

ON THE ^{14}C TO TRITIUM RELATIONSHIP IN THE NORTH ATLANTIC OCEAN

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ABSTRACT. Nuclear-weapon produced ^{14}C (or bomb ^{14}C) in the ocean can be traced by simultaneous tritium observations. Data are presented on the general relationship of bomb ^{14}C and tritium in the North Atlantic. For the period 1965 to 1973, the excess ^{14}C to tritium ratios in the surface water vary, systematically, over a factor of 10: the ratios monotonically increase with time, and decrease with latitude, particularly so for the later observations. The sub-surface water ratios show that the mid- and low-latitude water below about the 15°C isothermal horizon ($\sim 500\text{m}$ depth) originates from higher northern latitudes, rather than being renewed by local vertical mixing. It is further shown that in the North Atlantic, bomb ^{14}C did not penetrate beyond the horizon where the presently observed ^{14}C concentration is $\Delta^{14}\text{C} = -75\%$. Observed concentrations up to about -40% can be corrected for a bomb contribution if the tritium concentration is known because the bomb ^{14}C to tritium concentration ratio is rather uniform in this range. A surface water ^{14}C concentration versus time curve is presented for the period since 1957. This curve is based on a North Atlantic mixing model and is fitted to the ^{14}C observations. Making use of a previously published tritium versus time curve obtained by the same model, a time curve for the average excess ^{14}C to tritium ratio in North Atlantic surface water is given. This curve reproduces the observations well. The presented data and theoretical curves show the usefulness of simultaneous ^{14}C and tritium observations for mixing studies and to provide corrections for bomb ^{14}C in sub-surface ^{14}C data in the North Atlantic.

INTRODUCTION

One of the principal applications of ^{14}C is to measure time in large-scale convective and mixing processes in the deep and bottom waters of the world ocean (eg, Broecker and others, 1960; Bien, Rakestraw, and Suess, 1960; Stuiver, 1976; Ribbat, Roether, and Münnich, 1976; Broecker, 1979) on the basis of the distribution of natural ^{14}C . Further up in the water column, however, and in the vicinity of deep-water formation areas, the natural distribution has been disturbed by addition of ^{14}C that has originated from the nuclear weapon testing of the past two decades, and, preceding the nuclear era, to a lesser extent also by ^{14}C dilution effected by the burning of fossil fuel (Suess-effect) (Suess, 1955).

Nuclear weapon produced, or bomb, ^{14}C has always been accompanied by tritium. The natural tritium concentration of ocean water has been small (Dreisigacker and Roether, 1978), so that virtually all oceanic tritium is bomb produced. Therefore, tritium measurements, simultaneous with ^{14}C measurements, are an excellent tool to define the spatial limits of bomb ^{14}C in the ocean. Furthermore, such simultaneous data allows correction for the bomb ^{14}C component, as long as this component is sufficiently small (Roether, 1972; Broecker, 1979).

In the following, we explore the general correlation of ^{14}C and tritium for the North Atlantic. We present simultaneous ^{14}C and tritium data on surface and sub-surface samples taken since 1965. The surface water bomb ^{14}C to tritium ratios show systematic areal variations and an increase in time. The time increase is explained using a North Atlantic

mixing model of the near-surface water (Weiss, Roether, and Dreisigacker, 1979).

Simultaneous ^{14}C and tritium measurements

Our group has been collecting ^{14}C data in the North Atlantic since about 1965, both on surface and sub-surface waters, virtually all on research cruises of the West German *F S Meteor*. From the beginning, the ^{14}C sampling was always accompanied by tritium sampling. ^{14}C sub-surface sampling was made, using 50L stainless steel water samples¹, and since 1971, using 270L stainless steel Gerard-Ewing samplers (Roether, 1971), and surface water was pumped. Since 1971, hydrographic data are available for all ^{14}C samples (Roether and others, in press). Previously, no such data were taken and, therefore, admixture of water from shallower depths cannot be excluded with certainty. From the ^{14}C water samples, CO_2 was extracted aboard ship and taken to the laboratory for ^{14}C measurement, using procedures described previously (Ribbat, Roether, and Münnich, 1976). The tritium sample handling and measurement also is described elsewhere (Roether and Weiss, 1978; Weiss, Roether, and Bader, 1976).

The observed correlation

Figure 1 summarizes the surface water observations (for positions, see map, fig 2). The data were obtained during 1965-66, 1969, and 1972-73. The 1972 data are from the US GEOSECS program (Östlund, Dorsey, and Brescher, 1976). For each of the 3 periods, the data have been connected by V-shaped curves with their corners directed towards the upper right, which are supposed to show the general trend in this data. The higher latitude data fall near the (lower) right end of the respective curves, and equatorial values at the (upper) left end. The 1965/66 and 1969 data are for the eastern Atlantic (near or east of 30°W). The 1972 GEOSECS data south of 60°N are for the western Atlantic; eastern Atlantic data for this period (*Meteor*, 1973), fall on a different curve.

The thin straight lines represent lines of constant bomb ^{14}C to tritium ratio, *ie*, points along one such line represent water that has received bomb ^{14}C and tritium in a constant ratio but in varying amounts. The plot of figure 1 allows amount and ratio to be distinguished. Specifically, the curves show that, while the bomb ^{14}C concentrations are highest at mid-latitudes, the bomb ^{14}C to tritium ratios decrease monotonically with latitude (eg, in 1969 $\sim 30\text{‰}/\text{TU}$ at 10°S to 10°N , and $\sim 8\text{‰}/\text{TU}$ at 48°N to 60°N). The 1972/73 data furthermore show appreciably larger ratios in the western than in the eastern Atlantic.

The origin of the thin lines was chosen as -50‰ $\Delta^{14}\text{C}$ and 0.2 TU, which are the presumed pre-nuclear surface water concentrations. The 0.2 TU have been derived theoretically (Dreisigacker and Roether, 1978). The -50‰ are somewhat uncertain, not the least because the Suess effect in the surface water amounted to about 10‰ (see Broecker, 1979). However, a change of 10‰ would have little effect on the bomb ^{14}C to tritium ratio that is read from figure 1.

¹ Courtesy of H Kautsky, Hamburg.

Figure 3 shows ^{14}C versus tritium plotted for selected depth profiles, for which much less simultaneous data are available. Most of the data are from the US GEOSECS program (*Knorr* 30, 1972). ^{14}C and tritium measurements are from different depths for this cruise. An interpolation procedure, explained in the caption to figure 3, was therefore used to estimate corresponding ^{14}C and tritium concentrations. While the interpolation introduces some uncertainty, the basic features certainly are unchanged. The surface water points are distinguished by the latitude of the respective profile being shown at these points. Some of the shallow-water points are connected by broken lines.

In figure 3, the wide variation of the surface water data points (see fig 1) continues into the near-surface water. However, towards mid-depths there is a convergence of the data points towards a unique curve and towards a nearly uniform bomb ^{14}C to tritium ratio. From inspection of the accompanying hydrographic data it appears that the more uniform ratios are bounded by about the 15°C horizon. In the waters below this horizon, ^{14}C to tritium ratios, by comparison with figure 1, support the notion that these waters, in lower to mid-latitudes, originate from higher latitudes rather than being renewed by local vertical mixing. Otherwise, bomb ^{14}C to tritium ratios would have to be larger. Moreover, the water is an apparent two-component mixture, with the bomb

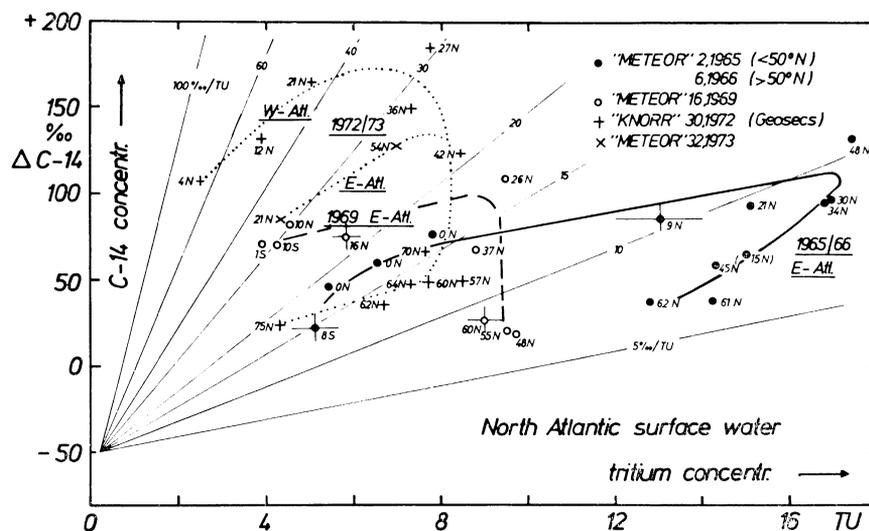


Fig 1. ^{14}C plotted versus tritium in North Atlantic surface water, 1965 to 1973. Latitude is shown with each data point, sampling positions; see figure 2. *Meteor* data points (cruises no. 2, 6, 16, and 32) are unpublished; *Knorr* data points are from Östlund, Dorsey, and Brescher (1976). Error bars (± 1 sigma) are given for some of the older Heidelberg data. ^{14}C in Δ -notation (Broecker and Olson, 1961) and tritium in TU ($1 \text{ TU} = [\text{T}]/[\text{H}] \cdot 10^{15}$). The full, broken and dotted lines (for 1965/66, 1969, and 1972/73) are drawn by eye to indicate the general trend in the data. Thin straight lines labelled 5 to $100\text{‰}/\text{TU}$ give lines of addition of bomb ^{14}C and tritium in a constant ratio. The 15°N data point, 1965, is from an upwelling area and may, therefore, be untypically low in ^{14}C . For further explanation, see text.

component free end member having characteristics of -75‰ and 0 TU. The so extrapolated $\Delta^{14}\text{C}$ value apparently is reasonably constant in the North Atlantic (see also, Broecker, 1979). In other words, the horizon at which the bomb component vanishes in the North Atlantic has a quite uniform ^{14}C concentration of about -75‰ . This apparent uniformity is explained by the relatively fast turnover of the North Atlantic intermediate and deep water: Broecker (1979) shows that the turnover time of the deep water in the northwest Atlantic is only about 100 years, and the shallower strata certainly have rather a faster turnover. Within 100 years, radioactive ^{14}C decay amounts to $\sim 12\text{‰}$, a concentration change that is small on the ordinate scale of figure 3. This means that water "old" enough to be essentially free of tritium (*ie*, ≥ 25 years), is still so young that radioactive ^{14}C decay can only produce ^{14}C concentration differences that are small compared to the bomb ^{14}C increase in the near-surface waters of the North Atlantic, Figure 3 indicates that for ^{14}C concentrations below about -40‰ , a uniform bomb ^{14}C to tritium ratio can be used to extrapolate back to the pre-bomb concentration, from a simultaneous tritium measurement. Such extrapolations are given elsewhere (Roether and others, in press).

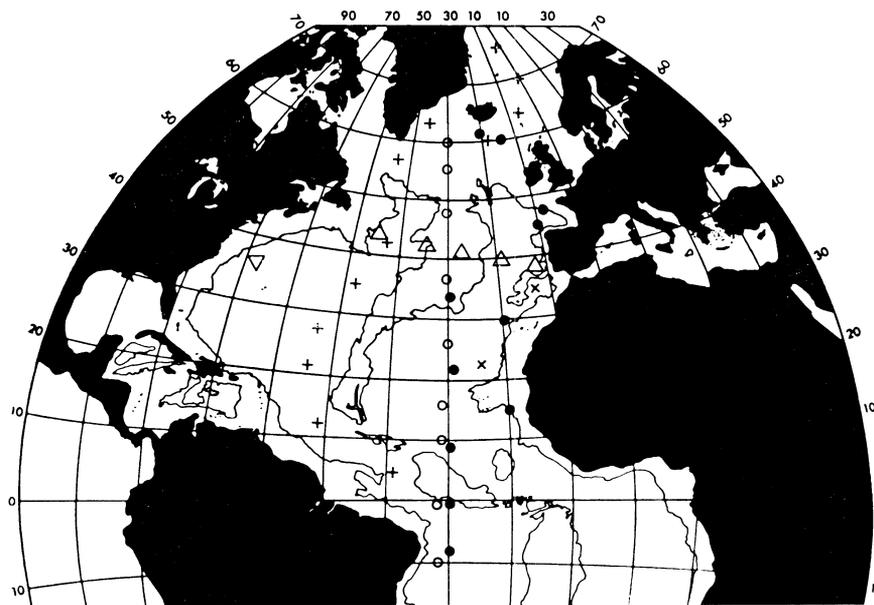


Fig 2. Station positions in the North Atlantic for data in figures 1 and 3.

- Meteor 2, 1965 and 6, 1966
- Meteor 16, 1969
- ▽ Knorr 9, 1970
- △ Meteor 23, 1971
- + Knorr 30, 1972
- × Meteor 32, 1973

In the waters above the 15° C horizon, or for ^{14}C concentrations above about -40‰ , the data points of figure 3 indicate the waters to be mixtures of surface water with the local water at the said 15° horizon (see broken lines for shallow water data at 12° N and 21° N). Unfortunately, at the GEOSECS stations at 12° N and 4° N, there are only few ^{14}C and tritium measurements available above 15° C, and the applicability of the mentioned interpolation procedure is restricted because subsurface tritium maxima are indicated in the data. It can be seen from figure 3 that such maxima are in fact implied also by the present interpretation.

In summary, figure 3 suggests two mixing regimes to exist separated by about the 15° C horizon, *ie*, one of lateral advection or mixing from

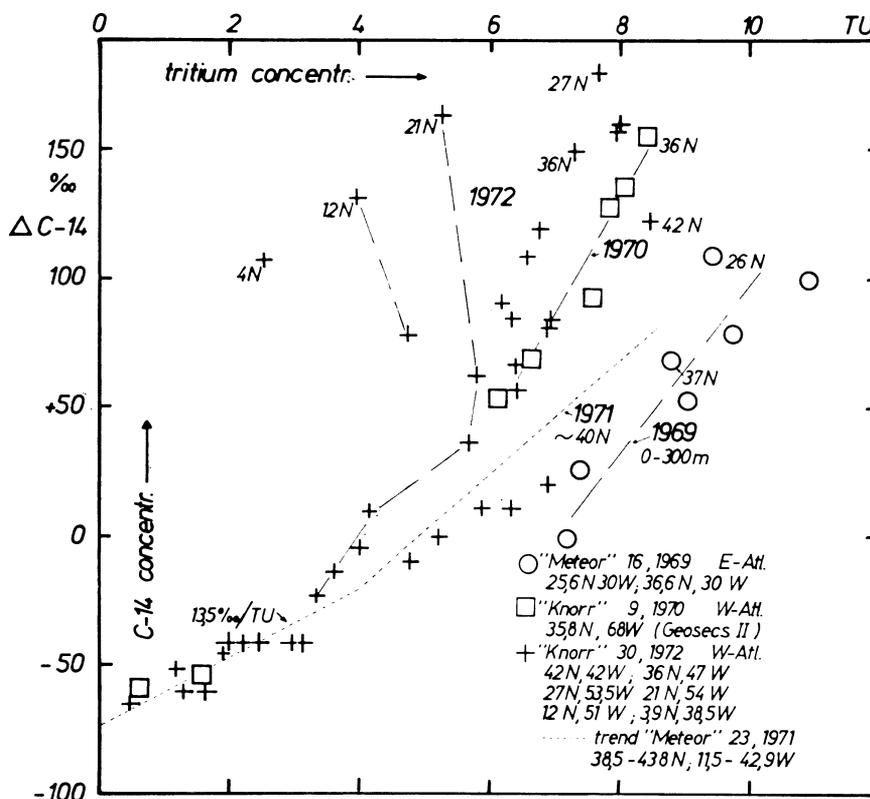


Fig 3. ^{14}C plotted versus tritium for North Atlantic depth profiles. *Knorr* 1970 tritium values from Roether and Münnich (1972); ^{14}C data are unpublished. For *Meteor* 1971 data, see Roether and others (in press). The latitude is shown with all surface water values. ^{14}C data for *Knorr* 30, 21° N, were taken from Stuiver (1978). Units and origin of other data as in figure 1; for station positions, see figure 2. For the *Knorr* 1972 sub-surface data, ^{14}C and tritium measurements were not made on the same water. Simultaneous concentrations were estimated by interpolating the property which has the better depth resolution to a measured value of the respective other property; interpolation was made linear by salinity or temperature; for shallow depths, for which neither salinity nor temperature is available for the ^{14}C data, interpolation was linear with depth.

higher latitudes below, and one of more local, and perhaps vertical, mixing above. The present data base is so limited, however, that this conclusion, and even more so, the latitudinal extension of the feature, must remain tentative.

Model bomb ^{14}C concentrations and bomb ^{14}C to tritium ratios in North Atlantic surface water

Quite apart from the problem of available data, a quantitative interpretation of ^{14}C to tritium ratios is difficult because bomb ^{14}C to tritium ratios in surface water vary both areally and temporally, and the ratios are further changed by radioactive tritium decay once a parcel of water has been removed from the surface water layer. As a first step towards a quantitative treatment, in the following we give a theoretical time curve for the bomb ^{14}C to tritium ratio in the North Atlantic surface water, including an account of radioactive tritium decay away from the ocean surface. The treatment is for concentrations averaged areally in the North Atlantic. Whereas for tritium the areal variation is small (Dreisigacker and Roether, 1978), areal variation for ^{14}C is appreciable (fig 1). Thus, if deductions are made from the results of the following treatment, effects of areal ^{14}C variation must always be considered.

A continuous time curve of the bomb ^{14}C concentration in North Atlantic surface water was calculated following a procedure applied previously to tritium (Dreisigacker and Roether, 1978; Weiss, Roether, and Dreisigacker, 1979), in which the input from the atmosphere is converted into surface water concentrations by means of an oceanic two-box mixing model (fig 4). ^{14}C from the atmosphere, at rate Q , enters the upper box (average depth d_1 [m]) that represents the surface water. From

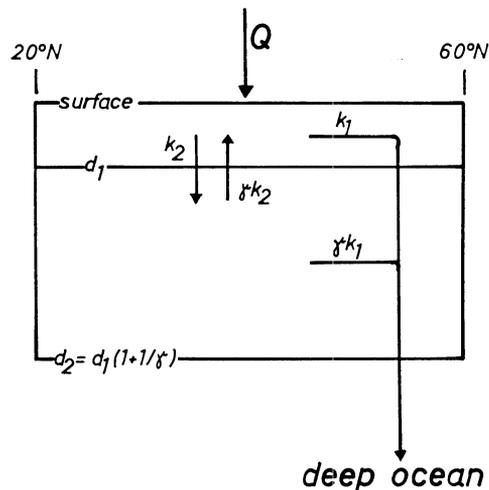


Fig 4. North Atlantic two-box mixing model, 20° N to 60° N (Dreisigacker and Roether, 1978; Weiss, Roether, and Dreisigacker, 1979). For parameter values, see table 1; for explanation, see text.

there it is mixed down into a sub-surface water box 2; both boxes release ^{14}C into the deep ocean. The input is specified as

$$(1) \quad Q(t) = w \cdot (c_0 - c_1),$$

where w = transfer coefficient, $\text{mol CO}_2/\text{m}^2 \cdot \text{year}$

c_0 = atmospheric ratio [$^{14}\text{C}/^{12}\text{C}$], relative to ^{14}C standard

c_1 = surface water ratio.

This leads to a time change of the box 1 concentration at a rate

$$(2) \quad k_0 = \left(\frac{\partial \ln(c_0 - c_1)}{\partial t} \right) Q = \frac{w}{\Sigma \text{CO}_2 \cdot d_1} (\text{year}^{-1}),$$

where ΣCO_2 = total dissolved anorganic carbon in surface water, 2.00 mol/m^3 .

Table 1 gives the model parameters and the governing equations. The equations were integrated numerically using forward differentiation in time and a quarter-year time step. Atmospheric concentrations, $c_0(t)$, were taken from Broecker, Peng, and Stuiver (1978). The transfer coefficient was not specified but rather obtained by fitting to surface water observations². The calculated bomb component is added to an assumed pre-nuclear surface water concentration of -50 per mil. The resulting surface water concentration versus time curve is shown in figure 5A (lower panel). The observations to which the curve is fitted were taken from figure 1. In view of the areal variability and limited areal coverage, averages cannot be specified unambiguously. However, the averages are sufficient to define the magnitude of the bomb-induced concentration increase in the surface water.

Figure 5B (upper panel) reproduces a published surface water curve for tritium (Dreisigacker and Roether, 1978) that was obtained using the same procedure. From the two surface water curves, bomb ^{14}C to tritium concentration ratios were calculated and are included in figure 5B. The curve labelled time-integrated ratio gives the ratios defined by

$$(3) \quad \int_{1952}^t C_{\text{bomb } ^{14}\text{C}}(t') \cdot dt' \quad / \quad \int_{1952}^t C_{\text{tritium}}(t') \cdot e^{-\lambda \cdot (t-t')} \cdot dt'$$

where $\lambda = 0.0564/\text{year}$, radioactive tritium decay constant. Mid-year

² The fit requires $k_0 = 0.088/\text{year}$, or via equation (2), $w = 33 \text{ mol/m}^2 \cdot \text{year}$, which transfer coefficient is about 50 percent larger than estimates for the real ocean (Peng and others, 1979). The larger transfer arises in the model formally because the removal from box 1 by internal transfers into box 2 and into the deep ocean acts on the *average* box concentrations. In nature, the transfers occur largely in higher latitudes ($\geq 4^\circ \text{N}$), where, according to figure 1, the concentrations are lower than average. The model removal, thus, is larger than the real one, and this is compensated by a larger than real atmospheric input. Additionally, in the real ocean, ^{14}C is advected from the South Atlantic, which inflow is not taken into account in the model, so that, again, a rather higher atmospheric input is formally required. It appears that the actual excess input (~ 50 percent) is very reasonable. It has been shown for the case of tritium (Dreisigacker and Roether, 1978), that the time dependence of the model concentrations primarily depends on that of the atmospheric input, rather than on the model parameter values. The same should hold for ^{14}C , so that, despite the above, the model ^{14}C concentrations should be realistic.

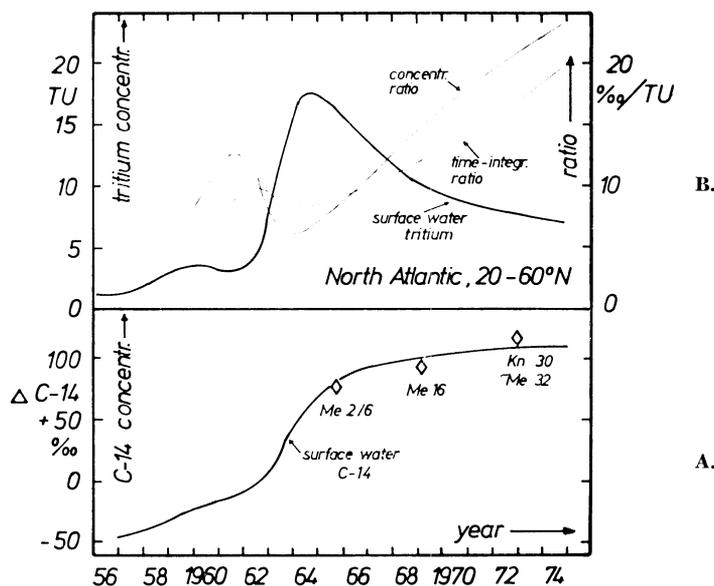


Fig 5A (lower panel). Theoretical ¹⁴C concentration versus time curve obtained using the model of figure 4. The three observational points (*Meteor 2/6*, *Meteor 16*, and *Knorr 30/Meteor 32*) were read from figure 1.

B (upper panel). Tritium versus time curve for North Atlantic surface water (from Dreisigacker and Roether, 1978), and bomb ¹⁴C to tritium ratio. Concentration ratio is the ratio obtained from the two surface water curves; time-integrated ratio from eq (3).

TABLE I
Parameters of the North Atlantic two-box mixing model*

a) Parameter values

Model area (20°-60° N)	26.9 × 10 ⁶ km ²
Depth of surface-water box (d ₁)	187m
Depth of sub-surface box (d ₂)	534m
Volume of surface-water box	5.03 × 10 ⁹ km ³
Volume of sub-surface box	14.36 × 10 ⁹ km ³
Rate constant for deep water formation (k ₁)	1/30 year ⁻¹
Rate constant for internal exchange k ₂	1/2.5 year ⁻¹
Box volume ratio (γ)	0.35

b) Governing equations (radioactive ¹⁴C decay is neglected)

$$\frac{\partial c_1}{\partial t} = k_0 \cdot (c_0 - c_1) + k_2 \cdot (c_2 - c_1) - k_1 \cdot c_1$$

$$\frac{\partial c_2}{\partial t} = \gamma \cdot k_2 \cdot (c_1 - c_2) - \gamma k_1 \cdot c_2$$

* Figure 4 (Weiss, Roether, and Dreisigacker, 1979) and governing equations.

^{14}C concentrations and ratios are listed in table 2. The *time-integrated* ratio is the ratio found in a sub-surface water mass to which surface water is added at a constant rate with no water removed, as in the case of the limit of slow renewal by surface water and fast internal mixing. The straightforward ratio is the limiting case for fast renewal. Figure 5b shows that the time-integrated ratios do not vary much during 1962 to 1968. In recent years, both ratios increase by about 8 percent/year, *ie*, somewhat faster than would correspond to radioactive tritium decay only (5.6 percent/year).

In the range of low bomb component, or of the lower mixing regime as discussed above, the observed ^{14}C to tritium ratios (fig 3) are smaller than the values read from figure 5B (13.5 versus 15 to 20‰/TU, 1971). This arises because the surface waters that feed the water in question originate from latitudes north of about 40° N, where ^{14}C to tritium ratios have been lower than average (fig 1). For water originating from low latitudes, on the other hand, the 15 to 20‰/TU should be a lower limit for the actual ratios. However, the observed ratios in the range of low bomb component for the lower latitude stations do not differ significantly from those for the higher latitude stations. It follows that the argumentation for a general higher latitude origin of this water of the previous section can be upheld also in the face of radioactive tritium decay, which is taken into account in the curves of figure 5B.

TABLE 2
Model ^{14}C concentrations in North Atlantic surface water
(areal average 20° to 60° N), and bomb ^{14}C to tritium ratios,
1957 to 1974*

	^{14}C conc Δ ‰**	Bomb ^{14}C to tritium ratio ‰/TU	Time-integrated ratio ‰/TU†
1957	-43		
58	-36		
59	-28	6.8	5.6
60	-20	8.9	7.0
61	-13	12.6	8.6
62	-1	11.0	9.8
63	+26	6.5	8.9
64	57	6.1	8.3
65	78	7.6	9.0
66	87	9.4	9.2
67	94	11.3	10.1
68	98	13.6	11.2
69	101	15.4	12.3
70	104	17.1	13.5
71	106	18.3	14.7
72	107	19.9	16.0
73	108	21.4	17.4
74	109	22.7	18.9

* The early values are uncertain because the pre-nuclear ^{14}C concentration is only approximately known. Tritium concentrations from Dreisigacker and Roether (1978).

** Mid-year, assumed pre-nuclear concentration -50‰.

† Eq (3).

CONCLUSION

It appears that in the North Atlantic, simultaneous ^{14}C and tritium measurements are potentially useful to reconstruct the distribution of natural radiocarbon wherever the bomb ^{14}C component is small. Moreover, bomb ^{14}C to tritium ratios can give information on source areas for mid-depth waters in mid and lower latitudes. We have explored the general pattern of the bomb ^{14}C to tritium ratios, and have given a quantitative account of a special aspect of it, *ie*, the variation in time of the areally averaged ratios. We regard our discussion to be of a phenomenologic nature. A truly three-dimensional and time dependent treatment will be necessary for a quantitative assessment.

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DISCUSSION

Siegenthaler: Can you make an estimate of pre-bomb surface ^{14}C concentrations by means of a correlation between ^{14}C and bomb tritium values?

Roether: In order to determine the excess ^{14}C /tritium ratios one has to know (or assume) a pre-bomb surface water ^{14}C value. Particularly, the northern North Atlantic is a region where a better assessment of the pre-nuclear surface-water ^{14}C concentration would be desirable. I assume that something like shell data are the way to go.