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Pycnocline and Deep Mixing in the Black Sea: Stable Isotope and Transient Tracer Measurements

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The peculiar mixing processes at the pycnocline and deep regions of the Black Sea are reviewed. In addition to the wind stirring and convective mixing, active in the upper pycnocline, the other important mechanism that results in limited ventilation of the anoxic waters of the Black Sea is the Mediterranean dense water inflow from the Bosphorus, modified by the entrainment of surface and intermediate waters, introduced into the interior through double-diffusive intrusions. This inflow, aided by the surface Ekman flux divergence, boundary processes and internal wave breaking, is the main mechanism for the mixing and renewal of the sub-pycnocline waters in the Black Sea interior. A review of these mixing processes is complemented by results from isotope measurements with improved accuracy and reduced noise compared to earlier experiments. Measurements of the stable isotopes oxygen-18 (^{18}O) and deuterium (^{2}H) confirm the origin of the water masses in the Black Sea and in the Turkish Straits, including the sea of Marmara, to be a continuous mixture with variable fractions of salty Mediterranean waters with inflowing fresh waters. Tritium (^{3}H) measurements confirm very little penetration of the transient signal to the sub-pycnocline and deep waters of the Black Sea, in comparison to the better ventilated Mediterranean waters filling the lower layer of the Marmara Sea. The comparison of stable and transient tracer isotopes shows the effects of fresh waters originating from the north-western shelf, and the difference between the renewal mechanisms of the Marmara and Black seas, as well as those between the upper and lower pycnocline of the Black Sea.

Keywords: mixing; connection; double diffusion; isotopes; tracers; ventilation; Black Sea

Introduction

The Black Sea (see recent reviews by Özsoy & Ünlüata, 1997, 1998) is the largest land-locked basin in the world with a positive surface water budget, located in a region that is sensitive to climatic contrasts and changes (Özsoy, 1999). Its effective isolation, despite the existence of the two-way exchange through the Turkish Straits (Oğuz *et al.*, 1990; Gregg & Özsoy, 1999; Gregg *et al.*, 1999), has led to stagnant conditions and anoxia in its deep waters; the estimated average age of water increasing to more than two thousand years near the bottom (Östlund, 1974; Grasshoff, 1975). The warm and saline Mediterranean water is the historical source of the Black Sea deep waters, evolving since the end of the last ice age (Stanley & Blanpied, 1980) or possibly in the last 7200 years, when the Black Sea ' lake ' was abruptly flooded (Ryan *et al.*, 1998).

In the surface waters above the main pycnocline of the Black Sea, cooling, convective motions, wind mixing, advection by currents and surface waves create turbulent mixing characterized by a wide range of turbulence scales. Because of the low level of tidal forcing, the only principal agents to create turbulence at the Black Sea pycnocline and in deeper waters are surface winds and internal waves (Gregg & Özsoy, 1999). Excess freshwater inputs into the basin, released only through the two-layer stratified flow through the Turkish Straits have stabilizing effects on vertical mixing. The lack of sufficient ventilation to counter organic matter fluxes from the highly productive surface waters of the Black Sea results in the present anoxic conditions below its pycnocline.

The principal mechanism of sub-pycnocline ventilation is the lateral intrusions at the basin boundary, especially near the Bosphorus. The plume, carrying the dense Mediterranean water into the Black Sea, first spreads out on the wide continental shelf, through a delta-like topography, where it is mixed with the overlying colder and less saline waters of the Black Sea (Latif *et al.*, 1991; Özsoy *et al.*, 2001), finally cascades down the continental slope at the shelf edge (Buesseler *et al.*, 1991; Murray *et al.*, 1991; Özsoy *et al.*, 1991). From then onwards, the anomalous waters entering the slope region drive double-diffusive convection adjacent to the source (Özsoy *et al.*,

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1993; Özsoy & Beşiktepe, 1995; Özsoy & Ünlüata, 1997, 1998) and ventilate the waters of the lower pycnocline.

There has been a surge of interest in the Black Sea in the last decade as a result of the environmental threats focused in the region. A great number of new findings and a new level of understanding have consequently emerged from these research activities. Isotopes have been widely used in the recent past for mixing and transport studies (e.g. İzdar & Murray, 1991; Murray, 1991; Swart, 1991*a*,*b*; Fabry & Fröhlich, 1992; Baxter, 1999), complementing the other types of measurements.

We give a brief review of the vertical mixing processes in the pycnocline and deep waters of the Black Sea, based on a synthesis of experimental results. Simultaneous measurement of the stable isotopes ^{18}O, ^{2}H and ^{3}H tracer are then discussed in slightly greater detail in order to complement this synthesis.

Experimental methodology

The temperature and salinity analyses leading to the present synthesis were obtained from a number of oceanographic surveys described in Özsoy and Ünlüata (1997, 1998). The isotope measurements were carried out in the western and southern Black Sea and the neighbouring Marmara Sea. The seawater samples at oceanographic stations were collected by the RV *Bilim* 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. The station positions and other details of the sampling and analyses are described in Özsoy *et al.* (1997) and Rank *et al.* (1999).

The laboratory analyses of stable isotopes were carried out by mass spectrometry (^{18}O measured on MAT 250, precision $< \pm 0{\cdot}01\%$ delta SMOW units, H_2O and CO_2 equilibration during sample preparation, and ^{2}H on MAT 251, precision $< \pm 0{\cdot}1\%$, 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 $\pm 0{\cdot}7$ TU. Liquid scintillation with electrolytical enrichment was either carried out on a Packard Tricarb instrument (250-ml sample, 20-ml vials) with a precision of $\pm 0{\cdot}2$ TU, or on an ALOKA LB3 instrument (800-ml sample, 140-ml vials) with a precision of $\pm 0{\cdot}1$ TU, depending on the size of the water sample.

The role of Bosphorus inflow in intermediate depth mixing

The introduction of Mediterranean water into the Black Sea occurs in stages. The dense water first passes a sill outside of the Bosphorus exit, then flows through a channel and over a wide continental shelf where it evolves by entraining ambient water several times greater than its original volume, and finally cascades down the continental slope after flowing over a distance of 50 km from the Bosphorus exit (Latif *et al.*, 1991). Along the steep continental slope, the entrainment is increased by an order of magnitude and the anomalous waters lose almost all their contrast with ambient waters (Özsoy *et al.*, 2000). However, their remaining anomalous properties are sufficient to act as sources to drive double-diffusive convection adjacent to the slope (Özsoy *et al.*, 1993; Özsoy & Beşiktepe, 1995; Özsoy & Ünlüata, 1997, 1998) and to ventilate the lower pycnocline. Both the above measurements, as well as modelling, have shown that the anomalous waters and the convection driven by them are limited to depths above 500 m (Stanev *et al.*, 1999; Özsoy *et al.*, 1993, 2001). The post-Chernobyl radioisotopes (Buesseler & Livingston, 1997), ^{14}C (Östlund, 1974; Östlund & Dyrssen, 1986) and tritium (Top *et al.*, 1991) measurements are consistent with the above results, showing very little penetration below a depth of about 500 m, and the increasing age of waters near the bottom.

Turbulence measurements on the Black Sea shelf outside the influence of the bottom flow and the mixed layer have shown (Gregg & Özsoy, 1999) diapycnal diffusivity values that were amongst the lowest observed in the world ocean, suggesting a background, internal wave field that appeared to be the only source of turbulence in the absence of wind mixing and tides. The continental slope anomalous source of double-diffusive convection is enacted in this kind of an environment.

The Black Sea is one of the few places where double-diffusive convection contributes to mesoscale and basin-scale mixing processes in the sub-pycnocline waters. The diffusive regime of double-diffusive convection occurs when a destabilizing temperature gradient competes with a statically stable salinity gradient (e.g. when both temperature and salinity increase with depth). Laboratory convective layers under similar conditions of a salinity gradient heated from below have been studied extensively (Turner, 1968; Huppert & Linden, 1979; Fernando, 1987), and many oceanographic and limnological examples are known (Turner, 1969; Fernando, 1989). Double-diffusive convection is often triggered by horizontal contrasts (e.g. boundaries or fronts) and by buoyancy sources (e.g. sources/sinks of heat or salt) in stratified environments with two diffusing properties, leading to interweaving layers characterized by a series of alternating diffusive/fingering interfaces (Turner, 1973, 1978; Huppert & Turner, 1980; Tsinober *et al.*, 1983; Tanny & Tsinober, 1988; Jeevaraj & Imberger, 1991).

The Black Sea is, in fact, a unique place to observe the large-scale effects of double-diffusive convection, where it occurs in connection with (1) the warm, saline Mediterranean waters intruding from the Bosphorus, (2) a diffusive interface capping the bottom convection layer, and (3) vertical fluxes in the deep waters.

The historical evolution of the deep waters has resulted in a peculiar distribution of the ' density ratio ' $R_\rho = (\beta dS/dz)/(\alpha dT/dz)$, such that the entire water column is formally in the diffusive instability range, changing from minimum values ($R_\rho > 2$) at the top (below the mixed layer) and bottom (above the bottom convective layer), to marginally stable values ($R_\rho \rightarrow \infty$) at a depth of about 500 m (Özsoy *et al.* 1993).

The dense bottom water of Mediterranean origin (modified in the Turkish Straits and across the continental shelf) cascading down the continental slope degenerates into discrete layers (depth range: 100–500 m) created by double-diffusive convection penetrating into the interior (Özsoy *et al.*, 1993; Özsoy & Beşiktepe, 1995). The unstable interior, in the sense of double diffusion, is expected to enhance the resulting convection (e.g. Turner, 1978), in addition to the effects of sidewall buoyancy source (heat and salt) introduced into the stratified environment (e.g Tsinober *et al.*, 1983; Tanny & Tsinober, 1988).

Boundary mixing (e.g. Garrett, 1990; Woods, 1991), in this case driven by double-diffusive convection (Özsoy *et al.*, 1993; Özsoy & Beşiktepe, 1995), appears to be the dominant ventilation mechanism across the permanent halocline of the Black Sea, as it has been amply demonstrated by the transport of shelf-derived materials, including dissolved nutrients, Chernobyl tracers and inorganic particulates (Buesseler *et al.*, 1991; Codispoti *et al.*, 1991; Buesseler & Livingston, 1997; Özsoy *et al.*, 1991, 1993). A schematic illustration of the mechanism of ventilation driven by the Mediterranean inflow, and aided by the other processes of wind-forced and boundary-driven ventilation, is given in Figure 1.

The relatively rapid penetration of tracers across the halocline, contrasting with almost none in the deep water, has been confirmed by tritium (Top *et al.*, 1991; Top, 1999; Rank *et al.*, 1999) and carbon-14 (Östlund, 1974, 1986) measurements. Plume parameterization of vertical mixing in general circulation models (Staneva & Stanev, 1997; Stanev *et al.*, 1999) has significantly improved the simulation of ventilation and tracer distributions in the Black Sea, yielding similar results.

The bottom convection layer, covering the entire abyssal plain of the Black Sea, with a thickness of about 450 m, is the largest known example in the world ocean. A destabilizing geothermal heat flux of about 40 mW m^{-2} at the bottom acts against an otherwise stable salinity stratification. The transport between the bottom convective layer and the overlying waters occurs through a single diffusive interface. The observed features and the parametric setting suggest that the rate of development of the convective layer is far from the typical initial phase of growth proportional to $t^{1/2}$ (e.g. Turner, 1968; Huppert & Linden, 1979; Fernando, 1987, 1989) and corresponds with the long time limit of the ' low stability regime ' of Fernando (1987, 1989) and Fernando and Ching (1991), in which the interfacial entrainment and, therefore, layer growth become negligibly small (Özsoy *et al.*, 1991; Özsoy & Beşiktepe, 1995). The heat flux at the stable Black Sea interface ($Ri^\star \cong 600$), estimated from the Huppert & Linden (1979) and Fernando (1989) models, was about 5–8 times larger than the geothermal heat flux from the bottom, since these models typically overestimate fluxes when the Richardson number $Ri^\star = \delta b\, h_s / w_s^2 > 240$ (Fernando, 1989, δb is the buoyancy step, h_s is the layer thickness and w_s is a vertical velocity scale for convection), and most probably do not apply in the long time limit, not adequately investigated in the laboratory studies on which they are based.

The bottom convective layer is expected to have a similar age to the deep waters (a few thousand years). The observed homogenization of water properties vertically and across the basin inside the convective layer is expected to occur within about 40 years, which could have a bearing on the homogeneity of deposition/diagenesis in the bottom sediments (Özsoy *et al.*, 1991; Özsoy & Beşiktepe, 1995).

Based on the comparison of flux ratios above the diffusive interface and the water column, double diffusion is most likely to be the main vertical transport mode for heat and salt in the deep waters extending from the lower part of the pynocline to the diffusive interface above the bottom convective layer (Özsoy *et al.*, 1991; Murray *et al.*, 1991; Özsoy & Beşiktepe, 1995). The observed ' S-shape ' of the potential

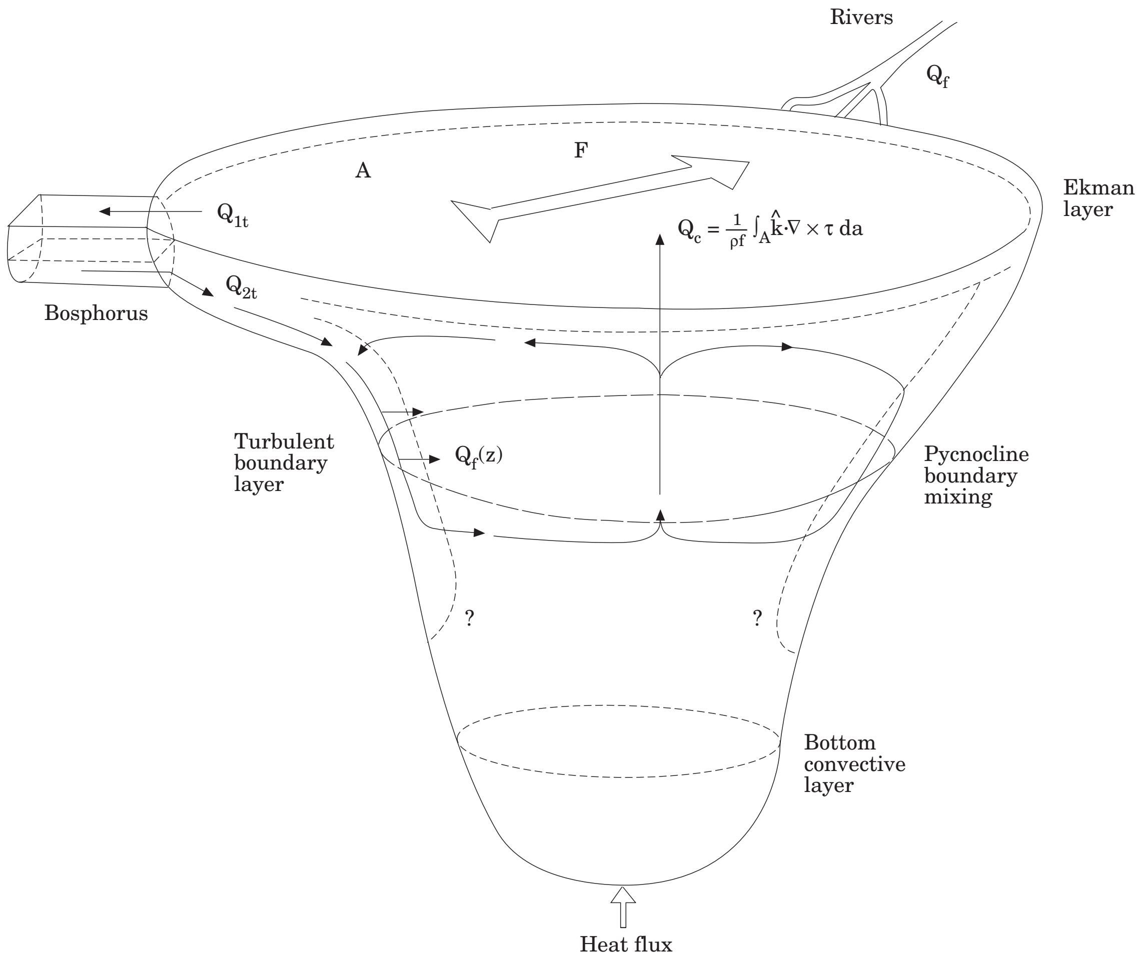

FIGURE 1. The scheme of vertical recirculations and ventilation of the Black Sea interior (after Özsoy *et al.*, 1993).

temperature *vs* salinity (θ–S) curves in the Black Sea deep waters also leads one to suspect that it has been generated by the different diffusivities of salt and heat (Mamayev, 1975, 1995).

The stable isotopes ^{18}O and ^{2}H, and tritium (^{3}H)

Mixing between the Mediterranean and the Black Sea waters is well reflected in the stable isotope data, with sharp contrasts across the pycnocline and constant levels in deep waters (Figure 2). Freshwater influence from major rivers (Danube, Dniepr and Dniestr) in the north-western shelf region of the Black Sea results in decreased $\delta^{18}O$ and $\delta^{2}H$ values near river mouths and along the shelf (Rank *et al.*, 1999). In general, our results are similar to those of Swart (1991*a*, *b*) and Gat *et al.* (1996), but contain much less noise, perhaps as a result of the differences in the methodologies used.

Earlier measurements of tritium in deep waters have been problematic because of the very low values, which are of the order of the detection limit. It was because of this reason that two data points with relatively large values measured in deep water raised questions about the possibility of the surface signals reaching the bottom of the Black Sea (Top & Clarke, 1983). Later measurements (Top *et al.*, 1991) reported that there was no measurable tritium in deep water below a depth of 500 m, consistent with the synthesis based on salinity, temperature and more recent tracer measurements. It is also true that the analytical precision of measurements have been improved greatly over the years.

Large gradients in tritium occur across the pycnocline in the Black Sea (Figure 2). Below the pycnocline, tritium decreases rapidly below 500 m to very low values near the bottom. Although we only have data from one deep station, it is evident that the concentrations decrease to almost zero in deep water, within the precision of the measurements. Our ^{3}H

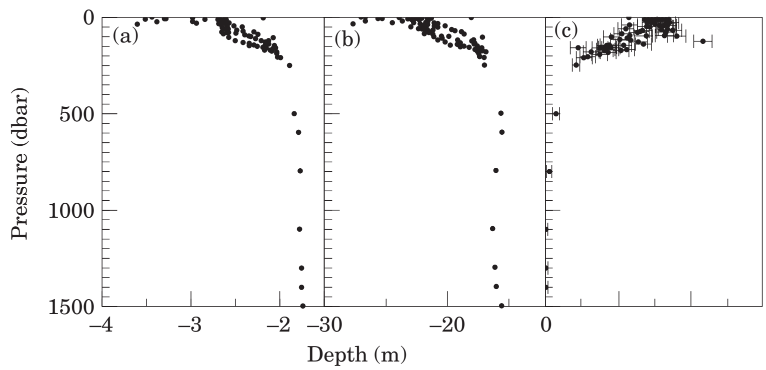

FIGURE 2. Vertical distribution of stable isotopes and tritium in the Black Sea with respect to depth: (a) $\delta^{18}O$; (b) $\delta^{2}H$; (c) ^{3}H. Error bars in tritium plot show standard deviation resulting from measurement uncertainty.

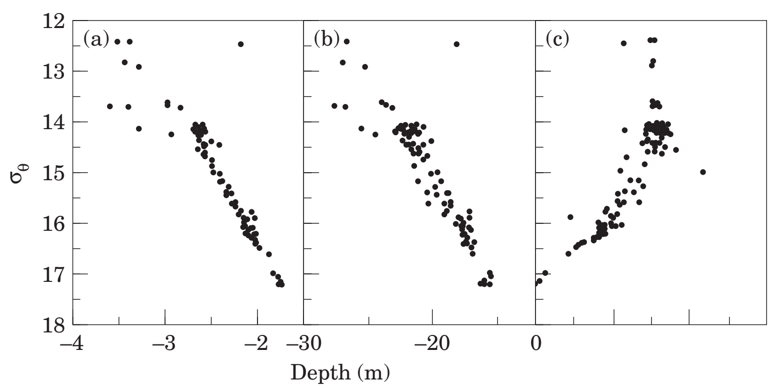

FIGURE 3. Profiles of stable isotopes and tritium as a function of σ_θ density in the Black Sea. (a) $\delta^{18}O$; (b) $\delta^{2}H$; (c) ^{3}H.

measurements appear to have less scatter than the earlier measurements, and clearly demonstrate physical variations.

Density dependence

Because mixing between the surface and deep waters is limited by stratification, the dependence between chemistry and physical mixing of scalar properties produces self-similar profiles (Tuğrul *et al.*, 1992; Özsoy & Ünlüata, 1997), especially in the lower part of the pycnocline where turbulence rapidly declines. The linear dependence of stable isotopes *vs* σ_θ density (Figure 3) eliminates the effects of dynamical displacements, and implies that the water-mass evolution has followed the history of contact with the atmosphere. This is an independent verification of the hypothesis of deep water mass formation by a mixture of Mediterranean and near-surface, relatively freshwater masses, based on temperature–salinity analyses alone (Özsoy *et al.*, 1991; Özsoy & Beşiktepe, 1995) and the late quaternary history of the Black Sea (Stanley & Blanpied, 1980; Boudreau & Leblond, 1989).

In contrast, the transient tracer tritium *vs* σ_θ density in Figure 3 indicates a significant change in tritium gradient with respect to density below the pycnocline. The fact that the break appears in the transient tracer and not in the stable isotopes specially indicates 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 $\sigma_\theta \cong 16{\cdot}3$. This result must reflect different mechanisms of renewal for intermediate and deep waters explored in a number of earlier interpretations (Özsoy *et al.*, 1991; Özoy & Beşiktepe, 1995), i.e. turbulent wind stirring in the surface mixed layer, diminishing rapidly at the pycnocline, Mediterranean water lateral intrusions below the pycnocline until a depth of about 500 m ($\sigma_\theta \cong 17$), and only double diffusion between 500 m and the bottom convective layer. Indeed, below 500 m, there is very little tritium.

Tritium transient in the surface waters

The tritium in Danube waters (Figure 4) and in precipitation (not shown) (Rank, 1992, 1995, 1996 indicates a transient decrease following the

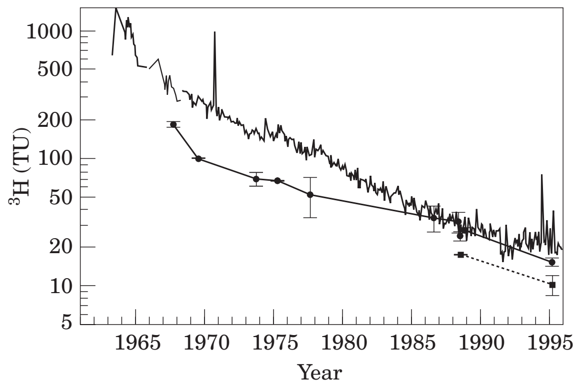

FIGURE 4. Surface ^{3}H concentrations measured in the Black Sea and Marmara Sea surface layers (0–20 m depth), superposed on a plot of ^{3}H measurements in the Danube River. Danube samples were obtained at Reichsbruecke (before December 1991) and Nussdorf-Schleuse (after December 1991) in Vienna, Austria. ——— Danube River, Vienna; —●— Black Sea surface (0–20 m depth); · · ·■· · · Marmara Sea surface (0–20 m depth).

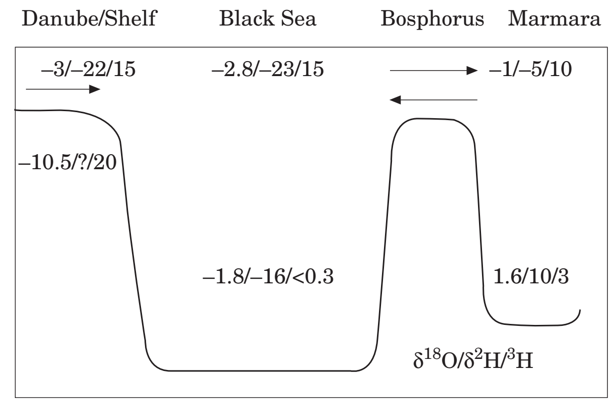

FIGURE 5. Idealized surface (depth 0–20 m) and bottom (depth >500 m in the Black Sea, and depth >50 m in the Marmara Sea) concentrations of isotopes, δ^{18}O/δ^2H/^{3}H, in the Danube River (Vienna), the north-western shelf area, the greater part of the Black Sea and the Marmara Sea.

1963–1964 maximum, in agreement with other observations elsewhere (e.g. IAEA, 1992). A compilation of past measurements has been used to calculate 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 Figure 4.

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, on 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 was larger in the 1960s and 1970s (Figure 4), when the inputs were considerably larger. These transient features suggest vertical redistribution of tritium by mixing across the main pycnocline, starting from the initially uncontaminated conditions, although this is not easy to establish with confidence from earlier measurements, which lack sufficient resolution and accuracy in intermediate and deep waters.

In the case of the adjoining 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.*, 1993, 1994).

Exchanges

The average surface and deep-water properties and the inter-basin exchange characteristics are schematized in Figure 5, based on average surface levels and those measured below a depth of 500 m in the Black Sea and 50 m in the Marmara Sea.

The Danube water in Vienna (Rank, 1992, 1994, 1996) has a δ^{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‰. We note limited variability in measurements between 1988 and 1995 (Swart, 1991*a*,*b*; Özsoy *et al.*, 1997). The freshwater inflow, specifically from the Danube River, entering the sea with a low δ^{18}O, has a significant influence in the north-western shelf region, where it lowers the δ^{18}O. The deep water of the Black Sea, with a historical Mediterranean contribution, is enriched in terms of δ^{18}O. In contrast, the recent source of tritium at the surface has not been able to penetrate into the deep water, 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.

Isotopic correlations

Figure 6 correlates all three isotopes studied. In Figure 6(a) we observe that all of the data points in the two interconnected Marmara and Black Sea basins lie along the same straight line, slightly displaced from the meteoric water line, and with only a minor change in slope between basis, suggesting that these waters were formed by a continuous mixture of freshwater

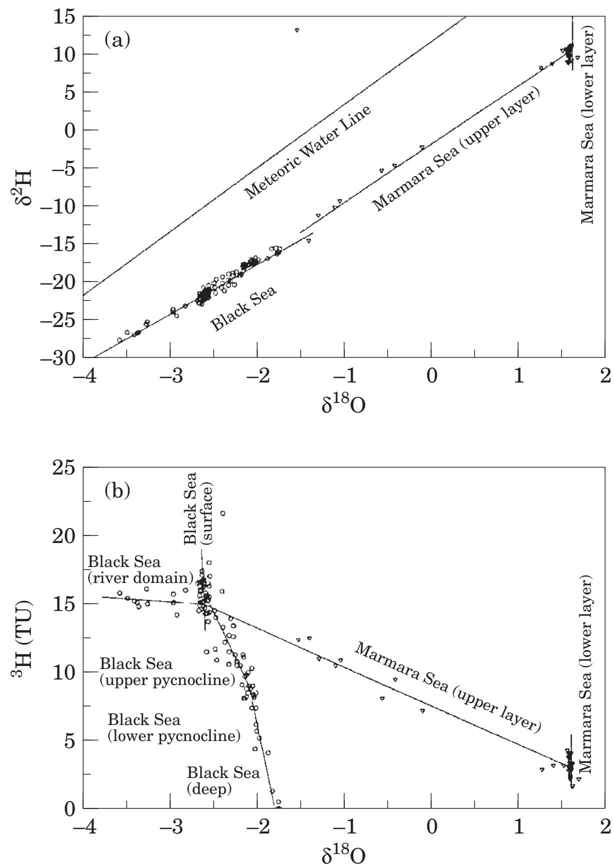

FIGURE 6. (a) δ^{18}O *vs* δ^2H and (b) δ^{18}O *vs* ^{3}H for the Black Sea (○) and Marmara Sea (▽), 1995. (Units of oxygen δ^{18}O and deuterium δ^2H are per mille psu, units for tritium ^{3}H are TU.)

inputs and Mediterranean water. Mediterranean water in the Marmara Sea has slightly higher δ^2H compared to the eastern Mediterranean (Gat *et al.*, 1996).

In the oxygen-18 *vs* tritium plot [Figure 6(b)], 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 density 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 of this cluster, and belonging to the north-western shelf region, is helpful in identifying riverine effects. Fresh water is tagged with 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.

Conclusions

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

Stable isotope anomalies in precipitation and river runoff, characterizing 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 an almost linear correlation between oxygen and deuterium for all water samples, showing that the waters in both basins are continuously proportional mixtures of surface freshwater inputs to the Black Sea and 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, and thereby pointing to 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 determines 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, owing to the difference in their respective source functions and differences in the renewal mechanisms of 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. The comparison of tritium with stable isotopes can characterize all distinct water masses in the two basins.

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