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CORRECTION OF ACCELERATOR MASS  
SPECTROMETRY  $^{14}\text{C}$  AGES MEASURED  
IN PLANKTONIC FORAMINIFERA:  
PALEOCEANOGRAPHIC IMPLICATIONS

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**Abstract.** Carbon 14 dates obtained by accelerator mass spectrometry (AMS) on foraminiferal samples from deep-sea sediment cores must be corrected for the difference in  $^{14}\text{C}$  composition between the atmosphere and the sea surface. In the modern ocean, the "apparent age" of carbonate shells formed in surface waters varies between 300 and 1200 years and depends mainly on latitude. The time variation of this parameter during climate oscillations of the last 40,000 years may have been significant: there should have been small changes for most of the ocean between 40°S and 40°N, but an increase of the apparent age by several hundred years should be expected at high latitudes in response to subpolar/subtropical front movements. The North Atlantic is likely to have experienced the most significant changes, due to large variations in the mode and rate of North Atlantic Deep Water production. These hypothetical changes may be measured by coupled AMS  $^{14}\text{C}$  dating of contemporaneous planktonic foraminifera and terrestrial organic matter (pollen, charcoal, wood, etc.) which occur in the same core or are stratigraphically linked by the same volcanic ash layer. The  $\Delta(^{14}\text{C}$  atmosphere,  $^{14}\text{C}$  sea surface) can be viewed as a new paleoceanographic tracer which may provide additional information about high latitude surface waters complementary to those obtained with  $^{13}\text{C}/^{12}\text{C}$  and Cd/Ca ratios measured in planktonic foraminifera.

INTRODUCTION

The recent development of mass spectrometers coupled with small electrostatic accelerators has made possible direct

measurement of the  $^{14}\text{C}/^{12}\text{C}$  ratio of fossil samples (see Litherland [1980] for a review of accelerator mass spectrometry [AMS] and Arnold et al. [1987] for the example of the Gif-sur-Yvette facility). The main advantage of this new method is a reduction of the amount of carbon needed for radiocarbon dating by a factor of 1000 to 100,000 (between 1 mg and 15  $\mu\text{g}$  of carbon can be measured, for very small samples see Vogel et al. [1986]). In particular, this makes it possible to measure  $^{14}\text{C}$  ages on monospecific samples of 1000-2000 hand picked planktonic foraminifera and eliminates the risk of contamination by older detrital carbonates [Andree et al., 1984; Duplessy et al., 1986; Bard et al., 1987a, b, 1988b; Broecker et al., 1988a, b](E. Bard et al., Sea-level estimates during the last deglaciation based on  $\delta^{18}\text{O}$  and accelerator mass spectrometry  $^{14}\text{C}$ -ages measured in *Globigerina bulloides*, submitted to *Quaternary Research*, 1988d) (hereinafter Bard et al., 1988d). The AMS of  $^{14}\text{C}$  is now an important tool for paleoclimatologists studying isotopic and faunal/floral records of the last 50,000 years in deep-sea sediment cores.

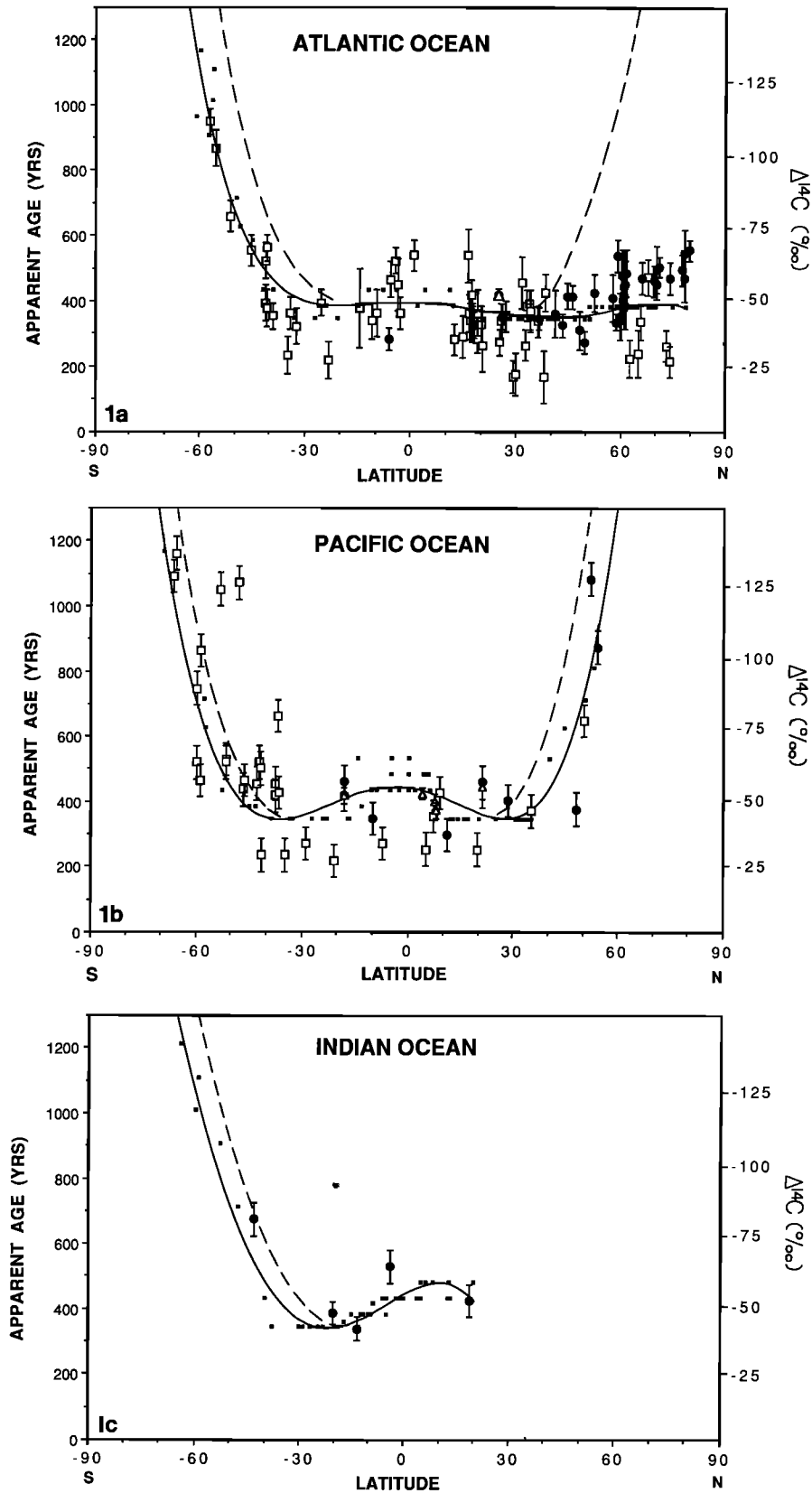
In order to compare marine AMS  $^{14}\text{C}$  ages with chronologies obtained from terrestrial plant matter, all  $^{14}\text{C}$  ages obtained from planktonic foraminifera must be corrected for the slight  $^{14}\text{C}/^{12}\text{C}$  difference between atmospheric  $\text{CO}_2$  and  $\Sigma\text{CO}_2$  of waters at the sea surface. Surface water is not in isotopic equilibrium with the atmosphere; instead, a steady state balance is maintained between the input of  $^{14}\text{C}$  from the atmosphere and its removal by advection and radiodecay in the water column. The age difference between the atmosphere and the surface ocean is termed the apparent age or reservoir age of the surface water [Suess and Revelle, 1957; Craig, 1957; Arnold and Anderson, 1957; Stuiver and Polach, 1977].

In the following discussion, I describe a method to determine this age correction, which is crucial for correct interpretation of AMS  $^{14}\text{C}$  dating of deep-sea cores. To evaluate the present-day apparent age, I first consider and remove the anthropogenic imprint on the  $^{14}\text{C}$  distribution in the atmosphere and ocean (due to fossil fuel burning [Suess, 1955] and thermonuclear  $^{14}\text{C}$  production [Rafter and Fergusson, 1957]). I then discuss the present-day apparent age distribution with respect to surface

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hydrography. Finally, I evaluate the possible magnitude and timing of the apparent age variations during the last 40,000 years.

As recommended by Stuiver and Polach [1977], the radiocarbon activities described in this paper are expressed as  $^{14}\text{C}$  ages (in years B.P.) and on the  $\Delta^{14}\text{C}$  scale (in ‰).

#### RECONSTRUCTION OF PREINDUSTRIAL SEA SURFACE APPARENT AGES

Two sources of anthropogenic carbon have affected the natural  $^{14}\text{C}$  content of modern surface water. The first is a gradual dilution of the  $^{14}\text{C}$  content of the atmosphere by injection of  $^{14}\text{C}$ -free  $\text{CO}_2$  formed since the last century by the combustion of old carbon. This slow change, often referred to as the "Suess effect," had a maximum magnitude of about  $-25\text{‰}$  in the average 1950 troposphere [Cain and Suess, 1976] and of at most  $-10\text{‰}$  in surface ocean waters [Druffel and Suess, 1983]. The second anthropogenic alteration is the input of synthetic  $^{14}\text{C}$  formed by large-scale testing of thermonuclear weapons which began in the late 1950s. Between 1962 and 1964, the  $^{14}\text{C}$  activity of the northern troposphere increased by about  $1000\text{‰}$  relative to its prebomb value [Nydal and Lovseth, 1983].

Consequently, to reconstruct the prebomb surface ocean, it is necessary to compile data obtained from sea water samples collected before the late 1950s [Burling and Garner, 1959; Fonselius and Ostlund, 1959; Bien et al., 1960; Broecker and Olson, 1961; Broecker, 1962; Rafter and O'Brien, 1970, 1972], from shells of molluscs collected alive before the bomb testing [Broecker and Olson, 1961; Mangerud and Gulliksen, 1975; Guillier et al., 1982], and from banded corals, which preserve a record of the radiocarbon content of surface water through time [Druffel and Linick, 1978; Druffel and Suess, 1983; Toggweiler, 1983; Glynn et al., 1983; Druffel, 1987]. Following Broecker et al. [1985a], the  $^{14}\text{C}$  measurements obtained before 1957-1958 are considered as roughly free of bomb-produced  $^{14}\text{C}$ . Certain coral data suggest that 1955 should be a more suitable date to place the acceptance limit for the prebomb  $^{14}\text{C}$  data. However, the eventual pre-1957 departures from the prebomb levels are always small ( $<10\text{‰}$ ) and an error of 2 years on growth band counting cannot always be ruled out (coral data obtained for Belize and Uva Island show

a statistically significant ( $1\sigma$  criterion) departure between 1955 and 1957 but other coral  $^{14}\text{C}$  data obtained for Florida, Fanning Island, and Fiji do not). Moreover, Figure 1 shows that there is no discrepancy between the coral data (obtained by averaging the available  $\Delta^{14}\text{C}$  measurements on growth bands from the 1930-1957 interval) and the sea water and mollusc data. Although the scatter in the data is large ( $35\text{‰} \approx 300$  years), one could also argue that the results obtained from shells are somewhat different from the sea water measurements in the North Atlantic. This difference could be attributed either to a slight pre-1957 anthropogenic contamination of the sea-water samples or to a natural coastal  $^{14}\text{C}$  effect on the shells.

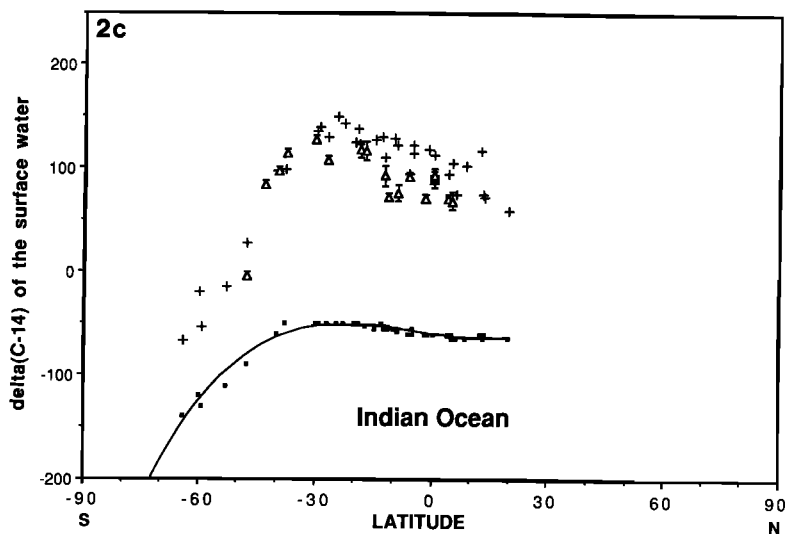
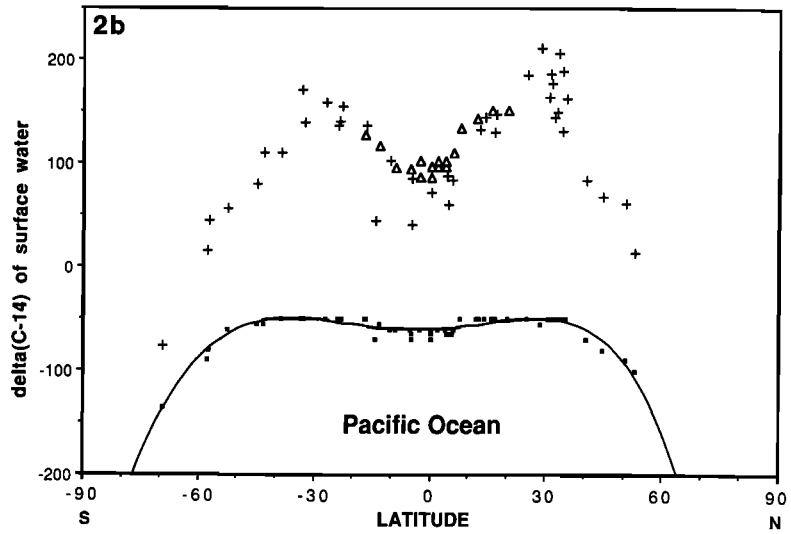
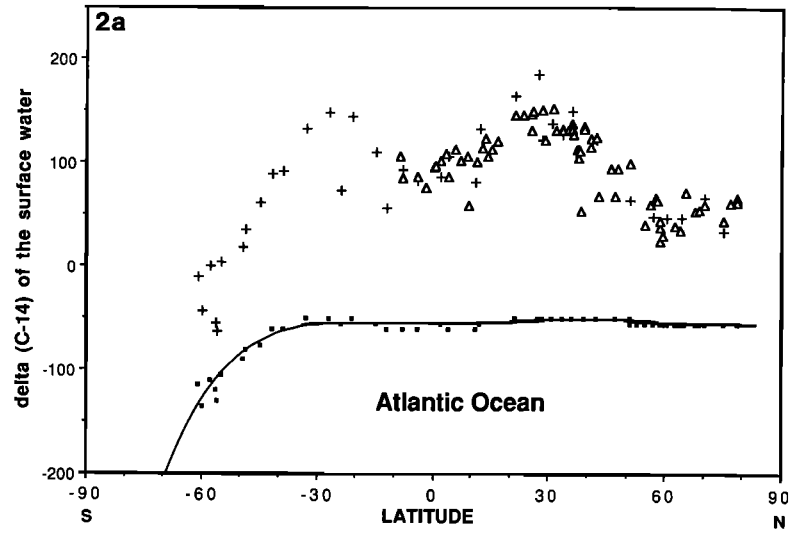
In addition to these raw data, I have also used  $^{14}\text{C}$  values of the sea surface used by Broecker et al. [1985a] for their calculation of the thermonuclear radiocarbon inventory in the oceans. These prebomb levels were obtained from the available surface prebomb  $^{14}\text{C}$ -data and from vertical profiles of tritium and radiocarbon measured as part of the GEOSECS, Transient Tracers in the Ocean (TTO), North Pacific Experiment (NORPAX), and Indien Gaz Exchange (INDIGO) oceanographic programs (for the GEOSECS program see Stuiver and Ostlund [1980, 1983] and Ostlund and Stuiver [1980]; for the TTO program see Ostlund [1983]; for the NORPAX program see Quay et al. [1983]; for the INDIGO program see Ostlund and Grall [1988] and Bard et al. [1987c, 1988a, c]).

To obtain the preanthropogenic  $^{14}\text{C}$  levels, the prebomb data must be corrected for the injection of  $^{14}\text{C}$ -free  $\text{CO}_2$  into the atmosphere from the burning of fossil fuel. This correction is at most  $+10\text{‰}$  in mid-latitude surface waters [Druffel and Suess, 1983] and should be negligible for the surface waters strongly influenced by upwelling of old subsurface water (in the southern ocean for example). To a first approximation, I assume that the Suess effect varies linearly between  $-10\text{‰}$  and  $0\text{‰}$  for samples between  $\Delta^{14}\text{C} = -30\text{‰}$  and  $-140\text{‰}$ , respectively. This correction is of the same order of magnitude as the typical error associated with the early  $^{14}\text{C}$  measurements ( $1\sigma$  between 5 and  $15\text{‰}$ ). Therefore, any assumption concerning the latitudinal variation of the Suess effect has little influence on the conclusions drawn in this paper.

The reconstruction of the preanthropogenic  $^{14}\text{C}$  age of the surface water is shown as a function of latitude for the three oceans in Figure 1. To a first approximation, the apparent age remains near 300-400 years between  $40^\circ\text{S}$  and  $40^\circ\text{N}$  latitude. For high latitudes in the southern ocean and in the North Pacific the correction curves deviate by up to 1200 years. High-latitude surface waters are old because of upwelling of subsurface water, whose  $\Delta^{14}\text{C}$  is not reset to atmospheric values. This is a consequence of the impedance in air-sea exchange and of the short residence time of surface waters in areas of deep and intermediate water formation.

The latitudinal variation of the  $^{14}\text{CO}_2$  exchange between the atmosphere and the ocean surface is also reflected in the transient profiles obtained as part of the oceanographic programs which followed the major input of thermonuclear  $^{14}\text{C}$  into the atmosphere in the early 1960s: GEOSECS (1972-1978), NORPAX (1979), TTO (1980-1981), and INDIGO (1985-1987). The transitory profiles shown in Figure 2 contain the same features as the prebomb steady state profiles: surface  $\Delta^{14}\text{C}$  values for latitudes higher than  $40^\circ$  in the southern hemisphere and in the North Pacific Ocean are also much lower than the values between  $40^\circ\text{S}$  and  $40^\circ\text{N}$ . As illustrated by Broecker et al. [1985a], the  $\Delta^{14}\text{C}$  minimum

Fig. 1. (Opposite) reconstruction of the natural  $^{14}\text{C}$  apparent age of the surface water for the three oceans (1a, Atlantic; 1b, Pacific; 1c Indian), showing (closed circles) mollusc measurements ( $1\sigma$  error bars), (open triangles) coral measurements (these values are preanthropogenic ages obtained by using the mean of several  $^{14}\text{C}$  measurements on growth bands), (open squares) sea water  $\Sigma\text{CO}_2$  measurements ( $1\sigma$  error bars), and (closed squares) reconstruction derived from the prebomb levels of Broecker et al. [1985a]. The solid curves are the polynomial least squares fits and the dashed curves represent hypothetical 18,000 years B.P. gradients. As noted by Broecker et al. [1985a] the New Zealand results for the south Pacific Ocean pose a problem in that the  $\Delta^{14}\text{C}$  values for a surface water traverse run in March 1958 average about  $60\text{‰}$  higher than those for samples collected on a nearly identical traverse in December 1958. This problem can account for the two anomalous apparent age values seen in Figure 1b (at about  $48^\circ\text{S}$  and  $53^\circ\text{S}$  from [Rafter and O'Brien, 1972]).



observed for the equatorial zones is linked to mixing with upwelling water.

Except for the North Atlantic, the latitudes of low <sup>14</sup>C age corrections are between 40°N and 40°S. This coincides with the subtropical/subpolar boundary, which corresponds to zero wind stress curl and separates the subpolar regions showing upward Ekman pumping from the subtropical waters characterized by downward Ekman pumping [Hellenman and Rosenstein, 1983]. To a first approximation, the equatorward expansion of seasonal sea ice in the southern ocean also follows the subtropical/subpolar front [Gordon and Taylor 1975].

The low <sup>14</sup>C concentrations in high-latitude surface waters are directly related to both upwelling and mixing with old subsurface waters and to the limiting effect of sea ice on atmosphere-ocean CO<sub>2</sub> exchange. The limiting effect of sea ice is particularly important because maximum seasonal sea ice extent occurs during the winter when high wind speeds would otherwise dramatically increase the atmosphere-ocean exchange coefficient for CO<sub>2</sub> (see Liss and Merlivat [1986] for an updated discussion). In addition, because of sea salt expulsion, the formation of sea ice increases the density of the underlying water, thereby reducing the stability of the ocean surface layer. This leads to convection in the upper 400-600 m [Gordon and Taylor, 1975], and possibly to deep convection directly [Martinson et al., 1981]. Both processes enhance mixing with deeper <sup>14</sup>C-depleted waters.

In contrast, no surface <sup>14</sup>C gradient is present between 40° and 70°N in the North Atlantic Ocean. The apparent natural <sup>14</sup>C age of these surface waters is almost constant at about 400-500 years (Figure 1a). Similarly, the surface Δ<sup>14</sup>C was also almost constant at about 70‰ from 1972 to 1981 (Figure 2a). However, the Δ<sup>14</sup>C values obtained for the zone 20°-40°N (approximately 140‰) are not the same as for higher latitude (approximately 70‰). This is the result of the North Atlantic Deep Water (NADW) formation, which through vertical advection holds down the surface Δ<sup>14</sup>C values despite the fact that large water column inventories of bomb <sup>14</sup>C are observed for the high-latitude North Atlantic [Broecker et al., 1985a]. North Atlantic surface waters are unique in their <sup>14</sup>C content, whereas the other high-latitude oceans exhibit strong <sup>14</sup>C gradients (Figures 1 and 2). Constant <sup>14</sup>C values are principally due to the Gulf Stream-North Atlantic Drift current system, which brings mid-latitude surface and intermediate waters to 50° and 60°N and across the Iceland-Faroe-Shetland Ridge into the Norwegian Sea. Unlike the southern ocean, the North Atlantic is characterized not by upwelling of <sup>14</sup>C-depleted waters but by sinking of high-salinity surface and subsurface waters. As discussed by Warren [1983], the process of NADW formation is self-perpetuating in that more upper layer water is drawn into the North Atlantic as the surface water sinks. This influx of warm, salty water drives the high evaporation rates that increase the density of surface waters and result in NADW sinking. In the case of the North

Atlantic the line of zero curl of wind stress has a clear southwest northeast orientation. This boundary between subpolar and subtropical waters corresponds to the position of the Gulf Stream-North Atlantic Drift system.

#### IMPLICATIONS FOR <sup>14</sup>C DATING OF PLANKTONIC CARBONATES IN DEEP SEA SEDIMENT CORES

AMS <sup>14</sup>C dating of planktonic foraminifera from deep-sea cores is used as a tool to compare the timing of climatic events on the land and in the ocean [Duplessy et al., 1986; Bard et al., 1987a; Broecker et al., 1988a, b]. To apply the correction for the air/sea surface <sup>14</sup>C/<sup>12</sup>C difference, the foraminiferal species which most accurately reflects the isotopic composition of the surface ΣCO<sub>2</sub> pool must be chosen. Our knowledge of calcification depths and isotopic composition of planktonic foraminifera comes from plankton tows [Fairbanks et al. 1982] and sediment traps [Fairbanks et al., 1982; Curry et al., 1983; Klas, 1987].

Postdepositional processes can also modify AMS <sup>14</sup>C dates obtained from foraminiferal samples. For example, Broecker et al. [1988a, b] have observed relatively large differences in AMS <sup>14</sup>C ages (up to 2000 years) obtained from different species of planktonic foraminifera at the same depth in deep-sea sediment cores. These apparent inconsistencies are likely the result of a combination of sedimentological processes (such as bioturbation and differential dissolution), with differences in calcification depths between the planktonic foraminiferal species providing additional effects [Bard et al., 1987d; Berger, 1987; Broecker et al., this issue (a)].

After dating selected foraminiferal species, the curves in Figure 1 can be used to determine the apparent age which must be subtracted from the <sup>14</sup>C measurements. The scatter in the preindustrial ages provides an estimate of the natural variability of this correction. This variability (± 100 years) should be kept in mind when correlating climatic events and discussing slight <sup>14</sup>C age differences between cores.

#### TIME VARIATION OF THE APPARENT AGES OF THE SURFACE WATER

All studies concerning AMS <sup>14</sup>C dating of planktonic foraminifera have assumed that the <sup>14</sup>C steady-state between the surface ocean and the atmosphere was constant prior to the recent anthropogenic changes. However, the last 40,000 years are characterized by important climatic and oceanographic changes linked to glaciation cycles. The influence of these climatic changes on the <sup>14</sup>C steady state must be understood in order to date and correlate older climatic events in deep-sea cores from different locations.

The steady state for <sup>14</sup>C in the mixed layer (≈75 m thick) can be described by the following equations which are derived for a simple box model of the carbon system [Oeschger et al., 1975; Heimann, 1978; Siegenthaler et al., 1980]:

$$k_{am}N_aA_a f_{am} - k_{ma}N_mA_m f_{ma} - \frac{K}{h_m}N_m \frac{\partial A_d}{\partial z}(z=0) - \lambda N_m A_m = 0 \quad (1)$$

The first two terms in (1) represent the exchange with the atmosphere, the third term describes the diffusive exchange with the deep-sea, and the last term expresses the loss of <sup>14</sup>C by radiodecay.

Fig. 2. (Opposite) prebomb and postbomb Δ<sup>14</sup>C levels of the sea surface for the three oceans (1a, Atlantic; 1b, Pacific; 1c Indian). The lower curves shows the reconstruction of the prebomb levels from Broecker et al. [1985a]. The data points above the lower curves represent the postbomb data obtained during the following programs: GEOSECS (plus, World Ocean 1972-1978), TTO (open triangles, North Atlantic 1980-1981), NORPAX (open triangles, North Pacific 1979), INDIGO (open triangles, Indian Ocean 1985-1986).

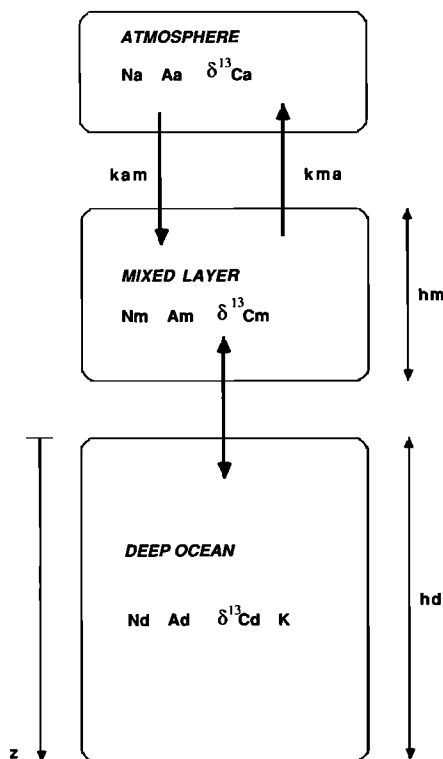


Fig. 3. Schematic diagram of the box model; see text for the parameter list.

$A_d(z)$  describes the steady state distribution of  $^{14}\text{C}$  in the deep-sea ( $K(d^2A_d/dz^2) - \lambda A_d = 0$  see Oeschger et al., [1975]) and is given by:

$$A_d(z) = A_m \frac{\cosh(\sqrt{\lambda K}(z-h_d))}{\cosh(\sqrt{\lambda K}h_d)} \quad (2)$$

In the case of solution equilibrium, the concentration of  $\text{CO}_2$  at the ocean-atmosphere interface can be written as  $\alpha P_a$  [Siegenthaler and Munnich, 1981]. Consequently, the following equation is obtained:

$$k_{am}N_a = w_{am}S_{oc}\alpha P_a \quad (3)$$

Note that a, m, and d are the indices for the different reservoirs of the model: the atmosphere, mixed layer and deep ocean respectively (see Figure 3). See the notation section for definitions of other variables.

To obtain the ratio  $A_m/A_a$ , which can be converted to an apparent age of the surface water, (1) can be solved:

$$A_m/A_a = f_{am}^2 \left/ \left( f_{ma}^2 + \frac{N_m(\sqrt{\lambda K} \tanh(\sqrt{\lambda K}h_d) + \lambda h_m)}{\alpha w_{am}S_{oc}P_a h_m} \right) \right. \quad (4)$$

To obtain the ratio  $A_d/A_m$ , which can be subsequently expressed as the mean  $^{14}\text{C}$  age difference between the oceanic mixed layer and the deep sea, (2) is integrated over depth:

$$A_d/A_m = \frac{\tanh(\sqrt{\lambda K}h_d)}{\sqrt{\lambda K}h_d} \quad (5)$$

If there is no exchange between the mixed layer and deep ocean ( $K=0$  in (4)), the mixed layer would reach isotopic equilibrium with the atmosphere, and there would be a positive difference between the sea surface and atmosphere of approximately  $+15\text{‰}$  (however because the  $^{14}\text{C}/^{12}\text{C}$  ratio is normalized to a fixed  $\delta^{13}\text{C}$  value of  $-25\text{‰}$  before radiocarbon age calculation, this difference would not lead to a younger age for the mixed layer with respect to the atmosphere).

Having derived (4) and (5), I will next consider the parameters in these equations which may have varied during the last 40,000 years and their probable influence on the apparent age of surface water.

#### Change of the Average Mixing Time of the Ocean

Because atmospheric  $^{14}\text{C}$  activity is maintained at a steady state level by a balance between cosmic ray formation and loss into the deep sea, global changes in the average mixing time of the ocean may have altered the atmospheric radiocarbon composition. Several authors have found anomalous deglacial trends (constant apparent ages, age inversion, etc.) in  $^{14}\text{C}$  chronologies obtained on land and in the ocean [Oeschger et al., 1980; Andree et al., 1986a; Bard et al., 1987b, 1988d; Broecker et al. 1988a]. While simple sedimentological interpretations such as sedimentation rate changes, bioturbation artifacts, and sediment reworking cannot always be ruled out, these authors suggested that changes in the mixing of the global ocean could be responsible for these small deviations in the  $^{14}\text{C}$  trend (see also Keir [1983] for a modeling study). For the purposes of this paper, I note that the age difference between the atmosphere and the surface ocean remains roughly constant even if a significant reduction or increase of the global mixing rate of the ocean occurs. Indeed, varying  $K$  between 2000 and 8000  $\text{m}^2 \text{yr}^{-1}$  would only increase the  $\Delta^{14}\text{C}$ -age(a,m) by +70 years. In contrast, the difference between the surface ocean and the deep ocean is strongly affected: by varying  $K$  from 2000 to 8000  $\text{m}^2 \text{yr}^{-1}$  the  $\Delta^{14}\text{C}$ -age(m,d) changes from 2300 to 900 years. This is the basic rationale for measuring AMS  $^{14}\text{C}$  age differences between benthic and planktonic foraminifera in deep-sea cores: through these age differences, the history of ocean ventilation rates may be determined [Broecker et al., 1984; Andree et al., 1986b; Shackleton et al., 1988].

#### Change of the Ocean Surface Area

During the last glaciation, the area of ocean surface available for  $\text{CO}_2$  exchange was reduced by the extension of sea ice and by the sea level drop due to storage of water in the continental ice caps. The maps produced as part of the Climate: Long Range Investigation, Mapping and Prediction (CLIMAP) project [1976, 1981] indicate a decrease of the free ocean surface of between 10 and 20%, which results in an increase of the  $\Delta^{14}\text{C}$ -age(a,m) between +45 and +100 years respectively.

#### Change of the Wind Speed

A significant variation of the  $\text{CO}_2$  exchange coefficient between the atmosphere and the ocean may have occurred in

response to the increase in wind speed during glacial periods [Petit et al. 1981]. This may have enhanced the exchange of  $\text{CO}_2$  between the atmosphere and the ocean, leading to a decrease of the  $^{14}\text{C}$  difference between the air and the sea surface. If the average piston velocity between the atmosphere and the mixed layer is increased by 50%, the  $\Delta^{14}\text{C}$ -age(a,m) will decrease by -140 years. This effect would be opposite to that of reduced sea surface area for the case of the last glacial maximum (LGM).

#### *Change of Temperature*

Since the piston velocity ( $w_{am}$ ) is a function of the molecular diffusivity of  $\text{CO}_2$  and of the kinematic viscosity of water, a variation of  $w_{am}$  is expected in response to the global decrease of sea surface temperature during the LGM. Indeed, between  $0^\circ\text{C}$  and  $30^\circ\text{C}$ , the piston velocity increases by about a factor of 2. However, the solubility of  $\text{CO}_2$  also shows significant variations with temperature: between  $0^\circ\text{C}$  and  $30^\circ\text{C}$ , the solubility decreases by about a factor of 2. Because the two temperature variations work in opposite directions, the product  $\alpha w_{am}$ , often referred as the exchange coefficient for  $\text{CO}_2$ , does not change by more than 10% between  $-2^\circ\text{C}$  and  $30^\circ\text{C}$  [Monfray, 1987]. Variation of the kinetic fractionation factors by lowering the temperature by  $15^\circ\text{C}$ - $20^\circ\text{C}$  would only lead to a reduction of the reservoir age by about 20-40 years (experimental data for the temperature dependence of the fractionation factors can be found in the work of Mook et al. [1974] and Inoue and Sugimura [1985]). Consequently, these variations amount to negligible changes in the mean  $^{14}\text{C}$  age difference between the atmosphere and the mixed layer due to deglacial temperature changes.

#### *Change in the Reservoir Sizes*

Variation of the relative masses of carbon in the atmosphere and mixed layer has an influence on the  $^{14}\text{C}$  apparent age. Unfortunately, only the atmospheric changes for the glacial/interglacial transitions are substantiated with direct evidence [Delmas et al., 1980; Neftel et al., 1982; Barnola et al., 1987]. Assuming as an upper limit a constant value for  $N_m$ , it is found that the apparent age is increased by about +150 years when the  $\text{CO}_2$  content of the atmosphere is reduced by 30%. A more complicated model, including at least ocean overturning, and marine and terrestrial biosphere, would be needed in order to simulate the glacial atmospheric  $p\text{CO}_2$  and calculate the ratio  $N_a/N_m$  [see Sarmiento and Toggweiler, 1984; Siegenthaler and Wenk, 1984; Broecker and Takahashi, 1984].

#### *Change in the Production Rate of $^{14}\text{C}$ by Cosmic Rays*

Numerous radiocarbon dates of tree rings have shown that the production rate of  $^{14}\text{C}$  has not been constant in the past and may have responded to changes in the geomagnetic dipole moment and in the solar modulation of cosmic rays which are responsible for the  $^{14}\text{C}$  formation [Stuiver, 1961; Bucha et al., 1970; Neftel et al., 1981; Damon and Linick, 1986]. These two causes are thought to have changed the absolute activity of the atmosphere by less than  $100\text{‰}$  and  $25\text{‰}$ , respectively. After a change of the  $^{14}\text{C}$  production rate in the atmosphere, the steady state age difference between the atmosphere and the mixed layer remains the same, and only the

transient reequilibration between the two reservoirs must be considered. An analog to this transient change of the atmospheric  $^{14}\text{C}$  activity can be found in the recent injection of anthropogenic  $^{14}\text{C}$  into the atmosphere, which almost doubled the northern troposphere  $^{14}\text{C}$  concentration in the early 1960s [Nydal and Lovseth, 1983]. Figure 4 represents the decrease of the  $^{14}\text{C}$  difference between the atmosphere and the mixed layer following the pulse like input of thermonuclear  $^{14}\text{C}$  (based on atmospheric measurements, [Nydal and Lovseth, 1983], and banded coral measurements [Druffel and Linick, 1978; Druffel and Suess, 1983]). In addition, recent measurements of bomb  $^{14}\text{C}$  in the Indian ocean [Bard et al. 1987c, 1988a, c] show that this difference is now about  $100\text{‰}$  (" $\approx$  800 years"), only twice the preindustrial value. This is down from a difference of nearly  $1000\text{‰}$  two decades previous, an order of magnitude higher than the actual difference expected from solar or geomagnetic dipole moment changes. Because the duration of this transient is of the order of a few decades, it is thus completely negligible on geological time scales.

#### *Change in the Position of the Subpolar/Subtropical Boundaries*

Because the polar  $^{14}\text{C}$  gradients are extremely steep (equivalent to about 40 years per degree of latitude), even slight changes in their position may have significantly altered the chronologies established for high-latitude cores. If the position of the sea surface  $^{14}\text{C}$  gradient in the southern ocean is assumed to change with the position of the subpolar/subtropical front, there may be an underestimation of the  $^{14}\text{C}$  correction during the glacial periods at a specific location. On the basis of diatom and Radiolaria assemblage variations, the Atlantic southern polar front is believed to have moved equatorward by  $8^\circ$  [Hays et al., 1976; Burckle, 1984]. This corresponds to a difference of about 300-400 years between the Holocene and the glacial  $^{14}\text{C}$  correction for sites located in the  $^{14}\text{C}$  gradient (see the dashed curves in Figure 1). For the Indian and the Pacific oceans, the amplitude of the southern polar front movements was at most about  $5^\circ$ - $7^\circ$  of latitude [Hays et al., 1976] (J. J. Morley, Variations in high-latitude oceanographic front in the southern Indian Ocean, Submitted to *Nature*, 1988), equivalent to correction differences of about 200-300 years. Precise corrections of southern ocean AMS  $^{14}\text{C}$  chronologies of the last deglaciation [Bard et al., 1988b] are made more complicated by the fact that the deglacial variations of the southern polar front are not precisely dated.

The variation of the North Atlantic correction curve may have been greater. During the LGM, the northern polar front was roughly zonal at about  $40^\circ\text{N}$  [Ruddiman and McIntyre, 1981; Bard et al., 1987a] and NADW formation was presumably reduced [Curry and Lohmann, 1983; Boyle, 1984; Broecker et al., 1985b; Mix and Fairbanks, 1985; Berger and Vincent, 1986; Boyle and Keigwin, 1987; Oppo and Fairbanks, 1987]. As a direct result of NADW reduction, a nutrient rich (low  $^{13}\text{C}$ , high Cd/Ca) water mass occupied the North Atlantic below approximately 2 km. The deepwater-surface water  $^{14}\text{C}$  difference for the Atlantic Ocean has recently been documented by means of coupled AMS  $^{14}\text{C}$  dating of benthic and planktonic foraminifera [Broecker et al., this issue (b)]. The results show that the deep-surface age difference was slightly higher during the Glacial period than during the Holocene. In addition, Keffer et al. [1988] suggest that during the LGM the North Atlantic line of zero curl of wind stress ran



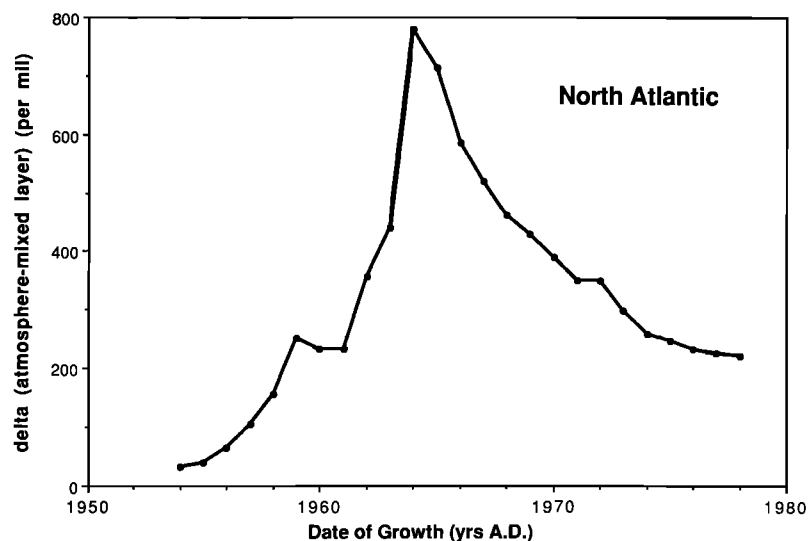


Fig. 4. Difference between the  $^{14}\text{C}$  values in the atmosphere and those in the surface water (corals) during the postbomb years [after Druffel and Suess, 1983]. Bomb  $^{14}\text{C}$  was considered as a spike in the northern hemisphere troposphere only after 1964.

more zonally toward Gibraltar at about  $35^{\circ}$ - $40^{\circ}\text{N}$  and that consequently, the North Atlantic took on more typical polar ocean characteristics like those of the southern ocean. They attribute such a change to the orographic effect of the thick North American ice cap (see also [Manabe and Broccoli, 1985]).

As a result of these hydrographic changes, the apparent age of the North Atlantic surface, which is now roughly constant at about 400-500 years, may have had a South Atlantic-type  $^{14}\text{C}$ -gradient due to a direct response to upwelling and sea-ice distribution (dashed curve, Figure 1a). Consequently, the apparent age of surface sea water would have been greater than 400 years. Like the South Atlantic today, the  $^{14}\text{C}$  ages of high-latitude North Atlantic planktonic foraminifera would have appeared older than the contemporaneous  $^{14}\text{C}$  ages of continental organic matter by as much as 800-1000 years during periods of southward displacement of the polar front. If this hypothesis is true, the inception of the last deglaciation which is dated by AMS dating of planktonic foraminifera at about 14,500 years B.P. [Duplessy et al., 1986; Bard et al., 1987a,b,d, 1988d; Broecker et al., 1988a] (the raw  $^{14}\text{C}$  data of Duplessy et al. [1986] have been corrected for bioturbation effect by Bard et al. [1987b, d]) could be too old by  $\approx 500$  years: the last deglaciation may well have started about 14,000 years B.P.

A way to quantify the reservoir age variation through time would be to measure coexisting terrestrial organic matters (pieces of wood, pollen, charcoals, etc.) and planktonic foraminifera in deep-sea cores raised near the continental coasts. Another way to measure fossil reservoir age would be to compare the  $^{14}\text{C}$  age of a short and well-defined event on land and in deep-sea sediments. Quasi-instantaneous events like the deposition of volcanic ash layers provide such an opportunity. In particular AMS  $^{14}\text{C}$  dating of the Icelandic Vedde ash layer ( $\approx 10,600 \pm 50$  years B.P. on land, i.e. the middle of the Younger Dryas cold event [Mangerud et al., 1984]) should be of great paleoceanographic interest, because North Atlantic circulation during the Younger Dryas climatic

event was presumably glacial-like and characterized by a temporary reduction of the NADW formation rate in response to the injection of meltwater from the decaying continental ice-sheets [Broecker et al., 1985b; Boyle and Keigwin, 1987; Broecker et al., 1988a]. The age difference between "continental" and "oceanic" AMS  $^{14}\text{C}$  dates of the same event should provide a unique test of this hypothesis.

#### CONCLUSION

Knowledge of the apparent  $^{14}\text{C}$  age of carbonate shells formed in ocean surface waters is important for correcting deep-sea sediment chronologies obtained by AMS  $^{14}\text{C}$  dating of planktonic foraminifera. For most of the ocean surface, the variation of this correction due to changes of climatic/oceanographic parameters, which occurred during the last Glacial-Holocene transition, is not likely to have exceeded 100 years, i.e. of the order of the statistical errors associated with radiocarbon dating. Poleward of  $40^{\circ}$ , the variation of the age correction becomes more important as a result of polar/subpolar front movements. In particular, a glacial/interglacial change of several hundred years is expected in the North Atlantic in response to oceanographic changes associated with the last deglaciation. In order to quantify these hypothetical  $^{14}\text{C}$  variations, it is necessary either to date terrestrial organic matter directly associated with sea surface carbonates or to date terrestrial organic matter and planktonic foraminifera contemporaneous with a well-correlated chronologic marker such as a volcanic ash layer.

#### NOTATION

- $\Delta^{14}\text{C}$ -age(i,j) radiocarbon age difference between reservoirs i and j.  
 $N_i$  mass of carbon in reservoir i (for example  $N_a$  for atmosphere,  $N_m$  for mixed layer, etc.).  
 $A_i$  mean  $^{14}\text{C}/\text{C}$  ratio in reservoir i.  
 $k_{ij}$  exchange coefficient from box i to box j.

- $f_{ij}$  kinetic fractionation factor for  $^{13}\text{C}$  from box  $i$  to box  $j$ .  
 $K$  eddy diffusivity for the deep ocean.  
 $z$  depth in the deep ocean.  
 $\lambda$  decay constant of  $^{14}\text{C}$ .  
 $h_m$  mixed layer thickness.  
 $h_d$  mean deep ocean depth.  
 $w_{am}$  piston velocity for  $\text{CO}_2$  exchange.  
 $S_{oc}$  free ocean surface.  
 $\alpha$  solubility of  $\text{CO}_2$ .  
 $P_a$  atmospheric partial pressure of  $\text{CO}_2$ .

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