea and the Sea of Marmara

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Received 14 October 1997; revised 13 May 1998; accepted 19 May 1998

Abstract

The stable isotopes ^{18}O and ^{2}H, together with the transient tracer ^{3}H can be used to identify origins and characteristics of distinct water masses in the Black Sea, and in its smaller neighbour, the Marmara Sea, connected between them and to the Mediterranean Sea through the Straits of Bosphorus and Dardanelles, respectively. Exchange through the system and the water mass formation and evolution are characterised, based on the isotope measurements. Spatial coverage including the shelf and deep regions, with improved accuracy and reduced noise provide-definitive assessments of pycnocline and deep mixing in the system.

1. Introduction

The Black Sea is the largest land-locked marine basin in the world. A limited, two-layer stratified exchange takes place through the Turkish Straits System (the Dardanelles and Bosphorus Straits and the Sea of Marmara) providing connection to the Mediterranean Sea. The excess of fresh water from large rivers (e.g. the Danube) and excess precipitation relative to evaporation has a stabilising effect on stratification. Anoxic conditions exist in waters below the strong pycnocline, despite the supply of oxygen-rich Mediterranean water through the Bosphorus.

Ventilation in the upper pycnocline is driven by turbulent convection, assisting the formation of the cold intermediate water (CIW). On the other hand, ventilation in the lower pycnocline, down to a depth of 500 m, is dominated by cascading of the shelf-mixed Mediterranean water across the continental slope (Buesseler et al., 1991; Murray et al., 1991; Özsoy et al., 1991), enhanced by double diffusive convection

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0265-931X/99/$ – see front matter
PII: S0265-931X(98)00094-0

(Özsoy et al., 1993; Özsoy & Beşiktepe, 1995; Özsoy & Ünlüata, 1997, 1998). The importance of this ventilation mechanism is evident from the rapid transfer to greater depths of post-Chernobyl radioisotopes (Buesseler & Livingston, 1997), consistent with the results of earlier ^{14}C (Östlund, 1974; Östlund et al., 1986) and tritium measurements (Top et al., 1991). The residence time of deep waters increases with depth, to about 1000 years at 500 m, reaching 2000 years near the bottom (Östlund et al., 1986).

The surface waters of the Marmara Sea are transitional in character (Ünlüata et al., 1990), with a mean residence time of a few months, while its relatively uniform deep waters have a residence time of 6 years, influenced by seasonal intrusions of Mediterranean water from the Dardanelles Strait (Beşiktepe et al. 1993, 1994), renewing the western basin bottom waters in winter, and mid-depth waters during the warmer part of the year.

Radiotracers have increasingly been used for investigating mixing and transport in the Black Sea in the last decade (e.g. İzdar and Murray 1991; Murray, 1991; Fabry & Fröhlich, 1992). Our analyses are based on simultaneous measurements of stable isotopes and tritium in the Black Sea and the Marmara Sea regions (Özsoy et al., 1997). The results provide additional support for some of the above hypotheses on mixing.

Oxygen ^{18}O and deuterium ^{2}H are stable isotopes freely existing in nature. Their variations in a given medium result from the hydrological cycle of water, subject to repeated evaporation and condensation. With the heavy isotopes retained more easily in surface waters following evaporation, the isotopic ratios in precipitation water ideally approach the 'Meteoric Water Line', i.e. a linear relationship between the two isotopes (e.g. IAEA, 1992). The stable isotope levels in marine waters are considerably different from those in riverine and precipitation waters (the reservoir effect), displacing the oxygen–deuterium relationship away from meteoric characteristics. Stable isotope measurements in the Black Sea (Swart, 1991a, b) emphasise hydrological contributions and the relative roles of surface sources to the existing stratification.

On the other hand, tritium, a radioactive element, is characterized by a half-life of 12.4 years with measurable levels in the hydrosphere resulting from nuclear weapons tests in the late 1950s reaching a maximum in 1963–64, and no apreciable signature from Chernobyl. It is therefore used as a transient tracer for following water masses. Its removal in the surface marine environment appears to proceed faster than its decay rate, as a result of mixing.

2. Measurements and methods

The sea water samples at oceanographic stations (Fig. 1) were collected by the R/V BİLİM of the IMS-METU in March–April 1995, and analysed to determine concentrations of ^{18}O, ^{2}H and ^{3}H isotopes. Of the 11 stations in the Black Sea, six were sampled down to a depth of 150 m, four stations extended to 250 m, and deep samples down to 1500 m could only be obtained at a single station. The three stations in the Marmara Sea extended to 1200 m.

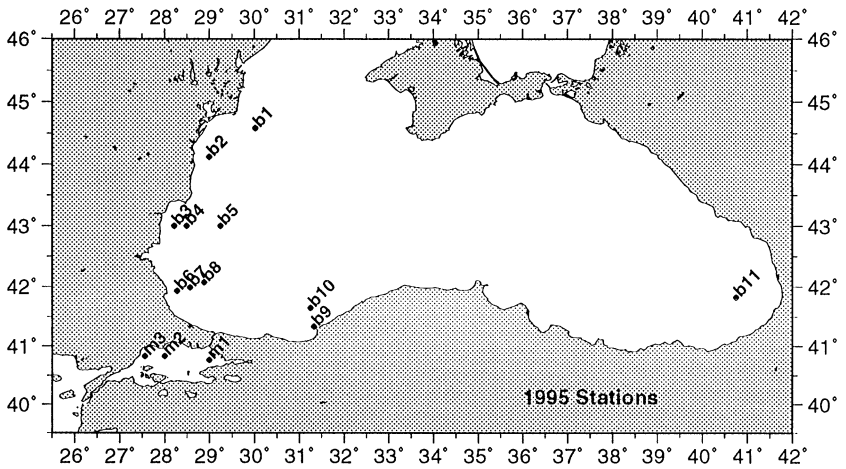

Fig. 1. Black Sea and Marmara Sea stations in 1995.

The laboratory analyses of stable isotopes were carried out by mass spectrometry (^{18}O measured on MAT 250, precision $< \pm 0.01\%$ delta SMOW units, H_2O and CO_2 equilibration during sample preparation, and ^{2}H on MAT 251, precision $< \pm 0.1\%$, H_2O and H_2 equilibration, with Pt as catalyst). In ^{3}H measurements, liquid scintillation without enrichment was carried out on ALOKA LB3 (70 ml sample in 140 ml vials), with a precision of ± 0.7TU. Liquid scintillation with electrolytical enrichment was either carried out on a Packard Tricarb instrument (250 ml sample, 20 ml vials) with a precision of ± 0.2TU, or on an ALOKA LB3 instrument (800 ml sample, 140 ml vials) with a precision of ± 0.1TU, depending on the size of the water sample.

3. Tracer history as a function of inputs

The levels of stable isotopes in surface waters show rather small seasonal and interannual variations. For example, the Danube water in Vienna (Rank, 1992, 1995, 1996) has a $\delta^{18}O$ value of about $-11.5‰$, and a range from -10.3 to $-13.3‰$, with the minimum corresponding to the Alpine snowmelt period. The average value near the mouth of the Danube is about $-10.5‰$. The average values within the upper 20 m waters of the two seas, given in Table 1, reflect differences in riverine and atmospheric sources, and in mixing with the underlying waters in the two regions. We note limited variability in these values, in comparison to other measurements in 1988 and 1993 (Swart, 1991; Özsoy et al., 1997).

In contrast, the tritium in Danube waters (Rank, 1992, 1995, 1996) (Fig. 2) and in precipitation (not shown) (Rank, 1992, 1995, 1996) indicates a transient decrease following the 1963/64 maximum, in agreement with other observations elsewhere (e.g. IAEA, 1992). In Table 2, a compilation of past measurements is used to calculate

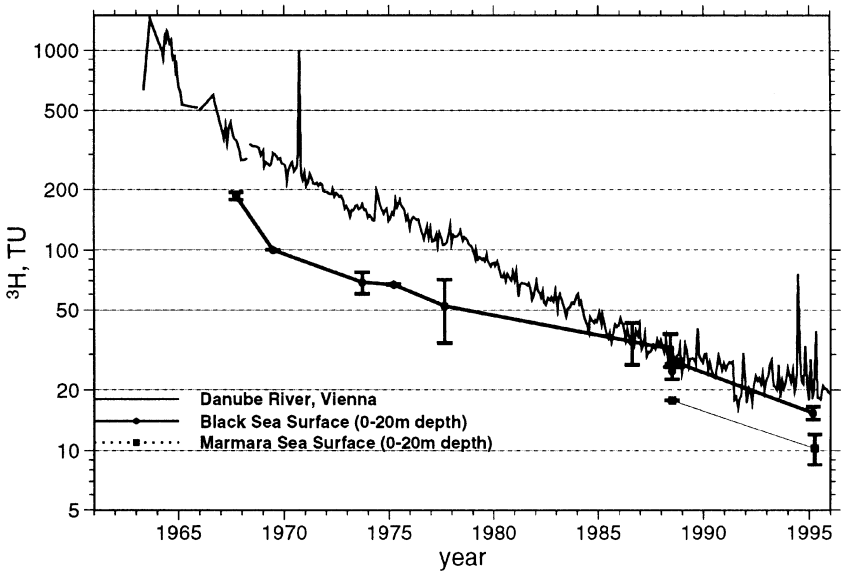

Fig. 2. Surface ^{3}H concentrations measured in the Black Sea and Marmara Sea surface layers (0–20 m depth), superimposed on a plot of ^{3}H measurements in the Danube River. Danube samples were obtained at Reichsbruecke (before Dec 1991), and Nussdorf-Schleuse (after Dec 1991) in Vienna.

Table 1
Measurements of δ^{18}O and δ^2H in surface waters (0–20 m depth) of the Black Sea and Marmara Sea

Area	Year	Month	δ^{18}O		δ^2H		Number of observations
			Avg	σ	Avg	σ	
Black Sea	1995	2	− 2.84	0.34	− 23.03	2.17	24
Marmara Sea	1995	3	− 0.93	0.48	− 5.50	8.03	8

average surface concentrations in the Black Sea and the Sea of Marmara, confirming a similar decrease in marine waters since the earliest measurements in 1967, as shown in Fig. 2.

The present input to the Black Sea is about 10–15 TU from precipitation (1995) and 20–25 TU from Danube water. This is at present of the same order of magnitude as the ^{3}H of Black Sea surface waters (15–17 TU). The comparison of riverine sources and surface marine observations indicate this difference to be larger in the 1960s and 1970s (Fig. 2), when the inputs were considerably larger. These transient features suggest vertical redistribution of tritium by mixing across the main pycnocline, starting from initially uncontaminated conditions, although this is not easy to establish with confidence from the earlier measurements, which lack sufficient resolution and accuracy in intermediate and deep waters.

Table 2
Measurements of ^3H in surface waters (0–20 m depth) of the Black Sea and Marmara Sea

Area	Year	Month	^3H (TU)		Number of obs.	Source
			Avg	σ		
Black Sea	1967	9	185.0	8.0	1	D. Rank, Mamaia coast
Black Sea	1969	–	100.0	–	1	Östlund (1974)
Black Sea	1973	9	68.9	8.6	23	Nikitin and Vakulovsky (1996)
Black Sea	1975	4	67.0	0.4	1	Top and Clarke (1983)
Black Sea	1977	8	52.6	18.4	21	Nikitin and Vakulovsky (1996)
Black Sea	1986	7	34.8	8.3	29	Nikitin and Vakulovsky (1996)
Black Sea	1988	5	32.0	6.0	30	Nikitin and Vakulovsky (1996)
Black Sea	1988	6	24.8	2.2	7	Z. Top, personal comm.
Black Sea	1988	9	27.2	1.3	1	D. Rank, Mamaia coast
Black Sea	1995	2	15.4	1.1	24	present measurements
Marmara S.	1988	6	17.75	–	1	Z. Top, personal comm.
Marmara S.	1995	3	10.26	1.78	8	Present measurements

Table 3
Measurements of $\delta^{18}O$ and δ^2H in deep waters of the Black Sea (depth > 500 m) and Marmara Sea (depth > 50 m)

Area	Year	Month	$\delta^{18}O$		δ^2H		Number of obs.
			Avg	σ	Avg	σ	
Black Sea	1995	3	− 1.77	0.03	− 15.87	0.28	7
Marmara Sea	1995	3	1.58	0.03	10.26	0.69	30

In the case of Marmara Sea waters, there is a larger difference in surface tritium concentrations with respect to the input functions. Despite the smaller size of the Marmara Sea, mixing with the underlying Mediterranean waters yields a further dilution in the surface waters. The main part of this mixing is believed to occur at the Bosphorus Strait and its exit region in the Marmara Sea (e.g. Ünlüata et al., 1990; Beşiktepe et al., 1994).

In deep water below the pycnocline, the stable isotopes in both seas are relatively constant. Average properties below a depth of 500 m are listed in Table 3, showing the different hydrological origins of these waters.

In comparing deep concentrations of tritium, it is necessary to exercise some care, since the modern measurements show very low levels in the Black Sea, and outliers, mainly occurring in the old data are not reasonably connected with reality, and are most likely to be the result of contamination of samples. Therefore, in Table 4, we only compare average values from Top and Clarke (1983) and those from Leg 4 of the 1988 R/V Knorr cruise (Top et al., 1991; Top, 1999), with the results of the present study. The 1975 measurements of Top and Clarke (1983) did show slightly higher values near the bottom, which could not be reproduced in recent measurements in

Table 4
Measurements of ^{3}H in deep waters of the Black Sea (depth > 500 m) and the Marmara Sea (depth > 50 m)

Area	Year	Month	^{3}H (*TU*)		Number of obs.	Source
			Avg	σ		
Black Sea	1975	4	0.22	0.50	5	Top and Clarke (1983)
Black Sea	1988	6	0.24	0.28	60	Z. Top, pers. comm.
Black Sea	1995	3	0.36	0.51	5	Present measurements
Marmara S.	1988	6	3.82	0.52	6	Z. Top, pers. comm.
Marmara S.	1995	3	3.12	0.60	30	Present measurements

1988 and 1995. On the other hand, the measurements of Nikitin and Vakulovsky (1996) have unacceptably high values and a large variance in deep water (Özsoy et al., 1997), and therefore they were not taken into consideration. The deep measurements in the Black Sea are rather scarce, and the differences between mean concentrations in Table 4 are not statistically significant in view of the variance associated with the physical distribution in deep water (Top, this volume) and with those of the analysis methods.

In the Marmara Sea, the difference between the Top et al. (1991) tritium measurements in 1988 and the present measurements (Table 4) is not accountable for by the decay process alone (a decay factor of 0.7 in 6.7 years between the 1988 and 1997 measurements), if one assumes a constant Mediterranean influence. The measured value in 1995 is considerably higher than the estimation based on decay. This confirms that the renewal of deep waters by inflowing Mediterranean water has a transient effect opposite to decay within a period comparable to the average age of Marmara Sea deep water.

4. Distribution of stable isotopes and tritium

4.1. Stable isotopes — oxygen and deuterium

Stable isotope measurements basically reflect the mixing of two main water masses in the regions of interest, i.e. the Mediterranean waters, with a positive anomaly characterising it with 'concentration basin' characteristics, versus the surface waters of the Black Sea, reflecting 'dilution basin' characteristics.

In Fig. 3, the profiles of the stable isotopes δ^{18}O and δ^2H in both seas have well defined vertical structures, displaying sharp contrasts and rapid increases across the pycnocline. Relatively constant levels exist in the deep waters of either sea.

In the Black Sea, a smooth transition occurs across the the pycnocline, between the surface and deep levels. Excluding vertical displacements of the pycnocline, the only sharp deviation from the common pattern occurs near the surface, where the freshwater influence from major rivers (Danube, Dniepr & Dniestr) in the northwestern shelf region can clearly be identified and followed along the western Black Sea

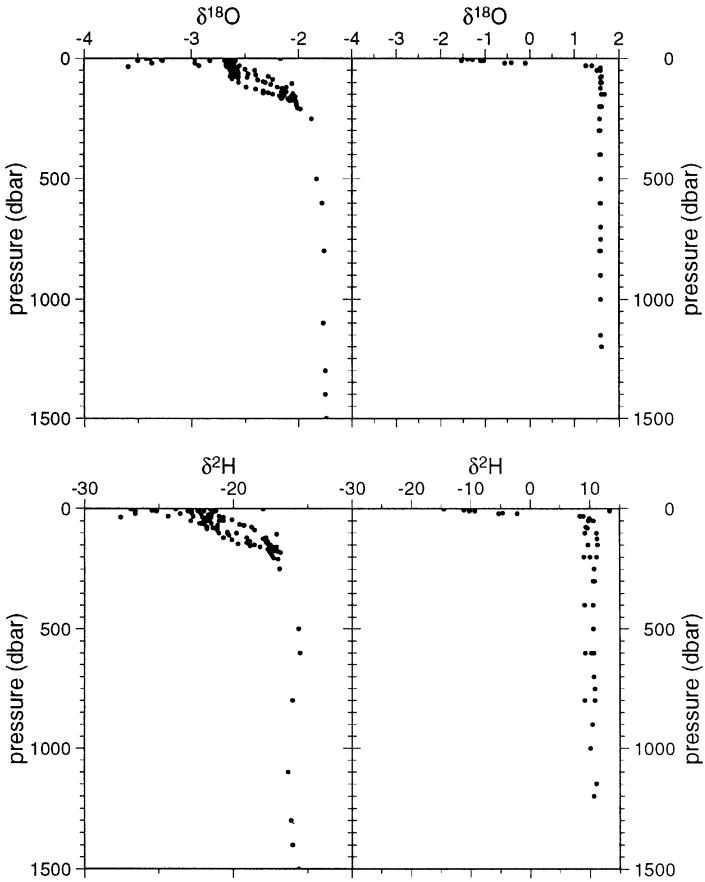

Fig. 3. Vertical distribution of stable isotopes in 1995; left: Black Sea; right: Marmara Sea; top: $\delta^{18}O$ measurements; bottom: $\delta^{2}H$ measurements.

continental shelf, with decreasing $\delta^{18}O$ and $\delta^{2}H$ values near the river mouths (Özsoy et al., 1997).

There are large horizontal and vertical variations within the thin (20 m) upper layer of the Marmara Sea, as a result of rapid entrainment and mixing between the Black Sea and Mediterranean waters in this small, confined domain. The deep water of Mediterranean origin, underlying the thin surface layer, has uniform concentrations, in agreement with its physical properties (Beşiktepe et al., 1996). On the other hand, large horizontal and vertical gradients occur within the surface layer, as a result of mixing across the pycnocline, as the upper layer flow passes through. We also observe that the sub-halocline (Mediterranean water) $\delta^{2}H$ variance is slightly larger than the $\delta^{18}O$ variance, in contrast with the eastern Mediterranean (Gat et al., 1996), interpreted as successive evaporation and precipitation in concentration basins such as the Mediterranean. The reverse situation in the Marmara Sea within a small range, could

be a result of modifications of Mediterranean water in the Aegean Sea, or simply of less accuracy in deuterium measurements relative to oxygen.

Although direct comparisons with historical data on stable isotopes could not be made, similar patterns of distribution are evident in the measurements of Swart (1991a, b), and Gat et al. (1996), though our measurements appear to have much less noise.

4.2. Tritium

The tritium distributions in the Black Sea and the Marmara Sea are displayed in Fig. 4, where error bars for individual measurements indicate the accuracy of the measurements.

Again, large gradients in tritium occur across the pycnocline in both seas. In the Black Sea, there is a region below the pycnocline, where the tritium decreases rapidly below 500 m to very low values near the bottom. Although we only have data from one deep station in 1995, it is evident that the concentrations decrease to almost zero in deep water, within the precision of the measurements. In the Marmara Sea only very small variations typical of the efficiently ventilated Mediterranean region occur below a depth of 25 m.

The tritium distribution in the lower layer of the Marmara Sea has a maximum at mid-depths and possibly a slight increase near the bottom. These slight variations in the small basin indicate local changes in relative age, depending on seasonal, transient renewals by intrusions of Mediterranean water (Beşiktepe et al., 1993, 1994) which are also evident in the salinity and temperature fine structures. We observe that δ^2H and ^{3}H have slightly larger variability compared to δ^{18}O in the subhalocline waters. Our ^{3}H measurements appear to have less scatter than the earlier measurements, and clearly demonstrate physical variations.

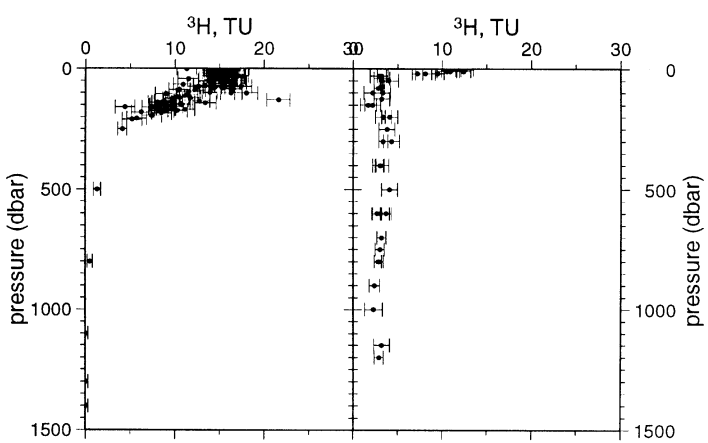

Fig. 4. Depth profiles of ^{3}H measurements, 1995. left: Black Sea; right: Marmara Sea; error bars show 1σ resulting from measurement uncertainty.

4.3. Density dependence

In the strongly stratified environments of the Black Sea and Marmara Sea, the pycnocline limits mixing between the surface and deep waters. Because the source functions for hydrology and chemistry are applied near the surface, the diffusion of scalar properties across the pycnocline by turbulent diffusion processes, rapidly attenuated with depth, results in similar profiles of scalar variables (Tugrul et al., 1992; Özsoy & Ünlüata, 1997). To remove the effects of vertical displacements associated with the dynamics, stable isotopes and tritium are plotted with respect to σ_θ density in Fig. 5.

The almost perfect linear correlation of stable isotopes versus σ_θ density in Fig. 5 suggests that each water mass in the Black Sea with a stable isotope characterisation of atmospheric contact can be assigned a proportional signature in density (i.e. temperature and salinity). This implies that the water mass evolution has followed the history of contact with the atmosphere. This is an independent verification of a hypothesis of deep water mass formation by a mixture of Mediterranean and near-surface, relatively fresh water masses, based on an interpretation of temperature–salinity relationships (Özsoy et al., 1991; Özsoy & Beşiktepe, 1995), and the late quaternary history of the Black Sea (Stanley & Blanpied, 1980; Boudreau & Leblond, 1989). It is also verified that the pycnocline displacements in Fig. 3 are linked with dynamical effects, eliminated as a consequence of the collapse to linear correlation in Fig. 5.

In contrast, the transient tracer tritium versus σ_θ density in Fig. 5, indicates a significant change in tritium gradient with respect to density below the pycnocline. In particularly the fact that the break appears in the transient tracer and not in the stable isotopes shows that bomb tritium from the atmosphere has not yet been able to penetrate into the deeper Black Sea, and that the efficiency of penetration sharply decreases below the pycnocline, at $\sigma_\theta \simeq 16.3$. This result must reflect different mechanisms of renewal for intermediate and deep waters, as explored in a number of earlier interpretations (Özsoy et al., 1991; Özsoy & Beşiktepe, 1995), i.e. turbulent mixing

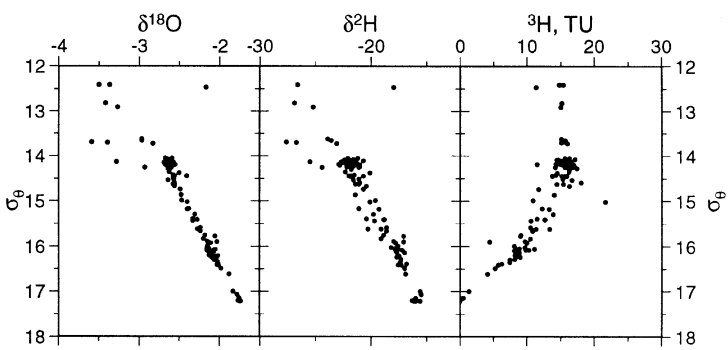

Fig. 5. Profiles of stable isotopes and tritium as a function of σ_θ density in the Black Sea. left: $\delta^{18}O$; center: δ^2H; right: ^3H

above the pycnocline, and rapidly diminishing turbulent mixing replaced with lateral intrusions of Mediterranean water below the pycnocline, extending down to a depth of about 500 m or $\sigma_\theta \simeq 17$. Indeed, below 500 m, there is very little tritium.

4.4. Basin characteristics

All three isotopes in the Black Sea and the Sea of Marmara are well correlated with water mass characteristics, decreasing rapidly below the pycnocline. The basin characteristics shown schematically in Fig. 6, are based on the average isotope levels measured near the surface in both seas, and those measured below a depth of 500 m in the Black Sea and 50 m in the Marmara Sea.

The fresh water inflow, specifically from the Danube river, entering the sea with a low $\delta^{18}O$, has a significant influence in the northwestern shelf region, where it lowers the $\delta^{18}O$ which is typically slightly higher in the surface waters elsewhere. Enrichment of ^{18}O at the surface of the Black Sea occurs by evaporation and as a result of mixing with the underlying waters. Tritium in the surface layer of the Black Sea is high as a result of the atmospheric supply. The deep water of the Black Sea, with a historical Mediterranean contribution, is enriched in terms of $\delta^{18}O$. In contrast, the recent source of tritium at the surface has not been able to penetrate into the deep waters, the ^{3}H values decreasing rapidly below a depth of 500 m. In the Marmara Sea, the ^{18}O and ^{2}H characteristics of surface waters evolve rapidly at the transitions across the Bosphorus and Dardanelles Straits, and from west to east by mixing with underlying Mediterranean waters. The subhalocline Marmara waters essentially bear the same signature as the Mediterranean waters.

4.5. Isotopic correlations and mixing

The relationships between stable isotopes and tritium reveal specific characteristics of the water masses in the region.

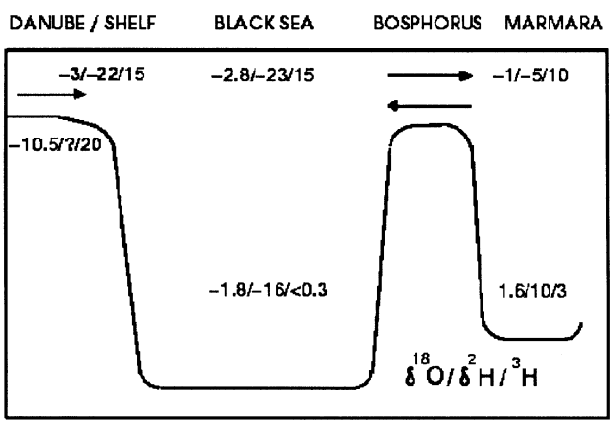

Fig. 6. Idealised surface (depth 0–20 m) and bottom (depth $>$ 500 m in the Black Sea, and depth $>$ 50 m in the Marmara Sea) concentrations of isotopes, $\delta^{18}O/\delta^{2}H/^{3}H$, in the Danube river (Vienna), the northwestern shelf area, the greater part of the Black Sea, and the Marmara Sea.

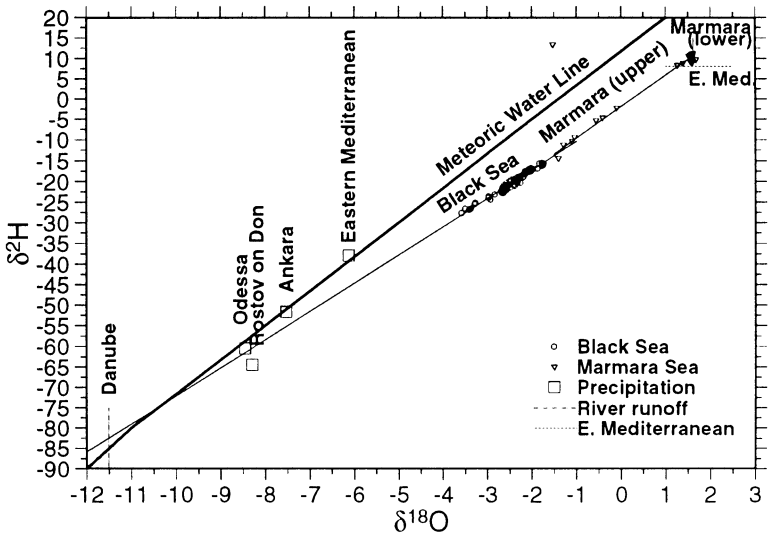

Fig. 7. $\delta^{18}O$ versus $\delta^{2}H$ in the Black Sea and Marmara Sea, compared to the sources. The Danube $\delta^{18}O$ measurements are after Rank (1995, 1996), the precipitation measurements at stations are after IAEA (1992) and the average values of Eastern Mediterranean precipitation measurements is after Gat et al. (1996). The typical Eastern Mediterranean range of stable isotope measurements represented by a dashed line are after Gat et al. (1996).

Fig. 7 establishes the $\delta^{18}O$ versus $\delta^{2}H$ relationship, in comparison to the meteoric water line, the inputs from rivers and precipitation. A least squares fit to the Black Sea data intersects the meteoric water line in the lower left hand side of the figure, and joins with Marmara Sea waters with a specific isotope signature at the other end. A continuous alignment of the data points between fresh water and Mediterranean water characteristics suggests that these waters were formed by a continuous mixture of fresh water inputs and Mediterranean water, as we could also verify through the density plots of Fig. 5.

Mediterranean water in the Marmara Sea has slightly higher $\delta^{2}H$, compared to the Eastern Mediterranean (Gat et al., 1996). The larger variation in $\delta^{2}H$ as compared to $\delta^{18}O$ in the lower layer of the Marmara Sea, contrasting with the results of Gat et al. (1996), could be the result of local characteristics or measurement accuracy.

Fig. 8 displays the main results of the present study, correlating all three isotopes studied. In the upper plot, a slightly enlarged version of Fig. 7, we observe that all the data in the two interconnected basins lie along the same straight line, with a minor change in slope.

In the oxygen versus tritium plot (lower part of Fig. 8), Marmara and Black Sea data are located along different branches, as a result of differences in residence time of the deep waters in the two basins. The transient tracer tritium indicates age, while the oxygen isotope is linearly related to stratification, and measures distance from the conditions determined by surface hydrology. The near-surface waters in the Black Sea form a cluster of rather uniform values; while a group of points displaced to the left from this cluster, and belonging to the northwestern shelf region is helpful in identifying

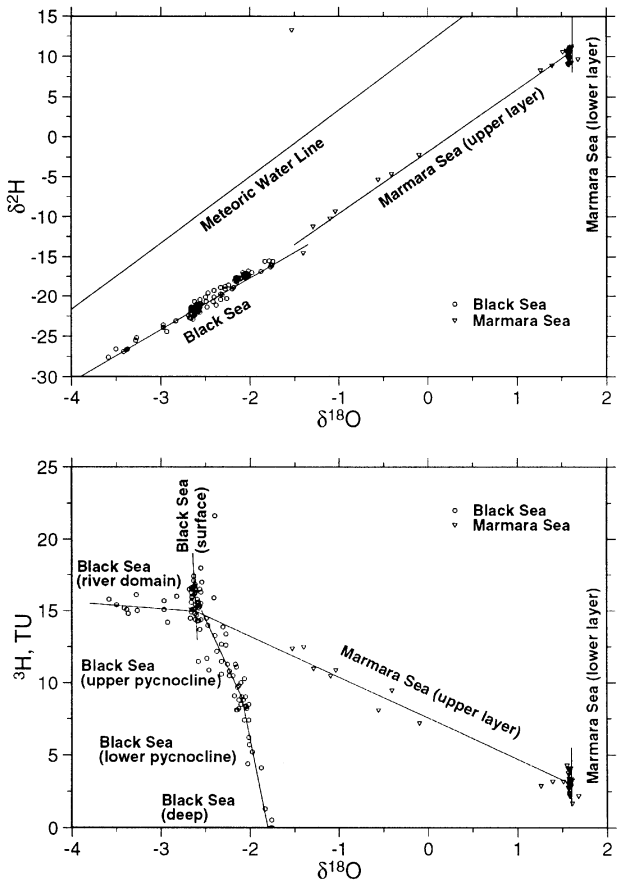

Fig. 8. Top: $\delta^{18}O$ versus $\delta^{2}H$, bottom: $\delta^{18}O$ versus ^{3}H for the Black Sea and Marmara Sea, 1995.

riverine effects. Fresh water is tagged by the oxygen isotope, but not by tritium, which is similar in all surface samples. Below the surface, tritium decreases and oxygen increases with depth. In the Black Sea, we observe that there is a well defined change of slope occurring immediately below the pycnocline, clearly showing that two different mechanisms are responsible for the renewal of the water masses above and below it.

In the Marmara Sea, the linear relationship (Fig. 8) has constant slope above the pycnocline, while there is greater variation of ^{3}H but relatively constant $\delta^{18}O$ in the Mediterranean water below the pycnocline.

5. Conclusions

The use of stable isotopes together with transient tracers in the Black Sea and Marmara Sea has yielded new results which help to understand the mixing and renewal in these semi-enclosed basins of greatly differing characteristics.

Stable isotope measurements in precipitation and river runoff, characterising fresh waters and the atmosphere, are relatively constant in time, and their ratios help identify mixing between waters with different origins. In the region studied, there is nearly a linear correlation between oxygen and deuterium for all water samples, showing that the waters in both basins are continuously proportional mixtures of surface fresh water inputs to the Black Sea and the salty waters of the Mediterranean.

In the Marmara Sea, stable isotopes at the surface evolve rapidly at the transitions across the Bosphorus and Dardanelles Straits, and in an east to west direction in the Marmara Sea interior by mixing with the lower layer Mediterranean waters. Both isotopes show very little change below 500 m in the Black Sea, and below 25 m in the Marmara Sea, consistent with hydrographic observations, showing common sources.

On the other hand, tritium is decreasing in precipitation, river runoff and the surficial waters of the Black Sea and Marmara Sea, consistent with fallout and decay. Mixing with underlying waters determine the delayed response of the Black Sea and Marmara Sea surface waters, which differ in their settled characteristics. The surface and deep waters in both Seas are separated by a sharp pycnocline identified in all three tracers. Stable isotopes have a continuous linear dependence on density, while tritium shows a discontinuous dependence indicating different renewal mechanisms for deep and intermediate waters.

River (freshwater) influence is identified along the western Black Sea continental shelf, on the basis of isotope correlations. Tritium measurements show a rapid decrease across the Black Sea pycnocline, and constant levels below the pycnocline in the Marmara Sea. Comparison of tritium with stable isotopes can characterise all distinct water masses in the two basins.

Acknowledgements

This paper is a result of cooperation, under project no. 302-K4-TUR-7333, part of a Coordinated Research Programme (CRP) of the International Atomic Energy Agency, entitled 'The Application of Tracer Techniques in the Study of Processes and Pollution in the Black Sea'. We were greatly encouraged by K. Fröhlich, and M. S. Baxter of the IAEA for our contribution to the Programme. We thank the Turkish Scientific and Technical Research Council and the NATO Science for Stability Program for funding research in the Black Sea, which enabled us to carry out the work. We are also grateful to Zafer Top of the University of Miami, for providing his 1988 data for comparative evaluations, and for his help with the manuscript. Many other individuals, including the scientists of the IMS-METU, and the captain and crew of the R/V BİLİM, have made valuable contributions during the course of the study.

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