PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

The Use of Anthropogenic Tritium and Helium-3 to Study Subtropical Gyre Ventilation and Circulation [and Discussion]

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The use of anthropogenic tritium and helium-3 to study subtropical gyre ventilation and circulation

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With tritium and helium-3 (^{3}He) data from the Transient Tracers in the Ocean (TTO) expedition and from two other contemporaneous cruises, a synoptic picture of the ventilation and circulation of the subtropical North Atlantic is built. We will see clear evidence of gyre circulation in the tritium–^{3}He age distributions on the shallower isopycnals, permitting estimates of the rates of circulation averaged over timescales from months to decades. The entry points of fluid into the main thermocline and the pathways of exchange with the upper ocean on seasonal to decade timescales appear clearly. It is the time-averaged transport processes on those timescales that are important to the uptake of carbon dioxide by the ocean. The overall relation between tritium and ^{3}He within the subtropics exhibits a systematic hook-like pattern that is consistent with 'strong' ventilation of the gyre thermocline; a fluid parcel entering the gyre thermocline making only about one circuit around the gyre before being ventilated.

Finally, we present a time-series of ^{3}He measurements made over a period of two years near Bermuda. The mixed layer is demonstrably supersaturated in this isotope throughout a large part of the year, requiring a gas-exchange flux of this isotope to the atmosphere. Model results are presented that permit the calculation of the *in situ* solubility isotope ratio anomaly for helium (as affected by bubble injection and gas exchange), and that can be used to estimate the upward flux of this isotope. Because only a small fraction of this flux can be produced in the mixed layer, this helium must be 'mined' from the main thermocline. The computed flux is consistent with the long-term evolution of the inventories of tritium and ^{3}He within the main thermocline. This flux has implications regarding the vertical transport of material within and from the permanent thermocline. A single observation of what may be one of the processes responsible for this upward flux is discussed.

1. On the use of the tritium–^{3}He age

The detonation of nuclear weapons in the atmosphere more than two decades ago produced several hundred kilograms of the radioisotope tritium. By observing the passage of this isotope into and through the ocean, we have an opportunity to characterize the various mechanisms whereby the ocean transports substances. Such information permits us to make inferences about the rates of physical, chemical and biological processes in marine systems. Unfortunately, application of the above strategy is limited by the quantity of data available (both in space and time) that documents the passage of this isotope, and in particular by imperfect knowledge of the precise manner in which tritium has entered the ocean. By simultaneously determining its stable, inert daughter product ^{3}He, however, additional information is gained. This is evident from the distributions of these isotopes in the upper main thermocline of the subtropical North Atlantic, where significant gradients of ^{3}He are observed where the tritium distribution is

relatively homogeneous. The timescale of significant radiogenic ^{3}He accumulation is short relative to the recirculation timescale of the upper waters of the gyre, which in turn is short relative to the time since the tritium transient.

A useful concept in discussing the ^{3}He distributions is the tritium–^{3}He age τ, defined as

$$\tau = 17.96 \ln (\xi/\theta), \tag{1}$$

where τ is in years, θ is the tritium concentration, and ξ is the sum of the tritium and the excess ^{3}He concentrations. In a sense, this 'age' may be regarded as the elapsed time since gas equilibration with the atmosphere. It represents the time information implicit in the two tracers while at the same time largely eliminating the dependence on 'initial conditions'. Timescales in the range of 0.1–10 years can be measured.

This simple concept, however, may be complicated by mixing. To quantify the effects of mixing, one can derive an advection–diffusion equation for τ. In the case of two-dimensional isopycnic flow with constant, isotropic turbulent diffusion, one can combine the advection–diffusion equations for tritium and ^{3}He to obtain

$$\tau_t = \kappa \nabla^2 \tau - \boldsymbol{u} \cdot \nabla \tau + 1 - \tilde{v} \cdot \nabla \tau, \tag{2}$$

where κ is the isopycnal diffusivity, $\boldsymbol{u}$ is the isopycnal velocity, and ∇ is the isopycnal gradient operator (Jenkins 1987). The subscript t implies partial differentiation with respect to time; $\tilde{v}$ is a 'mixing pseudo-velocity' arising from mixing of tritium and ^{3}He, defined as

$$\tilde{v} = \kappa \nabla \ln (\xi\theta).$$

One can readily identify the important terms in (2) by comparison with the chronological term (1) associated with the increase of age with time. The magnitude of the mixing terms, on timescales less than a decade, and on surfaces which outcrop to the south of the line of zero windstress curl ($\sigma_\theta \leqslant 27.1$), is of order 10 % or less for the eastern subtropical Atlantic (Jenkins 1987). On longer timescales, and where mixing may dominate over advection, the age becomes a much more model-dependent quantity. In general, because of the sense of the tracer gradients, τ underestimates the 'true age'. The sense of this age compression can be seen in figure 1, which is a plot of τ against the advective timescale, $T_a = X/u$ for a variety of one-dimensional advective–diffusive flow simulations. The model was forced by using the theoretical surface tritium time history (from Dreisigacker & Roether 1978), and scaled with the radiotracer Peclet number

$$Pe = u^2/\kappa\lambda, \tag{3}$$

(after Jenkins 1982) where the length scale is given by u/λ.

Typical values of κ and u for the upper main thermocline within the gyre interior are $\kappa = 10^3$ m^2 s^{-1} and $u = 10^{-2}$ m s^{-1}, respectively (see, for example, Armi & Stommel 1983; Jenkins 1987), which with a tritium decay probability $\lambda = 1.77 \times 10^{-9}$ s^{-1} lead to $Pe \simeq 50$. Thus one would conclude for areas away from the turbulent boundary regions, that τ is accurate to order 10 % for timescales less than a decade. For timescales of a few decades, the bias is of order 20–30 %. Moreover, the slope of the relation in figure 1 diverges from the isoline slope in the region of the advancing tritium front (where $\tilde{v}$ is largest). Thus, in frontal regions and where mixing is more important, the interpretation is more complex.

The immediate difficulty in assessing the impact of mixing on τ in such a comparison lies in the choice of the advective age as the correct measure of the timescale. The concept of such a

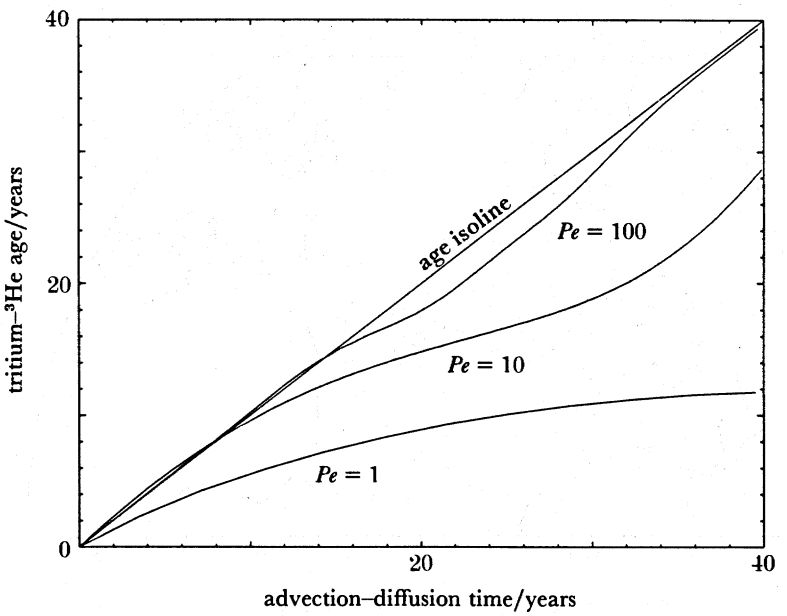

FIGURE 1. Comparison of tritium–^{3}He ages with advection timescale for a one-dimensional model with constant velocity and diffusion (see text). Three values of the Peclet number, $Pe = u^2/\kappa\lambda$, are compared to the ideal (equal age) isoline.

'box-car' time is questionable in the presence of diffusion, and a more appropriate measure may be $T = (1/T_d + 1/T_a)^{-1}$, where $T_d = X^2/\kappa$ is the diffusive timescale. This consideration is necessarily crude, but suggests that part of the apparent disparity may lie more in the concept of the physical timescale rather than the nonlinearities in τ. It is apparent, however, that τ is a model-dependent quantity and hence must be used with caution. It is nevertheless a useful quantity for gaining qualitative insight into gyre ventilation and circulation.

2. ISOPYCNIC MAPS OF TRITIUM AND THE TRITIUM–^{3}He AGE

The Transient Tracers in the Ocean (TTO) expedition, in combination with some other cruises occurring within the same time frame (1979–81), has provided the first opportunity to map the distribution of the tritium–^{3}He age (τ) on isopycnic horizons over significant portions of an ocean basin. Included here are four maps (figures 2–5) of tritium and τ on representative density horizons. The tritium and ^{3}He data were linearly interpolated onto the appropriate horizons only when sample points existed within a specified density range (i.e. ± 0.125 for $\sigma_\theta = 26.50$ and 26.75, ± 0.100 for $\sigma_\theta = 27.40$, and ± 0.025 for $\sigma_2 = 37.05$). The tritium–^{3}He age was computed from the interpolated data. The tritium data were normalized to a standard date of 1 January 1981, but the tritium–^{3}He age data were not adjusted, on the assumption that the tritium–^{3}He age is in approximately steady state. This is justified for the shallower surfaces where the advective timescale is much less than the time elapsed since the tritium transient (i.e. where tritium is relatively uniform), but is suspect on deeper horizons.

The $\sigma_\theta = 26.50$ surface

Although the tritium distribution (figure 2*a*) on this surface is relatively homogeneous (it varies by only 30% over the entire gyre), the tritium–helium age shows substantial variation.

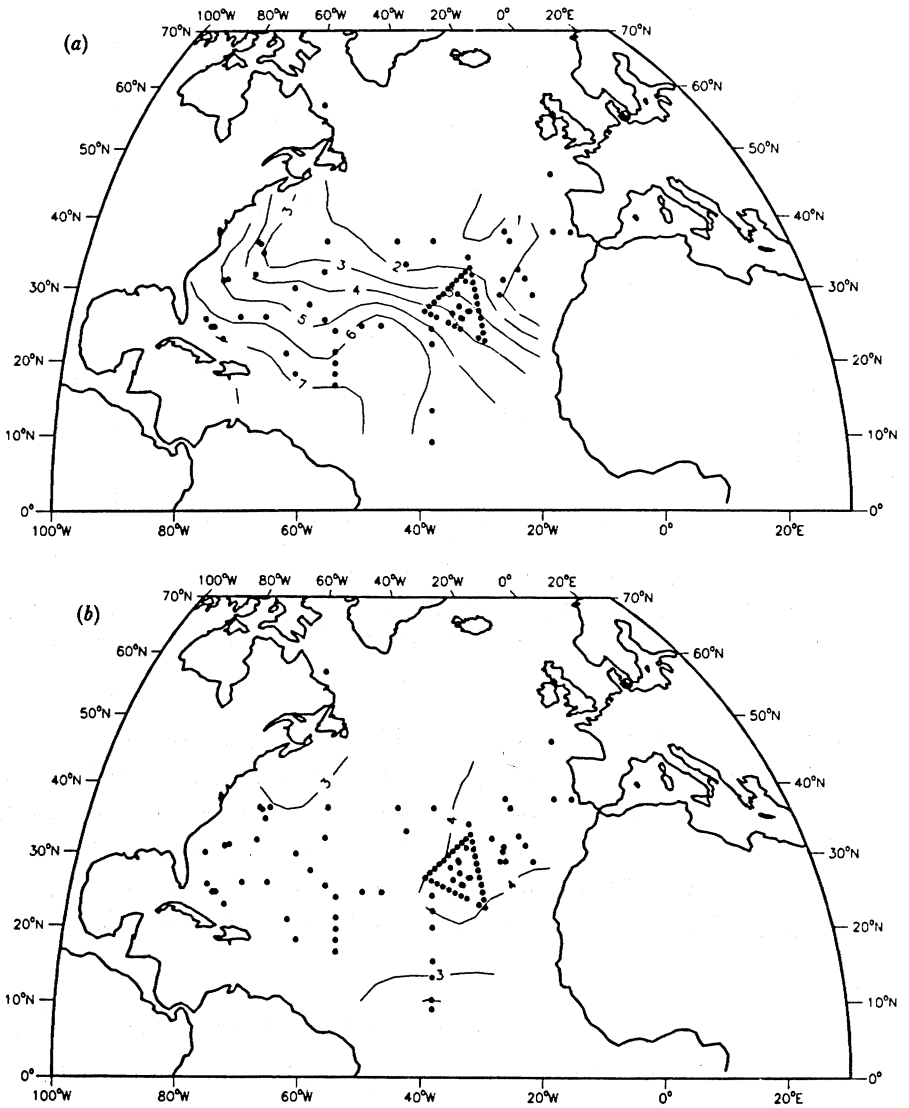

FIGURE 2. Maps of (*a*) tritium–^{3}He age in years and (*b*) tritium concentrations in TU on the $\sigma_\theta = 26.50$ isopycnal for the North Atlantic.

Analytical resolution for the tritium–helium age is about 0.1 year, so the gradients are well resolved. Within the Sargasso Sea, this surface corresponds roughly to the subtropical mode water ('Eighteen Degree Water'). In the northeast corner of the gyre, the ages are small (where this isopycnal shoals into the mixed layer), and increase southwestward at a rate equivalent to a velocity of 1.0 cm s^{-1}. The direction and magnitude of this velocity are close to the β-spiral velocities obtained by Armi & Stommel (1983) for this horizon (cf. Jenkins 1987).

What is more interesting, however, is the larger-scale shape of the isochrons, and what they imply about pathways of ventilation into the interior. The entry of young (ventilated) fluid in the northeast corner of the gyre is a phenomenon readily predicted on the basis of Ekman pumping (see, for example, Leetma & Bunker 1978) and the pattern of wintertime outcropping of the isopycnal (see, for example, Levitus 1982). The overall shape of the isochrons in fact

bears some resemblance to the outcrops (cf. Jenkins 1987, figure 1). Specifically, note the general southeastward trend in the central and eastern half of the gyre, and the southwestward bowing in the northwestern region of 'tight recirculation' (the parlance of Worthington 1976; see especially p. 93, figure 41). In the extreme northwest corner of the Sargasso Sea, the 3 and 4 year isochrons turn sharply northward, a sign of the northward flow of older waters in the Gulf Stream. In this region, the isopycnal is at 300–400 m depth. Along the northern rim of the gyre (east of perhaps 75° W), however, the tritium–^{3}He age decreases downstream. This is particularly remarkable considering that the isopycnal is deeper than 500 m in this region. It is possible within the resolution of the station spacing that this decrease is a result of the tight recirculation of waters ventilated further downstream. In any case, considering the large eastward transports (Worthington 1976), one must conclude that ^{3}He is effectively removed, and hence by inference, that all other surface forced properties are reset. This is clear evidence of the strong ventilation proposed by Sarmiento (1983) and by Jenkins (1987).

In the southern part of the gyre, the 6 and 7 year isochrons exhibit a interesting structure. There is evidence of the entry of younger water to the south and east (around 10° N, 40° W), despite the lower tritium concentration. This is the invasion into the gyre of the northward-flowing relatively tritium-impoverished equatorial–southern waters. The confluence of this equatorial water with the broader-scale gyre recirculation component near the Antilles further bends these isochrons southward, causing them to run parallel to the South American coast. It is clear that the simple interpretation of a flow field normal to the isochrons can be misleading in these circumstances, and that mixing must be considered.

The $\sigma_\theta = 26.75$ surface

This isopycnal is close to the one studied by Broecker & Östlund (1979), and lies within the main thermocline at a temperature of approximately 15 °C. During the TTO expedition, it outcropped near 60° N, and deepened southward and westward, reaching a maximum depth of over 700 m in the Sargasso Sea. Although tritium is still more-or-less uniform within the subtropical gyre, the 3 tritium units (TU) isopleth has crept northward by several hundred kilometres on the southern boundary and along the South American coast, and further southward along the North American coast. This is a reflection of the longer ventilation and circulation timescales of this horizon relative to those on the 26.50 isopycnal.

The zero-age region (figure 3) appears in the Irminger Sea beginning a westward march toward the Labrador Sea as one progresses to denser isopycnals. As on the $\sigma_\theta = 26.50$ horizon, the age increases smoothly southwestward in the eastern part of the gyre at a rate and direction in agreement with β-spiral velocities. The tongue-like extension of the 8 year isochron (near 20° N, 40° W) is suggestive of the upper thermocline circulation scheme suggested by Worthington (1976; p. 75, figure 29). Again as on the 26.50 surface, there is evidence of strong 'rim ventilation', seen in the downstream decrease in the tritium–helium age. Along the northwestern edge of the Sargasso Sea, the isochrons again show a northerly swing: the slope waters are in fact older than the Sargasso Sea waters on the same horizon. This suggests that on the shallow surfaces, significant ventilation does not occur across the Gulf Stream through the slope waters. This is consistent with float observations and other tracer gradients (Bower *et al.* 1985). However, as on the 26.50 surface, the downstream decrease of the tritium–^{3}He age requires that strong ventilation must occur, perhaps associated with the tight recirculation.

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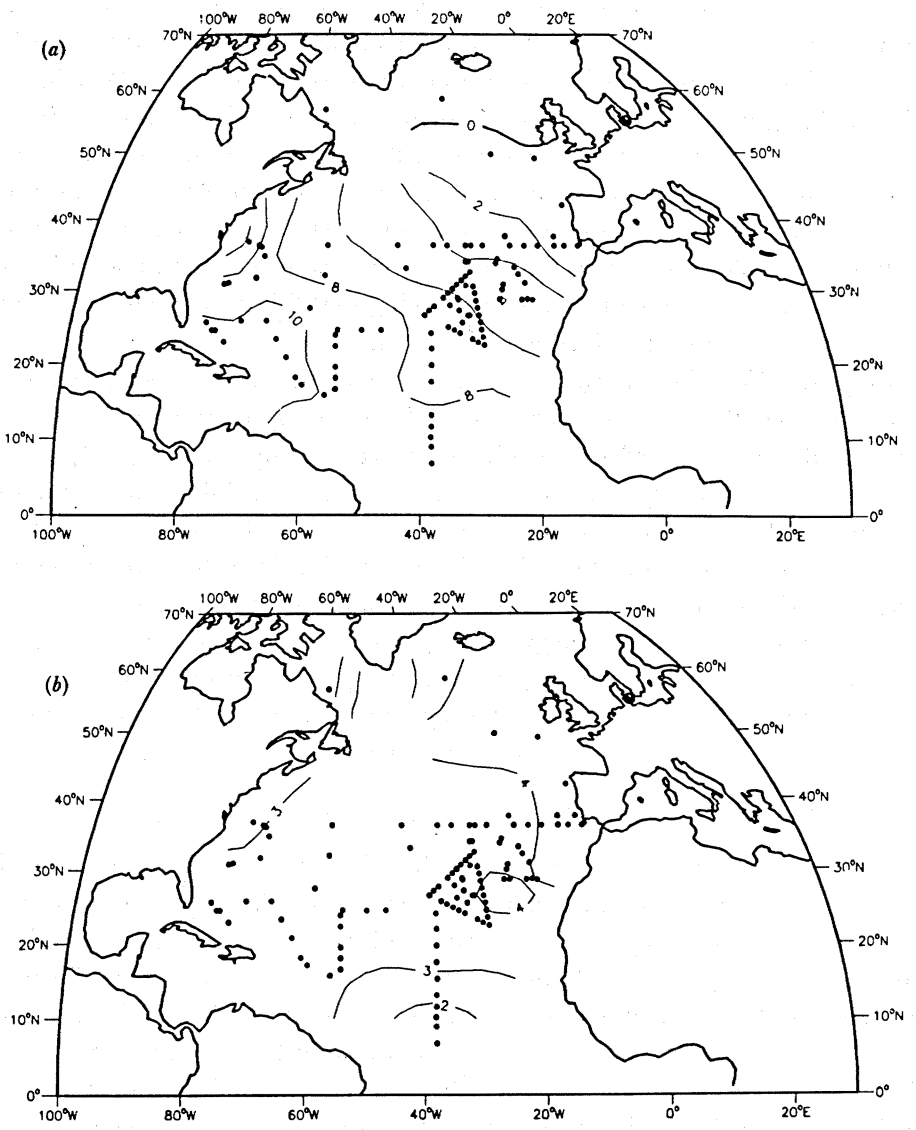

FIGURE 3. Maps of (*a*) tritium–^{3}He age in years and (*b*) tritium concentrations in TU on the $\sigma_\theta = 26.75$ isopycnal for the North Atlantic.

The $\sigma_\theta = 27.40$ surface

Unlike the previously discussed horizons, this isopycnal (figure 4) outcrops in the wintertime to the north of the line of zero windstress curl (see, for example, Jenkins 1987, figure 1). The zero-age region, and by inference the fluid-entry point, has rotated westward. From a minimum depth and age within the Labrador Sea the layer deepens to 1000 m within the subtropics where it lies generally within the main oxygen minimum. Tritium concentrations are very high near the Labrador and Newfoundland coasts, and decrease away from the Labrador Sea and southward. The 10 year isochron now lies entirely north of the subtropics, whereas the isochrons between 20° and 40° N are zonally aligned. Tritium gradients are similarly aligned within this region, and of lesser concentration than the horizons above. The

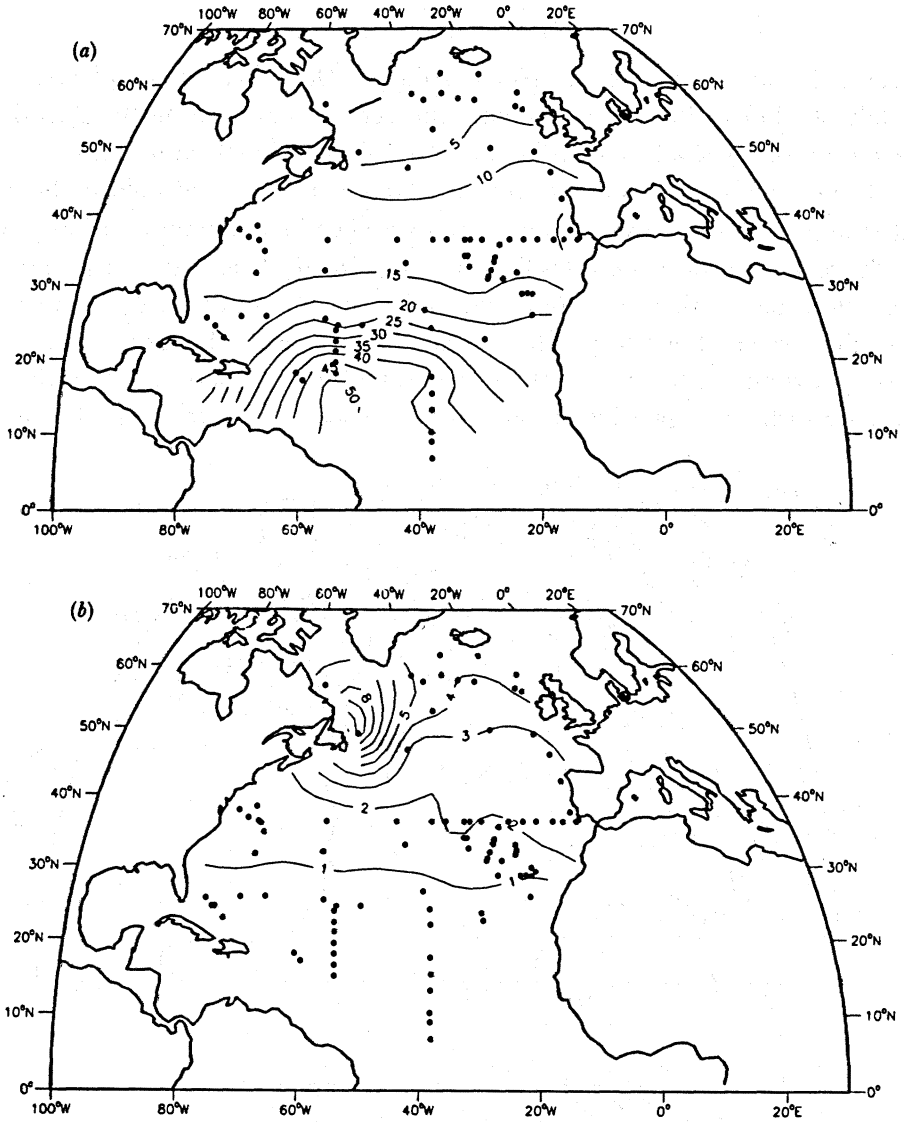

FIGURE 4. Maps of (*a*) tritium–^{3}He age in years and (*b*) tritium concentrations in TU on the $\sigma_\theta = 27.40$ isopycnal for the North Atlantic.

greater age, lower tritium concentrations and lack of significant gyre-like imprint on the isochrons is a reflection of the sluggish ventilation and circulation on this horizon.

To the south, evidence of entry of older water can be seen along the South American coast, both in the isochrons and in the presence of high-silica waters. This is probably Antarctic Intermediate Water. Some of the apparent age may be because of the transport of primordial ^{3}He with this water. From the observed ^{3}He–silicate relation for these waters (Jenkins & Clarke 1976), and the distribution of silicate, it is possible to correct in large part for this influence. This adjustment has been delayed pending compilation of the tritium and ^{3}He data from the TTO–TAS (Tropical Atlantic Study). Clearly, the tritium–^{3}He age in this region should not be interpreted literally, but rather within the framework of model calculations.

The $\sigma_2 = 37.05$ surface

This surface lies at the top of the North Atlantic Deep Water complex, at a depth of approximately 3000 m within the subtropics. One sees a rotation from the essentially zonally oriented isochrons of the shallower surfaces to a quasi-meridional slant. This is evident in the 10 year isochron, which extends down from the Greenland Sea and follows the east coast of Greenland (the present data are not adequate to resolve its circumnavigation of Cape Farewell). The 20 year isochron penetrates far down the North American coast, and the 30 year isochron parallels the South American coast, indicating the southward penetration of young water along the western boundary. This surface lies above the boundary current strikingly demarcated by tritium (cf. Jenkins & Rhines 1980; Olson *et al.* 1986), but the tritium isopleths in the figure are strongly suggestive of southward transport along the edge. In the light of the

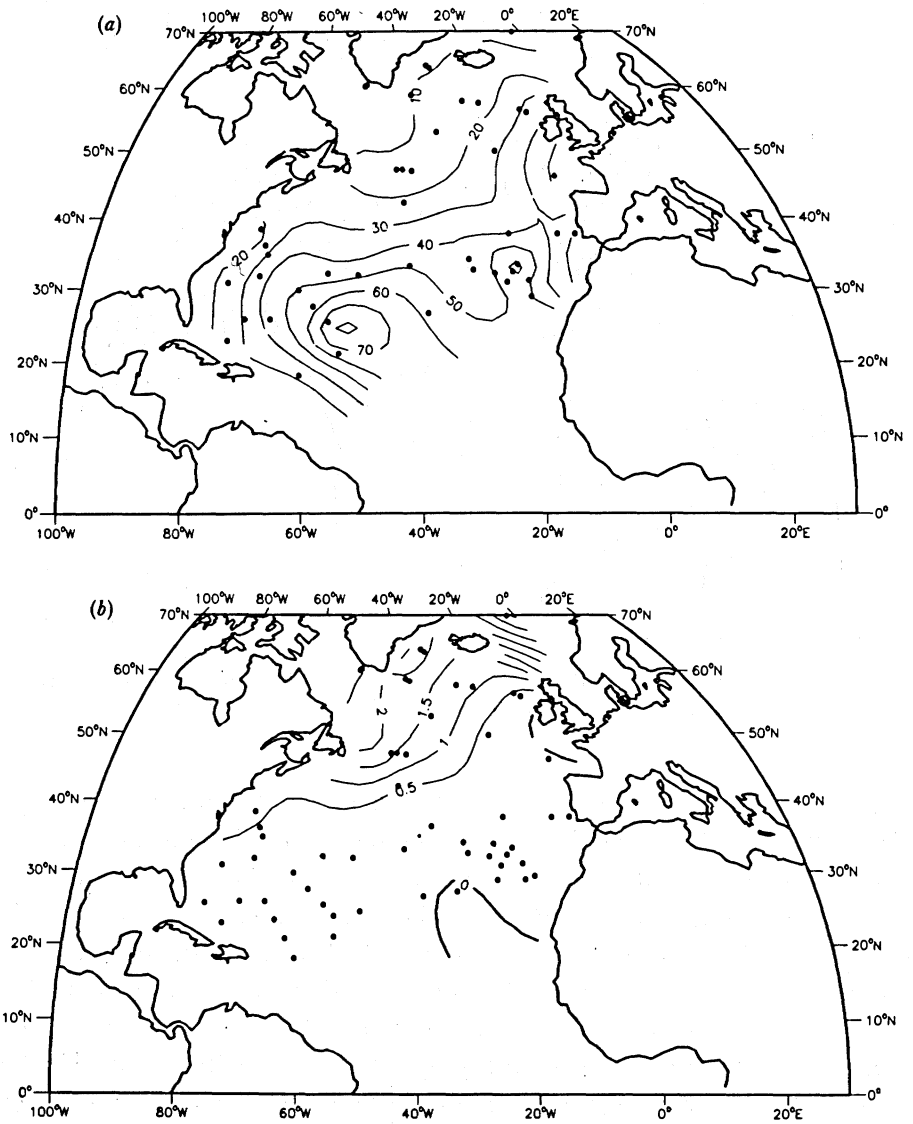

FIGURE 5. Maps of (*a*) tritium–^{3}He age in years and (*b*) tritium concentrations in TU on the $\sigma_2 = 37.05$ isopycnal for the North Atlantic.

large degree of dilution which occurs during the southward journey (Jenkins & Rhines 1980), the ages seen here are not 'box-car' in nature. Current meter measurements suggest smaller transit times. Rather, the age represents some complex mixing age resulting from the ablation and entrainment of interior abyssal waters along the pathway southward. The zonal gradients thus represent the signature of a process of ventilation of the deep interior by shedding of young water from the western boundary. There is a striking similarity in the general shape of the isochrons and the silica isopleths. The large-scale intrusion of apparently older, silica-rich water near 25° N, 50° W needs to be examined more carefully. In particular, one needs to correct for the presence of primordial ^{3}He that has been advected up the South Atlantic from the circumpolar regions.

Summary

The large-scale distributions of tritium and tritium–^{3}He age on a few representative density horizons within the North Atlantic have been presented, and begin to indicate the nature and pathways of ventilation. These distributions are consistent with what we know about gyre circulation, but present us with evidence of very strong ventilation occurring in the north-western segment of the subtropical gyre, as well as the previously supposed entry point of fluid in the northeastern section. We can see the westward march of the outcrop entry point from the Bay of Biscay for the shallowest surface, to the Labrador Sea, and then northward to the Greenland Sea for the deepest horizon. Interior gradients of the tritium–^{3}He age appear to be an important diagnostic feature of ventilation processes in the North Atlantic basin.

3. The tritium–^{3}He scatter plot

Some information can be extracted about the general character of the gyre circulation by examining the overall relation between tritium and ^{3}He for the entire data set. Figure 6*a* is a plot of nearly 2000 data points from the combined TTO–NAS and NATS data sets for the gyre region. The β-triangle data were not included because they were not sufficiently contemporaneous; although the tritium can be normalized for decay, it is not entirely clear how to correct the ^{3}He for the two-year offset between the cruises.

In general, the data fall along a characteristic hook-shaped curve, with the near-surface data lying near zero ^{3}He, and high (3–4 TU) tritium. The thermocline waters rise upward to a maximum of 2–3 TU in ^{3}He, and eventually ramp backward toward zero tritium and ^{3}He in the range 0.5–1.0 TU. It should be noted that the bulk of the data scattering below this curve (in the range 2–3 TU tritium) are in the slope waters. Further, the relation is much tighter on a region-by-region basis.

Figure 6*b* is a plot of results for the one-dimensional model with constant diffusivity (κ) and velocity (u) discussed in §1. The different curves are from varying Peclet number, as defined by (3). This Peclet number must be distinguished from the expected *local* Peclet number of order 100. This can be seen by noting that $L = u/\lambda$, the natural length scale that arises from the non-dimensionalization of the advection–diffusion equations, is of order 5000 km for the upper thermocline. Such a Peclet number thus characterizes the gyre as a whole, and should not be interpreted literally. In a crude sense, it may be regarded as the number of circuits that a parcel of water makes around the gyre before it is ventilated. Comparison with the data indicates that a Peclet number of order one or less provides the best match with the data. This indicates that the gyre is quite strongly ventilated, probably along the northern rim, and that

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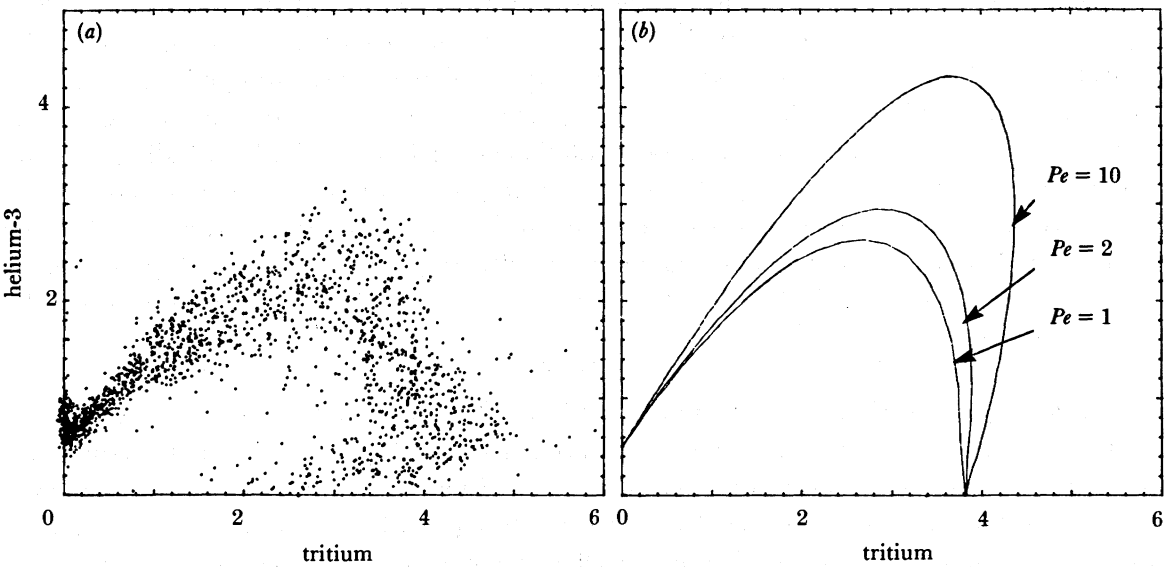

FIGURE 6. The relation between tritium and ^{3}He for (*a*) the North Atlantic TTO and NATS data, and for (*b*) one-dimensional model calculations for various Peclet numbers.

on average, a parcel of water does not make more than one trip around the gyre before it is influenced by ventilation. This is consistent with the high ventilation rates proposed by Sarmiento (1983) and by Jenkins (1987), and with the general character of the τ maps discussed in the previous section.

Another way to regard the scatter plot in figure 6*a* is to recall that the maximum North Atlantic surface tritium in the gyre occurred in the mid-1960s, and was about 18 TU. If a parcel of this water were isolated at that time (i.e. by subduction without excessive subsurface mixing), this water would appear on the plot at 7.8 TU (tritium) and 10.2 TU (^{3}He)! Even allowing for a twofold dilution (to bring the tritium down to the observed range) still gives a ^{3}He value about a factor of two greater than any values observed in the North Atlantic.

4. A TIME-SERIES OF ^{3}He MEASUREMENTS NEAR BERMUDA

The evolution of the transient-tracer distributions in time provides in principle the most useful information about transport processes in the ocean. In addition to quasi-synoptic maps of the tracer distributions obtained at intervals approaching a decade or more, one can obtain relatively frequent sampling at a few sites. Such a time-series is the Hydrostation S near Bermuda, which has been occupied on a nominally bi-weekly basis (with some gaps) for more than 30 years. Over the past decade, sampling has occasionally been done at this station for tritium–^{3}He, and since early 1985, on a regular, more frequent (nearly monthly) basis. The primary goal of this programme is to monitor the longer-term evolution of these two isotopes within the Sargasso Sea and its main thermocline. The more recent effort has also concentrated on the seasonal-timescale processes affecting their distribution. This work is providing important information about the vertical cycling and fluxes of ^{3}He in the upper part of the water column.

Figure 7 is a contour plot of the helium-isotope-ratio anomaly in the upper 1000 m (figure 7*a*) and the upper 250 m (figure 7*b*) for the period January 1985 to January 1987. The isotope-ratio anomaly is defined in the usual fashion as

$$\delta(^3\mathrm{He}) = (R_x/R_a - 1) \times 1000‰, \tag{4}$$

where R_x and R_a are the sample and atmospheric isotopic ratio (^{3}He:^{4}He) respectively. Analytical accuracy is of the order 1.5‰, and sampling depths are indicated in figure 7*b* by small crosses. The lighter isotope is less soluble in water, so that sea water in equilibrium with the atmosphere is characterized by $\delta(^3\mathrm{He}) \approx -16‰$. (This isotope effect has been determined as a function of temperature and salinity to considerable precision by Benson & Krause (1980).)

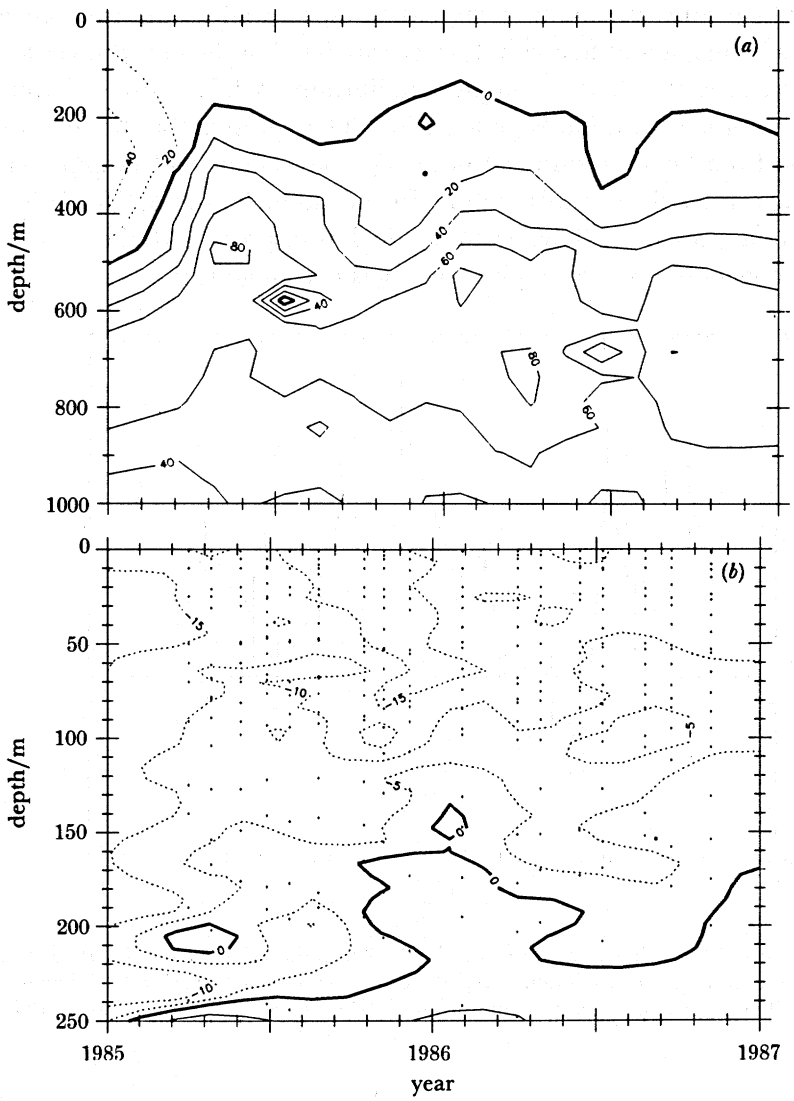

FIGURE 7. A time-series of $\delta(^3\mathrm{He})$ measurements for the years 1985 and 1986 at Station S near Bermuda. The data are contoured in units of per mille for (*a*) the upper 1000 m, and (*b*) the upper 250 m. In (*b*), sample positions are indicated by small + signs. Note the deep maximum at about 700 m depth, and the fact that the static solubility equilibrium value is $\delta(^3\mathrm{He}) \approx -16.5‰$.

The most significant features of the ^{3}He distributions shown in figure 7 are the maximum within the thermocline (common throughout the subtropics) and the vertical excursions of the isopleths on a seasonal basis. The former results from the rollover due to increasing isolation and decreasing tritium with depth. The latter arises in large part from the seasonal changes in the depth of the mixed layer. It is noteworthy that the affected ^{3}He isopleths extend well below the maximum penetration of winter convection for these years, suggesting the significant role of horizontal (or rather isopycnic) processes in the vertical transport of material in the upper ocean.

Examination of figure 7*b* reveals that the $\delta(^3\mathrm{He})$ is supersaturated during a large part of the annual cycle. This is especially evident in figure 8, which is a plot of the mixed layer mean $\delta(^3\mathrm{He})$ as a function of time. The error bars are standard deviation of the mean for each point, and include analytical and systematic error estimates, as well as the random error due to sampling. On the average, about five samples were analysed in the mixed layer for each point. Included for reference is the 'static isotopic equilibrium' $\delta(^3\mathrm{He})$ obtained from Benson & Krause (1980). The supersaturation is greatest in winter, presumably because of the greater flux of this isotope from below resulting from deep convection, and despite expectations of greater gas-exchange rates during this season. In the summer, the $\delta(^3\mathrm{He})$ is lower, because seasonal stratification presents a barrier to vertical transport, and the extremely shallow mixed layer is effectively outgassed despite lower gas-exchange rates.

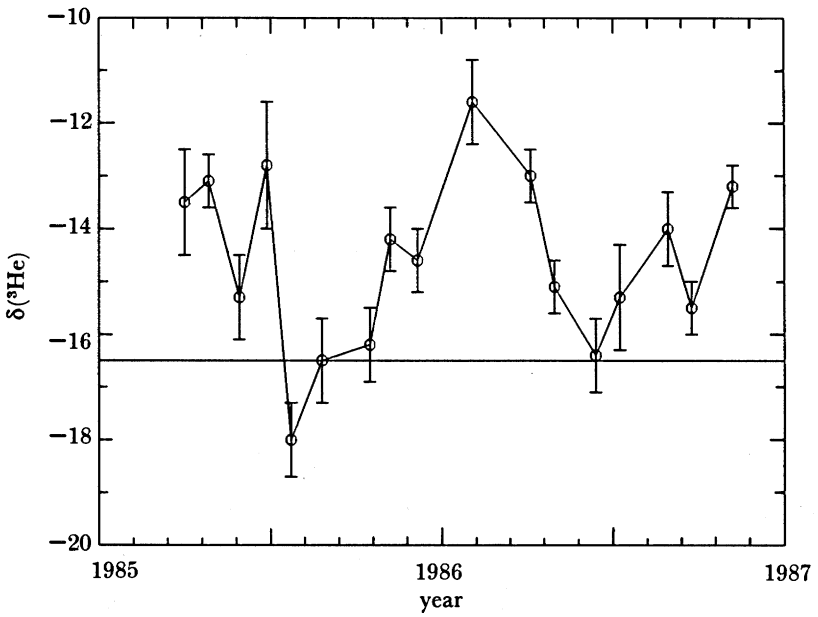

FIGURE 8. The average mixed-layer $\delta(^3\mathrm{He})$ against time for the Bermuda series. Generally, about five samples from the mixed layer were averaged for each point, and the error bars indicate the standard deviation of the mean for each average. The approximate static equilibrium $\delta(^3\mathrm{He})$ is shown here as a horizontal line for visual reference.

The existence of this supersaturation in ^{3}He implies a flux from the sea to the atmosphere, and knowing the gas-exchange rate, it should be possible to estimate the annual average of this flux. From climatological wind data (see, for example, Bunker 1975), and an observed relation between the mean gas-exchange rate and mean wind velocity (Roether 1986), a climatological

time history of gas exchange rate can be constructed to an accuracy of order 20%. The more difficult task is to obtain a realistic estimate of the equilibrium isotopic ratio for helium to use as a baseline for estimating the actual ^{3}He supersaturation.

Although seawater equilibrium $\delta(^3\mathrm{He})$ has been determined with extreme accuracy (Benson & Krause 1980), the concentration of helium in the mixed layer is not in equilibrium. In general, air injection maintains North Atlantic surface waters about 5% supersaturated in helium (Jenkins & Clarke 1976). Because, as will be discussed below, the processes responsible are potentially fractionating, the resultant $\delta(^3\mathrm{He})$ may deviate substantially from the equilibrium value determined in the laboratory (see Fuchs *et al.* 1987).

For the purposes of the present calculations one can identify two differing modes of air injection. The first, hereafter referred to as 'trapping', involves the complete dissolution of bubbles forced so far downward by turbulence or wave action that hydrostatic pressure forces collapse and total injection of gases. This process is non-fractionating, but in dynamic balance with gas loss by gas exchange, the resultant supersaturation of two species is given by

$$\Delta_1/\Delta_2 = \alpha_2 D_2^{0.5}/\alpha_1 D_1^{0.5}, \tag{5}$$

where Δ, α, and D are the fractional supersaturation, Ostwald solubility factor and molecular diffusivity respectively (see Fuchs *et al.* 1987). The $\delta(^3\mathrm{He})$ resulting from 5% helium supersaturation at 25 °C would be -18.8‰, compared to the 'static equilibrium' value of -16.3‰.

The second mode of air injection consists of 'partial injection', which occurs when bubbles undergo only slight dissolution, and escape before significant gas loss (or fractionation of the gas within the bubble) occurs. Such a mode of injection may be important in the swaths of bubbles created by whitecaps. The transport of gas from the bubble to the water is diffusively limited within the boundary layer, introducing an additional fractionation, dependent on the two thirds power of the molecular diffusivity (Levich 1962). Thus for a simple mass balance, one obtains

$$\Delta_1/\Delta_2 = \alpha_2 D_1^{0.167}/\alpha_1 D_2^{0.167}, \tag{6}$$

which differs from the other mode significantly. For the example discussed above, the corresponding $\delta(^3\mathrm{He})$ would be -14.4‰. Considering the observations (figure 8), the difference in 'baseline' between the two endmembers would result in factor of three difference in the predicted saturation anomaly in ^{3}He, and hence the flux.

These two modes of air injection are clearly endmembers, and what actually occurs will be a complex spectrum of intermediate degrees of bubble destruction, which will change as a function of wind speed and wave spectra. Within the framework of this discussion, we regard the problem as simply a linear mixture of the two modes, recognizing the limitations of this approach. It remains, however, to discern with adequate accuracy, the relative contributions of these modes. To do this, an additional constraint is needed.

The above equations may be applied to different gases. For neon, the predicted ratios of saturation anomaly (relative to helium) are 1.03 and 0.81 for the trapped and partial components respectively. An additional complication, however, results from radiative heating and mixing induced supersaturations, which are significant for neon. This may be seen (figure 9*a*) in a regression of ΔNe against ΔHe for the GEOSECS Atlantic data (GEOSECS 1987), where the intercept is significantly non-zero ($\Delta\mathrm{Ne}_0 = 1.1 \pm 0.3$%). To account for these effects, we employ a one-dimensional model similar to the one used successfully to simulate the seasonal

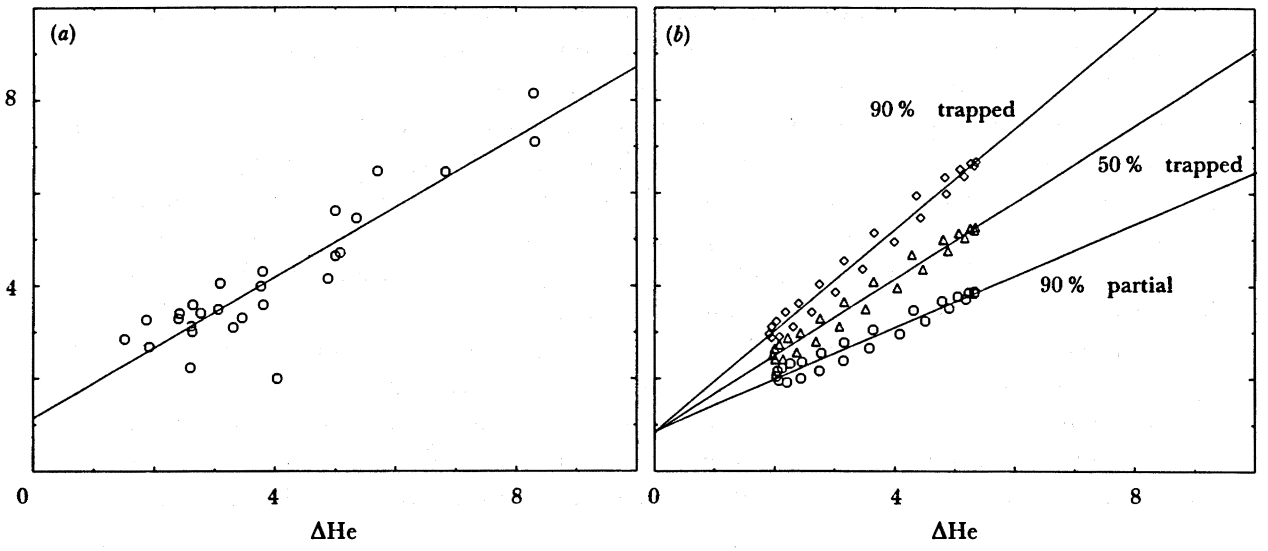

FIGURE 9. The relation between excess neon and excess helium are shown for (*a*) the GEOSECS Atlantic upper ocean data, and (*b*) a numerical seasonal mixed-layer simulation. The regression line in (*a*) corresponds to $\Delta\mathrm{Ne} = (1.1 \pm 0.3) + (0.76 \pm 0.07)\ \Delta\mathrm{He}$, with a linear correlation coefficient of 0.91 for 27 points. The three curves shown in (*b*) are for three representative mixtures of partial and trapped modes of air injection. Note the hysteresis evident in the model data points due to seasonal heating effects on ΔNe.

cycling of oxygen (Musgrave *et al.* 1988) and argon (Chou 1985). The only addition to this model is a wind-dependent air injection component given by

$$W_i = W_0 \cdot u_{10}^{\gamma} (f_t + f_p \cdot \Gamma),$$

where W_i is the injection rate of the gas, W_0 is an adjustable parameter related to the flux of bubbles (and the same for all gases), u_{10} is the wind speed, γ is an adjustable power-law dependence, f_t and f_p are the relative fractions of the trapped and partial components, and is constrained by $f_t + f_p = 1$. Finally, Γ is a normalized diffusivity factor associated with the boundary-layer diffusive limitation for gas transfer from the bubbles, given by

$$\Gamma = D^{0.667} / D_0^{0.667},$$

where D_0 is the molecular diffusivity of helium at 20 °C.

Aside from the relative fractions f_t and f_p, the functional dependence of air injection employed here employs only two unknown parameters. These were adjusted to obtain agreement with observations of the mean helium supersaturation (*ca.* 4.5 %) and its annual range (from *ca.* 2 % in summer to *ca.* 6 % in winter) observed in this time-series. It is encouraging that the optimal case exhibited a cubic dependence on the wind speed, in agreement with acoustic observations of bubble populations (Crawford & Farmer 1988). Within the above constraints, the relation between ΔNe and ΔHe was explored for various mixtures of partial and trapped air injection components. Three regression lines are shown in figure 8*b* for a range of extreme cases. Note that the zero ΔHe intercept for all these is within the errors of that obtained for the GEOSECS observations. From the regression slope and its uncertainty from the GEOSECS data, the optimal mix of partial against trapped components and its uncertainty is determined to be $f_p = 0.63 \pm 0.13$.

This mixture is not inconsistent with the strong attenuation with depth of bubble populations

(e-folding depths of order 1 m) observed by Crawford & Farmer (1988), because this would imply a large probability of bubble escape before collapse.

The model can then be used to compute the 'dynamic equilibrium' $\delta(^3\mathrm{He})$ in the mixed layer as a function of time. This, and its uncertainty is shown in figure 10*a*, along with a sinewave fit to the observations. Combined with the climatological gas-exchange rate (see, for example, Musgrave & Jenkins 1987), the annual flux of ^{3}He can be computed (figure 10*b*). The mean gas-exchange flux from the mixed layer is thus computed to be 21 ± 6‰ m d^{-1}. The uncertainty includes contributions due to uncertainty in the dynamic equilibrium $\delta(^3\mathrm{He})$ and a 20% uncertainty in the gas-exchange rate.

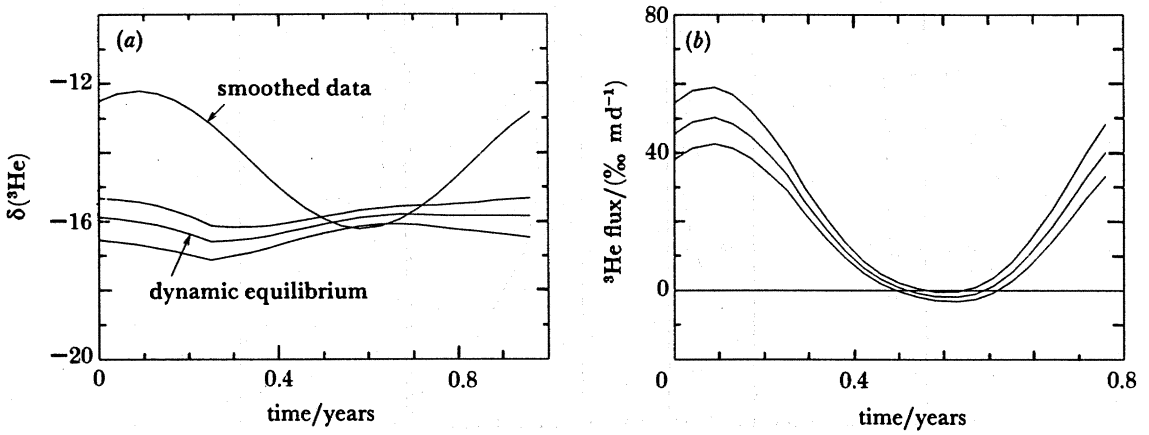

FIGURE 10. (*a*) Comparison of the range of dynamic equilibrium $\delta_0(^3\mathrm{He})$ (lower three curves) predicted by the upper-ocean model, and as restricted by the ΔNe–ΔHe results with a sinusoidal fit to the time-series data. (*b*) Predicted time history of the ^{3}He flux from the ocean to the atmosphere based on the model $\delta_0(^3\mathrm{He})$ and climatological gas-exchange rates obtained from the Roether (1986) windspeed-piston velocity relation and climatological wind data (Bunker 1975).

'Local' production of ^{3}He within the upper ocean can be estimated to be about 10% of the observed flux (in the upper 100 m), so that the upward flux of this isotope from the waters below is 19 ± 6‰ m d^{-1}. This flux represents the 'mining' of tritiugenic ^{3}He from the main thermocline. By comparison of the 1986 profiles with tritium and ^{3}He data obtained from this station in early 1977, it is possible to compare this flux to the long-term changes in the inventory of these isotopes in the main thermocline. The equivalent change rates in the tritium and ^{3}He inventories in the upper 700 m (i.e. above the oxygen minimum) are 12 and 4.5‰ m d^{-1} respectively. The combined flux agrees well with mixed-layer flux estimate.

What does this flux imply about vertical transport? Using the observed vertical gradient of ^{3}He in the upper main thermocline (figure 7*a*) of 0.17‰ m^{-1}, one would require a vertical turbulent diffusivity of approximately 10^{-3} m^2 s^{-1}, a number that is unreasonably large by any standard (see, for example, Jenkins 1980). It is, of course, unreasonable to demand a local balance between the observed mixed layer flux of ^{3}He and the thermocline inventory changes. The tritium–^{3}He age maps discussed earlier, however, point to the Gulf Stream recirculation region as an important window of ventilation for the shallow isopycnals, so that we may indeed be observing part of the process of ventilation.

The data do contain an additional hint as to the nature of one possible mode of return of ^{3}He to the surface layer. Figure 11 is a comparison beween two reoccupations of the Bermuda

Site (stations 593 and 597) in July and August 1986. The earlier station (solid line) shows anomalous structure compared to the later one (broken line), which is more typical of the series. The anomalous data show significant amounts of nitrate in the bottom of the euphotic

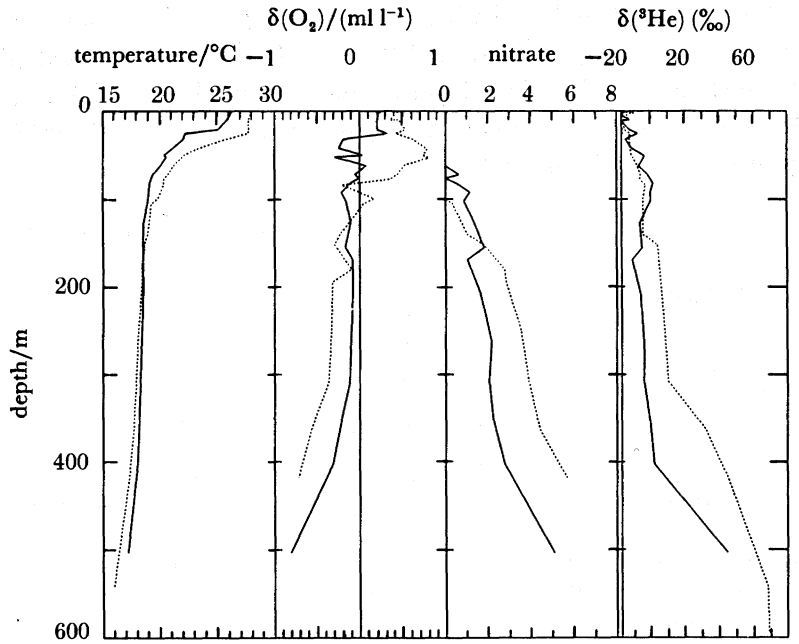

FIGURE 11. Property profiles for two stations from the time-series at Bermuda. Station 593 (7 July 1986, solid line) exhibits anomalous profiles compared with a more typical station (Station 597, 25 August 1986, broken line). Note especially the anomalous spike of nitrate and ^{3}He in the lower part of the euphotic zone (75–100 m), and the comparatively young subtropical mode water below.

zone, accompanied by substantial amounts of excess ^{3}He. The amount of ^{3}He associated with this 'spike' would require in excess of two years of isolation to accrue, indicating that the spike must have come from below. Further, the lifetime of nitrate within the euphotic zone is sufficiently short (less than a day), that the injection must have occurred within the very recent past. Integrating the vertical distribution of the excess ^{3}He in the upper 150 m (the depth accessable by winter convection), one estimates that this spike can account for 20–30 % of the annual flux of ^{3}He. Thus one would require only a few of these events to support the observed flux.

It is noteworthy, however, that the peculiar conditions observed at station 593 also are accompanied by a unique structure in the subtropical mode water below. The mode water for this station is characterized by anomalously young tritium–^{3}He age (about half of normal), low nitrate and apparent oxygen utilization (AOU) (again about half of normal), and low potential vorticity. These combine to suggest the presence of a column of freshly ventilated mode water that has been advected around with the gyre-scale circulation. The timing, at least, appears consistent with this. Thus one may be tempted to assert that the introduction of ^{3}He and nutrients into the upper waters may be related to some dynamical feature of this entity. However, in the absence of more complete data (in space and time), this remains rank speculation.

However, by combining large-scale mapping of these isotopes in the thermocline, with the

time-series of data that is evolving, we are beginning to observe and quantify the geochemically important processes of ventilation of the ocean.

I am indebted to my highly capable research staff (especially D. Lott, M. Pratt, S. Birdwhistel and M. Davis) for carrying out the demanding tasks associated with the measurements. Samples for TTO were acquired through the hard work of the PACODF group at Scripps Institution of Oceanography, for NATS with the help of C. Wunsch, D. Roemmich and H. Stommel, and for the Bermuda series by the excellent staff of the Bermuda Biological Station (including A. Knap, T. Jickells and R. Sherrif-Dow). Work on the above was funded by NSF grants no. OCE79-21378, OCE81-17998 and OCE85-01171 respectively.

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PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

Discussion

D. J. Webb (*Institute of Oceanographic Sciences, Godalming, Surrey, U.K.*). The changes produced by the near-surface mixing event discussed are very similar to those produced by events at the edge of a continental shelf. As the site mentioned is near to Bermuda, is it not possible that the mechanism responsible was connected with the island?

W. J. Jenkins. Studies have been made to determine the biological and physical effects of the island, and I do not recall any significant perturbations at this distance, except evidence of dynamic effects in near bottom current meter records. What leads me to suspect that the observed shallow entrainment phenomenon may be an open-ocean process is its apparent association with the deep 'ventilated' structure. Similar structures seem to have been observed in the open ocean (along 55° W) by McCartney *et al.* (1980).

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L. Merlivat (*Université Pierre et Marie Curie, Paris, France*). On the graph of ^{3}He–tritium showing the experimental data of the TTO samples, it appears that for a tritium content equal to zero, the ^{3}He content is not zero. Could Dr Jenkins comment on the origin of this ^{3}He amount, which seems to be associated with zero-tritium waters?

W. J. Jenkins. There are three components of the 'zero-tritium' ^{3}He to which Dr Merlivat refers. A simple extrapolation of the tritium against ^{3}He relation to zero tritium yields an intercept close to 0.5 TU. Part of this (perhaps half or more) may be due to the complete decay of pre-anthropogenic tritium. Work by Roether and co-workers at Heidelberg estimate pre-anthropogenic tritium concentrations in surface waters that are not drastically different from this. The remainder of this ^{3}He, the second component, is likely to be from the invasion of primordial helium via Antarctic intermediate and bottom waters. I interpret the ultimate source of this helium to be at least outside of the North Atlantic because it falls on the mixing curve.

The third component is undoubtedly indigenous injection of primordial ^{3}He from local sources, i.e. the Mid-Atlantic Ridge. We have seen evidence of such injections near the Charlie–Gibbs Fracture Zone (although on an irregular basis), and have observed them directly in the TAG hydrothermal area and the Kane Fracture Zone. Geometric considerations lead me to the conclusion that these sources are not a significant interference with the tritium–^{3}He story in the upper 1500 m in most of the Atlantic, but elsewhere, we must be careful.

W. Roether (*University of Bremen, F.R.G.*). I was very interested to hear that Dr Jenkins has observed mixed-layer excess ^{3}He so consistently. In our own data we seemed to find excesses stochastically in summer observations. Our conclusion was that the finding pointed to transient outcropping of thermocline waters, because the usual time-averaged vertical mixing coefficients

were clearly unable to support such excesses. If a large-enough data base could be collected, it might be possible to quantify the outcropping (or whatever it is) effect. Furthermore, I wonder whether Dr Jenkins accounted for the 2‰ depression in the equilibrium $\delta(^3\mathrm{He})$ values that our work suggests?

W. J. Jenkins. My results do not disagree with Professor Roether's observations. In fact they support them. The individual data points in the mixed layer show scatter that is considerably larger than analytical, and that I would attribute (as Professor Roether does) to random injections of tritiugenic ^{3}He from below. The summer data, in fact, are rather close to static equilibrium. The error bars on the mean mixed layer $\delta(^3\mathrm{He})$ include the observed scatter. Where our interpretations diverge, however, is in the modelling of the 'dynamic equilibrium helium isotope effect'. Whereas Professor Roether's work with Fuchs favoured a greater trapped component, and hence projected a 2‰ depression in the $\delta(^3\mathrm{He})$, my work points to a dominance of partial bubble injection, resulting in much less of a depression. However, our differences may well be geographic: Professor Roether's work (as I recall) is from an area where bubble trapping may be dominant. I would not contend that my data and model apply outside of the western subtropics. It is clear, however, that further work would be of considerable value.

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES