

the right on the Sr-Ca peak. This sample was determined to be pure aragonite by XRD (7). The dashed line in Fig. 5 is the Fourier transform of the Sr K-edge EXAFS spectrum for coral AP-14. A linear combination of ~60% Sr substituted in aragonite and ~40% Sr in a SrCO₃ phase approximates ($\pm 15\%$) the EXAFS of AP-14 and all of the MA corals. The remaining two AP corals did not show as large a SrCO₃ shoulder on the doublet at ~4 Å, suggesting a more homogeneous distribution of their Sr in the aragonite structure.

Plummer and Busenberg (6) showed that there was a wide miscibility gap in the aragonite-SrCO₃ solid solution from a SrCO₃ mole fraction of 0.0058 ± 0.0003 to 0.875 ± 0.011 at 25°C. Aragonite with more than ~5000 ppm of Sr substituted for Ca is therefore metastable. For the two-phase corals, our estimate of 4500 ppm of Sr at the Ca structural sites in aragonite (60% of 7500 ppm) and 3000 ppm in the SrCO₃ (40% of 7500 ppm) agrees with thermodynamic analysis of the laboratory aragonite-SrCO₃ system (6). Most scleractinian corals grow between 25° and 29°C, a temperature range close to the 25°C of the synthetic system (6). The two AP corals exhibiting smaller SrCO₃ shoulders apparently formed as metastable high-Sr aragonite.

It seems reasonable that the SrCO₃ domains are primary features that formed during biomineralization. The domains are present in specimens that were collected live, and solid-state diffusion or dissolution-precipitation is difficult or slow in seawater at ambient temperatures. The size and distribution of the SrCO₃ domains is not known, and neither is the mechanism by which corals sequester more Sr in aragonite skeletons than expected at equilibrium with seawater, although Plummer and Busenberg (6) suggest that precipitation kinetics cause nonequilibrium incorporation of Sr in many marine carbonates.

These findings indicate that Sr does not substitute solely for Ca at its structural sites in MA and AP, species that have not been directly calibrated for Sr/Ca SST paleothermometry. Further investigation of the SrCO₃ in these and other species will advance our understanding of the mechanisms of Sr incorporation into scleractinian coral.

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8. The materials for XAS analysis were ground to fine powders of 300 to 400 mesh size. The standards were mixed in Duco cement, and spread as a thin layer on Al foil. Beam line 4-1 at SSRL was used with a Si(111) (for Ca) and Si(220) (for Sr) monochromator. The stored electron beam energy was 3 GeV at a current of 50 to 90 mA. The monochromator was detuned to reduce harmonics, and the slits were set at 1 mm by 20 mm to give ~1- and 4-eV resolution at the Ca and Sr K-edges, respectively. The data were gathered in fluorescence mode for the corals and in absorption mode for the standards. The Ca K-edge experiment was performed in the electron yield mode, as noted in the data where appropriate.
9. The Fourier transform peaks were identified with the use of the software Shells for EXAFS-Interactive (B. Rupp, Lawrence Livermore National Laboratory) to calculate bond distances and coordination numbers. Unit cell parameters are available in R. W. Wyckoff, *Crystal Structures* (Interscience, New York, 1963), vol. 1, for all compounds except SrO₂, for which we made an estimate for the fractional parameter u of 0.391.
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Pulses of Rapid Ventilation in the North Atlantic Surface Ocean During the Past Century

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The Suess Effect is a gradual decrease of the ¹⁴C/¹²C ratio in tree rings from 1870 to 1950 that is attributed mostly to the burning of ¹⁴C-free fossil fuels for energy. In contrast, pre-nuclear bomb high-precision measurements of ¹⁴C/¹²C ratios in banded corals from the North Atlantic Ocean do not exhibit the expected Suess Effect. Instead, large biennial-to-decadal shifts in the ¹⁴C/¹²C ratios appear throughout the coral records. This pattern indicates that rapid pulses of increased mixing between surface and subsurface waters occurred in the North Atlantic Ocean during the past century.

Since the late 19th century, humans have burned substantial amounts of fossil fuels for energy. As a result of this combustion, the concentration of CO₂ in the atmosphere has increased by at least 25% (1). This increase represents about half of the CO₂ that has been emitted into the atmosphere; most of the other half is in the oceans. The decline in $\Delta^{14}\text{C}$ (2) values of atmospheric CO₂, due to CO₂ release from fossil fuels and to cosmic ray flux variability, has been measured in tree rings [–20 per mil (3)] and is known as the Suess Effect (3, 4). Because atmospheric CO₂ exchanges with dissolved inorganic carbon (DIC) in the surface ocean, $\Delta^{14}\text{C}$ measurements in the upper ocean can be used to quantify the main sink for fossil-fuel CO₂. Coralline aragonite has been shown to accrete calcium carbonate

with the same $\Delta^{14}\text{C}$ signature as that in DIC in the surrounding seawater (5, 6). The $\delta^{18}\text{O}$ (7) values in banded corals have been shown to correlate inversely with sea surface temperature (SST) (8–10) and to correlate directly with the salinity and $\delta^{18}\text{O}$ of seawater (11, 12). Here I report high-precision $\Delta^{14}\text{C}$ measurements of banded corals from three sites in the North Atlantic Ocean and use the results to calculate the water mass renewal (or ventilation) rate in the North Atlantic over the past 100 years.

Cores of *Diploria strigosa* were collected from surface waters (at a depth of 5 m) near North Rock (32°29'N, 64°41'W; 14 km north of Bermuda), and *Montastrea annularis* corals were cored from Pickles Reef off Key Largo, 100 km south of Miami, and from The Rocks Reef (24°57'N, 80°33'W), 1 km south of Plantation Key in Florida. Biennial (2-year) bands were cut from the

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Bermuda and Pickles Reef coral, and 5-year bands were cut from The Rocks coral (13). Most of the samples were analyzed for $\Delta^{14}\text{C}$ with the use of conventional gas counting (13); a few samples were analyzed with accelerator mass spectrometry (AMS) at the University of Arizona (14). The $\Delta^{14}\text{C}$ measurements from conventional counting have 1σ uncertainties (counting statistics and laboratory errors) of 2.5 to 3.0 per mil. Uncertainties for the AMS measurements were 2.8 to 3.9 per mil (15).

The $\Delta^{14}\text{C}$ values for pre-1955 coral bands from Bermuda and Florida varied widely throughout the records (-34 to -64 per mil) (Fig. 1A). The depletion of $\Delta^{14}\text{C}$ in the surface ocean as compared with $\Delta^{14}\text{C}$ in atmospheric CO_2 [-2 to -25 per mil (3)],

known as the reservoir effect (16), was maintained by mixing of subsurface waters with lower ^{14}C into the surface ocean. Bermuda $\Delta^{14}\text{C}$ values before 1940 had a slightly higher average value (-43.9 ± 4.8 per mil; $n = 33$) than those from Florida (-47.4 ± 4.5 per mil; $n = 25$). There were three times when the $\Delta^{14}\text{C}$ values of the Florida coral were significantly lower than those of the Bermuda coral: 1890-1898, 1919-1924, and 1949-1952. Between 1947 and 1953, $\Delta^{14}\text{C}$ values at the two Florida sites averaged -58.2 ± 4.4 per mil ($n = 5$), which is indicative of a late Suess Effect (-11 per mil) and is comparable to a model-calculated trend (see Fig. 1A). Only two $\Delta^{14}\text{C}$ values for the Bermuda coral were low (average -52.4 ± 0.8 per mil between 1945

and 1948), which is opposite of the Florida trend and indicates the lack of a Suess Effect. Carbon-14 produced by atmospheric testing of nuclear weapons in the 1950s and early 1960s began to be evident in these corals between 1954 and 1956 (5, 17). Overall, the coral $\Delta^{14}\text{C}$ records showed a large amount of short-term variability, especially between 1910 and 1935, when the range of values within corals varied by up to 20 per mil. The range of $\Delta^{14}\text{C}$ values observed in the Bermuda and Florida corals (30 per mil) is as large as the global range of pre-bomb surface water values between the tropics, which are influenced by low- ^{14}C upwelled waters and the mid-gyre regions (18).

The stable oxygen isotope values ($\delta^{18}\text{O}$)

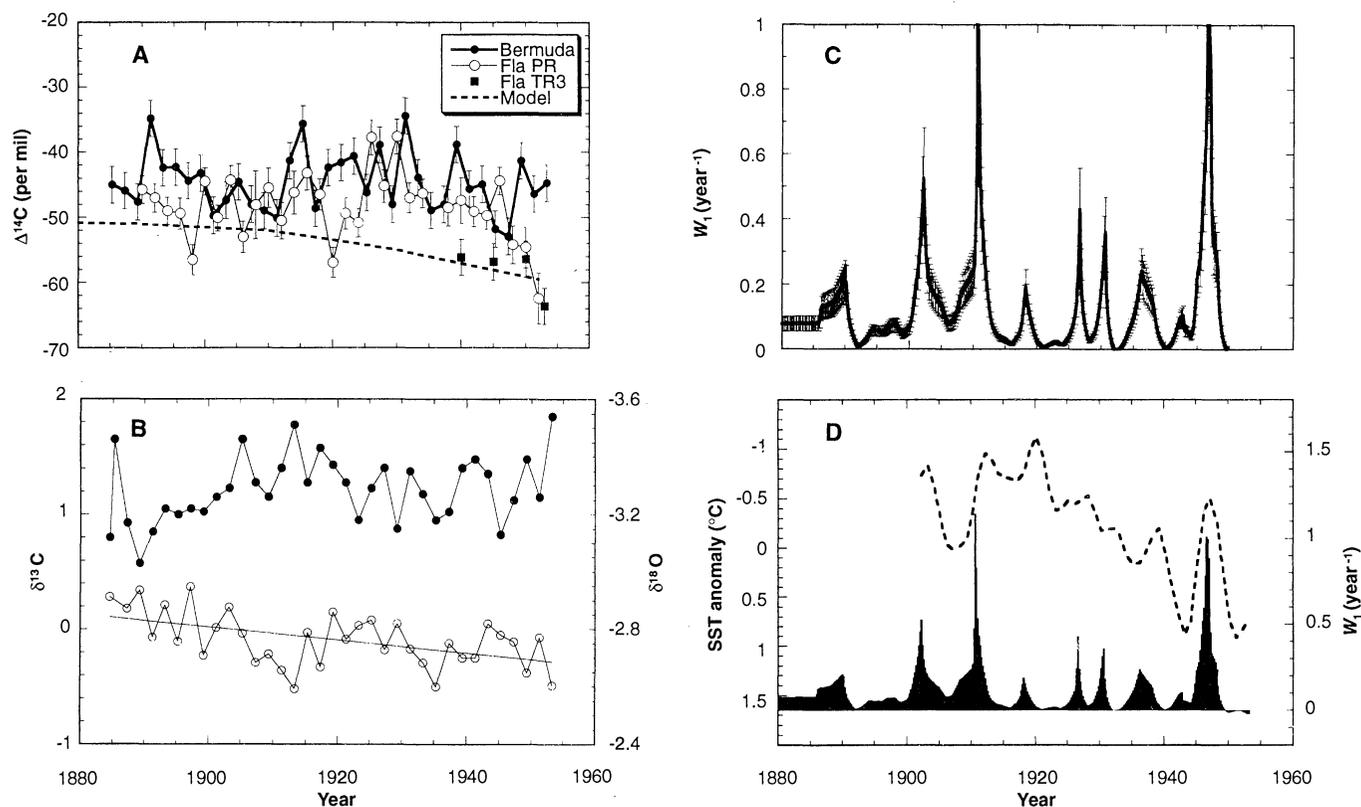


Fig. 1. (A) $\Delta^{14}\text{C}$ of biennial coral bands from Bermuda and the two Florida Keys sites [Pickles Reef (PR) and The Rocks (TR3)] in the North Atlantic Ocean. Duplicate analyses are plotted as the weighted average $\Delta^{14}\text{C}$ value. The uncertainty of year assignments for these corals is about ± 1 to 2 years by 1885 (13). The model calculations of $\Delta^{14}\text{C}$ in the mixed layer are also shown and were done with the use of bidecadal tree-ring (atmospheric) $\Delta^{14}\text{C}$ values and a box-diffusion model of the oceans, assuming constant atmospheric CO_2 concentration, constant biospheric reservoir sizes, and constant gas exchange rates (37). (B) $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ results from biennial coral samples from the Bermuda coral. Solid circles, $\delta^{18}\text{O}$; open circles, $\delta^{13}\text{C}$. Line indicates least-squares fit of $\delta^{13}\text{C}$. Samples were crushed in methanol and roasted at 375°C in vacuo just before analysis. Samples were analyzed with a light-isotope ratio mass spectrometer (Micromass 602E) at Woods Hole Oceanographic Institution in the laboratory of L. Keigwin and have an uncertainty of ± 0.1 per mil for both $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$. (C) Results of the inverse model that show W_1 in the Sargasso Sea between the surface box B and the shallowest subsurface box of $\sigma_\theta = 26.4$ for the time period 1885-1955.

Periods of high W_1 are the result of low $\Delta^{14}\text{C}$ values that appear in the Bermuda coral record [see (A)]. The uncertainty indicated by error bars represents 30% of the W_1 values. (D) W_1 as in (C) (filled curve) and time series of the winter SST anomaly east of Newfoundland [the SST values are averaged over the region 52° to 40°N , 50° to 30°W and smoothed with a five-point binomial filter (dashed line)] [after (33)]. Periodicities within the individual records were investigated by means of spectral and cross-spectral analyses. Fourier transforms of the correlation functions of each record were done with Arand Crospec software provided by P. Howell and based on the Blackman-Tukey method (38). I used the detrended annual SST anomaly [figure 6a in (33)] and annually averaged W_1 values from (C) for the period 1902-1953 ($n = 52$). The data were lagged by $n/3$. The spectral density for both the W_1 and SST anomaly data displays most of their variance at the 9-year cycle, which is significant to the 95% confidence level. In order to demonstrate similarities between the two records, cross-spectral analyses were performed and the 9-year period was coherent to the 95% confidence level; the SST anomaly lagged W_1 by 2 years.

for the Bermuda coral (Fig. 1B) ranged from -3.55 to -3.05 per mil and for most of the record (1895–1955) varied inversely with the $\Delta^{14}\text{C}$ values (Fig. 1A). A least-squares fit of $\Delta^{14}\text{C}$ versus $\delta^{18}\text{O}$ revealed an inverse correlation (linear correlation coefficient $r = -0.31$; $n = 30$), which is significant at the 90% confidence level ($\alpha = 0.90$). An inverse correlation ($r = -0.68$, $\alpha = 0.95$; $n = 9$) was also found between annual $\delta^{18}\text{O}$ and $\Delta^{14}\text{C}$ values lagged by 1 year for the post-1971 bands from this coral (17). This association suggests that low SSTs or high salinities (indicated by high $\delta^{18}\text{O}$) or both accompany periods of low $\Delta^{14}\text{C}$ values in the Sargasso Sea and are controlled by atmospheric forcing of the ocean surface. The $\delta^{18}\text{O}$ results (lagged by 2 years) for the Florida coral (19) also showed an inverse correlation with $\Delta^{14}\text{C}$ data ($r = -0.45$, $\alpha = 0.98$; $n = 28$). This association and the inverse correlations between average annual wind speed and average annual SST, and between annual average wind speed and annual water mass renewal rate at Bermuda (17), suggest that a decrease in the Bermuda coral $\Delta^{14}\text{C}$ value is caused by increased mixing between surface and subsurface waters (≤ 400 m) during the formation of subtropical-mode water (18°C and 36.5 per mil salinity) (20) in late winter in the northern half of the Sargasso Sea.

The $\delta^{13}\text{C}$ (7) values for the Bermuda coral decreased by about 0.4 per mil from 1885 to 1955 (Fig. 1B); this may represent evidence of excess CO_2 (from fossil fuel and biomass burning with a $\delta^{13}\text{C}$ of -26 to -28 per mil) in the surface ocean. Combined with an additional decrease of 0.4 per mil from 1955 to 1983 (17), values decreased overall by 0.8 per mil over a 100-year period. The $\delta^{13}\text{C}$ values of DIC in surface waters of the Pacific decreased by about 0.4 per mil from 1970 to 1990 (21). Similarly, DIC $\delta^{13}\text{C}$ in surface waters near Bermuda decreased by 0.018 ± 0.005 per mil year $^{-1}$ from 1984 to 1993 (adjusted for interannual variation) (22). This value is close to the average decrease observed from 1955–1983 in the Bermuda coral of 0.014 ± 0.003 per mil year $^{-1}$ (17). In a separate Bermuda coral, a decrease in $\delta^{13}\text{C}$ of 0.5 per mil from 1880–1950 (0.007 per mil year $^{-1}$) (23) was measured, which is similar to the decrease in the Bermuda coral that I observed of 0.006 per mil year $^{-1}$ (Fig. 1B). In contrast, the $\delta^{13}\text{C}$ values of the Florida coral remained constant over the past 100 years (19). Coral $\delta^{13}\text{C}$ should be used cautiously as a proxy for water mass DIC $\delta^{13}\text{C}$ values, because physiological processes in the coral polyp involve large isotopic fractionations (10, 24, 25).

The lack of a Suess Effect implies that some process other than gas exchange with the atmosphere is controlling the $\Delta^{14}\text{C}$ values of the surface waters in the North Atlantic. Exchange of CO_2 between atmosphere and surface ocean has too long a time constant (5 to 10 years) to be responsible for the huge shifts in $\Delta^{14}\text{C}$ values. Lateral advection of water is unlikely to have caused large $\Delta^{14}\text{C}$ shifts, because the $\Delta^{14}\text{C}$ signature of the Gulf Stream is similar to that of Bermuda. One process that could account for the shifts is changes in vertical circulation.

To evaluate this effect, I used a multi-box, isopycnal mixing model to estimate the ventilation rate (the water mass renewal rate, W_i) of the upper 700 m. This model is similar to that used to evaluate the W_i in the Sargasso Sea from 1950 to 1983 with post-bomb $\Delta^{14}\text{C}$ values (17). The model has an atmospheric box (A), and annual $\Delta^{14}\text{C}$ values of atmospheric CO_2 are set equal to values reported for tree rings (3). The rate constant of the output of $^{14}\text{CO}_2$ from the sea surface to the air, k_1 , is a function of the mixed layer depth Z (135 m), the ΣCO_2 in surface waters (2.12 mol of CO_2 m^{-3}), and the CO_2 gas exchange rate I (18.9 mol of CO_2 m^{-2} year $^{-1}$), which is a function of the gas exchange piston velocity V_p (in meters per day), according to

$$k_1 = \frac{V_p \times 0.019 \text{ mM} \times 365 \text{ days year}^{-1}}{Z \times \Sigma\text{CO}_2} \text{ (year}^{-1}\text{)} \quad (1)$$

where $V_p = \text{WS} \times 0.9995 - 3.47$ (the average annual wind speed WS is 6.2 m s^{-1}), the CO_2 concentration in seawater is 0.019 mM, and $I = V_p \times 0.019 \text{ mM} \times 365 \text{ days year}^{-1}$ (26, 27). The input rate of $^{14}\text{CO}_2$ from the air to the sea surface, k_{-1} , depends on the partial pressure of CO_2 in the surface ocean ($p\text{CO}_{2s}$) and that in the air ($p\text{CO}_{2a}$) and on the ^{14}C concentration in the atmosphere at time t [$A(t)$], which includes the Suess Effect (3), and in surface waters [$B(t)$]

$$k_{-1} = \{A(t)/0.983 - B(t)\} \times (p\text{CO}_{2s}/p\text{CO}_{2a}) \times k_1 \text{ year}^{-1} / [A(t)/0.983 - B(t)] \times 0.983 \text{ (year}^{-1}\text{)} \quad (2)$$

where 0.983 is the isotopic fractionation factor when CO_2 is transferred from the atmosphere to surface waters, and $p\text{CO}_{2s}/p\text{CO}_{2a} = 1$. The three surface ocean boxes are B (Bermuda), S (slope water entrained in the Sargasso Sea), and GS (Gulf Stream); 15% of the water in B is replaced with water from S and GS each year (with a

ratio of 3/17 for S/GS) (28), and the same amount is lost from the sides of B to conserve mass. The observed coral $\Delta^{14}\text{C}$ values for B and GS are the Bermuda and Florida (Pickles Reef) records, respectively (Fig. 1A), and the pre-bomb $\Delta^{14}\text{C}$ level for S is set to -90 per mil (29). The seven subsurface boxes D_i contain ^{14}C concentrations $D_i(t)$ through $D_7(t)$ that are homogeneously mixed along surfaces of constant density [$0.1\sigma_\theta$ (density anomaly σ at potential temperature θ) wide from 26.4 to 27.0]. Convective mixing between surface waters (B) and the isopycnal boxes (D_i) is assumed to occur throughout the year at a rate W_i (in year $^{-1}$) to simulate late winter mixing in the northern Sargasso Sea. Steady-state $\Delta^{14}\text{C}$ values for D_i to D_7 ranged from -49.3 to -53.5 per mil (30).

A forward model was run to calculate the $^{14}\text{CO}_2$ concentration in B at time $(t + \Delta t)$ and is equal to $B(t)$ plus the $^{14}\text{CO}_2$ transfer (per liter of seawater) into and out of box B.

$$B(t + \Delta t) = B(t) + 0.15 \times \Delta t \times [0.85 \times \text{GS}(t) + 0.15 \times \text{S}(t) - B(t)] + \Delta t \times [k_{-1} \times A(t) - k_1 \times B(t)] + W_1(t) \times \Sigma S_i \times \Delta t \times [\Sigma D_i(t) - B(t)] \quad (3)$$

where $\Delta t = 0.2$ year (0.1 year in earlier model) and S_i is W_i/W_1 , the relative change of the ventilation rates on each isopycnal (S_1 through S_7 are 1.0, 0.8, 0.65, 0.49, 0.36, 0.24, and 0.15) (31). When the forward model (Eq. 3) was solved using a steady-state, pre-1900 $\Delta^{14}\text{C}$ value of -43.4 per mil, a constant annual value for $W_1(t)$, of 0.08 year $^{-1}$ was obtained (32). Results of the forward calculation revealed that a constant $W_1(t)$ of 0.08 year $^{-1}$ was too low during several periods when the surface $\Delta^{14}\text{C}$ values in the Bermuda corals were lower than about -48 per mil; they are 1889–1890, 1901–1903, 1907–1912, 1925–1926, 1929–1930, 1935–1938, and 1945–1948.

Numerous variables were tested in the forward model to determine their effects on the $B(t)$ record. An instantaneous reduction in the mixed layer depth from 135 to 50 m caused a 7-per-mil rise in $\Delta^{14}\text{C}$ of $B(t)$ in 2 years. Also, if all mixing with subsurface isopycnals had ceased in the forward model, then the $\Delta^{14}\text{C}$ of the surface $B(t)$ would have increased 6 per mil in 4 years. Likewise, if W_1 had increased instantaneously by a factor of 6, then a 5-per-mil decrease of $\Delta^{14}\text{C}$ in surface box $B(t)$ would have occurred in 1 year.

I then ran an inverse calculation to determine the W_1 record that was needed to reproduce the Bermuda $\Delta^{14}\text{C}$ record for the period 1885–1955, using

$$W_1(t) = \frac{B(t + \Delta t) - B(t) - X(t)}{[\sum D_i(t) \times \sum S_i - B(t) \times \sum S_i] \times \Delta t} \quad (4)$$

where

$$X(t) = 0.15 \times \Delta t \times [0.85 \times GS(t) + 0.15 \times S(t) - B(t)] + \Delta t \times [k_{-1} \times A(t) - k_1 \times B(t)] \quad (5)$$

Subsurface $\Delta^{14}C$ values were calculated with the use of

$$D_i(t + \Delta t) = D_i(t) - W_i \times D_i(t) \times \Delta t + B(t) \times W_i \times \Delta t + (\%I_i \times W_i) \times [I - D_i(t)] \quad (6)$$

where %I is the fraction of water in D_i exchanged with a deep box, I, to maintain steady-state concentrations of ^{14}C (% I_1 to % I_7 values are 0.096, 0.115, 0.14, 0.169, 0.198, 0.237, and 0.285); box I was missing from the earlier model.

The resultant record for the period 1885–1955 (Fig. 1C) contained seven periods when W_1 was greater than twice the steady-state value of 0.08 year⁻¹. These periods coincided with the low $\Delta^{14}C$ values in the Bermuda coral record. They were 1889, 1901–1903, 1908–1912, 1926–1927, 1930–1931, 1935–37, and 1945–1948. When the Bermuda $\Delta^{14}C$ measurements were ≤ -50 per mil, W_1 values were >1 , which means that the water in box B was being replaced completely more than once per year (Fig. 1C). The periodic pulses of high W_1 values in Fig. 1C suggest that subsurface depths in the ocean were ventilated faster during these times. The W_1 values calculated for the early 1960s were also high (17), which suggests that these pulses of high ventilation occurred throughout the entire past century. Conversely, high $\Delta^{14}C$ values (> -40 per mil) in the Bermuda coral caused the W_1 values to be near or equal to zero (Fig. 1C); this occurred during 1891–1892, 1915–1916, 1921–1924, 1932–1933, and 1939–1941.

Sensitivity analyses were conducted on the inverse model to ascertain the change required in each of the factors influencing the W_1 record. A change in the average annual WS by $\pm 12\%$ (from 5.5 m s⁻¹ to 7.0 m s⁻¹) altered the CO₂ exchange rate I from 14.1 to 24.5 mol m⁻² year⁻¹. The peaks and valleys in the W_1 record occurred at the same time, however, and the W_1 values changed by $<30\%$. The influence of the Gulf Stream $\Delta^{14}C$ time history was tested by running the inverse model with a constant GS value (-47.4 per mil for 1880–1940). The change in the W_1 record was $<5\%$. An average $\Delta^{14}C$ value (-43.4 per mil for 1885–1955) was used for $B(t)$ in place of the actual record, and the resultant

W_1 record was 0.08 ± 0.04 year⁻¹ over the entire 1885–1955 period. These tests show that the Bermuda $\Delta^{14}C$ variability is responsible for the large peaks in the W_1 record (Fig. 1C).

The decadal time-scale decreases of the winter SST anomaly east of Newfoundland (33) match the pulses of high W_1 obtained here (Fig. 1D). A cross-spectral analysis of these two records (between 1902 and 1953) reveals that the 9-year period is coherent to the 95% confidence level (the SST anomaly lags W_1 by 2 years). This decade time-scale variability is superimposed on a gradual warming trend of SST from the 1920s to 1940 in the North Atlantic (33, 34) and a decrease in the W_1 values (see Fig. 1D). The record of the North Atlantic Oscillation (NAO), which is the difference in winter sea-level air pressures between the high at Lisbon, Portugal, and the low at Stykkisholmur, Iceland, displays decadal shifts that are linked to changing wind patterns over the North Atlantic (35). A cross-spectral analysis of NAO values (35) and annual W_1 values between 1885 and 1983 (17) reveals that the 6-year period is coherent to the 95% confidence level (no lag). These correlations suggest a link between the pulses of high W_1 , which represent increased subtropical-mode water ventilation, and both low NAO values and lower SST values in the area northwest of the Sargasso Sea [and likely off the east coast of the United States (36)].

The absence of a predictable Sues Effect in the surface waters of the North Atlantic demonstrates the importance of circulation as opposed to gas exchange on the ^{14}C history of surface waters. The large range of $\Delta^{14}C$ values present in the North Atlantic during the past century shows that the vertical mixing in the upper few hundred meters of the water column can be rapid and variable and that it may be related to changes in North Atlantic surface properties and the NAO in general. These dramatic shifts of surface ocean $\Delta^{14}C$ signatures represent the uncertainty (± 200 apparent ^{14}C years) that needs to be incorporated into a reservoir age correction for the North Atlantic surface ocean [400 years (37)].

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