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Is there an imbalance in the global budget of bomb-produced radiocarbon?

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Abstract. Several recent studies of the global inventory of radiocarbon produced by above ground nuclear weapons testing have brought into question our understanding of the global cycle of bomb-produced radiocarbon. Radiocarbon produced from these explosions has provided a unique test for global carbon cycle models used in the analysis of emission scenarios for carbon dioxide. We employ a globally aggregated model for the global cycles of carbon and its isotopes (^{13}C and ^{14}C) to examine these studies, and find several modeling approximations or assumptions which could be responsible for the differences between analyses. In light of the considerable uncertainty in both model-based and data-based estimates of global inventories, we conclude that the global budget of bomb-produced radiocarbon cannot be shown to be out of balance. Uncertainties limit the utility of ^{14}C as a tracer for determining the flow of carbon dioxide within the atmosphere-ocean-terrestrial-biosphere system of carbon cycle. Our model-based analyses suggest that improved analysis of past nuclear tests and their production of radiocarbon, as well as additional measurements of ^{14}C in the biosphere and oceans, could reduce uncertainties in model studies of the evolution of ^{14}C in the carbon cycle system.

Introduction

Concerns have been raised by two independent model studies of the evolution of the bomb ^{14}C budget [*Broecker and Peng*, 1994; *Hesshaimer et al.*, 1994] which highlight an inconsistency in their model-estimated budget for radiocarbon. Using estimates based on a schematic coupled atmosphere-ocean model as well as a compilation of measurements of tropospheric radiocarbon composition, the estimate of bomb ^{14}C in the stratosphere, and a compilation of bomb detonation dates and strengths, *Hesshaimer et al.* [1994] estimated the combined inventories (i.e., the sum of atmosphere, ocean and biosphere) of bomb-produced radiocarbon at the time of the Geochemical Ocean Sections Study (GEOSECS, 1972-1978). They found a major discrepancy between the calculated and observed atmospheric concentration of ^{14}C and suggested that the ocean uptake estimates based on observations made during the GEOSECS survey be reduced by 25% and a similar reduction be added to the model-based estimates for ocean uptake of anthropogenic CO_2, in order to eliminate this discrepancy. Meanwhile, *Broecker and Peng* [1994] estimated oceanic and biospheric inventories and inferred the stratospheric inventories by assuming the total bomb radiocarbon to be constant between 1964 and 1990. *Broecker and Peng* [1994] found that the inferred stratospheric inventories did not agree with the limited observations available for stratospheric radiocarbon, implying an error in one or more of the model inventories or the source term for bomb ^{14}C production. More recently, *Broecker et al.* [1995] revised the global oceanic bomb ^{14}C inventory for 1975 based on the GEOSECS data survey spanning the period from 1972 to 1978. The revised estimate of *Broecker et al.* [1995] turned out to be consistent with their earlier estimate [*Broecker et al.*, 1985]. On the basis of their revised findings they concluded that oceanic uptake estimates of bomb ^{14}C could not be reduced by 25% as suggested by *Hesshaimer et al.* [1994]. Using the schematic model of *Broecker and Peng* [1994], *Broecker et al.* [1995] also recalculated the bomb ^{14}C budget from 1965 onward, and found that they were able to balance the bomb ^{14}C budget within the limits of perceived uncertainties, whereas *Hesshaimer et al.* [1994] started their calculations in 1950 and found their model estimated bomb ^{14}C budget was outside of their limits of uncertainty. On the basis of schematic model estimates, *Broecker et al.* [1995] reached the conclusion that for times earlier than 1965, the stratosphere and perhaps tropospheric inventories were uncertain because of inhomogeneties in the distribution of bomb radiocarbon during the first years after the test ban was implemented [*Broecker et al.*, 1995].

Duffy and Caldeira [1995] have injected additional analyses into this debate. They used an enhanced version of the Geophysical Fluid Dynamics Laboratory (GFDL) ocean general circulation model (OGCM) [*Duffy et al.*, 1995] to estimate the ocean uptake of bomb ^{14}C and found that their OGCM-estimated ocean inventory after 1975 is smaller than the schematic model estimates of *Hesshaimer et al.* [1994] and *Broecker and Peng* [1994]. *Duffy and Caldeira* [1995] presumed that the reason for this difference is that the general

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circulation model includes important physical processes, which are omitted from the simpler models. On the basis of their OGCM results of the ocean inventory of bomb ^{14}C, *Duffy and Caldeira* [1995] concluded that the apparent imbalance in the global budget of bomb ^{14}C is smaller than estimated by other studies [*Hesshaimer et al.*, 1994, and *Joos*, 1994] and is not significantly different from zero. A simulation of bomb ^{14}C uptake and transport with an earlier version of the GFDL ocean OGCM by *Toggweiler et al.* [1989] preceded *Duffy and Caldeira*'s [1995] study; however, the bomb ^{14}C inventory estimated by *Toggweiler et al.* [1989] failed to match the observation-based estimates of *Broecker et al.* [1985], leading to the conclusion that the description of transport processes in the earlier version did not accurately represent the ocean uptake of bomb ^{14}C [*Toggweiler et al.*, 1989].

In this paper we reexamine the bomb ^{14}C inventories in the oceans and terrestrial biosphere, in the context of previous studies, using our globally aggregated model of carbon cycle [*Jain et al.*, 1995, 1996; *Kheshgi et al.*, 1996], which we describe in the following section. Using observed inventories for the stratosphere and troposphere, we estimate the bomb ^{14}C budget over the period 1950-1990. We then compare our model-estimated inventories for bomb ^{14}C in the oceans and terrestrial biosphere to form a global bomb ^{14}C budget, which is compared to observed data and the results of other carbon cycle models. Finally, the relative merits of both data-based and model-based estimates of ^{14}C inventories are discussed.

The Model

The model system used in this study consists of an upwelling diffusion ocean model described in detail by *Jain et al.* [1995, 1996] and a six-box biosphere model described in detail by *Kheshgi et al.* [1996]. Recirculation of polar bottom water is included in the model's deep ocean. A marine biosphere source term is included in the deep sea associated with the oxidation of the organic debris exported from the mixed layer, where it is produced by photosynthesis. The ocean transport parameters, upwelling velocity w and eddy diffusivity k, are calibrated by matching the preindustrial ^{14}C distribution in the deep ocean. The values of k and w required to match the preindustrial vertical profile of ^{14}C are 4700 m^2 yr^{-1} and 3.5 m yr^{-1} [*Jain et al.*, 1995]. The carbonate equilibrium in the mixed ocean layer is calculated from the full set of chemical equations as described by *Peng et al.* [1987]. The air-sea gas exchange flux, for the calibrated values of k and w, was estimated from the ^{14}C balance equation of the surface reservoir for the preindustrial system [*Jain et al.*, 1995]; the resulting global mean gas exchange rate at preindustrial CO_2 concentration of 278 ppmv is 17 mol m^{-2} yr^{-1}.

The biosphere model is a six-box, globally aggregated model coupled to the atmosphere box of the carbon cycle model. The six boxes are ground vegetation, nonwoody tree parts, woody tree parts, detritus, mobile soil (turnover time 70 years), and resistant soil (turnover time 500 years). The net primary productivity of the ground vegetation and nonwoody tree parts is assumed to follow a logistic law, whereas the other exchange rates obey first-order kinetics. The photosynthesis rate is calculated by increasing carbon dioxide concentration in the atmosphere by a logarithmic law and the proportionality factor $\beta = 0.395$ [*Jain et al.*, 1996]. The exchange rates are temperature dependent according to an Arrhenius law. The temperature is calculated by an energy balance model, a part of the integrated science assessment model (ISAM) [*Jain et al.*, 1994].

This carbon cycle model was originally developed as a tool to predict the likely future changes of the atmospheric abundance of CO_2 in response to scenarios of the future use of fossil fuels, deforestation, and expansion of agriculture land [*Jain et al.*, 1995, *Kheshgi et al.*, 1996]. This schematic model for the carbon cycle is constructed to be consistent with our current understanding of the global carbon cycle. Testing of the capability of the model to represent this understanding has been based, in part, on the analysis of tracer records such as those for ^{13}C and ^{14}C [*Jain et al.*, 1996]. In addition, an intercomparison [*Enting et al.*, 1994] with 17 other carbon cycle models found this carbon cycle model response to lie in the middle of the range of other model responses. For the last two Intergovernmental Panel on Climate Change (IPCC) assessments [*IPCC*, 1995, 1996], our carbon cycle model has been used to assess the future scenarios. While schematic models for the global uptake of carbon and carbon isotopes do not contain the spatial resolution available in current OGCMs, the simplicity of schematic models enables users to avoid modeling deficiencies common in current OGCM-based studies of ocean carbon cycle, which often contain simplifying approximations, neglect to use available data to constrain modeled processes, and contain spurious errors. Moreover, recent studies have shown [*Kheshgi and White*, 1996; *Joos et al.*, 1996] that the global oceanic uptake of carbon produced by OGCMs with carbon transport modeled as a tracer can be represented by a nonlinear convolution model, which is consistent with the depiction of the global carbon cycle in our schematic model.

Modeling Bomb ^{14}C Inventories

Atmospheric nuclear testing in the 1950s and early 1960s introduced radiocarbon into the stratosphere and troposphere, from where it was later transported into the oceans and terrestrial biosphere. In order to calculate the global budget of bomb ^{14}C, we need to have information about the total amount of bomb ^{14}C produced by nuclear testing and the bomb ^{14}C inventories in the key reservoirs: the atmosphere (stratosphere and troposphere), the oceans, and the terrestrial biosphere. These inventories are inferred from certain measurable time-dependent quantities such as atmospheric ^{14}C concentration and ocean surface water concentrations of $\delta^{13}C$, $\Delta^{14}C$, and dissolved inorganic carbon (DIC). However, some time-dependent quantities are not measurable, e.g., biospheric bomb ^{14}C in the terrestrial biosphere, while for others the data is not sufficient to derive accurate inventories, e.g., stratospheric and oceanic bomb ^{14}C concentration and biospheric and oceanic $\delta^{13}C$. Therefore most studies rely on models to augment data to estimate inventories.

Our approach is as follows. Prior to extensive nuclear weapons testing, i.e., 1950, the model calculates the atmosphere, biosphere, and ocean distribution of ^{14}C (in atoms) with prescribed fossil and land use emissions and natural ^{14}C production [*Jain et al.*, 1995, 1996; *Kheshgi et al.*, 1996], resulting in the decline of ^{14}C often referred to as the Suess effect [*Suess*, 1955]. After initiation of extensive nuclear weapons testing, the distribution of ^{14}C in the ocean and biosphere is calculated with the prescribed average observed tropospheric $\Delta^{14}C$ concentrations in per mil [*Broecker et al.*,

1985, 1995; *Tans*, 1981]. This is the deviation of ^{13}C-normalized concentration of ^{14}C from that of the oxalic acid standard as defined at the U.S. National Bureau of Standards (now National Institute of Standards and Technology), expressed in per mil as [*Stuiver and Pollach*, 1977]

$$\Delta^{14}C = \left[\delta^{14}C - 2(\delta^{13}C_i + 0.025)(1+\delta^{14}C)\right] \times 1000‰ \quad (1)$$

where

$$\delta^{14}C = \frac{\left(^{14}C/C\right)}{\left(^{14}C/C\right)_{standard}} - 1 \quad (2)$$

To convert $\Delta^{14}C$ into $\delta^{14}C$, and then to ^{14}C inventories, we use model-estimated atmospheric and oceanic $\delta^{13}C$, atmospheric CO_2 concentration, and oceanic DIC values. In a separate study [*Jain et al.*, 1996], we have shown that our model-calculated atmospheric $\delta^{13}C$ and CO_2 trends as well as oceanic DIC and $\delta^{13}C$, match observed records well within the range of observational uncertainty.

Figure 1 shows the model-estimated bomb ^{14}C inventories for the ocean and biosphere reservoirs, which are taken to be the difference between the ^{14}C inventories and their respective values in 1950. Our model-estimated ocean inventory for January 1, 1975, shown in Figure 1 is 326 x 10^{26} atoms, which is about 7% higher than the revised observation-based estimate of ocean inventory of 305x10^{26} atoms [*Broecker et al.*, 1995]. *Broecker et al.* [1995] assume that the uncertainty in this estimate is of the order of ±10%, but do not provide any basis for this uncertainty estimate. Our analysis suggests that the observed data, particularly for the prenuclear time, used by Broecker et al. for ^{14}C inventory calculations, could lead to an inventory error much higher than ±10%. The largest source of uncertainty in the observation-based estimates of ocean inventory is the prenuclear surface water and deep ocean $\Delta^{14}C$ profiles that are required to subtract the natural component from the bomb ^{14}C component. As was pointed by *Broecker et al.* [1985], the prenuclear $\Delta^{14}C$ profiles in different oceans could lead to inventory errors ranging from 5 to 18%. Note that *Broecker et al.* [1995] used the same prenuclear values as *Broecker et al.* [1985]. Another source of uncertainties is oceanic DIC that is required to convert the excess bomb $\Delta^{14}C$ values to the number of excess ^{14}C atoms. Instead of using the actual profiles of DIC, *Broecker et al.* [1995] simplified the calculations by selecting an average DIC concentration for that part of the water column contaminated with bomb ^{14}C. On the basis of the DIC profiles for the five stations in the temperate regions of three oceans (Atlantic, Indian, and Pacific), *Broecker et al.* [1995] found that the integrated bomb ^{14}C values are on the average about 2% higher than those using the mean concentration for DIC, and they argued that this procedure does not introduce inventory errors exceeding 2%. As we show later, our model produces the same difference when we assume constant DIC. Observed atmospheric $\Delta^{14}C$ values also introduce uncertainty in the calculation of the ocean bomb ^{14}C. However, compared to the other uncertainties discussed above, those associated with atmospheric $\Delta^{14}C$ histories are small. The concentrations of $\Delta^{14}C$ (in per mil) are estimated to be accurate to ±4‰ [*Stuiver and Östlund*, 1980; *Östlund and Stuiver*, 1980]. Our model results show that an error of ±4‰ in atmospheric $\Delta^{14}C$ produces an error of ±1% in ocean inventories. Another source of uncertainty arises from $\delta^{13}C$ values, which are required to convert $\Delta^{14}C$ into $\delta^{14}C$, as shown in equation (1). *Broecker et al.* [1995] assumed $\delta^{13}C$ = 0‰ in their calculations. Since the change in atmospheric in $\delta^{13}C$ over the period from 1950 to 1975 is less than 1‰ [*Friedli et al.*, 1986; *Keeling et al.*, 1995] and the change in ocean $\delta^{13}C$ is even smaller than the change in atmospheric $\delta^{13}C$ [*Kroopnick*, 1985], our model results show that the inaccuracies associated with $\delta^{13}C$ are only ±0.6%. On the basis of the various uncertainties discussed above, *Broecker et al.* [1995] estimates of ocean inventory of bomb radiocarbon could be in error by much as ± 20%.

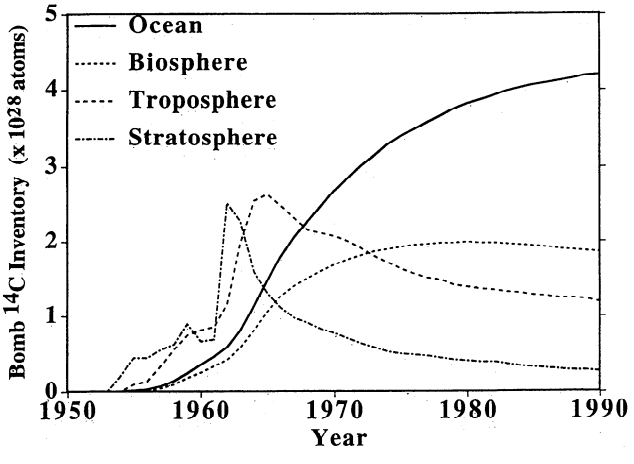

Figure 1. Bomb ^{14}C inventories as a function of time, calculated as the difference from the respective ^{14}C inventories in 1950. The terrestrial biosphere and ocean inventories are calculated with prescribed tropospheric $\Delta^{14}C$ concentration, while the tropospheric inventories are calculated from the observed $\Delta^{14}C$ concentration. The stratospheric values shown in the figure are from *Hesshaimer et al.* [1994] and match the observed data for the period 1963-1969 [*Telegadas*, 1971] and for the year 1990 [*Nakamura et al.*, 1992].

Observation-based estimates of stratospheric inventories are available for the period 1963-1969 [*Telegadas*, 1971] and for the year 1990 [*Nakamura et al.*, 1992]. To extend the observation-based estimates, *Hesshaimer et al.* [1994] tuned their one-box stratosphere model to reproduce the observed stratospheric inventory; their model results from 1950 to 1990 are shown in Figure 1. Similar analyses have been done with the Lawrence Livermore National Laboratory (LLNL) two-dimensional model of the global atmosphere (D. E. Kinnison, private communication, 1996]. Figure 1 also shows the tropospheric inventory calculated from the observation-based estimates of $\Delta^{14}C$ for the atmosphere [*Broecker et al.*, 1985].

Model Intercomparison

Our model-estimated changes in the terrestrial biosphere and ocean inventories for the period 1965-1990 are compared in Table 1 with the estimates of previous model studies by *Broecker et al.* [1995], *Siegenthaler and Joos* [1992], *Hesshaimer et al.* [1994], *Broecker and Peng* [1994], and *Duffy and Caldeira* [1995].

Our model-estimated change in ocean inventory (289 x 10^{26} atoms) for the period 1965-1990 is within 4% of the model

estimates of 287 x 10^{26} atoms by *Siegenthaler and Joos* [1992] (taken from *Joos* [1994]), 299 x 10^{26} atoms by *Hesshaimer et al.* [1994], and 300 x 10^{26} atoms by *Broecker et al.* [1995], as seen in Table 1. However, as also shown in Table 1, our model-estimated value of 289 x 10^{26} atoms is about 17% lower than the 337 x 10^{26} atoms calculated by *Broecker and Peng* [1994] and about 9% higher than the 268 x 10^{26} atoms calculated by *Duffy and Caldeira* [1995]. It is important to note that the five models other than our own listed in Table 1 were tuned to match estimates of ocean bomb ^{14}C inventory based on the GEOSECS data. Our model, however, was calibrated to match estimates of the preindustrial atmosphere and ocean depth structure, yet it is also able to reproduce estimates of bomb ^{14}C ocean inventories within the range of other models' uncertainties.

Since all other model estimates of bomb ^{14}C ocean inventories summarized in Table 1 are based largely on the GEOSECS-based estimates, we now consider effects of the GEOSECS-based estimates on model results. The reason for the higher value of *Broecker and Peng* [1994] is that their model was calibrated for the GEOSECS value of 370 x 10^{26} atoms, which is about 20% higher than the revised observed inventory of 305 x 10^{26} atoms [*Broecker et al.*, 1995, Table 2].

Duffy and Caldeira's [1995] estimate of ocean bomb ^{14}C inventory is close to the observed GEOSECS data as of January 1, 1975 because their OGCM was tuned to agree with the ocean bomb ^{14}C inventory estimated from the GEOSECS data by *Broecker et al.* [1995]. However, as seen in Table 1, *Duffy and Caldeira*'s [1995] OGCM-based estimate of ocean bomb ^{14}C inventory change over the period 1965-1990 is lower than schematic model studies. We have examined their model assumptions carefully and conclude that the following approximations in their calculations could reduce their OGCM-estimated ocean bomb ^{14}C inventory change over the period 1965-1990:

1. *Duffy and Caldeira* [1995] do not simulate ^{12}C and ^{13}C. Therefore they use an arbitrary scale to express the $^{14}C/^{12}C$ ratio and to convert the arbitrary model ratios into the standard $\Delta^{14}C$ as

$$\Delta^{14}C\ (‰) = \delta^{14}C_{arbitrary} \qquad (3)$$

Table 1. Changes in Bomb ^{14}C Inventories for Terrestrial Biosphere and Ocean Reservoirs Estimated by Different Studies Over the Period From mid-1965 to mid-1990

Study	Change, x 10^{26} atoms	
	Ocean Inventory	Terrestrial Biosphere Inventory
This Study	289	92
Broecker *et al.* [1995]	300	39
Broecker and Peng[1994]	337	39
Hesshaimer *et al.* [1994]	299	60
Siegenthaler and Joos [1992]	287[a]	99[a]
Duffy and Caldeira [1995]	268	75[b]

[a]Taken from *Joos* [1994].

[b]Adopted from *Joos* [1994], which is the average of results from four schematic carbon cycle: *Hesshaimer et al.* [1994], *Broecker et al.* [1985], *Siegenthaler and Oeschger*[1987], and *Siegenthaler and Joos* [1992].

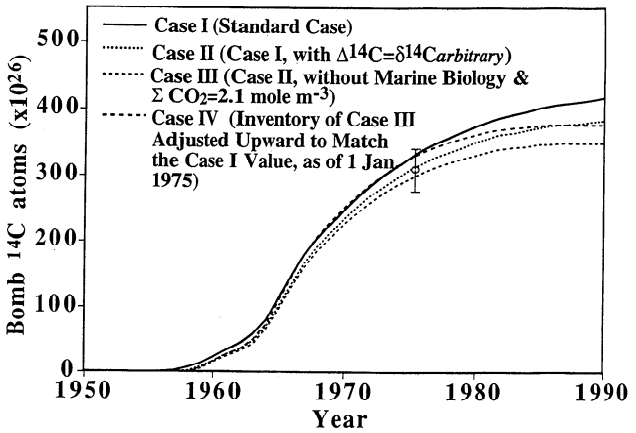

Figure 2. Model-estimated bomb ^{14}C ocean inventory for four different cases, calculated as the difference from the respective ^{14}C inventory in 1995: case I, calculated ocean inventory includes the effect of marine biology and actual profile of ΣCO_2; case II, same as case I but with $\Delta^{14}C=\delta^{14}C_{arbitrary}$; case III, same as case II but without marine biology and ΣCO_2 =2.1 mol m^{-3}; and case IV, inventory of case III adjusted upward to match the case I value for January 1, 1975. The *Broecker et al.* [1995] estimated value, as of January 1, 1975, of 305 x 10^{26} atoms is also shown in this figure.

where

$$\delta^{14}C_{arbitrary} = (R_{arbitrary} - 1)\ 1000 \qquad (4)$$

$$R_{arbitrary} \equiv \text{Arbitrary Model } ^{14}C/^{12}C \text{ ratio} \qquad (5)$$

Equations (1)-(4) imply that they underestimated $\delta^{14}C$ and therefore also underestimated the ocean inventory of bomb ^{14}C as we discuss below.

2. The ocean inventory calculations of bomb ^{14}C from the $\Delta^{14}C$ excess over prenuclear time requires the concentration of oceanic DIC for the part of the water column that contains bomb ^{14}C. *Duffy and Caldeira* [1995] assume an average DIC concentration of 2.15 mol m^{-3}, the data-based global estimate of surface water concentration for January 1, 1975, which does not vary with time or space during the entire period of their simulation [*Duffy et al.*, 1995]. *Broecker et al.* [1985, 1995] note that this procedure introduces inventory error of about 2% at the time of the GEOSECS survey. We have also found a similar difference in our model results, as discussed below, when assuming constant DIC.

3. *Duffy and Caldeira* [1995] do not take into account the ^{14}C transported in particulate organic matter. Particulate settling of organic matter originating in the ocean mixed layer and decaying in deeper waters (the "biological pump") leads to higher total carbon concentration in the deep waters than in the surface ocean waters.

4. *Duffy and Caldeira*'s [1995] model transports the $^{14}C/^{12}C$ ratio, not the ^{14}C atoms [Duffy *et al.*, 1995]. This means that anthropogenic emissions of carbon dioxide to the atmosphere, which lead to additional transport of ^{14}C via the net flux of CO_2 into the ocean, are neglected.

We have used our model to make estimates for the sizes of the corrections 1-4 discussed above. In Figure 2, case I shows our model-estimated ocean inventory for the standard case that

includes all the effects omitted by *Duffy and Caldeira* [1995]. This figure shows that by assuming $\Delta^{14}C$ (per mil) = $\delta^{14}C_{arbitrary}$ (correction 1), our model-estimated ocean inventory as of January 1, 1975, reduces from our standard case (case I) of 326 x 10^{26} atoms to 308 x 10^{26} atoms (case II), i.e., roughly 6% lower than our standard case. Our model results show that assuming DIC of 2.15 mol m^{-3} without a biological pump (corrections 2 and 3) drops our model ocean inventory for January 1, 1975, from 308 x 10^{26} atoms (case II in Figure 2) to 296 x 10^{26} atoms (case III in Figure 2), i.e., a further 3% reduction compared to our standard case I. To this 3%, the contribution of marine biological pump is 1%, and the contribution of DIC is 2%. However, the model-estimated inventory for the period 1965-1990 dropped from 296 x 10^{26} atoms to 267 x 10^{26} atoms, which is close to *Duffy and Caldeira*'s [1995] estimate for the same period (Table 1). Note that in case III the rate of increase of ocean inventory decreases after 1975, and ocean inventory in the late 1980s becomes nearly constant, which is similar to *Duffy and Caldeira*'s [1995] results. This is caused by keeping DIC constant at the 1975 value of 2.15 mol m^{-3}, while in reality DIC increases with time. Thus after 1975 the transport of ^{14}C due to anthropogenic sources is ignored, which slowes the rate of increase of ocean bomb ^{14}C inventory after 1975.

Duffy and Caldeira's [1995] approach was to tune the model transport parameters to bring the model ^{14}C inventory into agreement with the inventory calculated with the data-based estimate for January 1, 1975. Following their approach, we also adjusted upward our model-estimated inventory as of January 1, 1975, of 296 x 10^{26} atoms (case III in Figure 2) to our standard case value of 326 x 10^{26} atoms (case IV in Figure 2). Figure 2 shows that the ocean inventory for case IV matches well with case I only up to January 1, 1975. However, the calculated bomb ^{14}C inventory in the year 1990 drops to 375 x 10^{26} atoms (case IV in Figure 2) from our current estimated value of 418 x 10^{26} atoms, about a 10% drop. Moreover, Figure 2 also shows that in cases III and IV the change of ocean inventory over the period from 1975 to 1990 is approximately the same. This suggests that without taking into account the effects of marine biology and DIC, we might be able to reproduce inventories for the GEOSECS years but not for the years after 1975.

We conclude that the OGCM used by *Duffy and Caldeira* [1995] underestimated the ocean inventory change by roughly 10%; by taking into account the corrections (≈10%) discussed above, their OGCM-estimated inventory change for the period 1965-1990 could increase from 268 x 10^{26} atoms to 296 x 10^{26} atoms, which is well within the range of results estimated by other schematic models (see Table 1).

Our model-estimated bomb ^{14}C inventory change for the terrestrial biosphere over the period 1965-1990 is 92 x 10^{26} atoms. While our model-estimated biospheric inventory is close to that of *Siegenthaler and Joos* [1992] (taken from *Joos* [1994]), it is much higher than the other studies shown in Table 1. The reason for this difference is that this study and that of *Siegenthaler and Joos* [1992] are the only two studies listed that include modeled effects of changes in land use and CO_2 fertilization on the biosphere; these lead to an additional uptake of bomb ^{14}C by the terrestrial biosphere. This comparison highlights the great diversity, and disparity, of processes included in terrestrial biosphere carbon cycle models. Also since there are no observation-based estimates of the terrestrial biosphere's inventory of bomb ^{14}C and we are not certain about the nature of the global biosphere's carbon cycle, we cannot conclude that our biospheric model results (and those of Siegenthaler and Joos) are more accurate than other model studies listed in Table 1.

While the model-estimated bomb ^{14}C inventories of the terrestrial biosphere listed in Table 1 vary over a wide range from 39 x 10^{26} to 99 x 10^{26} atoms, the ocean inventory estimated by the different models deviate little (except for the minor differences discussed above). This is because the ocean estimates are constrained by observations [*Broecker et al.*, 1985, 1995], whereas the inventories of bomb ^{14}C in the terrestrial biosphere are not. From this model intercomparison we can conclude that it is likely that model estimates of bomb radiocarbon inventory for terrestrial biospheric will remain uncertain as long there are no sound observational constraints on models for the terrestrial carbon cycle.

Global Budget of Bomb ^{14}C

In order to test the accuracy of the derived global budget of bomb radiocarbon, we have compared the estimated cumulative inventories (sum of troposphere, stratosphere, ocean, and biosphere inventory shown in Figure 1) with the cumulative production of bomb ^{14}C as a function of time (Figure 3). In this study we have not attempted to estimate the production of bomb ^{14}C from individual nuclear tests but used the *Hesshaimer et al.* [1994] approach instead. Following their approach, estimates of ^{14}C production per year are assumed to be directly proportional to the total megatonnage (Mt TNT) that year; as discussed below, this is not a good assumption. Estimates of the constant of proportionality (*Pc*) found in the literature vary between 1 x 10^{26} and 2 x 10^{26} atoms $(Mt\ TNT)^{-1}$ [UNSCEAR, 1993]. *Hesshaimer et al.* [1994] obtained a value of *Pc* of 1.05 x 10^{26} atoms $(Mt\ TNT)^{-1}$ by demanding that their model-estimated tropospheric and oceanic stratospheric inventories agree with observed inventories before 1963. Figure 3 shows that using *Pc* = 1.09 x 10^{26} atoms $(Mt\ TNT)^{-1}$, our derived total inventory also matches the cumulative production until 1963, but thereafter the total inventory exceeds the cumulative production of bomb ^{14}C. Our derived inventory remains nearly constant after 1970 at a value of 740 x 10^{26} atoms. There appear to be at least three reasons for the difference. First, there could be error in the production estimates using the *Hesshaimer et al.* [1994] approach. Carbon 14 is produced by reactions (with nitrogen) of neutrons that escape into the atmosphere from the exploding nuclear devices. Therefore the amount produced is dependent on the specific device, including its design, its fission-fusion yield split, and where the explosion occurred (i.e., surface or atmospheric explosion). Thus the constant of proportionality depends on the individual bomb test; assuming a constant value for *Pc* is not necessarily valid. Error in current estimates of bomb ^{14}C production from individual explosions could vary between 10% and 50% ^{14}C [*Wuebbles*, 1995] due to use of simple relationships to estimate the amount produced. Second, as was suggested by *Broecker et al.* [1995], there could be error in the observed stratospheric inventories during the first 1-2 years after the test ban was implemented. To test this, we estimated the cumulative production of ^{14}C that matches our estimated total inventory in year 1965, instead of 1963 (Figure 3). This has been achieved by multiplying the total megatonnage in each year by 1.20 x 10^{26} atoms per $(Mt\ TNT)^{-1}$. Figure 3 shows that our estimated combined inventory in 1990 is now only 3% higher than the revised cumulative production, well within the range of uncertainties in the observed and

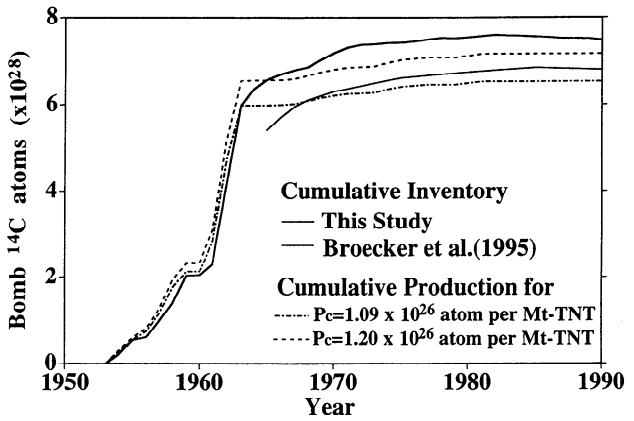

Figure 3. Estimated cumulative bomb ^{14}C inventories and the cumulative production as a function of time. The cumulative bomb ^{14}C inventories are the sum of troposphere, stratosphere, terrestrial biosphere, and ocean inventories as shown in Figure 1. Cumulative production is calculated for Pc=1.09 and 1.20 atoms (Mt TNT)$^{-1}$ that matches the total inventories in years 1963 and 1965, respectively. For comparison, we have also plotted the total inventories of *Broecker et al.* [1995] for the period 1965-1990.

estimated inventories. A third reason for the imbalance in the bomb carbon budget could be the uncertainty in the biosphere inventory discussed earlier. The lack of direct measurements that can contribute to observation-based estimates of the terrestrial biospheric uptake of radiocarbon, along with the wide range of results for alternative models, as compared in Table 1, leads us to believe that the terrestrial biospheric inventories are a large source of uncertainty in the global budget of bomb ^{14}C.

In light of the uncertainties associated with the bomb radiocarbon budget, *Broecker et al.* [1995], concluded on the basis of their model results that there is no inconsistency for the period after 1965, as the global inventories estimated by their model remain nearly constant at a value of 670 x 10^{26} atoms. However, it is important to note that our model also produces a nearly constant combined inventory (Figure 3), even though our model-estimated combined inventory in 1990 is about 9% higher than *Broecker et al.*'s [1995]. Table 1 shows that the difference between our results and the *Broecker et al.* [1995] model results is mainly due to biospheric inventories. Therefore the leveling-off of total bomb ^{14}C inventory at a near constant value does not provide a strong evidence for the consistency in bomb ^{14}C budget, since there are considerable uncertainties in ^{14}C global inventories.

Concluding Discussion

Results from various published schematic models and an OGCM have been compared. It is found that ocean inventory estimated by various schematic models over 1965-1990 compare well with each other. However, the OGCM [*Duffy and Caldeira*, 1995] results are about 8-10% lower than the schematic model results. This paper clearly shows that there are a number of approximations made in OGCM calculations that would increase the OGCM-estimated ocean bomb ^{14}C inventory and cause the results of the OGCM to fall in line with estimates of the schematic models. While many of the OGCM flaws have been identified by intercomparing model results in this study, a head-to-head comparison of OGCM and upwelling-diffusion ocean carbon cycle models (with the carbon isotopes ^{12}C, ^{13}C, and ^{14}C all independently conserved) would constitute the next logical step beyond the present study.

In contrast to ocean inventory, we found large discrepancies in the different model estimates for the bomb ^{14}C inventory of the terrestrial biosphere. In addition to uncertainties in the biospheric inventories, there remain key uncertainties that limit the usage of the radiocarbon budget as a test for model descriptions of global carbon cycle intended for application to global climate change issues. These uncertainties include (1) Uncertainty in the pre-nuclear surface water and deep ocean $\Delta^{14}C$ values, (2) Uncertainty in the stratospheric inventory due to insufficient record of atmospheric measurements especially during at times close to the enactment of the nuclear weapons test treaty, and (3) Uncertainty in the production rates of bomb-produced radiocarbon.

We do not find a discrepancy, a so-called imbalance, in model closures of the global radiocarbon budget given our expectation of these large uncertainties. Nor do we expect, however, that the ability of a global carbon cycle model to close the global radiocarbon budget is alone a strong confirmation of a global carbon cycle model, again given the large uncertainties involved. More definitive statements about the effects of bomb radiocarbon budget on the flow of the anthropogenic carbon dioxide within the carbon cycle system will require a reduction of these uncertainties. Because of the limited number of measurements of the prebomb surface water concentration of ^{14}C and the stratospheric concentrations, there remain questions about the distribution of bomb ^{14}C. Improved analyses of nuclear weapons tests and their production of bomb radiocarbon could have a significant effect on reducing uncertainties in model studies of the evolution of bomb radiocarbon in the atmosphere-ocean-biosphere system. This would provide a better capability for using ^{14}C as a tracer for determining the flow of carbon dioxide within the carbon cycle. Furthermore, direct measurements of global net primary productivity, plant carbon content, or soil carbon content could reduce the discrepancies in the different model estimates for the bomb ^{14}C inventory of the terrestrial biosphere. A careful assessment of the extent to which these uncertainties could be narrowed and the concomitant use as a constraint on projections of future carbon cycle would be a useful basis for further analysis of historical radiocarbon data.

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