



Radiocarbon-based circulation age of the world oceans

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[1] The distributions of deep ocean $\Delta^{14}\text{C}$ data are often used to illustrate the rate of deep ocean circulation. The associated conventional ^{14}C ages show a difference of 1000 years between the North Atlantic and the Southern Ocean and a difference of another 1000 years between the Southern Ocean and the North Pacific. These differences may be interpreted directly and mistakenly as the timescale of circulation. The characterization of the deep ocean circulation being millennial is common. Using objectively gridded, natural $\Delta^{14}\text{C}$ “data” of Key et al. (2004), I recast $\Delta^{14}\text{C}$ in terms of circulation by accounting for (1) long-surface ocean reservoir ^{14}C ages and (2) two sources of deep water formed in the North Atlantic and around Antarctica. The new distribution of “circulation ^{14}C ages” is more consistent with the deep ocean being characterized by a centennial timescale than a millennial timescale. Also, the role of the southern sourced deep water is now made more obvious. By accounting for the two important controls on oceanic ^{14}C that are not well known outside the field of chemical oceanography, the new map will be useful as an illustration of global deep ocean circulation to the wider scientific community and as a pedagogical tool to new students in Earth sciences and oceanography.

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1. Introduction

[2] The deep ocean has a very large capacity to hold carbon and heat, both of which are critical in controlling near-surface climate and biology. The timescale of deep ocean circulation is therefore a key parameter in the global climate and ecological systems. Also, it is of central importance in the debate surrounding purposeful sequestration of anthropogenic carbon in the deep ocean because the rate of leakage of sequestered carbon out to the atmosphere depends critically on how rapidly the deep ocean circulates. However, the timescale of deep ocean circulation is not easily determined. Nonconservative tracers such as nutrients phosphate (PO_4^{3-}) and nitrate (NO_3^-) clearly identify the general path of the deep water flow but not its rate. Nutrients are most depleted in the North Atlantic and most enriched in the North Pacific, with the Circumpolar Deep Water (CDW) having intermediate values. The progressive enrichment from the North Atlantic shows accumulation of nutrients as sinking particulate organic matter is remineralized along the path of the global overturning circulation.

[3] Obtaining the rate of deep water flow requires a “clock” such as radiocarbon (^{14}C). Its entry from the atmosphere, where it is produced, into the deep ocean is reasonably well understood, and its radioactive decay rate or half-life is well established. This means that water parcels,

once in the ocean interior and isolated from the atmosphere, will have no source of ^{14}C but only loss by decay at a known rate. Therefore the amount of ^{14}C that has decayed within the ocean is a measure of time since losing contact with the atmosphere. Another example of a clock is chlorofluorocarbons (CFCs) in the oceans. These entirely anthropogenic gases were introduced into the atmosphere at monotonically increasing rates since the 1950s until the advent of the Montreal Protocol enforcement in the 1990s. Because CFCs come into equilibrium in seawater relatively quickly on the timescale of weeks, the dissolved CFC concentrations in seawater can be related to the times when atmospheric CFCs had the appropriate partial pressures to yield those dissolved concentrations. However, CFCs are not entirely appropriate to study the global deep water circulation because they have not been in existence long enough to have penetrated the deep Indian and Pacific oceans. In the Atlantic Ocean, CFC-11 has been detected as far south as 10°S by the North Atlantic Deep Water (NADW) [Smethie et al., 2000].

[4] The first global ocean survey of ^{14}C was made in the early 1970s by the Geochemical Ocean Sections Study (GEOSECS). In a seminal study analyzing the data, Stuiver et al. [1983] presented “replacement times” for the deep Pacific, Indian, and Atlantic of approximately 510, 250, and 275 years, respectively. These are essentially the circulation timescales based on the mass balance of ^{14}C using a simple box model of the world ocean. The relatively short Atlantic timescale has since been confirmed [Broecker et al., 1991].

[5] Yet the centennial timescale appears to be underappreciated in the wider community. The deep ocean circulation timescale is perhaps more typically characterized as

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millennial, sometimes in association with glacial, millennial climate changes [Crowley and North, 1991]. Introductory college textbooks on Earth sciences and oceanography often refer to the deep ocean timescale as millennial [Garrison, 2005; Skinner and Porter, 1995]. The lack of an easy method to appropriately illustrate the centennial flow is evident in a special report by the *Intergovernmental Panel on Climate Change (IPCC)* [2005] on carbon sequestration [IPCC, 2005]. It attempts to illustrate the centennial flow with a map of conventional ^{14}C age of natural radiocarbon (i.e., bomb component removed, as explained below) like that shown in Figure 1. However, conventional ages are known to be too old to illustrate ocean circulation. So, to explain the 2000-year-old water in the North Pacific, the IPCC [2005, p. 292] report is obliged to say that “radiocarbon age is not a perfect indicator of time since a water parcel last contacted the atmosphere.” The IPCC [2005, p. 292] report goes on to say that really, “the age of the North Pacific deep water is estimated to be in the range of 700 to 1000 years.”

[6] It is not well known outside the field of chemical oceanography that ^{14}C age at the surface ocean is much older than “zero.” The typical residence time of surface waters is much shorter than the roughly 10-year period that is required for carbon isotopic equilibration to become established [Broecker and Peng, 1982]. This means that the surface ocean does not have the same ^{14}C age as the atmosphere (i.e., not zero age), thus giving rise to the so-called “surface reservoir age.” When surface waters are transformed into deep waters and become isolated from the atmosphere, the radiocarbon clock begins to tick, but the ^{14}C age is already a few hundred years old. It is as if a piece of wood used in archaeology to obtain a ^{14}C date were already significantly “old,” when the plant from which the wood piece came died. As a result, the conventional ^{14}C age of the newly formed NADW is already 400 years (Figure 1). The oldest northeast Pacific deep water is older than 2400 years, with CDW having an intermediate age of about 1400 years.

[7] Without knowing the equilibration timescale of ^{14}C , one gets the false impression from Figure 1 that it takes 1000 years for NADW to reach the South Atlantic and another 1000 years for CDW to reach the North Pacific. Also, the role of southern sourced deep water, the Antarctic Bottom Water (AABW), is not obvious. It takes some knowledge about ^{14}C disequilibrium and deep water formation to understand correctly that maps like Figure 1 reflect more the timescale of deep ocean ventilation with respect to ^{14}C than of deep ocean circulation. Deep ocean ^{14}C age based only on decay since leaving the surface would give a clearer picture of circulation. Broecker et al. [1991] have done this for the Atlantic Ocean using GEOSECS data, but no such map for the global ocean is available today.

[8] Here I build on the works of Stuiver et al. [1983] and Broecker et al. [1991] and revisit the ^{14}C age of the global deep ocean by analyzing the gridded $\Delta^{14}\text{C}$ data of Key et al. [2004], which are based on measurements obtained mostly by the World Ocean Circulation Experiment (WOCE) and the Joint Global Ocean Flux Study (JGOFS) during the 1990s. The overall goal of this work is to recast water column $\Delta^{14}\text{C}$ in terms of world deep ocean circulation

rather than ventilation. A map of circulation ^{14}C age, defined in section 2 and distinguished from conventional ^{14}C age (Figure 1), will help to illustrate the timescale of deep ocean circulation.

2. Data and Methods

[9] The number of $\Delta^{14}\text{C}$ measurements obtained by WOCE is approximately 15,000 [Key, 1996; Key et al., 2002] compared to the roughly 2200 obtained by GEOSECS. These new measurements were made by both the large-volume technique [Key et al., 1991; Stuiver et al., 1974] and the small-volume technique that use an accelerator mass spectrometer [Key et al., 1996; McNichol et al., 1994]. The reproducibility of both methods is about 4‰ [Elder et al., 1998]. These global $\Delta^{14}\text{C}$ measurements cannot be used directly in this study for two reasons. First, despite the greatly increased number of the new data set, the global coverage still lacks sufficient spatial resolution to make objective mapping meaningful. See Matsumoto and Key [2004, Figure 1] for station locations. Data gaps must be filled before one can obtain an objectively mapped circulation ^{14}C age. Second, the new data from some parts of the world ocean are contaminated with the so-called “bomb ^{14}C .” This refers to the anthropogenic ^{14}C that was produced in the 1950s and 1960s by primarily the United States and the former Soviet Union, which conducted a large number of atmospheric thermonuclear weapon tests. This bomb ^{14}C reacts with O_2 in the atmosphere to form $^{14}\text{CO}_2$, which then enters the surface ocean by air-sea gas exchange and subsequently the deep ocean by circulation. In this study, the interest is not in this bomb ^{14}C but in the “natural” or “background” ^{14}C , which reflects the circulation and must therefore be separated. In the deep ocean this contamination is chiefly an issue in the North Atlantic, where NADW transports bomb ^{14}C down from the surface.

[10] An empirical, linear regression method that separates the bomb and natural components in the measured $\Delta^{14}\text{C}$ can also be used to fill gaps in data coverage. The method capitalizes on an excellent empirical relationship ($r^2 = 0.93$) that exists between potential alkalinity (PALK) and measured $\Delta^{14}\text{C}$ from water samples that are assumed to contain no bomb ^{14}C [Rubin and Key, 2002]. The absence of tritium indicates bomb ^{14}C -free waters; tritium was produced by nuclear weapon tests like ^{14}C but does not exist in the background unlike ^{14}C . This PALK method actually represents an improvement over a previous empirical method based on silica [Broecker et al., 1995]. The silica method had a poor, unexplained correlation with tritium-free $\Delta^{14}\text{C}$ measurements in the critical Southern Ocean. The PALK method overcomes this major problem but has its own, albeit less critical, in the far north Indian Ocean. In this area, seasonal biological productivity associated the Indian monsoon can be very high. Rubin and Key [2002] speculate that the north Indian may be unique in being an exception to the general notion that inorganic carbon particles do not play a significant role in transporting ^{14}C atoms to the deep ocean [Fiadeiro, 1982]. In terms of filling gaps in $\Delta^{14}\text{C}$ data coverage, the PALK method is able to do this by using alkalinity measurements from WOCE/JGOFS, which are more abundant than the $\Delta^{14}\text{C}$ measurements.

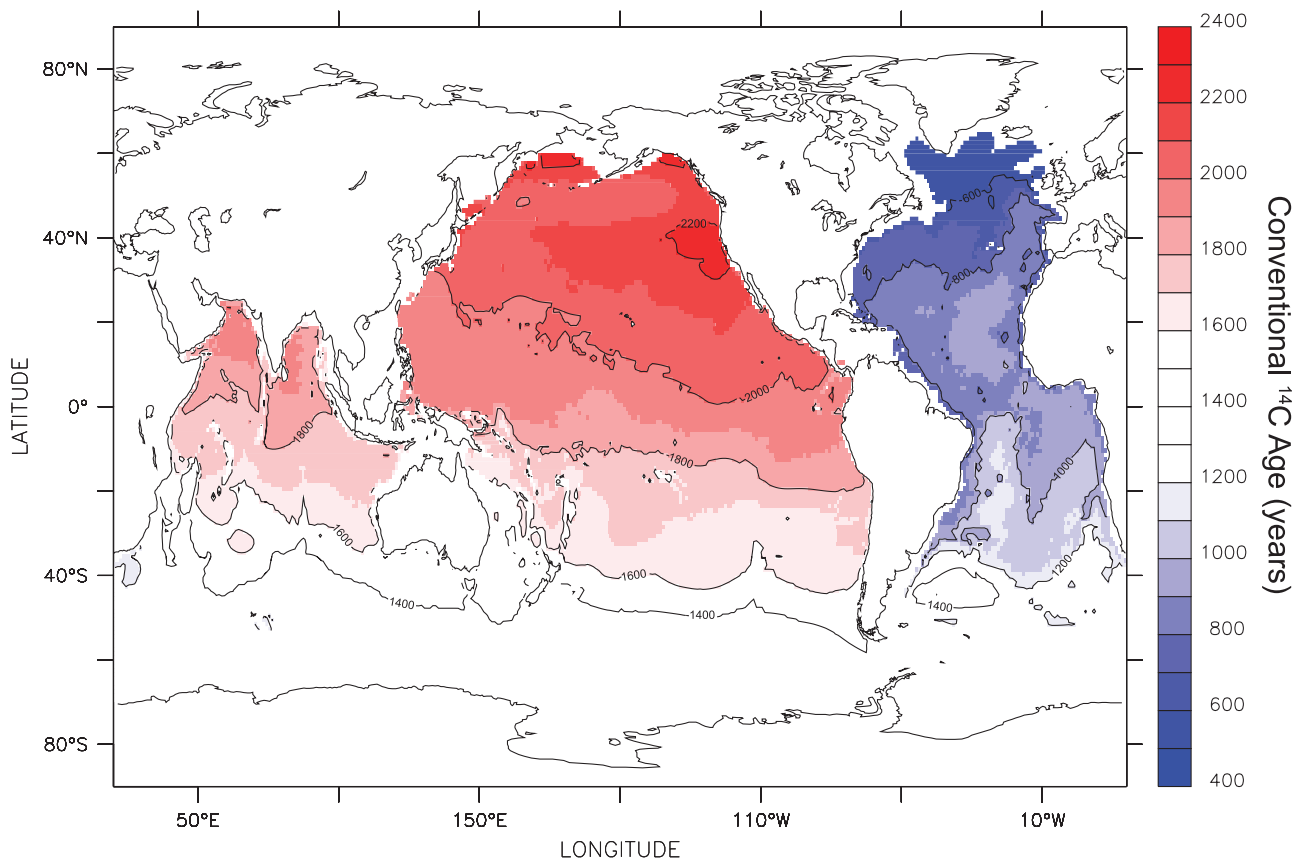


Figure 1. An objectively mapped conventional ^{14}C age of natural radiocarbon below 1500 m, following Matsumoto and Key [2004]. Unit is years.

[11] In this study, I use the PALK-derived, objectively gridded, natural $\Delta^{14}\text{C}$ data of Key *et al.* [2004] (available at Department of Energy site <http://cdiac.esd.ornl.gov/>). The excellent correlation guarantees that the use of proxy data would not compromise the main results of this study. So, for example, the deep North Pacific would have $\Delta^{14}\text{C}$ of about -250% and indicate a conventional ^{14}C age in excess of 2000 years (Figure 1) regardless of whether measured $\Delta^{14}\text{C}$ or derived $\Delta^{14}\text{C}$ is used. Rather, a potentially more important problem with the data set is objective mapping. Details are given by Key *et al.* [2004], but the use of correlation length scales of 1550 km and 740 km at various depth levels in the mapping procedure effectively smoothes features in high-gradient areas. Therefore I restrict my discussion to large, basin-scale features.

[12] There are two basic steps involved in converting natural $\Delta^{14}\text{C}$ in the deep ocean to ^{14}C age that reflects the time elapsed since leaving the surface. The first step determines the fractional contributions of the two recognized end-members of the deep ocean, NADW and AABW. To do this, I follow Broecker *et al.* [1998] in using a biogeochemical tracer PO_4^* . The tracer is essentially the initial surface (or preformed) PO_4^{3-} concentration, and it is defined as $\text{PO}_4^* = \text{PO}_4^{3-} + \text{O}_2/175 - 1.95 \mu\text{mol kg}^{-1}$ [Broecker *et al.*, 1985]. The constant 175 is the average elemental stoichiometry of oxygen to phosphate in particulate organic matter [Anderson and Sarmiento, 1994]. The constant 1.95 is arbitrary. The addition of PO_4^{3-} in subsur-

face waters by degradation of organic matter is corrected in PO_4^* using the apparent oxygen utilization (AOU). AOU is the amount of oxygen consumed accompanying organic matter degradation and is thus related to PO_4^{3-} by the stoichiometry of 175. The use of AOU requires the assumption that surface waters were saturated with respect to atmospheric oxygen; this is not always true but a reasonable assumption for a fast equilibrating gas like oxygen. Therefore PO_4^* is a quasi-conservative tracer. The end-member values for the ventilated, surface contributions to NADW and AABW are defined by Broecker *et al.* [1998]. The southern end-member value is $1.95 \pm 0.07 \mu\text{mol kg}^{-1}$ and represents the Antarctic surface waters in the Weddell Sea and Ross Sea that eventually entrain other waters to become AABW. The northern end-member value is $0.73 \pm 0.07 \mu\text{mol kg}^{-1}$, which pervades the North Atlantic deeper than 1500 m and north of 55°N .

[13] The fractional contributions of the two water masses in a water parcel can thus be determined by a simple, two-end-member mixing analysis. Specifically, the NADW fraction is determined as $f_n = (1.95 - [\text{PO}_4^*]) / (1.95 - 0.73)$ [Sarmiento and Gruber, 2006]. In this equation, the denominator is the dynamic range of PO_4^* , and the numerator is a measure of how close a water parcel is to the end-members. For example, if a water parcel has a PO_4^* of $1.35 \mu\text{mol kg}^{-1}$, then it is halfway between the two end-member values 0.73 and 1.95. It would thus represent a 50-50 mixture, and f_n would be 0.5. The phosphate and

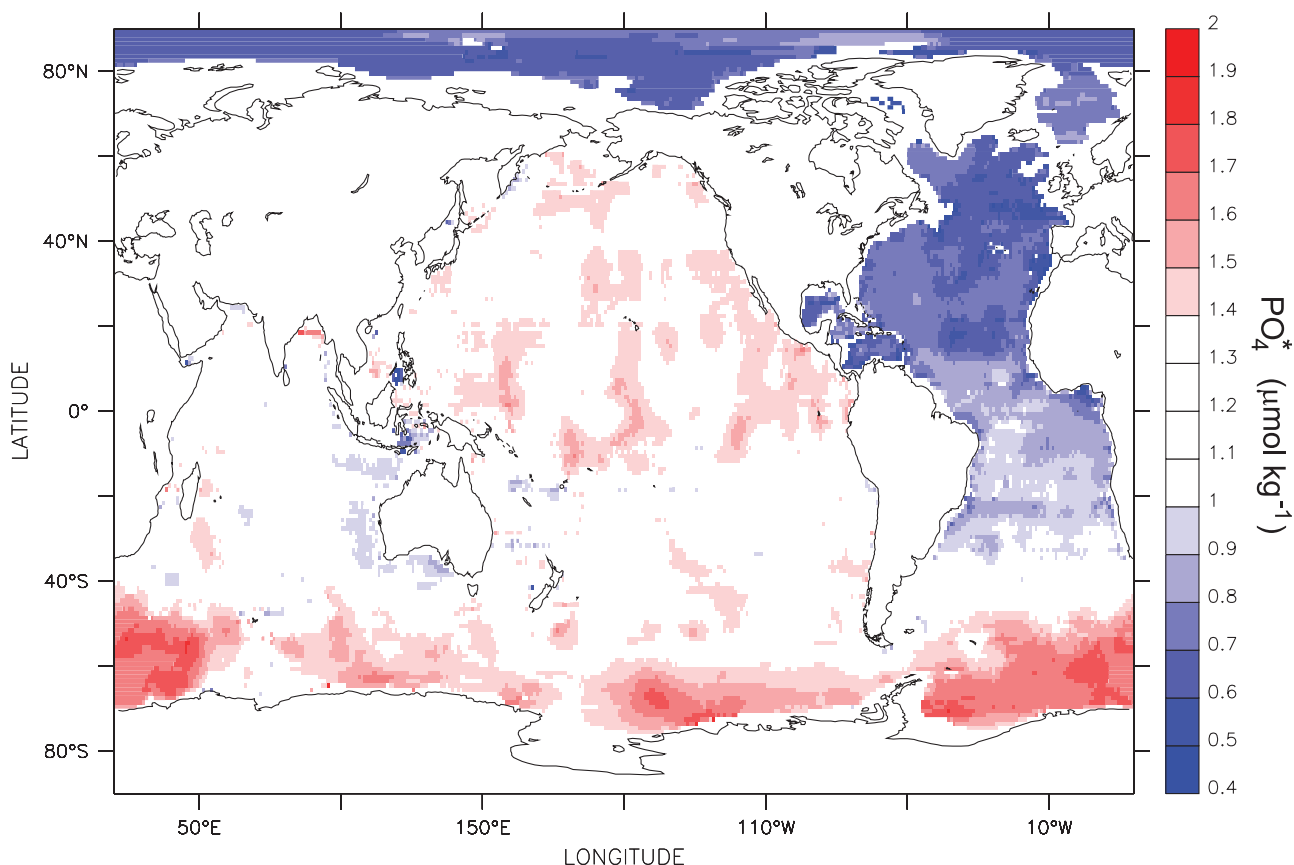


Figure 2. A map of PO_4^* below 1500 m following *Broecker et al.* [1998]. Unit is $\mu\text{mol kg}^{-1}$.

oxygen data sets that I used to make these calculations are the annual mean, Levitus nutrient, and oxygen climatologies [Levitus and Boyer, 1994; Levitus et al., 1993].

[14] In the second step, the reservoir ages of newly formed deep waters are set to zero, so that the radiocarbon clock will actually mark the time since these deep waters have become isolated from the atmosphere. Setting to zero effectively removes the surface reservoir age; it is akin to correcting for the old, contaminated ^{14}C in fresh wood piece in the above archaeology example. This is done for NADW by subtracting -67‰ , which represents $\Delta^{14}\text{C}$ of “newly formed NADW” and for AABW by subtracting -140‰ , which represents $\Delta^{14}\text{C}$ of the “Weddell Sea surface water” [Broecker et al., 1998]. As with PO_4^* end-members, the focus is on the ventilated waters that have recently contacted the atmosphere. The above $\Delta^{14}\text{C}$ values represent surface $^{14}\text{CO}_2$ disequilibrium and thus reservoir ages. It is possible that the southern value may be refined in the future because AABW is formed not just in the Weddell Sea, although it makes the greatest contribution to AABW [Orsi et al., 2002]. Also, the AABW formation is rather complicated and involves shelf processes and mixing of different water masses [Foster and Carmack, 1976]. Nevertheless, the chosen $\Delta^{14}\text{C}$ end-member values are likely quite reasonable, given that the global deep water formation rates estimated by Broecker et al. [1998], using them, and by Orsi et al. [2002], using CFC-11 saturation, are in very close agreement. The two studies used completely independent tools to represent the surface, ventilated waters.

Also, it makes sense that the surface $\Delta^{14}\text{C}$ in the perimeter of Antarctic is more negative than in the open North Atlantic, in part because Antarctic sea ice limits air-sea gas exchange and thereby increases the disequilibrium. Indeed, CFC-11 saturation shows higher values in the North Atlantic [Smethie and Fine, 2000] than in the Southern Ocean [Orsi et al., 2002].

[15] The reservoir age correction for a mixture of NADW and AABW is weighted by their fractions as determined by f_n . For example, if a water parcel is shown to have $f_n = 0.3$, then -118‰ ($-67 \times 0.3 - 140 \times 0.7$) is subtracted from the natural $\Delta^{14}\text{C}$. The residual $\Delta^{14}\text{C}$ reflects the deficit of ^{14}C due to radioactive decay that has occurred in the ocean interior after losing contact with the atmosphere. Here the ^{14}C age of this residual $\Delta^{14}\text{C}$ is denoted circulation ^{14}C age. In contrast, the ^{14}C age without the reservoir age correction (i.e., age of natural $\Delta^{14}\text{C}$) gives the traditional conventional ^{14}C age shown in Figure 1.

[16] For both ^{14}C ages, $\Delta^{14}\text{C}$ is converted to age using the formula $t = -8033 \ln(1 + \Delta^{14}\text{C}/1000)$ [Stuiver and Polach, 1977]. The constant 8033 is the inverse of decay constant λ ($1.21 \times 10^{-4} \text{ yr}^{-1}$) in the standard radioactive decay formula $A = A_0 e^{-\lambda t}$, where A denotes radioactivity. For example, $\Delta^{14}\text{C}$ of -150‰ would have a conventional age of about 1340 years.

[17] All analyses presented here are applied to vertically averaged seawater properties below 1500 m [Stuiver et al., 1983]. Because of the homogeneity of the deep ocean the

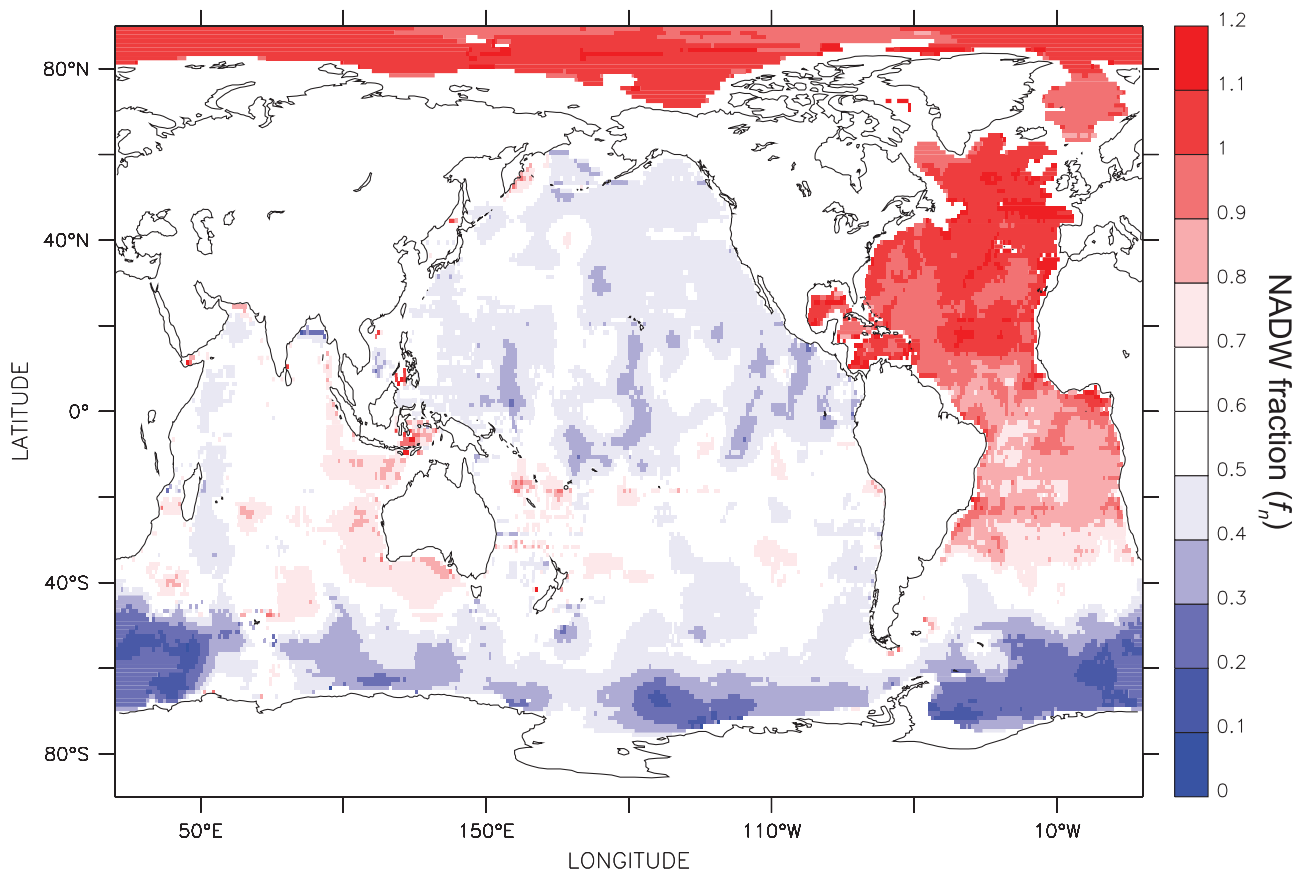


Figure 3. A map of the NADW fraction f_n of the deep water below 1500 m following *Sarmiento and Gruber* [2006]. The AABW fraction is given by $1 - f_n$. There are few isolated regions with $f_n > 1$ because there are some PO_4^* values in the North Atlantic less than the end-member value of $0.73 \pm 0.07 \mu\text{mol kg}^{-1}$.

analyses produce qualitatively very similar results when applied to depth levels of 2000, 3000, and 4000 m.

3. Results

[18] Figure 2 shows the deep ocean PO_4^* distribution using the Levitus data. It confirms previous analyses of this tracer in showing low PO_4^* concentrations in the Atlantic where NADW is the dominant deep water mass [Broecker *et al.*, 1991, 1998; Sarmiento and Gruber, 2006]. In the North Atlantic, PO_4^* approaches the NADW end-member value of $0.73 \mu\text{mol kg}^{-1}$ as expected. Likewise, PO_4^* approaches the AABW end-member value of $1.95 \mu\text{mol kg}^{-1}$ in the perimeter of Antarctica. Low values in the vicinity of Indonesia are likely an artifact of PO_4^* not being a truly conservative tracer.

[19] The associated map of f_n shows that in these regions close to the sites of deep water formation, the water is composed almost entirely of the respective end-members (Figure 3). That is, f_n in the North Atlantic is about 1 or 100% NADW. Much of the Atlantic is dominated by NADW. In contrast, the deep waters around Antarctic are dominated by AABW, although $f_n \approx 0$ (i.e., $1 - f_n \approx 1$) is not obvious in Figure 3, which reflects a water column average. However, it is discernable on discrete deeper levels, which are composed of nearly 100% AABW. Much

of the Indian and Pacific have somewhat more AABW than NADW.

[20] Figure 4 shows the desired map of circulation ^{14}C ages that corrects for the surface reservoir ages and accounts for two sources of deep water. A comparison of this map with the equivalent map of conventional ^{14}C age (Figure 1) map reveals a number of important differences. First, the circulation age is much lower everywhere than the conventional age. The difference in the Indian and Pacific is almost 1000 years. Second, the Atlantic circulation ^{14}C ages are clearly centennial, and young waters are sourced from both polar regions as we expect from the penetration of NADW and AABW. Third, the Southern Ocean has just as young circulation ^{14}C ages as in the North Atlantic. The conventional age in the Southern Ocean is about 1000 years older than in the North Atlantic.

[21] Table 1 summarizes these results by major oceans. The summary includes a sensitivity of the results to the choice of the northern boundary of the Southern Ocean as either 40°S or 50°S [Stuiver *et al.*, 1983]. The sensitivity is only tens of years in all basins (Table 1). Also, the uncertainty in the two end-member PO_4^* values ($\pm 0.07 \mu\text{mol kg}^{-1}$) was propagated through the set of analyses to show that the error in the final circulation ^{14}C age is ± 75 years. The summary is thus good to about 100 years. Additional errors that were not explicitly con-

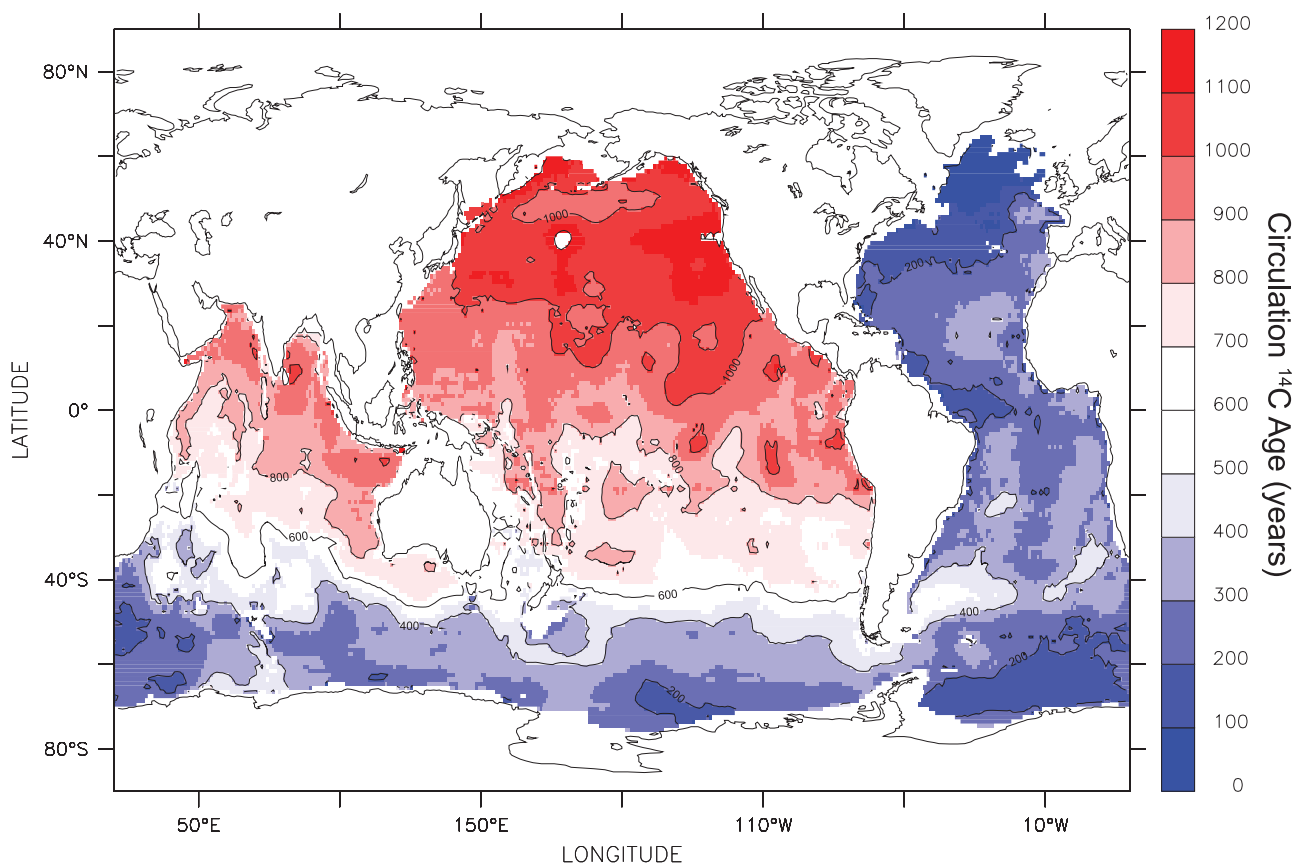


Figure 4. A map of circulation ^{14}C age below 1500 m. This is equivalent to conventional ^{14}C age (Figure 1) but accounts for surface ocean ^{14}C reservoir age and the different sources of deep water. Unit is years.

sidered here include those associated with objective mapping (long-correlation-length scales) and choice of the surface reservoir $\Delta^{14}\text{C}$ (taken straight from *Broecker et al.* [1998]).

4. Discussion and Summary

[22] Two modifications were made to the conventional ^{14}C age (Figure 1) to obtain the new circulation ^{14}C age of the deep ocean (Figure 4). First, the fractional contributions of NADW and AABW were determined using PO_4 (Figures 2 and 3). Second, the initial $\Delta^{14}\text{C}$ of newly formed deep waters were removed, so that their radiocarbon clocks were reset to zero (i.e., removed the reservoir ages). These two considerations allow deep ocean $\Delta^{14}\text{C}$ data to be recast in terms of circulation rather than ^{14}C ventilation.

[23] The circulation age reflects more accurately than the conventional age that there are two sources of deep water, especially in the Atlantic. Figure 1 shows a monotonic decrease in conventional age from the north to the south, as if there is only NADW in the deep ocean. In contrast, Figure 4 shows youngest circulation ages and thus deep waters sourced at both polar regions. The contribution of AABW is more obvious in Figure 4 all around Antarctica.

[24] The large reduction in ^{14}C age in Figure 4 compared to Figure 1 suggests that the deep ocean circulation is more accurately described as centennial [*Broecker et al.*, 1991; *Stuiver et al.*, 1983] rather than millennial. This does not entirely negate the notion of a millennial timescale because circulation ages of 1000 years are found in the North Pacific (Figure 4). However, basin-wide, average circulation ages (Table 1) are all less than 1000 years and contrast with the

Table 1. Summary of Results by Basin^a

	F _n Fraction	Conventional ^{14}C Age, years	Circulation ^{14}C Age, years
Atlantic	0.86 (0.91) ^a	984 (936)	288 (274)
Pacific	0.51 (0.51)	1891 (1922)	889 (916)
Indian	0.61 (0.62)	1627 (1680)	716 (769)
Southern Ocean ^b	0.38 (0.44)	1320 (1360)	295 (372)

^aThe values in the parentheses show the sensitivity of the results to the choice of 40°S as the northern boundary.

^bThe Southern Ocean is defined as south of 50°S.

erroneous first impressions that one gets from conventional ages (i.e., 1000 years in the Atlantic and 1000 years in the Pacific).

[25] The new circulation ^{14}C ages summarized in Table 1 are broadly consistent with the centennial replacement times of *Stuiver et al.* [1983]. The Atlantic circulation ^{14}C age of 288 years compares well with their 275-year replacement time. The new ^{14}C ages of 889 and 716 years for the Pacific and Indian are older than the equivalent replacement times of 510 and 250 years, respectively, but are in fact consistent because those replacement times of *Stuiver et al.* [1983] were calculated relative to CDW. Here, the CDW circulation ^{14}C age is given as 295 years.

[26] The new map of ^{14}C ages (Figure 4) accounts for important pitfalls when interpreting the conventional ^{14}C ages (Figure 1) in terms of circulation. Those pitfalls are not always obvious to those outside the field of chemical oceanography. Now there is a map that actually shows what the special IPCC [2005] report on carbon sequestration says about the age of the North Pacific deep water. In addition to meeting such a practical need and serving the wider scientific community, Figure 4 will be useful as a pedagogical tool to students taking introductory courses in Earth sciences and oceanography.

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