Debate over the ocean bomb radiocarbon sink: Closing the gap

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[1] There has over recent years emerged a controversy as to how much of the radiocarbon released into the atmosphere by nuclear weapons testing has been taken up by the ocean. Hesshaimer et al. [1994] made a case based on stratospheric and tropospheric measurements coupled with estimates of total bomb radiocarbon yield that it was not possible to explain both atmospheric observations and existing ocean-based bomb radiocarbon uptake estimates [Broecker et al., 1985, 1995]. They therefore proposed that estimates of the oceanic sink should be revised downward by about 25%. One reason for concern over this discrepancy is that the widely used wind speed dependent air-sea gas exchange parameterization of Wanninkhof [1992] is scaled to give an average exchange rate matching that given by the ocean bomb-radiocarbon budget. An example of an application of the Wanninkhof [1992] parameterization is in estimating ocean CO₂ uptake based on direct measurements of the air-sea pCO₂ difference [Takahashi et al., 1997]. Such estimates scale linearly with the air-sea gas exchange coefficient. Further, as has been highlighted by the Hesshaimer et al. [1994] study, an understanding of the global budget of radiocarbon is an important issue in and of itself. In this paper, a number of new approaches to assessing the size of the ocean bomb radiocarbon sink are explored, and estimates are given for the total ocean bomb radiocarbon inventory during both the mid-1970s and the mid-1990s. The revised estimates of bomb-radiocarbon ocean uptake yield a mid-1970s inventory in closer agreement with that proposed by Hesshaimer et al. [1994] than the inventory obtained by the Broecker et al. [1995] study.

INDEX TERMS: 0312 Atmospheric Composition and Structure: Air/sea constituent fluxes (3339, 4504); 4299 Oceanography: General: General or miscellaneous; 4599 Oceanography: Physical: General or miscellaneous; 4504 Oceanography: Physical: Air/sea interactions (0312); KEYWORDS: ocean bomb-radiocarbon budget


1. Introduction

[2] Controversy over the magnitude of the oceanic sink for bomb radiocarbon remains an important unresolved issue [Broecker et al., 1985; Sarmiento and Sundquist, 1992; Siegenthaler and Sarmiento, 1993; Hesshaimer et al., 1994; Joos, 1994; Broecker et al., 1995; Hesshaimer and Levin, 2000; Nydal, 2000]. On the apparent discrepancy between ocean-based and atmosphere-based approaches to estimating the magnitude of the ocean bomb radiocarbon sink, Hesshaimer et al. [1994] stated, “This problem needs to be resolved,” to which Broecker et al. [1995] responded, “We see no way by which this estimate [295–305 × 10^{26} atoms] could be reduced to the 225 × 10^{26} atom value suggested by Hesshaimer et al.”

[3] Assuming a production rate of 1.05 × 10^{26} radiocarbon atoms per megaton of TNT, and utilizing measurements of bomb-radiocarbon in the troposphere and stratosphere, Hesshaimer et al. [1994] argued that the Broecker et al. [1995] estimate of ocean bomb radiocarbon uptake for the mid-1970s [Broecker et al., 1985, 1995] led to a global budget which was seriously out of balance. On the basis of a simple global carbon cycle model, they argued that either a missing source of bomb radiocarbon equaling some 80×10^{26} atoms must be invoked, or that existing estimates of the ocean sink must be overestimated by this amount.

[4] The Broecker et al. [1995] (hereinafter referred to as B95) estimate of 305 ± 30 × 10^{26} atoms ocean uptake of bomb radiocarbon in 1975 (or, 295 × 10^{26} atoms on 1 January 1974, corresponding to the date adopted in the Hesshaimer et al. [1994] study) is based on an observed correlation between natural radiocarbon and dissolved silicate in waters assumed to be free of bomb-derived tracers. Extrapolation of this correlation to thermocline waters led these authors to an estimate of the natural (pre-bomb) radiocarbon distribu-
tion in the upper ocean at specific locations for which there existed both radiocarbon and silicate measurements. The bomb radiocarbon at each hydrographic station was then estimated as the difference between observed and natural radiocarbon profiles. If the B95 natural radiocarbon estimate at each station is correct, then the estimate of bomb radiocarbon at each station must, by definition, also be correct.

[5] However, the Broecker et al. [1995] approach has a number of sources of hard-to-quantify error. The first global ocean survey (Geochemical Sections, “GEOSECS,” mid-1970s) sampled radiocarbon only at a number of discrete locations, and coverage was far from uniform (the black dots in Figure 9 (in section 2.4) represent the GEOSECS station locations at which radiocarbon was measured). Therefore, in order to obtain a global inventory estimate, Broecker et al. [1995] chose to divide each ocean into 10-degree-wide latitude bands, and to assume the bomb radiocarbon estimates at stations falling within each band represented the mean for the entire band. It is likely, however, that tracer distributions are in places strongly non-uniform across a line of constant latitude. A second source of error in the B95 approach is in the estimation of the natural radiocarbon field. The average least squares error of the silicate-natural radiocarbon correlation is just over 13\%, which introduces an error to the predicted natural radiocarbon field which is of order 10\%. Furthermore, the linear silicate-\Delta^{14}C relation that holds in most of the ocean was observed to break down at the few high-latitude Southern Ocean GEOSECS stations, leading to a higher error in Southern Ocean bomb radiocarbon inventories.

[6] The error in the estimation of the natural radiocarbon field can be expressed as an error bar on the final inventory estimate, and this was done by B95. However, the error arising from the non-uniform station distribution, and the extrapolation over 10 latitude bands of station-based bomb radiocarbon estimates, is harder to quantify. This problem is addressed by two separate approaches in this paper.

[7] The first part of the paper is concerned with better constraining the ocean bomb radiocarbon inventory in the mid-1970s. Two separate approaches are taken. One method relies on an ocean model to separate bomb and natural radiocarbon in the ocean. Once waters have been sorted into bomb-contaminated and bomb-free, a multiple tracer correlation-based extrapolation method is used to infer the total (natural + bomb) radiocarbon concentrations throughout the ocean. A slight variation on the B95 silicate method is used to infer the natural radiocarbon distribution. Global gridded \(^1^\)\(^\text{C}\) fields are estimated for natural and total radiocarbon, and the global bomb radiocarbon field is then simply the difference between natural and total. The main error in this approach is the large scatter in the tracer-tracer correlations in the thermocline. The second method relies on using an ocean model to correct for the extrapolation error in the B95 method. In the second part of the paper, an estimate of the ocean bomb-radiocarbon distribution is made for the mid-1990s based on the recently collected World Ocean Circulation Experiment (WOCE) data-set.

2. Ocean Bomb Radiocarbon Inventory in the Mid-1970s: Method 1

[8] The problem of obtaining an estimate of the amount of bomb radiocarbon in the ocean is threefold. The first problem is to somehow separate waters into those “contaminated by the bomb” and “natural” (non-contaminated) waters. Having achieved this, one must then estimate the natural distribution in the “bomb contaminated” water. Finally, it is necessary to estimate the total (bomb + natural, or “observed”) distribution. The bomb radiocarbon distribution is then simply the difference between the natural and the total radiocarbon estimates. Such a process can either be done on a station-by-station basis (the B95 approach) and then extrapolated to a global estimate, or can be done directly on a global basis. The first approach taken in this paper is to seek global estimates of the natural and total radiocarbon ocean distribution in the mid-1970s.

2.1. Separation of Bomb-Contaminated and Natural Radiocarbon in the Ocean

[9] The question of how to distinguish between waters contaminated by bomb-produced radiocarbon and those which represent the natural background radiocarbon distribution in the ocean has long been a problem. The most widely adopted approach has been to use tritium (another bomb-derived tracer which does not have a natural background in the ocean) to infer the penetration depth of radiocarbon [Broecker et al., 1985, 1995; Rubin and Key, 2002]. It has been convincingly argued by Broecker et al. [1995] that tritium and bomb radiocarbon should have penetrated to similar depths in the ocean, despite the fact that tritium was incorporated into rain and very rapidly rained out over the Northern Hemisphere ocean, whereas radiocarbon had a much longer residence time in the atmosphere and became well mixed before crossing the air-sea interface as \(^1^\text{CO}_2\) molecules. The reason that strong similarities should be expected in the depth of penetration of these tracers in the ocean at a given time is because advection and diffusive processes will act to distribute two tracers with a similar input history over time. The main shortcomings of using tritium measurements to estimate the penetration depth of bomb radiocarbon are that existing tritium measurements are often at different hydrographic stations than radiocarbon, and there exist relatively few tritium data.

[10] In this paper, an alternative approach to the separation of bomb-free and bomb-contaminated waters is taken: The penetration depth of bomb radiocarbon is assumed to be very close to that of chlorofluorocarbons (CFCs). The global penetration depth of CFCs at the time of GEOSECS and WOCE is estimated based on the results from an ocean general circulation model (GCM). The model-predicted penetration depth is used to separate waters that are presumed free of a bomb-radiocarbon signal from those contaminated by the bomb signal. Details of the model are given in Appendix A.
[11] What basis is there for such an expectation? CFC-11 began accumulating in the atmosphere in significant amounts in the late 1950s to early 1960s. Bomb radiocarbon was first introduced into the atmosphere in the 1950s, and peaked in the mid-1960s. The main difference between the two tracers is in the shape of the input function, not the timescale of input: The bomb radiocarbon was injected into the atmosphere as a “pulse” which has gradually decayed up to the present day, whereas CFC-11 concentrations showed an exponential rise in the atmosphere, plateauing in the late 1990s. It is argued here that any tracer that is introduced to the ocean on a similar timescale should show a similar penetration depth in the ocean, regardless of the actual shape of the input function. It is essentially the same point made by Broecker et al. [1995]: The advective-diffusive processes in the ocean will result in two different magnitudes of signal for two different tracers with a similar input timescale and different input function, but the signals should be spread to similar depths.

[12] CFCs were not measured during GEOSECS; however, they were measured at unprecedented spatial resolution during WOCE. One way to test the postulation that the penetration depth of the two tracers should be similar is to look at WOCE CFC-11 and tritium data obtained from water in the same bottles. Such data exist for many of the WOCE lines. Shown in Figure 1 is a scatterplot of measured CFC-11 versus measured tritium from the same water samples for various Pacific Ocean cruises. Each color/symbol represents a different cruise line (no selection for depth or location was made on a given cruise track; all data are plotted). The fact that CFC-11 approaches zero at the same depth that tritium approaches zero indicates a similar penetration depth for the two tracers. In order to estimate the global distribution of penetration depth of bomb radiocarbon, an ocean general circulation model (GCM) that has been independently proven to accurately simulate the CFC-11 thermocline distribution and penetration depth was used [Peacock et al., 2004].

[13] The depth to which CFC-11 has penetrated in the model at the time of GEOSECS is very similar to the depth of penetration of tritium, as obtained from GEOSECS measurements. This can be seen in Figure 2, which shows measured GEOSECS tritium profiles from the Pacific, Indian, and Atlantic Oceans, and the Parallel Ocean Program (POP) 1975 CFC-11 penetration depth (defined as the depth at which the CFC-11 concentration falls to less than 1% of the mean surface value) interpolated onto the selected GEOSECS stations. There are order 100 good GEOSECS tritium profiles; of these, only two regions (five stations) were identified in which the match between simulated CFC-11 and measured tritium was poor: GEOSECS stations 223 and 224 and the northwest Pacific, at roughly 35°N (which coincide with the highly variable path of the Kuroshio extension), and stations 413, 416, and 417 in the northwest Indian Ocean (a region in which tracer gradients change dramatically over a scale not resolved by coarse-mesh models). Two of the “poor fit” stations are shown in Figure 2; the fit for the rest of the GEOSECS stations at which tritium was measured was uniformly extremely good.

[14] The excellent agreement between observed tritium penetration depth and simulated CFC-11 penetration depth is the first strong argument in favor of using results from a model CFC simulation to estimate the penetration depth of bomb radiocarbon in the ocean. Another important reason is that it is much easier to simulate CFCs than bomb radiocarbon, because the equilibrium time is days rather than years (thereby making it much easier to eliminate drift in simulated fields). Finally, there now exists a global CFC data set from WOCE at unprecedented spatial coverage. Because CFCs have no natural background, a model-data comparison is straightforward and unambiguous.

[15] The model provides an estimate of the penetration depth everywhere in the ocean. In order to select waters that are bomb contaminated and bomb free, the depth at which simulated CFC dropped to below 1% of the mean surface concentration at a given time was selected as the divider of the bomb influence. It should be noted that the definition of penetration depth adopted here is a different measure than that defined by B95 as “mean penetration depth.” The mean penetration depth is defined as being equal to the column inventory (mol/m²) divided by the surface concentration (mol/m³) at a given point [Broecker et al., 1995]. The Broecker definition is an integral property, which will always be smaller than the actual depth to which a tracer has penetrated (Figure 3). All references to “penetration depth” herein refer to the level at which the tracer drops to less than 1% of its surface concentration.

[16] Figure 4 shows the POP simulated CFC-11 penetration depth for 1975 and 1995. The deepest penetration is observed in the far North Atlantic, and the Weddell and Ross Seas in Antarctica. Most of the Southern Ocean is also characterized by a large penetration depth. There is a clear east-west gradient, most noticeable in the Pacific, where the eastern part of the basin has a considerably shallower tracer penetration than the western region. The subtropical gyres
stand out as local maxima in penetration depth; this is particularly pronounced in the North Pacific. There is an increase everywhere in penetration depth between the mid-1970s and the mid-1990s.

2.2. Estimating the Natural Radiocarbon Distribution in the Ocean

[17] In order to obtain an estimate of natural radiocarbon in bomb-contaminated waters, B95 exploited a strong correlation between silicate and natural radiocarbon ($\Delta^{14}C_N$) in the deep (non-bomb-contaminated) ocean. They showed that, importantly, in tritium-free thermocline waters, the same tight correlation also held. This observation led to the idea of using measured silicate to infer the pre-bomb distribution of radiocarbon in the bomb-contaminated thermocline.

[18] One source of error in the B95 study in applying this $\Delta^{14}C_N$-silicate relation was in the Southern Ocean, where there was a marked deviation away from the global ocean trend. This deviation only holds in the deep waters of the Southern Ocean, which are characterized by a high-silicate signature (>70 µmol/kg). These deep, high-silicate waters are characterized by a very homogeneous $\Delta^{14}C_N$ of between $-150$ and $-160\%$. The fact that they deviate from the linear relation that characterizes the rest of the world does not in any way make the silicate method redundant; rather, it requires that deep Southern Ocean $\Delta^{14}C_N$ be inferred using a different correlation than that of the rest of the world ocean.

[19] The approach taken here is to exploit the $\Delta^{14}C_N$-silicate correlation discovered by B95, but to make the following minor modifications: First, a very slightly different fit between $\Delta^{14}C_N$ and silicate was adopted (based on the best fit to the bomb-free waters after utilizing a model-based penetration depth estimate). Second, a different correlation

![Figure 2. Measured tritium as a function of depth for selected GEOSECS stations. Upper panel shows Pacific stations; middle panel shows Indian stations, and lower panel shows Atlantic stations. The large star marks the model-derived penetration depth of CFC-11 at the time that the GEOSECS measurements were made.](image-url)
was used for deep, high-silicate (>70 μmol/kg) waters in the Southern Ocean than that used for the rest of the world ocean. Finally, the Levitus annual mean silicate climatology (1 global gridded data set [Levitus, 1982]) was used to obtain a global three-dimensional (3-D) estimate of the natural radiocarbon distribution in the ocean. [20] Using the penetration depth obtained from the CFC-11 simulation in POP, all the GEOSECS data that fell into the "bomb-free water" category were selected, and are plotted, separated by ocean basin, in Figure 5. The best fit line to the data north of 40°S was almost identical to the B95 relation,  \[
\Delta^{14}C_N = -66.68 - (0.96 \times SiO_2),
\]  (1)

It should be noted that the separation into bomb-contaminated and bomb-free waters in B95 was based strictly on observed penetration depth of tritium; in this study it was based on model-simulated penetration depth of CFC-11 interpolated onto the GEOSECS stations. The best fit line to the Southern Ocean data was found to be  \[
\Delta^{14}C_N = -131 - (0.2 \times SiO_2),
\]  (2)

The natural radiocarbon fields estimated in this way at the ocean surface, 1000 m, 2000 m, and 3500 m are shown in Figure 6. Cross sections of the predicted natural radiocarbon distribution are shown in Figure 7. [21] In the Pacific cross section, the oldest waters are found north of about 20°N, and have a \(\Delta^{14}C_N\) of around −220‰. The Southern Ocean is very homogeneous below the uppermost layers, with a mean \(\Delta^{14}C_N\) concentration of between −150 and −160‰. The thermocline marks a very sharp gradient in \(\Delta^{14}C_N\), with surface-ocean values between −50 and −60‰ in most of the subtropical and tropical oceans. The Indian Ocean also shows a strong north-south gradient in the deep ocean \(\Delta^{14}C_N\), but the lowest values are only about −180‰, much higher than the average Pacific Ocean values at a corresponding latitude. The Atlantic is for the most part very "young" (\(\Delta^{14}C_N\) values of less than −125‰). The signature of Antarctic Intermediate Water is apparent at about 30°S and 1500 m depth. The signature of Antarctic Bottom Water is also apparent in its very depleted \(\Delta^{14}C_N\) signature between 40°S and the equator. The surface value lies between −60 and −70‰ in most of the subtropical and tropical ocean; the only significant deviation from this is in the Southern Ocean, particularly the Ross and Weddell Seas, where the surface \(\Delta^{14}C_N\) drops to as low as −150‰. The surface \(\Delta^{14}C_N\) estimate is between −60 and −80‰ for much of the surface ocean and is slightly (order 10‰) lower than many coral-based estimates of the pre-

Figure 3. Model-simulated CFC-11 in 1977 at roughly 35°S, 180°W (black line). The shaded area depicts the boundary of surface concentration and penetration depth computed following Broecker et al. [1995], as column inventory divided by surface concentration. The Broecker definition yields a depth which is far shallower than the actual depth to which the tracer penetrates.

Figure 4. Model penetration depth of CFC-11 in (top) 1974 and (bottom) 1994, defined as the depth at which the tracer concentration drops to 1% of its surface value. Contours show depths shallower than 1000 m in intervals of 100 m.
The reason for this can be seen in Figure 5; the best fit line in the surface (low silicate, low $\Delta^{14}C_N$) waters deviates slightly from a trend toward less $^{14}C$-depleted waters seen in the data. This is a small but systematic bias in the estimate of the natural radiocarbon field. The implication for global budget estimates is that this will cause a very small upward bias (that is, correction to the “true” pre-bomb surface-ocean value would lower the bomb$^{14}C$ inventory estimate very slightly).

2.3. Estimating the Total Radiocarbon Distribution at the Time of GEOSECS

In order to estimate the total (bomb + natural) radiocarbon distribution in the ocean at the time of GEOSECS, the best fit correlation (purely empirical) between total radiocarbon and various other tracers was sought; this fit was then used to extrapolate into regions with no data, based on annual mean climatologies.

The number of ocean radiocarbon measurements are few and widely scattered in space and time. However, there are tracers in the ocean for which there exist orders of magnitude more measurements, both spatially and temporally. The Levitus gridded climatology of the global annual-

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**Figure 5.** GEOSECS silicate data plotted against $\Delta^{14}C$. The data included in the plot are those in the bomb-free water, as computed based on the POP CFC-derived penetration depth. The black line shows the best fit for waters north of 40°S; the gray line shows the best fit to waters south of 40°S.

**Figure 6.** Predicted distribution of natural radiocarbon on depth surfaces 0 m, 1000 m, 2000 m, and 3500 m.
mean distribution of the tracers temperature (T), salinity (S), oxygen (O$_2$), and dissolved silicate (SiO$_2$) exists at horizontal resolution with 33 variable-thickness vertical levels [Levitus et al., 1998]. The temperature data set is based on over 4.5 million ocean temperature profiles collected over the last century. In contrast, at the time of the GEOSECS, the first global survey of ocean D$_{14}$C, D$_{14}$C depth profiles were obtained. The reason for seeking a global estimate of the total radiocarbon field in the mid-1970s is to get around the problem of extrapolating between very sparse data points across an ocean that may have strong zonal tracer gradients.

[24] It must be stressed that there is no reason to believe that any correlation that exists between total radiocarbon and other steady state tracers in the upper ocean should hold in the deeper, bomb-free waters. For this reason, a model-based estimate of penetration depth was used to provide a cutoff below which the total radiocarbon field is required to be identically equal to the natural radiocarbon field. (i.e., bomb radiocarbon at each grid point is set to zero below the model-derived penetration depth).

[25] A trial-and-error approach yielded the following function as that yielding the smallest prediction error for observed radiocarbon:

$$ \Delta^{14}C_{\text{total}} = \alpha \text{SiO}_2 + \beta \sqrt{\text{SiO}_2} + \gamma T + \epsilon O_2 + \varepsilon S + \zeta z^2 + \eta (\text{lat}) + \theta T^2, $$

where ($\alpha$, $\beta$, ..) are the regression coefficients, $z$ is depth, and lat is latitude. On the basis of the Levitus [Levitus et al., 1998] annual mean climatologies for temperature (T), salinity (S), oxygen (O$_2$), and silicate (SiO$_2$), the observed radiocarbon distribution at the time of GEOSECS was estimated based on the coefficients obtained from a least squares regression. It must be emphasized here that the

Figure 7. Predicted distribution of natural radiocarbon on depth-transects in the (top) Pacific, (middle) Indian, and (bottom) Atlantic Oceans. The separation was made based on equation (2) for the Southern Ocean south of 40°S and with silicate greater than 70 µmol/kg; equation (1) was used to infer natural radiocarbon in the rest of the world ocean. The prediction was made based on the GEOSECS data set.
prediction skill for the observed radiocarbon distribution was very low compared to that for natural radiocarbon. The mean least squares prediction error was 30%, and the $r^2$ value was a relatively poor 0.83. A number of sensitivity studies in which the size of the data set was randomly cut by 30% had a very minor effect on prediction skill. The error bar on the estimate of global distribution of observed radiocarbon, and hence on global distribution of bomb radiocarbon, is therefore large; it is also dependent on the absolute magnitude of the bomb radiocarbon signal (waters with a smaller bomb$^{14}$C signal have a correspondingly larger error bar). If the average bomb radiocarbon is taken to be 150%, this puts an error bar of 20–25% on the estimate of the bomb-radiocarbon field using this tracer-tracer correlation method. The estimated global total radiocarbon field is by far the most uncertain component in obtaining a global bomb radiocarbon inventory.

### 2.4. Bomb Radiocarbon at the Time of GEOSECS

[26] In order to obtain an estimate of the global bomb radiocarbon field in the mid-1970s, the estimated natural radiocarbon field was simply subtracted from the estimated total radiocarbon field. The estimate of bomb radiocarbon at various depth horizons in the ocean is shown in Figure 8. The estimate of spatial distribution of global ocean bomb radiocarbon inventory in the mid-1970s is given in Figure 9. The GEOSECS stations locations are shown by black dots. [27] Figure 9 exhibits significant variation in predicted bomb radiocarbon inventory along lines of constant latitude. In the Pacific and Atlantic Oceans in particular, the locations of the GEOSECS stations are such that there are few stations outside of the low latitudes that sample the low-inventory waters characteristic of upwelling regions along the eastern margins of the oceans. Furthermore, the entire Arctic region is seen to be characterized by relatively low inventories. The Arctic inventory was assumed by Broecker et al. [1995] to be equal to the average obtained from the two northernmost Atlantic stations. It appears from this estimate of the bomb radiocarbon field that the consequence of assuming that the locally derived GEOSECS inventories represent averages over entire latitudinal bands will be an overestimate of the net ocean inventory. This issue will be explored in greater detail in section 3. On the basis of this extrapolation method, the total inventory implied by the functions yielding the smallest prediction error was $224 \times 10^{26}$ atoms. The error bar associated with this prediction is
estimated to be 30% in $\Delta^{14}C$ units (from the multiple linear regression). This translates to a random error of order 20–25%, which is approximately $5 \times 10^{26}$ atoms.

[29] The inventories obtained by this tracer-correlation method are not necessarily equal to those obtained by B95 at the GEOSECS stations; the inventories predicted by this method are compared with those of B95 at the GEOSECS stations in Figure 10. If we assume that the natural radiocarbon profiles estimated by B95 at the GEOSECS stations are correct, then it follows that the B95 station-by-station inventories are also correct. Therefore, in regions where there is disagreement between the predictions of B95 and this study at the GEOSECS stations, it is likely that the B95 inventory is more-or-less correct and the inventory predicted by this study is in error.

[30] Figure 10 shows that in the Atlantic Ocean, the agreement between B95 and this study is very good everywhere except at the stations lying at around 30°N, where this study predicts an inventory that is slightly too low. In the Indian Ocean, the agreement is close except between 20°S and 40°S, where this study again underestimates the station inventories. A mismatch in the same direction is also seen in the Pacific between 20°S and 40°S. This study also predicts inventories that are higher than those of B95 in waters north of 30°N in the Pacific. On the basis of the station-by-station comparison, it seems likely that the estimate presented here contains a systematic downward bias. The mean offset between the inventory estimates obtained from the multiple linear regression and those of B95 at the GEOSECS stations is 15%. If this correction is assumed to represent a systematic bias, and a correction of this magnitude is applied to the $224 \times 10^{26}$ atoms obtained above, a “bias-corrected” estimate of $258 \times 10^{26}$ atoms is obtained. This is still significantly lower than the B95 estimate.

[31] However, the error bar on this estimate is frustratingly large. Is it possible to better constrain the magnitude of the error? To try and address this question, a completely different approach is taken to estimating the mid-1970s bomb radiocarbon inventory in section 3.

3. Ocean Bomb Radiocarbon Inventory in the Mid-1970s: Method 2

[32] 1. The first possible source involves using the silicate-$\Delta^{14}C_N$ relation to estimate the natural radiocarbon distribution. The error in the predicted natural radiocarbon field comes directly from the correlation between natural radiocarbon and silicate, and is judged to be of order 13%.

[33] 2. The second possible source relates to the range in collection dates of GEOSECS data. B95 show that the temporal evolution of the total bomb radiocarbon inventory is steep during the 1970s, with an increase of roughly 20% in just 5 years. The GEOSECS data were collected between July 1972 and April 1978. The B95 estimate of global inventory, taking into account the distribution in collection dates, is $305 \times 10^{26}$ atoms for 1 January 1975, or $295 \times 10^{26}$ atoms for 1 January 1974 (the date for which the Hessheimer estimate of $225 \times 10^{26}$ atoms as the best fit ocean sink was made). The error associated with the time-corrected estimate of the total bomb radiocarbon inventory is thought to be less than 5% [Broecker et al., 1995].

[34] 3. The third possible error source involves assumed penetration depth. In estimating the bomb radiocarbon penetration depth, tritium was commonly measured on different water samples than radiocarbon, and extrapolation/interpolation was therefore necessary in order to estimate the bomb radiocarbon penetration depth. However, this error is judged to be small (1–2%).

[35] 4. A final possible source relates to the extrapolation method for areas with no data. The most difficult error bar to assign is that involved with extrapolating to areas with no data coverage. The goal of section 2 was to improve on this error. However, the extrapolation method used itself had a very large error bar (20–25%). In this section we take a very different approach to estimating the error in extrapolating to the global ocean inventory based just on measurements from the GEOSECS stations. B95 took average inventories for 10 latitude bands in each ocean and assumed these were representative of the entire ocean over that latitude range. The averages for each latitude band were frequently based on just one or two stations. We have here chosen to test the error in this assumption of zonal homogeneity by analyzing the spatial distribution of tracers in the POP ocean GCM.

[36] The model simulation of CFCs has been shown in independent work [e.g., Peacock et al., 2004] to very closely agree with WOCE observations throughout the thermocline, giving significant confidence in the simulated 1990s spatial inventory of the CFCs. Anthropogenic CO$_2$ has also been simulated in the same model, and again shows good agreement with independent estimates of its oceanic distribution (unpublished work).

[37] The approach taken here was to subsample the model CFC-11 distribution in the year 2002, and anthropogenic...
CO₂ in 1975 at the locations of the GEOSECS stations. The mean inventory for each 10° latitude band was then computed in exactly the way done with bomb radiocarbon by B95: based on just extrapolation of the mean from the GEOSECS stations falling within each latitude band. This “station-based” inventory estimate was then compared with the true model inventory for each 10° latitude band in each ocean. The difference was then a correction factor that can be directly applied to the B95 bomb radiocarbon estimates. The model-predicted CFC-11 inventories in 2002 for each 10° latitude band of each ocean, and the CFC-11 inventories computed using just the mean CFC-11 from the GEOSECS stations that fall within each latitude band, are shown in Tables 1–3. The results for anthropogenic CO₂ are very similar, and for this reason are not shown. The reason for carrying out this analysis for both CFCs and anthropogenic CO₂ was to test the sensitivity of the results to both the timescale over which the tracer had been present in the ocean and the shape of the tracer input function. Neither are the same as for bomb radiocarbon; both CFCs and anthropogenic CO₂ gradually increased over time, but anthropogenic CO₂ began entering the ocean much earlier than did CFCs; CFCs plateau in the late 1990s, and anthropogenic CO₂ continues to rise. The bomb radiocarbon input to the ocean, in contrast, was an early spike and then gradual decay. It is assumed that the spatial distribution of bomb radiocarbon in the ocean should fall somewhere between the simulated CFC and anthropogenic CO₂ distributions. The CFC-11 sampling-bias was estimated based on data from 2002 because of the relative shapes of the bomb ¹⁴C and CFC-11 boundary conditions; because the CFC-11 rose much more slowly in the atmosphere, it was thought that it might take a somewhat

**Figure 10.** Bomb radiocarbon inventory at the GEOSECS stations in the (top) Atlantic Ocean, (middle) Indian Ocean, and (bottom) Pacific Ocean plotted as a function of latitude. The red crosses show the inventory estimates (10⁹ mol/m²) obtained in this study, interpolated onto the GEOSECS stations. The blue dots show the inventory estimates obtained in the Broecker et al. [1995] study at the same stations.
longer time to attain a spatial distribution similar to bomb\(^{14}\)C at an earlier time. However, it has been verified that the relative spatial gradients in the ocean change remarkably little with time in the model, so the actual year chosen does not affect the conclusions presented.

[38] Tables 4–6 give the mean bomb radiocarbon inventories computed by B95 for each 10-degree latitude band of each ocean, based on the mean values for the stations within each latitude band. The correction factors from both CFC and anthropogenic CO\(_2\) are applied to the B95 numbers, and the “corrected” B95 inventories given in the final two columns of each table. The magnitude of the correction is actually remarkably similar for both the CFC-based and the anthropogenic CO\(_2\)-based results. It was found that by far the largest error in the B95 method occurred in the Atlantic Ocean, and that the correction for the Pacific and Indian Oceans was significantly smaller. The total global correction required from the CFC-based estimate was 30.45 \times 10^{26} \text{ atoms}, and that from the CO\(_2\)-based estimate was 36.27 \times 10^{26} \text{ atoms}. Applying these corrections to the B95 estimate of 295 \times 10^{26} \text{ atoms} (for 1 January 1794) yields a corrected B95 bomb radiocarbon inventory of between 259 \times 10^{26} and 265 \times 10^{26} \text{ atoms}. The error bar on this estimate is judged, based on independent work examining the fit between observed and simulated CFC-11 and CO\(_2\), to be less than 10%.

[39] For completeness, it should be noted that the uncorrected (“raw”) total inventory computed by B95 was 329 \times 10^{26} \text{ atoms} for 1 January 1975. To this number a correction was applied in order to account for the shallow shelf regions (see Broecker et al. [1995] for details). For consistency, the “actual model CFC-11 inventories” in Tables 1–3 were computed for the same open-ocean fraction as computed by B95 in order that this bias and correction be consistent with that assumed by B95. It is therefore just the difference between the B95 and corrected inventories that should be subtracted from the B95 estimate for 1 January 1974.

### 4. Estimating the Ocean Uptake of Bomb Radiocarbon in the 1990s

[40] The first difficulty in estimation of the mid-1990s ocean bomb radiocarbon inventory is that only the Pacific Ocean data are currently publicly available. Therefore the approach taken was as follows: An estimate of the Pacific Ocean inventory was obtained by extrapolating across latitude bands based on station inventories; the ocean GCM results were then used to infer how partitioning of tracer between Atlantic, Pacific, and Indian Oceans has changed through time, and hence obtain a global bomb radiocarbon inventory estimate for the mid-1990s.

[41] The choice was made not to use the method of seeking correlations between the total radiocarbon field and tracers for which there exist annual mean climatologies (section 2) to obtain a bomb radiocarbon estimate for the mid-1990s for two reasons. First, on the basis of the experience with applying this tracer correlation method to the GESECS data, we believe the error associated with this method to be undesirably large (order 20–25% for the GESECS data). Second, very few WOCE radiocarbon
Table 4. Corrected Bomb Radiocarbon Inventory for the Atlantic Ocean, Divided Into 10° Latitude Bands

<table>
<thead>
<tr>
<th>Latitude</th>
<th>B95 Mean Inventory, 10^26 atoms/cm²</th>
<th>B95 Total Inventory, 10^26 atoms</th>
<th>% Correction (CFC-11)</th>
<th>% Correction (CO2)</th>
<th>Corrected Inventory (CFC-11), 10^26 atoms</th>
<th>Corrected Inventory (CO2), 10^26 atoms</th>
</tr>
</thead>
<tbody>
<tr>
<td>20°N–10°N</td>
<td>16.8</td>
<td>19.2</td>
<td>56.76</td>
<td>56.76</td>
<td>8.30</td>
<td>8.30</td>
</tr>
<tr>
<td>30°N–20°N</td>
<td>23.4</td>
<td>15.3</td>
<td>10.4</td>
<td>19.89</td>
<td>11.38</td>
<td>11.16</td>
</tr>
<tr>
<td>20°–30°N</td>
<td>21.7</td>
<td>15.3</td>
<td>13.17</td>
<td>2.73</td>
<td>17.31</td>
<td>18.48</td>
</tr>
<tr>
<td>10°N–20°N</td>
<td>4.2</td>
<td>2.8</td>
<td>9.26</td>
<td>15.79</td>
<td>2.54</td>
<td>2.36</td>
</tr>
<tr>
<td>10°S–10°S</td>
<td>4.2</td>
<td>2.6</td>
<td>24.59</td>
<td>30.7</td>
<td>1.90</td>
<td>1.8</td>
</tr>
<tr>
<td>30°S–20°S</td>
<td>9.2</td>
<td>6.2</td>
<td>9.71</td>
<td>15.28</td>
<td>5.6</td>
<td>5.25</td>
</tr>
<tr>
<td>40°S–30°S</td>
<td>10.3</td>
<td>7.7</td>
<td>9.62</td>
<td>11.84</td>
<td>6.96</td>
<td>6.79</td>
</tr>
<tr>
<td>50°S–40°S</td>
<td>8.2</td>
<td>6.00</td>
<td>2.69</td>
<td>−2.79</td>
<td>5.84</td>
<td>6.17</td>
</tr>
<tr>
<td>60°S–50°S</td>
<td>3.7</td>
<td>2.20</td>
<td>6.48</td>
<td>9.92</td>
<td>2.06</td>
<td>1.98</td>
</tr>
<tr>
<td>90°S–60°S</td>
<td>2.5</td>
<td>1.4</td>
<td>−4.2</td>
<td>−8.79</td>
<td>1.46</td>
<td>1.52</td>
</tr>
<tr>
<td>Total</td>
<td>111.4</td>
<td></td>
<td></td>
<td></td>
<td>90.97</td>
<td>84.49</td>
</tr>
<tr>
<td>Difference</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>20.43</td>
<td>26.91</td>
</tr>
</tbody>
</table>

The second column gives the inventory (atoms/cm²) that [Broecker et al., 1995] obtained for each 10° latitude band. The third column gives the total inventory obtained by B95. The implied percent correction to be applied to the station inventories from the model CFC-11 and anthropogenic CO2 distributions are given in the fourth and fifth columns. The B95 inventories with these corrections applied are given in the final two columns. The final two rows show the total inventories in each ocean obtained with the B95 original and corrected inventories, and the difference between the B95 original and corrected inventories.

A very extensive data set of bomb-radiocarbon measurements exists for the Pacific Ocean. The first step was to apply the “B95 method” (extrapolation over 10-degree latitude bands based on average station inventories) to the WOCE Pacific radiocarbon data. The natural radiocarbon was estimated based on the measured silicate profiles at each station. The total radiocarbon content at each station was simply the observed profile, and the bomb radiocarbon component at each station was computed as the difference between observed and natural profiles. The bias in assuming that station-based means were representative of zonal means was tested for each latitude band using the POP tracer distributions in exactly the same manner as described in section 3. It was found that the WOCE stations are sufficiently densely spaced that they do in fact very closely represent zonal mean concentrations in the Pacific. When the POP-based correction factors were applied, the difference in total inventory was found to be a mere 0.66%, and therefore the station-based inventory estimates are presented here without any correction factor.

The stations at which radiocarbon profiles were measured from the surface down to the estimated radiocarbon penetration depth (based on POP tracer simulations) are shown in Figure 11. Table 7 shows the average WOCE-era bomb radiocarbon inventories for each 10-degree latitude band in the Pacific, and the inferred total Pacific Ocean...
The WOCE data-based estimate of the Pacific bomb radiocarbon inventory during the mid-1990s is $158.71 \times 10^{26}$ atoms.

The second step was to proceed from this Pacific inventory to a global inventory estimate. Simulation of CFC-11 and anthropogenic CO$_2$ in POP reveal that the partitioning of tracers between the various ocean basins is remarkably constant with time, despite a continuous increase in total ocean inventory. Table 8 shows how the CFC-11 inventory is partitioned between the Atlantic, Pacific, Indian, and Arctic Oceans between the early 1970s and late 1990s. The partitioning for anthropogenic CO$_2$ was extremely similar, and is not shown. If it is assumed that this GCM partitioning also holds true for the real ocean (the reason we have to believe it does is that in the 1990s the model so accurately predicts the global CFC distribution, at least everywhere outside of the Southern Ocean), then it is possible to simply scale the Pacific inventory and thereby obtain global estimate. Applying a 44.99% scaling gives a global inventory estimate of $353 \times 10^{26}$ atoms for the mid-1990s.

This estimate is probably somewhat upwardly biased. The reason for this is that although there are far more WOCE stations than there were GEOSECS stations, the majority of these stations still lie in the open ocean. Accounting for a reduction in inventory in coastal regions is expected to pull this estimate downward by between 5 and 10%. Therefore our best estimate for the mid-1990s global inventory is somewhere between $320 \times 10^{26}$ atoms.

It should be noted that the bomb radiocarbon Pacific inventory estimated by B95 was 44.1% of their estimated global ocean value. In the corrected B95 estimates we present here for the mid-1970s, the mid-1970s Pacific estimate was 47.9% of the global corrected value. These numbers are in good agreement with the model-based estimates.

Table 6. Corrected Bomb Radiocarbon Inventory for the Indian Ocean, Divided Into 10° Latitude Bands

<table>
<thead>
<tr>
<th>Latitude</th>
<th>B95 Mean Inventory, 10$^9$ atoms/cm$^2$</th>
<th>B95 Total Inventory, 10$^{26}$ atoms</th>
<th>% Correction (CFC-11)</th>
<th>% Correction (CO$_2$)</th>
<th>Corrected Inventory (CFC-11), 10$^{26}$ atoms</th>
<th>Corrected Inventory (CO$_2$), 10$^{26}$ atoms</th>
</tr>
</thead>
<tbody>
<tr>
<td>10°N–20°N</td>
<td>5.9</td>
<td>2.7</td>
<td>15.63</td>
<td>11.11</td>
<td>2.28</td>
<td>2.40</td>
</tr>
<tr>
<td>Equator–10°N</td>
<td>5.2</td>
<td>3.2</td>
<td>5.13</td>
<td>8.82</td>
<td>3.04</td>
<td>2.92</td>
</tr>
<tr>
<td>10°S–equator</td>
<td>4.8</td>
<td>4.9</td>
<td>19.35</td>
<td>9.09</td>
<td>3.95</td>
<td>4.45</td>
</tr>
<tr>
<td>20°S–10°S</td>
<td>9.9</td>
<td>10.0</td>
<td>17.82</td>
<td>12.57</td>
<td>8.22</td>
<td>8.74</td>
</tr>
<tr>
<td>40°S–30°S</td>
<td>17.6</td>
<td>19.2</td>
<td>9.38</td>
<td>5.34</td>
<td>17.40</td>
<td>18.17</td>
</tr>
<tr>
<td>50°S–40°S</td>
<td>12.1</td>
<td>13.3</td>
<td>0.77</td>
<td>22.84</td>
<td>13.20</td>
<td>16.34</td>
</tr>
<tr>
<td>60°S–50°S</td>
<td>4.2</td>
<td>3.8</td>
<td>-15.06</td>
<td>-15.69</td>
<td>4.37</td>
<td>4.40</td>
</tr>
<tr>
<td>90°S–60°S</td>
<td>3.1</td>
<td>1.6</td>
<td>9.89</td>
<td>4.60</td>
<td>1.44</td>
<td>1.53</td>
</tr>
<tr>
<td>Total</td>
<td></td>
<td>73.5</td>
<td></td>
<td></td>
<td>67.27</td>
<td>72.61</td>
</tr>
<tr>
<td>Difference</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>6.23</td>
<td>0.89</td>
</tr>
</tbody>
</table>

*aSee caption to Table 4 for details.

Table 7. Estimated Bomb Radiocarbon Inventory for the Pacific Ocean Based on the WOCE Data

<table>
<thead>
<tr>
<th>Latitude</th>
<th>Area, 10$^{13}$ m$^2$</th>
<th>Mean Inventory at WOCE Stations, 10$^9$ atoms/cm$^2$</th>
<th>Total Inventory Mid-1990s, 10$^{26}$ atoms</th>
</tr>
</thead>
<tbody>
<tr>
<td>50°N–60°N</td>
<td>0.57</td>
<td>4.89</td>
<td>2.79</td>
</tr>
<tr>
<td>40°N–50°N</td>
<td>0.86</td>
<td>7.81</td>
<td>6.72</td>
</tr>
<tr>
<td>30°N–40°N</td>
<td>1.17</td>
<td>11.84</td>
<td>13.85</td>
</tr>
<tr>
<td>20°N–30°N</td>
<td>1.49</td>
<td>13.06</td>
<td>19.46</td>
</tr>
<tr>
<td>10°N–20°N</td>
<td>1.87</td>
<td>6.74</td>
<td>12.60</td>
</tr>
<tr>
<td>Equator–10°N</td>
<td>2.12</td>
<td>5.46</td>
<td>11.58</td>
</tr>
<tr>
<td>10°S–equator</td>
<td>1.75</td>
<td>6.46</td>
<td>11.31</td>
</tr>
<tr>
<td>20°S–10°S</td>
<td>1.72</td>
<td>9.37</td>
<td>16.12</td>
</tr>
<tr>
<td>30°S–20°S</td>
<td>1.53</td>
<td>13.15</td>
<td>20.12</td>
</tr>
<tr>
<td>40°S–30°S</td>
<td>1.37</td>
<td>12.79</td>
<td>17.52</td>
</tr>
<tr>
<td>50°S–40°S</td>
<td>1.20</td>
<td>12.77</td>
<td>15.32</td>
</tr>
<tr>
<td>60°S–50°S</td>
<td>1.03</td>
<td>9.39</td>
<td>9.67</td>
</tr>
<tr>
<td>90°S–60°S</td>
<td>1.00</td>
<td>1.66</td>
<td>1.66</td>
</tr>
<tr>
<td>Total</td>
<td></td>
<td></td>
<td>158.71</td>
</tr>
</tbody>
</table>

*aThe ocean has been divided into 10° latitude bands. The areas here are slightly different from those in Table 2; those in Table 2 are from the POP model, while the areas in this table were taken from B95.

Figure 11. Locations of WOCE Pacific stations at which radiocarbon measurements were made from the surface ocean down to the estimated bomb radiocarbon penetration depth.
Table 8. Percentage of CFC-11 in the Atlantic, Pacific, Indian, and Arctic Sectors of the World Ocean as a Function of Timea

<table>
<thead>
<tr>
<th>Year</th>
<th>Atlantic, %</th>
<th>Pacific, %</th>
<th>Indian, %</th>
<th>Arctic, %</th>
<th>Total Inventory, 10^8 moles</th>
</tr>
</thead>
<tbody>
<tr>
<td>1970</td>
<td>34.29</td>
<td>46.98</td>
<td>16.29</td>
<td>2.44</td>
<td>0.55</td>
</tr>
<tr>
<td>1974</td>
<td>34.46</td>
<td>46.79</td>
<td>16.33</td>
<td>2.42</td>
<td>0.996</td>
</tr>
<tr>
<td>1979</td>
<td>34.79</td>
<td>46.41</td>
<td>16.42</td>
<td>2.38</td>
<td>1.75</td>
</tr>
<tr>
<td>1984</td>
<td>35.29</td>
<td>45.89</td>
<td>16.53</td>
<td>2.29</td>
<td>2.55</td>
</tr>
<tr>
<td>1989</td>
<td>35.74</td>
<td>45.47</td>
<td>16.58</td>
<td>2.21</td>
<td>3.48</td>
</tr>
<tr>
<td>1994</td>
<td>36.26</td>
<td>44.99</td>
<td>16.63</td>
<td>2.11</td>
<td>4.27</td>
</tr>
<tr>
<td>1999</td>
<td>36.89</td>
<td>44.47</td>
<td>16.64</td>
<td>2.00</td>
<td>4.89</td>
</tr>
</tbody>
</table>

aTotal inventory is also shown.

[47] Nothing has yet been said about the error due to the non-synchronous timing of the data collection. This is because, for the mid-1990s, it is judged to be very small. On the basis of the 1-D model of Broecker and Peng [1994], the ocean radiocarbon inventory should be expected to plateau in the 1990s, implying that the magnitude of the correction due to non-synchronous sample collection is expected to be very small over this period.

[48] One final consistency check can be undertaken. The 1-D model of Broecker and Peng [1994] predicts that the increase in total ocean bomb radiocarbon inventory between 1975 and 1995 should have been approximately 30%. A mid-1990s ocean bomb radiocarbon predicted here is 320–350 × 10^26 atoms. Using the 1-D model as a guide, this implies a 1975 inventory of 224–245 × 10^26 atoms for 1974. This falls within the error bars of the corrected B95 inventory obtained from method 1 (224–258 × 10^26 atoms, with an error bar of 25%), and the estimate from method 2 (roughly 260 ± 26 × 10^26 atoms, with an error bar of order 10%).

5. Discussion

[49] It is worth saying a few words about a recently proposed alternative method by which to estimate the natural radiocarbon distribution in the ocean. This method is based on a global deep-ocean linear relation between “potential alkalinity” (a function of nitrate, alkalinity, and salinity) and Δ14C in Rubín and Key, 2002] (hereinafter referred to as the “PA” method).

[50] The PA authors claim that the advantages of this method over the silicate method are a slightly lower prediction error, a Southern Ocean relation that does not deviate from the global trend, and a more accurate rendering of surface-ocean pre-bomb radiocarbon concentrations. However, they go on to demonstrate that the predicted inventories of bomb radiocarbon obtained at the GEOSCECS stations are not very different from those obtained using the silicate method. A disadvantage of the PA method is that it is not as widely applicable as is the silicate method (nitrate, alkalinity, and salinity are all required to compute potential alkalinity; interpolation of three tracers to a common point will involve hard-to-quantify interpolation error). Furthermore, the deviation of the silicate-Δ14C trend from the global ocean B95 line in the Southern Ocean does not necessarily need to be considered a disadvantage: It simply becomes necessary to fit a different function to the high-silicate deep Southern Ocean waters. On the basis of the model-derived CFC penetration depth, the best fit correlation between silicate and natural radiocarbon for waters north of 40°S has an r2 value of 0.91. The r2 value for the best fit line between potential alkalinity and natural radiocarbon on these same waters is 0.90. This translates to an extremely small difference in prediction skill.

6. Summary and Conclusions

[51] In summary, two new methods have been applied to the problem of estimating the ocean bomb radiocarbon inventory. The first method was to seek correlations between tracers for which there exist global climatologies, and total observed radiocarbon, in order to try and infer the total radiocarbon field. Combined with the already proven silicate method to estimate the natural radiocarbon field, this yields a global estimate of the bomb radiocarbon component, albeit with a rather large error bar. The estimated ocean bomb radiocarbon inventory in the mid-1970s obtained from this correlation-based technique is judged to lie between 224 and 258 × 10^26 atoms, with an error bar of 20–25%.

[52] The second method was to use an ocean GCM to assess the magnitude of the correction required in estimating a truly zonal mean based on just a small handful of hydrographic stations for a given latitude band. The POP ocean model, in a configuration which has been independently shown to simulate remarkably closely the global WOCE CFC distribution in the ocean’s thermocline, was the model used to assess the magnitude of the correction. This method led to an ocean bomb radiocarbon inventory estimate of between 259 and 265 × 10^26 atoms for the mid-1970s. The error bar on this is estimated to be order 10%.

[53] In order to obtain an estimate of the mid-1990s bomb radiocarbon inventory, the Pacific Ocean inventory was first estimated using the B95 extrapolation method. Then the distribution in each ocean basin of CFC-11 and anthropogenic CO2 was computed as a function of time from POP, and it was found that despite a continual increase in total inventory, the percentage of total tracer in each ocean basin remained remarkably constant through time. On the basis of this observation, it was possible to take the estimate of the Pacific Ocean inventory in the mid-1990s (assumed to be known quite accurately, due to the very good spatial coverage in this ocean during the WOCE period) and obtain a global inventory estimate simply by scaling the Pacific number. The resulting estimate, corrected for shallow ocean margins, is 320–350 × 10^26 atoms as the ocean bomb radiocarbon inventory during the mid-1990s. Finally, it was shown that these results are self-consistent: The projected increase in ocean bomb radiocarbon inventory between the mid-1970s and mid-1990s is roughly 30%, in fair agreement with the projected increase based on the 1-D model of Broecker and Peng [1994].
most consistent with the available data. It is hoped that this paper conveys an appreciation of how tricky the problem of better constraining the ocean bomb radiocarbon inventory is from a data-based perspective.

Appendix A: Ocean Model Details

[55] The model used for the tracer simulations reported herein was a coarse-resolution configuration of the Parallel Ocean Program (POP), a z-coordinate ocean general circulation model (OGCM) with a variable free surface [Dukowicz and Smith, 1994]. Overall, the average grid spacing was about 200 km. There were 25 non-uniformly spaced vertical levels, varying from 12 m at the surface to 450 m at depth, and bathymetry was interpolated from the ETOP05 database.

[56] Owing to the coarseness of the horizontal grid, several state-of-the-art parameterizations of subgrid-scale mixing were implemented. The Gent-McWilliams (GM) formulation was used for eddy mixing of tracers [Gent and McWilliams, 1990] (Redi and Bolus diffusivities were both set at $8 \times 10^7$ cm$^2$/s; a maximum slope of 0.1 was used for the Redi and Bolus terms; for details of terms see http://climate.lanl.gov/Models/POP/UsersGuide.pdf). A grid-aligned anisotropic viscosity was used for momentum [Smith and McWilliams, 2003]. The KPP parameterization was used for vertical mixing of momentum and tracers [Large et al., 1994], with gravitational instabilities resolved by means of large diffusion coefficients (0.1 m$^2$s$^{-1}$). A polynomial UNESCO equation of state was used for calculating density.

[57] Surface forcing was formulated in a manner very similar to Large et al. [1997], using monthly mean fields from a variety of sources. Wind stresses, atmospheric temperature, humidity, and wind speed were obtained from a 1985–1988 climatology from the National Center for Atmospheric Research/National Centers for Environmental Prediction (NCAR/NCEP) reanalysis [Kalnay et al., 1996]. Cloud fraction (used for calculating longwave radiative flux) and shortwave radiation were from International Satellite Cloud Climatology Project (ISCCP) [Rossow and Schiffer, 1991], and precipitation was a blended Xie-Aarkin/MSU product [Xie and Arkin, 1997; Spencer, 1993]). Heat fluxes were calculated using the Large and Pond bulk formulae [Large and Pond, 1982]. Surface restoring of salinity and temperature to annual values with a timescale of 15 days was performed under climatological ice. There was also weak surface restoring of salinity in the open ocean with a timescale of 100 days. A time step of 1 hour was used.

[58] The model was initialized with Levitus mean annual temperature and salinity fields [Levitus and Boyer, 1994] and was then spun up for 500 years without passive tracers. This is certainly not long enough for the model to be in equilibrium with the surface forcing, but drifts were judged to be acceptably small. CFCs crossed the air-sea interface following standard Ocean Carbon-Cycle Model Intercomparison Project protocol [Dutay et al., 2002].

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