An Earth system model of intermediate complexity: Simulation of the role of ocean mixing parameterizations and climate change in estimated uptake for natural and bomb radiocarbon and anthropogenic CO_2

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Received 15 February 2005; revised 12 May 2005; accepted 3 June 2005; published 2 September 2005.

[1] We examine the sensitivity of simultaneous simulation of climate, natural 14 C, bomb ¹⁴C, and anthropogenic CO₂ uptake to the choice of three different ocean mixing schemes: horizontal/vertical mixing (HV), isopycnal mixing (ISO), and Gent-McWilliams mixing (GM) using an Earth system model of intermediate complexity, Integrated Science Assessment Model-2.5D (ISAM-2.5D). Our modeling results suggest that the HV scheme greatly underestimates the observed values of natural ¹⁴C in the deep ocean, while the ISO and GM schemes yield more realistic results by simulating increased amounts of natural ¹⁴C values through enhanced vertical diffusion and deep water formation. The GM scheme further improves the ISO-based natural ¹⁴C distribution in the Southern Ocean and the deep Pacific and Indian oceans through a more realistic simulation of the Southern Ocean circulation. The model simulated global uptake of anthropogenic CO₂ for the 1980s ranges between 1.8 and 2.3 PgC/yr, largely consistent with data-based estimates and OGCM results. The ISAM-2.5D simulated oceanic uptake of ¹⁴C and CO₂ is highest for the ISO scheme and lowest for the HV scheme, with the largest discrepancies occurring among different mixing schemes found in the Southern Ocean. However, no single mixing scheme is more successful than the others in simulating GEOSECS-measured uptake of bomb ¹⁴C and anthropogenic CO₂ for various ocean basins. Climate change is found to reduce CO_2 uptake by 7–9% and 6–8% for the 1980s and over the period 1765–1990, mainly as a result of decreased CO₂ solubility associated with increased sea surface temperatures. However, the effect of climate change on bomb ¹⁴C uptake is negligible.

Citation: Cao, L., and A. Jain (2005), An Earth system model of intermediate complexity: Simulation of the role of ocean mixing parameterizations and climate change in estimated uptake for natural and bomb radiocarbon and anthropogenic CO₂, *J. Geophys. Res.*, *110*, C09002, doi:10.1029/2005JC002919.

1. Introduction

[2] A good understanding of climate change, ocean circulation, the ocean carbon cycle, and feedback mechanisms between them is crucial for a reliable future projection of atmospheric CO₂ concentration and resultant climate change [*Prentice et al.*, 2001]. In order to make such predictions, 3-D Atmosphere-Ocean General Circulation Models (AOGCM) [e.g., *Maier-Reimer et al.*, 1996; *Sarmiento et al.*, 1998, 2000; *Orr et al.*, 2001], 2-D models of intermediate complexity [e.g., *Stocker et al.*, 1994; *Joos et al.*, 1999; *Plattner et al.*, 2001], and 1-D models [e.g., *Siegenthaler and Joos*, 1992; *Jain et al.*, 1995; *Harvey*, 2001] have been used to investigate the oceanic uptake of natural and bomb radiocarbon, anthropogenic CO₂ and/or the influence of climate change on their uptake. However,

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simulation results differ widely between models with a large part of discrepancies associated with differences in the representation of subgrid-scale ocean mixing processes. For example, in a recent study, *Matsumoto et al.* [2004] compared simulated natural radiocarbon, anthropogenic CO₂, and CFC-11 from 19 ocean carbon cycle models with data-based estimates and found large discrepancies between model results, which, may at least partly be attributed to the different parameterizations of ocean mixings.

[3] Three different parameterization schemes are usually used in ocean models to represent subgrid ocean mixing processes: horizontal/vertical mixing (HV), which mixes tracers along horizontal and vertical orientations [*Bryan*, 1987]; isopycnal mixing (ISO), which mixes tracers along and across surfaces of constant density [*Redi*, 1982]; and Gent-McWilliams mixing (GM), which, in addition to diffusing tracers along and across isopycnals, transports tracers by the estimated sum of large-scale and eddy-induced velocity [*Gent et al.*, 1995]. Many efforts have

been made to simulate oceanic carbon uptake using a hierarchy of models, typically with the HV scheme [e.g., Stocker et al., 1994; Jain et al., 1995; Sarmiento et al., 1998; Plattner et al., 2001; Harvey, 2001; Orr et al., 2001]. A few studies have investigated the sensitivity of oceanic carbon uptake for a certain carbon tracer to the choice between various subgrid ocean mixing schemes. Duffy et al. [1997] and England and Rahmstorf [1999] used Ocean General Circulation Models (OGCMs) to investigate the effect of ocean mixing parameterizations on the simulation of natural ¹⁴C as a way to assess modeled ocean circulation over long periods of time (centennial to millennial). Duffy et al. [1995a, 1995b] studied the role of ocean mixing schemes in the simulation of oceanic uptake for bomb-produced ¹⁴C, which is mainly used to evaluate modeled ocean mixing over shorter periods of time (decadal to centennial). However, the sensitivity of the coupled climate-ocean-carbon cycle system to different ocean mixing parameterizations has not been studied in a consistent manner. Therefore, as an extension of previous studies, the purpose of this study is to examine the concurrent simulation of different carbonrelated tracers and climate in a coupled modeling system for multiple sets of ocean mixing parameterization schemes. The novelty of this study is that we examine the effect of various ocean mixing schemes and climate change on the uptake of natural and bomb radiocarbon, and anthropogenic CO_2 simultaneously.

[4] The ISAM-2.5D (Integrated Science Assessment Model-2.5D) coupled climate-ocean-carbon cycle model used in this study utilizes an intermediate approach by retaining fairly sophisticated climate, chemistry, and carbon cycle, but simplifying the advanced dynamics of comprehensive General Circulation Models (AGCMs). Therefore ISAM modeling framework provides far greater computational efficiency and allows extensive explorations of key physical and chemical interactions among individual components of the earth system (atmosphere, ocean, and cryosphere) as well as between carbon cycle, climate change, and ocean thermohaline circulation. In terms of its applications, the model is capable of a wide range of studies having direct relevance to projections of future atmospheric CO₂ levels and associated changes in global climate and ocean circulation. For example, it has recently been applied to study the effect of climate change on ocean carbon sequestrations [Jain and Cao, 2005].

[5] The paper is organized as follows: in section 2 we briefly describe the coupled model used in this study. The detailed governing equations of the carbon cycle module are given in the appendix. The procedures of model spin-up in preindustrial time (year 1765 in our case) and transient simulations for the period 1765–2000 are given in section 3. Simulated climate is briefly described in section 4. Section 5 discusses the simulated oceanic uptake of natural and bomb ¹⁴C. Oceanic uptake of anthropogenic CO₂ is discussed in section 6. In section 7 we examine the effect of climate change on carbon uptake. Finally, discussions and conclusions are presented in section 8.

2. Model Description

[6] The model used in this study is an Earth system model of intermediate complexity. The model represents

key components of the climate system in a fully coupled system through exchange of energy, momentum, freshwater, and carbon without the use of flux adjustment. The geographical configuration of the model has a latitudinal resolution of 10° with each latitude band divided into one or more ocean and/or land bands in order to resolve major ocean basins and landmasses. The atmosphere is represented by an energy and moisture balance atmosphere module based on Weaver et al. [2001], which predicts surface air temperature, specific humidity, precipitation, and freshwater flux. Radiative forcing is calculated by a radiative transfer module [Jain et al., 2000]. The ocean module resolves major ocean basins: the Atlantic, Pacific, and Indian Ocean, which are connected through the zonally well-mixed Southern Ocean at about 40°S. Within each basin, the ocean dynamics is formulated largely on the basis of the zonally averaged 2-D ocean model of Wright and Stocker [1992] and Harvey [1992] but with some extensions. One major extension is that three different mixing schemes are used here to parameterize subgrid-scale ocean mixing: horizontal/vertical mixing (HV [Bryan, 1987]), isopycnal mixing (ISO [Redi, 1982]), and Gent-McWilliams mixing (GM [Gent et al., 1995]). The HV scheme parameterizes mixing along horizontal and vertical directions, while the ISO and GM schemes parameterize mixing along isopycnal and diapycal directions. In the GM scheme, in addition to the large-scale velocity determined from the geostrophic equations, an additional component of largescale velocity induced by mesoscale eddies is accounted for. Another extension is that vertical/diapycnal diffusivity is parameterized as a function of local Brunt-Vaisala frequency [Gargett, 1984; Hirst and Cai, 1994]. In addition, zonally averaged ocean bathymetry is included in the model to allow for realistic simulations of tracer uptake.

[7] The modeling framework also accounts for sea ice on the basis of a thermodynamic and dynamic sea ice module of *Semtner* [1976] and *Harvey* [1988]. In addition, a simple land module based on energy and moisture balance [*Wang and Mysak*, 2000] is also included.

[8] As a first step toward incorporating a comprehensive ocean carbon cycle model, the model currently includes an inorganic ocean carbon cycle component based primarily on the protocols of Ocean Carbon Cycle Model Intercomparison Project (OCMIP) [Orr and Dutay, 1999]. The carbon module simulates two carbon-related passive tracers: dissolved inorganic carbon (DIC) and radiocarbon (¹⁴C). The 14 C concentrations are modeled in terms of 14 C/ 12 C ratio. This modeling method is consistent with many previous studies of carbon cycle models [e.g., Toggweiler et al., 1989a, 1989b; Stocker et al., 1994; Jain et al., 1995; Duffy et al., 1997; England and Rahmstorf, 1999; Orr et al., 2001]. In the ocean interior, DIC and ${}^{14}C/{}^{12}C$ are governed by the processes of advection, diffusion, and convection (equation (A1)) under three different mixing schemes (HV, ISO, and GM). Following Harvey [1992] and Wright and Stocker [1992], the modeled temperature and salinity are also governed by the same processes. In this way, we are able to study the uptake of carbon, temperature, and salinity in a consistent manner.

[9] At the ocean surface both freshwater flux (equation (A2)) and air-sea exchange of CO_2 (equation (A3)) influence DIC concentration. Air-sea exchange of ${}^{14}C/{}^{12}C$ is



Figure 1. Atmospheric CO₂ evolution (ppm) and ${}^{14}C/{}^{12}C$ evolution (permil) for three latitude bands: $90^{\circ}-20^{\circ}S$, $20^{\circ}S-20^{\circ}N$, and $20^{\circ}-90^{\circ}N$ between 1765 and 2000 based on the data of *Enting et al.* [1994].

calculated as a proportion of the difference between the atmospheric and oceanic surface ¹⁴C/¹²C values (equation (A4)). This calculation follows the approach of Orr et al. [2001] but includes minor modifications. As opposed to using a constant surface DIC value in the calculation of the ¹⁴C air-sea exchange rate [e.g., *Toggweiler et al.*, 1989a; Orr et al., 2001], our model takes into account both the spatial and temporal variation of the surface DIC. For steady state simulation the observed surface DIC concentration is used, while for perturbation simulations the modeled change in surface DIC relative to preindustrial value is added to the observed value. The gas transfer velocity is the same for DIC and ${}^{14}C/{}^{12}C$ and is calculated as a nonlinear function of wind speed and sea surface temperature (equation (A5)) according to Wanninkhof [1992]. No air-sea exchange of DIC and ${}^{14}C/{}^{12}C$ occurs in regions covered by sea ice. ${}^{14}C$, in addition to ocean mixing and air-sea exchange, undergoes natural radioactive decay with a half life of 5730 years. To compare model results with observation and other studies, the model simulated ¹⁴C/¹²C ratios are converted into the standard Δ^{14} C notation in the unit of permil (equation (A6)).

[10] Currently, the model does not account for organic carbon cycle (biological carbon pump). Since the oceanic uptake of anthropogenic CO_2 is driven mainly by physical and chemical processes, the biological effect on ocean uptake of CO_2 is negligible [*Prentice et al.*, 2001]. As for radiocarbon, the error introduced by ignoring the biological pump in modeling ${}^{14}C/{}^{12}C$ is less than 10% [*Fiadiero*, 1982; *England and Maier-Reimer*, 2001] on our modeling results.

3. Model Simulations

[11] First, the coupled atmosphere-ocean-sea ice-land model is used to simulate steady state climate fields (e.g.,

sea surface temperature, sea surface salinity, and ocean circulation) based on the daily average solar insolation at the top of the atmosphere and prescribed wind stress at ocean surface. Next, the ISAM-2.5D carbon cycle module uses these steady state climatic fields to determine a steady state distribution of DIC and ¹⁴C. In order to investigate the influence of various ocean-mixing parameterizations on the simulation of ocean carbon cycle, three separate equilibrium states are determined from simulations using three different mixing schemes. In all three model spin-up cases, atmospheric ¹⁴C and CO₂ concentrations are held fixed at their preindustrial levels of 0.0 permil and 278 ppm, respectively. In this way we calculate three sets of steady state surface and deep ocean distributions of DIC and ¹⁴C. Each simulation is run to a state where the globally integrated air-sea flux of CO₂ is less than 1×10^{-4} PgC yr⁻¹, and the change in global averaged Δ^{14} C is less than 1×10^{-6} per mil yr indicating that the ocean carbon cycle has reached an approximate equilibrium. For each mixing scheme, it takes about 10,000 model years to reach this steady state for both DIC and ¹⁴C. These initial steady states provide the starting points for the following two sets of perturbation simulations over the period 1765 to 2000:

[12] 1. In the "with climate" (WC) set of simulations, climate change is allowed to influence the carbon cycle. The transient changes for the HV, ISO and GM schemes are defined as HV-WC, ISO-WC, and GM-WC. The ISAM-2.5D climate and carbon cycle components are forced with the historical data of atmospheric CO₂, atmospheric ¹⁴C/¹²C ratios, and the resulting radiative forcing due to changes in atmospheric CO₂ concentrations calculated using the radiative transfer component of the ISAM-2.5D [*Jain et al.*, 2000].

[13] 2. The "without climate" (WOC) set of simulations is run identically to WC case except that the climate is held constant at a steady state level and thus no effect of climate change on the carbon cycle is considered. The transient changes for the HV, ISO and GM schemes are defined as HV-WOC, ISO-WOC, and GM-WOC.

[14] From the results of these two sets of experiments we can investigate the effect of climate change on the oceanic uptake of anthropogenic CO_2 and bomb ¹⁴C. To diagnose model simulated oceanic uptake of bomb radiocarbon, model-estimated column inventory and penetration depth are compared with the observations. The bomb ¹⁴C column inventory is determined by integrating the difference between the postbomb and prebomb (year 1954) vertical distribution of ¹⁴C from the ocean surface to bottom of the ocean (see equation (B1)). To convert modeled ¹⁴C from permil into atoms, instead of using a constant DIC value [e.g., Toggweiler et al., 1989b; Stocker et al., 1994], the observed DIC profile plus the modeled DIC change relative to the preindustrial time is used. The penetration depth of ¹⁴C is defined as the ratio of bomb ¹⁴C inventory to surface bomb ¹⁴C concentration (see equation (B2)).

[15] The atmospheric CO₂ and ¹⁴C/¹²C data for the period 1765 to 2000 are taken from *Enting et al.* [1994] (Figure 1). The ¹⁴C/¹²C ratio data are available for three latitude bands (90°–20°S, 20°S–20°N, and 20°–90°N), whereas the atmospheric CO₂ data are global and annual mean data. As Figure 1 shows, the atmospheric ¹⁴C/¹²C data account for the "Suess effect" [*Suess*, 1955] between 1840 and



Figure 2. Basin mean profiles of ocean temperature (°C) in the (a) Atlantic, (b) Pacific, (c) Indian, and (d) Southern oceans from the model simulations based on the horizontal/vertical (HV), isopycnal (ISO), and Gent-McWilliams (GM) schemes and observations [*Levitus and Boyer*, 1994].

1954 (negative values), and the bomb effect thereafter (positive values).

4. Modeled Climate

[16] Since our focus in this study is the oceanic uptake of carbon, ISAM-2.5D simulated climate fields such as atmosphere temperature and precipitation, ocean temperature (Figure 2) and salinity are not discussed in detail here. Nevertheless, these variables have similar magnitudes and patterns for different ocean mixing schemes and compare reasonably well with the corresponding observations.

[17] To demonstrate the model's ability to simulate ocean dynamics, model simulated overturning stream functions for different ocean basins are shown in Figure 3. The modeled large-scale global ocean circulation is broadly consistent with both observations [*Schmitz*, 1995; *Ganachaud and Wunsch*, 2000] and OGCM results [e.g., *Duffy et al.*, 1997; *England and Rahmstorf*, 1999]. Deep water forms in both the North Atlantic (Figure 3a) and Southern Ocean (Figure 3d). The maximum strength of the North Atlantic Deep Water (NADW) formation is 16, 20, and 18 Sv for the HV, ISO, and GM schemes respectively, and the strength of the Antarctic Bottom Water (AABW) formation is 16, 22 and 20 Sv for HV, ISO, and GM schemes respectively. The

simulated strength of deep water formation in each case is consistent with the observational estimates of 15 ± 2 Sv in the North Atlantic and 21 ± 6 Sv in the Southern Ocean [Ganachaud and Wunsch, 2000]. Compared to the HV scheme, isopycnal mixing in the ISO and GM schemes reduces large diapycnal diffusive fluxes in high-latitude regions of steeply sloping isopycnal surfaces, thereby increasing the meridional density gradients and the strength of deep water formation [Knutti et al., 2000]. The most pronounced difference in simulated circulation between different schemes is observed between 50°S and 60°S in the Southern Ocean (Figure 3d), where a deep clockwise overturning Deacon cell [Döös and Webb, 1994] penetrating to a depth of about 3000m is seen in the HV and ISO simulations, but not in the GM simulation. In the GM simulation, the eddy-induced circulation cancels the largescale Deacon cell, a finding consistent with results from both eddy-resolving models [e.g., Döös and Webb, 1994; Karsten et al., 2002] and 3-D OGCMs using the GM scheme [e.g., Danabasoglu and McWilliams, 1995; Hirst and McDougall, 1998]. One important consequence of the disappearance of the Deacon cell is the reduction in the downward transport of tracers from the surface to deep ocean between 50° S and 60° S of the Southern Ocean. The absence of the Deacon cell also facilitates the northward



Figure 3. Model-simulated annual mean meridional overturning stream function (Sv, 1Sv = 10^6 m³ s⁻¹) in the (a) Atlantic, (b) Pacific, (c) Indian, and (d) global oceans based on the horizontal/vertical (HV), isopycnal (ISO), and Gent-McWilliams (GM) schemes. The direction of flow is clockwise around solid contours and anticlockwise around dashed contours.



Figure 4. Latitude-depth distribution of natural Δ^{14} C (permil) in the Atlantic Ocean based on (a) GLODAP data [*Key et al.*, 2004] and model simulations for (b) horizontal/vertical (HV), (c) isopycnal (ISO), and (d) Gent-McWilliams (GM) schemes.

intrusion of the AABW into other ocean basins (Figure 3d), indicating that the Southern Ocean will exert a stronger influence on tracer distributions in other ocean basins based on the GM scheme.

5. Radiocarbon Simulation

5.1. Simulation of Natural ¹⁴C

[18] In this section we discuss simulated oceanic uptake of natural ¹⁴C based on different mixing schemes. Radiocarbon undergoes radioactive decay with a half time of 5730 years. Therefore preindustrial time (year 1765 in our case) natural ¹⁴C is frequently used to assess model simulated ocean mixing processes over a timescale of hundreds to thousands of years [e.g., *Toggweiler et al.*, 1989a; *Jain et al.*, 1995; *Duffy et al.*, 1997; *England and Rahmstorf*, 1999]. We compare our model simulated ocean natural ¹⁴C distributions with the data from the Global Ocean Data Analysis Project (GLODAP). The data consist of synthesized data products from World Ocean Circulation Experiment (WOCE), the Joint Global Ocean Flux Study (JGOFS), and the Ocean Atmosphere Carbon Exchange Study (OACES) [*Key et al.*, 2004].

[19] The latitude-depth distribution of simulated Δ^{14} C for each mixing scheme along with Δ^{14} C data from GLODAP in the Atlantic, Pacific, Indian, and global oceans are presented in Figures 4–7. The simulated averaged Δ^{14} C depth profile for each ocean basin together with the corresponding GLODAP data are plotted in Figure 8.

[20] Figures 4–8 show that $\Delta^{f4}C$ values in the ocean interior are usually negative because of natural decay. More negative values indicate that a greater time has elapsed since



Figure 5. Latitude-depth distribution of natural Δ^{14} C (permil) in the Pacific Ocean based on (a) GLODAP data [*Key et al.*, 2004] and model simulations for (b) horizontal/vertical (HV), (c) isopycnal (ISO), and (d) Gent-McWilliams (GM) schemes.

the water mass was last in contact with the atmosphere. With regard to the model results for various mixing schemes, the simulated natural radiocarbon is most negative for the HV scheme, less negative for the GM scheme, and least negative for the ISO scheme. Relative to the HV scheme, the less depleted Δ^{14} C in the ocean interior for the ISO and GM schemes is mainly attributed to two factors. First, the increased deep water formation in both the North Atlantic and Southern oceans (Figure 3) increases the rate of ¹⁴C transport from the surface to the deep ocean. Second, the enhanced effective vertical diffusion associated with isopycnal mixing also facilitates the surface-to-deep ocean ¹⁴C transport, especially at high latitudes where isopycnal surfaces slope steeply. Nevertheless, the largest

differences between various mixing scheme results are found in the Southern Ocean, where the ISO-based Δ^{14} C is about 50 permil higher than that of the HV scheme (Figure 8d). This phenomenon is a result of enhanced vertical diffusion and deep water formation under the ISO scheme. On the other hand, the GM-based Δ^{14} C results are about 20 per mil lower than the ISO results (Figure 8d), which is mainly due to the reduced surface-to-deep ocean downward transport of ¹⁴C around 50°S to 60°S associated with the absence of the Deacon cell (Figure 3d).

[21] As compared to the GLODAP data, overall, the GM scheme simulates natural ¹⁴C values in various deep ocean basins more realistically than the other two schemes. In the entire deep Atlantic Ocean, the HV scheme (Figure 4b)



Figure 6. Latitude-depth distribution of natural Δ^{14} C (permil) in the Indian Ocean based on (a) GLODAP data [*Key et al.*, 2004] and model simulations for (b) horizontal/vertical (HV), (c) isopycnal (ISO), and (d) Gent-McWilliams (GM) schemes.

underestimates the observed Δ^{14} C, while the ISO and GM schemes greatly improve the Δ^{14} C simulation by enhancing Δ^{14} C values (Figures 4c, 4d, and 8a). As a result of enhanced northward intrusion of the AABW (Figure 3d), the GM scheme is also better in capturing the middepth minimum of Δ^{14} C in the Pacific (Figures 5d and 8b) and Indian oceans (Figures 6d and 8c). Again, the GM parameterization gives the most realistic simulation of Δ^{14} C in the Southern ocean, whereas the HV simulated Δ^{14} C is too low and the ISO simulated value is too high relative to observation (Figure 8d).

[22] The pattern similarity between simulated and databased natural ¹⁴C is quantified in Figure 9, which provides a statistical summary of how well matched simulated and observed patterns are in terms of correlation, root mean square (r.m.s) difference, and standard deviation [*Taylor*, 2001]. Figure 9 shows that correlation with observation is similar between different mixing schemes for each basin (about 0.75 to 0.8 for the Atlantic, Pacific, and Indian oceans and 0.85 for the Southern Ocean). In the Pacific and Southern Ocean, the use of the ISO and GM schemes reduces the overestimated standard deviation in the HV simulation and leads to smaller r.m.s difference, indicating the improvement of natural ¹⁴C simulation in these basins based on the ISO and GM schemes. However, in the Atlantic and Indian oceans the ISO and GM schemes underestimate the standard deviation, while the HV simulated standard deviation compares well with observations.



Figure 7. Latitude-depth distribution of natural Δ^{14} C (permil) in the global ocean based on (a) GLODAP data [*Key et al.*, 2004] and model simulations for (b) horizontal/vertical (HV), (c) isopycnal (ISO), and (d) Gent-McWilliams (GM) schemes.

Another point we observe is that although the GM scheme simulates a more realistic basin mean profile of natural ¹⁴C (Figure 8), it does not exhibit improved skill over the ISO scheme in simulating the pattern of ¹⁴C distribution as shown in Figure 9.

[23] There are only a few limited studies available in the open literature that have investigated the effect of various ocean mixing parameterizations on the simulation of natural Δ^{14} C. *England and Rahmstorf* [1999] studied HV, ISO and GM schemes, whereas *Duffy et al.* [1997] studied only HV and GM schemes. *England and Rahmstorf* [1999] found that relative to the HV scheme, the ISO simulated Δ^{14} C values are higher. Our model results also show that compared to the HV scheme, the ISO scheme leads to less depleted Δ^{14} C in the ocean interior (Figure 8). Both *Duffy et*

al. [1997] and England and Rahmstorf [1999] found reduced AABW and NADW formations when the GM scheme was used. This resulted in decreased transport of Δ^{14} C from the surface to the deep ocean and lowered ocean interior Δ^{14} C values. Moreover, in these two studies there was a much larger discrepancy between observed and modeled natural Δ^{14} C in the deep North Atlantic based on the GM scheme than based on the HV scheme. In contrast, we find an enhanced AABW formation when the GM scheme is used, which increases the surface-to-deep ocean Δ^{14} C transport and thus leads to higher values of Δ^{14} C as compared to the HV scheme. In addition, our model simulated Δ^{14} C in the North Atlantic for the HV scheme is somewhat lower than the observational data, but the GM scheme brings the simulated Δ^{14} C much closer to the



Figure 8. Basin mean profiles of natural ¹⁴C (permil) in the (a) Atlantic, (b) Pacific, (c) Indian, and (d) Southern oceans from the model simulations based on horizontal/vertical (HV), isopycnal (ISO), and Gent-McWilliams (GM) schemes and GLODAP data [*Key et al.*, 2004].

observations (Figure 4). This is due to the fact that in our simulation, relative to the HV scheme, the GM-estimated NADW formation is slightly higher and the vertical mixing is stronger.

5.2. Simulation of Bomb-Produced ¹⁴C

[24] In this section we discuss simulated oceanic uptake of bomb produced ¹⁴C for different mixing schemes with climate change cases (WC). Unlike natural ¹⁴C, which provides estimates of deep ocean mixing rates over long timescales, uptake of radiocarbon as a result of atmospheric nuclear bomb tests ("bomb radiocarbon") is generally used to assess ocean model's performance in the simulation of ocean mixing processes in approximately the upper one kilometer of ocean associated with a timescale of several decades [*Toggweiler et al.*, 1989b; *Jain et al.*, 1995; *Duffy et al.*, 1995a, 1995b].

[25] In terms of comparing model simulated oceanic uptake for bomb ¹⁴C with the GEOSECS measurements, it is worth emphasizing here that considerable uncertainties exist in the GEOSECS measurement-based estimates [*Broecker and Peng*, 1994; *Hesshaimer et al.*, 1994; *Broecker et al.*, 1995; *Jain et al.*, 1997; *Hesshaimer and Levin*, 2000; *Peacock*, 2004]. Using the observed correlation between natural radiocarbon and dissolved silicate,

Broecker et al. [1995] estimated the oceanic bomb carbon uptake based on the GEOSECS measurements. The uncertainty in the Broecker et al. [1995] estimation was assumed to be of the order of 10%, but the results did not provide any basis for this estimate of uncertainty. Jain et al. [1997] concluded that model-based estimates of bomb radiocarbon inventories were well within the uncertainty range, but suggested that the Broecker et al. [1995] estimates of ocean bomb radiocarbon inventory could be in error by as much as 20%. In a more recent study, *Peacock* [2004] pointed out a number of sources of hard-to-quantify error in the Broecker et al. [1995] approach. For instance, GEOSECS observations were made at individual stations of particular latitudes and longitudes. Therefore, in order to make the global inventory estimates, Broecker et al. [1995] chose to divide each ocean into 10° wide latitudinal bands and calculated the mean of the bomb radiocarbon inventory for the entire band on the basis of just a small handful of station data for each given band. According to *Peacock* [2004], it is likely that in some locations tracer distributions are strongly nonuniform across a line of constant latitude. Peacock [2004] revised the Broecker et al. [1995] estimation on the basis of a number of new approaches, primarily involving the use of OGCM-simulated CFC and anthropogenic CO₂. Peacock [2004] found that the corrected bomb radio-



Figure 9. Taylor diagram of pattern statistics [*Taylor*, 2001] describing the model performance in the simulation of natural ¹⁴C. Each statistic variable is normalized by the standard deviation of the corresponding observed field. Therefore the observed field, after it is normalized by itself, is always plotted at unit distance from the origin along the abscissa. For each point the radial distance from the origin to the point gives the pattern standard deviations, and the azimuthal positions give the correlation with observations. The distance between the observed point and any model's point is proportional to the centered RMS error. Abbreviations are as follows: A, Atlantic Ocean; P, Pacific Ocean; I, Indian Ocean; and S, Southern Ocean. Circles indicate HV scheme, triangles indicate ISO scheme, and diamonds indicate GM scheme.

carbon ocean uptake was in closer agreement with that proposed by *Hesshaimer et al.* [1994] than that obtained by *Broecker et al.* [1995]. In spite of these corrections, we strongly feel that the most accurate method to compare model results with GEOSECS observations is to carry out a point-to-point evaluation. However, our 2.5-D ocean model output, which represents the averaged value at a given latitude band, cannot be directly compared with individual GEOSECS station data. With this in mind, the comparison between modeled bomb ¹⁴C and the GEOSECS data along latitudinal bands discussed below should be taken as a first-order assessment of model performance in simulating ¹⁴C.

[26] One consistent pattern in Table 1 is that the HV scheme in each ocean basin yields the lowest bomb ¹⁴C inventory and penetration depth, whereas the ISO scheme yields the highest ¹⁴C inventory and penetration depth. The GM scheme-based values lie between the HV and ISO schemes. This pattern of oceanic uptake for bomb ¹⁴C can also be seen in the latitude-depth distribution of bomb-produced ¹⁴C of the global ocean in the year 1975 (the middle year of the GEOSECS survey) (Figure 10). Compared to the HV scheme, enhanced uptake of bomb-produced ¹⁴C is seen most clearly in both the North Atlantic and Southern Ocean for the ISO scheme. Relative to the ISO scheme, the GM scheme reduces the uptake of bomb ¹⁴C in the Southern Ocean because of the absence of the Deacon cell (Figure 3d).

[27] The largest differences in the simulated bomb ¹⁴C uptake between various mixing schemes occur in the Southern Ocean. In terms of column inventory, the ISO and GM schemes enhance HV-based ¹⁴C inventory by 8.7% and 7.2% in the Atlantic Ocean, 10.1% and 7.9% in the Pacific Ocean, 6.7% and 5.3% in the Indian Ocean, and 26.6% and 17.0% in the Southern Ocean. These results indicate that the simulated oceanic uptake of bomb ¹⁴C is most sensitive to different ocean mixing parameterizations in the Southern Ocean.

[28] Table 1 shows that compared to the GEOSECS measurement, no single mixing scheme is consistently better than the others in simulating ^{14}C inventory and penetration depth for different ocean basins. For instance, model simulated ¹⁴C inventories based on the ISO scheme in the Atlantic and Indian oceans are in better agreement with GEOSECS data than those based on the other two schemes, whereas the HV-based results in the Pacific and Southern oceans compare better with the observations. Although the ISO and GM schemes improve the simulated penetration depth in the Atlantic and Indian oceans by increasing the vertical transport of bomb ¹⁴C, this process leads to overestimations of observed penetration depth in the Pacific and Southern oceans. The pattern similarity of the bomb ¹⁴C inventory between model simulations and observations is summarized in a Taylor diagram (Figure 11),

Table 1. Comparison of the Model Simulated Bomb ¹⁴C Inventory and Penetration Depth Based on Various Mixing Schemes With the Data-Based Estimates^a

			This	Study			Data-Based Estimation			
		Inventory			Depth		Inver	Inventory		
	HV ^b	ISO ^b	GM ^b	HV ^b	ISO ^b	GM ^b	GEOSECS ^c	Peacock [2004] ^d	GEOSECS ^c	
Atlantic	6.9	7.5	7.4	256	488	452	11.4 (10.3-12.5)	8.9/9.7	543 (489-597)	
Pacific	8.9	9.9	9.6	370	505	439	8.4 (7.6–9.2)	7.7/8.1	317 (285-349	
Indian	7.5	8.0	7.9	218	268	266	10.6(9.2 - 11.7)	9.7/9.3	389 (350-428)	
Southern	9.4	11.9	11.0	293	609	445	6.6 (5.9-7.3)	7.2/6.7	353 (318-388)	
Global	8.4	9.4	9.1	308	486	414	9.0 (8.1-9.9)	8.2/8.3	386 (347-425)	

^aInventory is in 10^9 atmos cm⁻², and penetration depth is in m.

^bAbbreviations are as follows: HV, horizontal/vertical; ISO, isopycnal; GM, Gent-McWilliams.

Range of values is estimated on the basis of the 10% uncertainty assumed by Broecker et al. [1995].

^dThe two values are the corrected values of *Broecker et al.* [1995] based on CO₂ (first value) and CFC data (second value), respectively.



Figure 10. Model simulated latitude-depth distribution of bomb-produced ¹⁴C (permil) for global ocean in 1975 based on the (a) horizontal/vertical (HV), (b) isopycnal (ISO), and (c) Gent-McWilliams (GM) schemes.

which shows that no single mixing scheme is consistently superior to the others in simulating bomb ¹⁴C inventory in different ocean basins. All schemes exhibit approximately the same skill in the Atlantic Ocean, with a correlation coefficient of about 0.65. In the Indian Ocean, all schemes exhibit a high correlation (>0.95), but the HV scheme underestimates the standard deviation. In the Pacific and

Southern oceans, the HV-based bomb ¹⁴C is in closer agreement with observations than that based on the ISO and GM schemes, as indicated by the smaller r.m.s errors based on the HV scheme.

[29] These results indicate that no single scheme is able to simultaneously simulate well the upper ocean vertical mixing in different ocean basins. In addition to vertical mixing, another important factor limiting our ability to accurately simulate oceanic uptake of bomb ¹⁴C is the rate of air-sea exchange, which may be subject to uncertainties associated with its dependence on wind speed and high-latitude sea ice fraction. Moreover, as discussed above, uncertainties in the data-based estimates of bomb ¹⁴C uptake also limit our ability to evaluate model performance in a more reliable manner. This is especially true in the Southern Ocean, where uncertainty between model-based and data-based results is quite large, perhaps because of limited observations in this region. For example, Leboucher et al. [1999] estimated bomb ¹⁴C inventory using measured CFC-11 and CFC-12 and a mixing model in the Southern Ocean, and found that their results are roughly 5 times greater than that of Broecker et al. [1995].

[30] To illustrate model simulated bomb ¹⁴C after the GEOSECS time period, we compare model simulated basin



Figure 11. Taylor diagram of pattern statistics [*Taylor*, 2001] describing the model performance in the simulation of bomb ¹⁴C inventory. Each statistic variable is normalized by the standard deviation of the corresponding observed field. Therefore the observed field, after it is normalized by itself, is always plotted at unit distance from the origin along the abscissa. For each point the radial distance from the origin to the point gives the pattern standard deviations, and the azimuthal positions give the correlation with observations. The distance between the observed point and any model's point is proportional to the centered RMS error. Abbreviations are as follows: A, Atlantic Ocean; P, Pacific Ocean; I, Indian Ocean; and S, Southern Ocean. Circles indicate HV scheme, triangles indicate ISO scheme, and diamonds indicate GM scheme.



Figure 12. Basin mean profiles of bomb ¹⁴C (permil) in the (a) Atlantic, (b) Pacific, (c) Indian, and (d) Southern oceans from the model simulations based on the horizontal/vertical (HV), isopycnal (ISO), Gent-McWilliams (GM) schemes and GLODAP data [*Key et al.*, 2004]. The Pacific, Indian, and Southern oceans profiles represent bomb ¹⁴C for mid-1990s, while the Atlantic Ocean profile represents bomb ¹⁴C for mid-1980s.

mean bomb ¹⁴C profiles with the GLODAP data [Key et al., 2004] (Figure 12). The GLODAP bomb ¹⁴C profiles for the Pacific, Indian, and Southern oceans are available for mid-1990s, while the profile for the Atlantic Ocean is available for mid-1980s. Compared to the HV scheme, the ISO and GM schemes transport more bomb ¹⁴C from the upper 500m ocean to the ocean interior, resulting in less bomb ¹⁴C above 500 m and more bomb ¹⁴C below that (Figure 12). Figure 12 also shows that no single mixing scheme is consistently better than the others in simulating observed bomb 14C profile. For instance, the HV scheme greatly overestimates bomb ¹⁴C in the upper Southern Ocean, but simulates well below about 500 m. On the other hand, the ISO and GM schemes improve the simulated ¹⁴C profile in the upper ocean, but overestimate it below 500 m (Figure 12d).

[31] *Duffy et al.* [1995a, 1995b] investigated the effect of various ocean mixing parameterizations on oceanic uptake of bomb ¹⁴C using the GFDL OGCM. They found that relative to the HV scheme, the GM scheme decreases the overall vertical transport of bomb ¹⁴C because of the net effect of increased vertical diffusion and decreased advective transport and convective mixing. As a result, their GM-based ¹⁴C inventory and penetration depth, when averaged amongst all GEOSECS stations, are smaller than those

estimated from the HV scheme and compare worse with the GEOSECS data. In contrast to their studies, our model results show that the enhanced vertical mixing based on the GM scheme increases the modeled column inventory and penetration depth, and leads to better agreement with GEO-SECS data at the global scale (Table 1).

6. Uptake of Anthropogenic CO₂

[32] This section compares model-estimated anthropogenic CO₂ uptake for WC case with data-based estimates and OGCM results. Similar to the oceanic uptake of bomb ¹⁴C, the model-simulated anthropogenic CO₂ uptake for all basins is largest for the ISO scheme and smallest for the HV scheme, while the GM-based values lie between the two (Table 2). Among all ocean basins, the Pacific and Southern oceans are the largest contributors to the global uptake. Upwelling at the equatorial Pacific, along with deep water formation and convective mixing at the Southern Ocean are important processes that enrich interior water with anthropogenic CO₂ through more frequent contact with surface water.

[33] Among all ocean basins, simulated CO_2 uptakes determined by different schemes differ most in the Southern Ocean (Table 2). For instance, during 1765–1990 the

1989 and	1765-1990 ^a				
Table 2.	ISAM-2.5D Simulated	Oceanic CO_2 Uptake	(PgC) for Different Mixin	g Schemes and Experim	ents During the Period of 1980-

	1980-1989						1765 - 1990)		
	Global	Atlantic	Pacific	Indian	Southern	Global	Atlantic	Pacific	Indian	Southern
				This S	tudv					
HV-WC ^b	17.7	3.2	7.3	1.4	5.9	111.7	20.2	45.4	8.8	37.2
HV-WOC ^b	19.1	3.4	8.1	1.5	6.1	120.4	21.3	50.2	9.6	39.2
ISO-WC ^b	22.5	4.5	8.7	1.5	7.8	140.2	28.9	51.4	9.7	50.1
ISO-WOC ^b	24.5	4.9	9.5	1.7	8.5	151.7	31.3	58.0	10.6	51.7
GM-WC ^b	18.9	4.2	7.9	1.5	5.3	121.3	28.3	49.8	9.9	33.3
GM-WOC ^b	20.8	4.7	8.6	1.6	5.9	128.5	29.5	53.8	10.7	34.7
				Other S	tudies					
Orr et al. $[2001]^{c}$	15.0 - 22.0									
Prentice et al. [2001] ^d	13.0 - 25.0									
Mcneil et al [2003] ^e	12.0 - 20.0									

^aThe table also provides the data-based and OGCM-estimated global uptake for the period 1980–1989.

^bAbbreviations are as follows: HV, horizontal/vertical; ISO, isopycnal; GM, Gent-McWilliams; WC, with climate change; WOC, without climate change. ^cBased on four OGCM simulations (GFDL, Hadley, MPI, and IPSL).

^dBased on atmospheric CO₂ and O₂ measurements.

^eBased on CFC data.

simulated uptakes based on HV-WC and ISO-WC experiments differ by 12.9 PgC in the Southern Ocean, compared to differences of 8.7, 6.0, and 0.9 PgC in the Atlantic, Pacific, and Indian oceans, respectively. The difference in the Southern Ocean uptake between ISO and GM schemes is much more striking. Over the same period, simulated CO_2 uptakes based on ISO-WC and GM-WC experiments differ by 16.8 PgC in the Southern Ocean, compared to the differences of only 0.6, 1.6, and 0.2 PgC in the Atlantic, Pacific, and Indian oceans. This indicates that the Southern Ocean is the region where simulated CO_2 uptake is most sensitive to various ocean-mixing parameterizations.

[34] As shown in Table 2, the ISAM-2.5D simulated global uptake based on the HV-WC, ISO-WC, and GM-WC experiments for the 1980s vary between 17.7 and 22.5 PgC, as compared to CO_2 and O_2 measurement-based estimates (13.0-25.0 PgC) [Prentice et al., 2001], CFC data-based estimates (12.0-20.0 PgC) [McNeil et al., 2003] and OGCM-based results (15.0-22.0 PgC) [Orr et al., 2001]. As for the period between 1800 and 1994, modeled CO₂ uptake for the global ocean ranges between 117.2 and 146.5PgC, comparable to the data-based estimate of 118 \pm 19 PgC [Sabine et al., 2004]. In terms of regional oceanic CO₂ uptake, the ISAM-based latitudinal variations (Figure 13) are also largely consistent with that simulated by various OGCMs [Orr et al., 2001], with higher uptake occurring in the equatorial and high-latitude oceans. ISAM-2.5D simulated CO₂ uptake for the Southern Ocean is in the middle of the range of OGCM-simulated values. However, model simulated uptake in the North Atlantic and equatorial oceans are at the high end of OGCM results.

7. Effect of Climate Change on Bomb ¹⁴C and CO₂ Uptakes

[35] In this section, we investigate the effect of climate change on the uptake of bomb ^{14}C and anthropogenic CO₂ by comparing the results of WC and WOC experiments as described in section 3.

[36] Climate change has the potential to affect carbon uptake through changes in sea surface temperature (SST),

ocean circulation, sea ice cover, and freshwater flux. Our sensitivity study shows that for the period 1765–2000 the main factor that affects carbon uptake is the decrease in CO_2 solubility as a result of increasing SST due to global warming. For all mixing schemes, climate change reduces the oceanic uptake of anthropogenic CO_2 (Table 2). At the global scale, WC-based CO2 uptakes for the 1980s are about 1.4–2.0 PgC lower relative to WOC-based estimates (a reduction of about 7% to 9%) (Table 2). For comparison, Joos et al. [1999] estimated a reduction of about 2.5 PgC (about 12%) in CO₂ uptake over the same time period. Over the period 1765–1990, the global reduction in cumulative CO_2 uptake due to climate change varies from 7.2 to 11.5 PgC (a reduction of about 6% to 8%). Among all ocean basins, the reduction in CO₂ uptake is largest in the Pacific Ocean, ranging from 0.7 to 0.8 PgC for 1980–1989, and 4.0 to 6.6 PgC for 1765-1990. However, in contrast to CO₂ uptake, we found negligible influence of climate change on the oceanic uptake of bomb ¹⁴C. Differences in modelsimulated bomb inventory and penetration depth between WC and WOC experiments are less than 1%. This is because ¹⁴C inventory is determined by the integration of the excess ¹⁴C in the entire ocean and this integration is largely determined by the subsurface ¹⁴C value, which is not significantly influenced by changes in surface temperature. Therefore the inventory, unlike the air-sea flux of anthropogenic CO₂, is much less sensitive to changes in SST associated with climate change.

8. Discussion and Conclusions

[37] This study examines the sensitivity of simulated uptakes of natural ¹⁴C, bomb ¹⁴C, and anthropogenic CO₂ to three ocean mixing parameterizations: HV, ISO, and GM schemes, and the resultant climate change effect using an Earth system model of intermediate complexity, ISAM-2.5D.

[38] In the case of natural ¹⁴C, relative to the HV scheme the ISO and GM schemes simulated natural ¹⁴C distributions compare better with GLODAP measurements through enhanced vertical diffusion and deep water for-



Figure 13. Model simulated zonal mean uptake of anthropogenic CO_2 for the year 1990 based on the horizontal/vertical (HV), isopycnal (ISO), and Gent-McWilliams (GM) schemes. The vertical bars illustrate the OCGM-simulated range of CO_2 uptake for 60°S, 0°, and 60°N latitudes [*Orr et al.*, 2001].

mation. In particular, the GM scheme yields more realistic natural 14 C in the Southern Ocean because of a more realistic ocean circulation without the appearance of the Deacon cell. Moreover, the GM scheme also more realistically captures the observed middepth minimum of natural 14 C in the Pacific and Indian oceans as a result of enhanced northward intrusion of the Antarctic Bottom Water (AABW).

[39] In the simulation of uptake for bomb 14 C and anthropogenic CO₂, both climate and the carbon cycle are integrated simultaneously so that the effect of climate change on carbon uptake is included. The model estimated global bomb ¹⁴C inventories differ by about 12% between the highest (ISO-based) and the lowest (HV-based) estimated values, with the largest difference among various parameterizations found in the Southern Ocean (about 27%). Similar to ¹⁴C uptake, model simulated CO₂ uptake based on different schemes also differ the most in the Southern Ocean. No single mixing scheme is consistently better in simulating oceanic uptakes of bomb ¹⁴C in various basins as compared to the GEOSECS data. For instance, ¹⁴C inventories simulated by the ISO scheme in the Atlantic and Indian oceans shows closer agreement with GEOSECS data than those based on the other two schemes. However, the HV scheme yields better results in the Pacific and Southern oceans. This may be due to the fact that relative to the vertical mixing, the magnitude of bomb ¹⁴C uptake in the upper ocean is more dependent on the air-sea exchange, which may be subject to uncertainties associated with highlatitude sea ice fraction and its dependence on wind speed. However, it should also be noted that considerable uncertainties exist in the data-based estimates of oceanic uptake of bomb ¹⁴C, especially in the Southern Ocean, which

continue to hamper attempts to evaluate model performance in a more reliable manner.

[40] Model simulated global uptake of anthropogenic CO₂ ranges from 17.7 to 22.5 PgC for 1980–1989, a result largely consistent with the data-based estimates of 13.0–25.0 PgC [*Prentice et al.*, 2001] and the OGCM results of 15.0–22.0 PgC [*Orr et al.*, 2001]. Comparison of simulations with and without climate effect shows that climate change reduces CO₂ uptake by 1.4 to 2.0 PgC for 1980–1989 and 7.2 to 11.5 PgC for 1765–1990, mainly because of reduced CO₂ solubility as a result of increased SST. However, the effect of climate change on bomb ¹⁴C inventory and penetration depth is negligible.

[41] In addition, we find that isopycnal mixing has much larger effects on the simulation of ¹⁴C than that of temperature in the ocean interior. In our model, simulated deep ocean temperatures only differ by about 0.5°C to 1.0°C between different schemes (Figure 2). The fact that isopycnal mixing has larger effects on carbon than on temperature is consistent with those of previous studies [e.g., *Siegenthaler and Joos*, 1992; *Jain et al.*, 1995; *Harvey*, 2001], which also found that tracers like ¹⁴C with concentration contours crossing isopycnal contours will experience more enhanced vertical mixing than temperature that has isotherms virtually parallel to isopycnals.

[42] It is also important to note that the GM scheme gives the most realistic results overall, such as a more realistic Southern Ocean circulation and northward intrusion of the AABW, and a more realistic natural ¹⁴C. Moreover, compared to the HV and ISO schemes, the GM scheme is a more physically based subgrid-scale ocean mixing parameterization in that it takes into account both the diffusion along and across isopycnals and the effect of mesoscale eddies on large-scale circulation. Therefore the use of the GM scheme in our future model version is recommended, although its role in the coupled climate-carbon cycle modeling systems needs to be further studied with emphasis on the Southern Ocean.

Appendix A: Governing Equations for Ocean Carbon Cycle

[43] ISAM-2.5D ocean carbon cycle module simulates dissolved inorganic carbon (*DIC*) and radiocarbon (${}^{14}C$) that are governed by the following equation:

$$\frac{\partial C}{\partial t} = -\frac{1}{a\cos\phi} \frac{\partial(\cos\phi vC)}{\partial\phi} - \frac{\partial(wC)}{\partial z} + q_C^{conv} + q_C^{diff} + J + J_v - \lambda C$$
(A1)

In the above equation, tracer *C* represents *DIC* or ${}^{14}C/{}^{12}C$. ϕ is the latitude, a = 6371 km is the radius of the earth, and ν and *w* are the meridional and vertical velocity. The first and second terms at the right hand side of equation (A1) represent the changes in *DIC* and ${}^{14}C/{}^{12}C$ due to meridional and vertical advection, and q_C^{conv} represents the effect of convective overturning and q_C^{diff} represents the effect of diffusive mixing. λ is the radioactive decay constant, which is set to be $\ln 2/5730$ yr⁻¹ for ${}^{14}C$ and zero for *DIC*. *J* is the

[44] The effect of freshwater flux on $DIC J_v$ is calculated as

$$J_{\nu} = \frac{1}{\Delta z_1} DIC_g(E - P - r)$$
(A2)

where Δz_1 is the depth of first ocean layer, DIC_g is the globally averaged surface concentration of *DIC*. *E*, *P*, r is the evaporation, precipitation, and river runoff respectively.

[45] The air-sea exchange of *DIC* is calculated as

$$J = \frac{1}{\Delta z_1} K_w (C_{atmo} - C_{surf}) (1 - f_{ice})$$
(A3)

where f_{ice} is the fraction of ocean that is covered by sea ice. $C_{atmo} = \alpha p CO_2^{atmo}$ (α is the CO₂ solubility and $p CO_2^{atmo}$ is the atmospheric partial pressure of CO₂). C_{surf} is the aqueous [CO₂] concentration at ocean surface that is calculated from the chemical equilibria in the carbonate system. K_w is the gas transfer velocity calculated according to *Wanninkhof* [1992].

[46] The air-sea exchange of ${}^{14}C/{}^{12}C$ is calculated on the basis of *Orr et al.* [2001] with minor modifications

$$J = \frac{k_w \alpha p C O_2^{atmo}}{D I C_s \Delta z_1} (C_a - C_0) (1 - f_{ice})$$
(A4)

where C_a is the atmospheric ${}^{14}C/{}^{12}C$ ratio and C_0 is the oceanic ${}^{14}C/{}^{12}C$ ratio. C_a is held at 100 for control run and C_0 is initialized to be 85 everywhere. $DIC_s = DIC_{so} + DIC_{smc}$ where DIC_{so} is the observed surface DIC concentration taken from GEOSEC data and DIC_{smc} is the modeled change in surface DIC concentration relative to the preindustrial value, which is only applied in the perturbation run.

[47] The gas transfer velocity K_w for both *DIC* and ${}^{14}C/{}^{12}C$ is calculated on the basis of *Wanninkhof* [1992]

$$K_w = du^2 \left(\frac{S_c}{660}\right)^{-0.5}$$
 (A5)

where *u* is the observed surface wind speed [*Oberhuber*, 1988] and *Sc* is the Schmidt number determined as a function of sea surface temperature [*Wanninkhof*, 1992]. The proportionality factor d is determined through adjustment by obtaining a value of $K_w\alpha$ that matches the globally averaged CO₂ gas exchange rate of 0.061 mol C m⁻² yr⁻¹ given by *Broecker et al.* [1985]. In this way, we obtain a value of 0.50 for d in our model.

[48] Levels of ¹⁴C are not carried directly in permil but as a percent depletion relative to the atmosphere [*Toggweiler et al.*, 1989a, 1989b; *Orr et al.*, 2001]. Units in permil are obtained by

$$\Delta^{14}C = 10 \times \left({}^{14}C/{}^{12}C - 100\right) \tag{A6}$$

 Δ^{14} C based on equation (A6) is consistent with D definition given by *Stuiver and Pollach* [1977].

Appendix B: Calculation of Bomb ¹⁴C Inventory and Penetration Depth

[49] The inventory of bomb Δ^{14} C is defined as

Inventory =
$$\int_0^z \left[DIC \left(\Delta^{14} C - \Delta^{14} C_{1954} \right) \right] dz \qquad (B1)$$

where $\Delta^{14}C_{1954}$ and $\Delta^{14}C$ are the values of radiocarbon in year 1954 and any given year after that (the nuclear test begins in year 1955). $DIC = DIC_o + DIC_{mc}$ where DIC_o is the observed DIC concentration taken from GEOSEC data and DIC_{mc} is the modeled change in DIC concentration relative to preindustrial value.

[50] The penetration depth of bomb Δ^{14} C is defined as the ratio of bomb 14 C inventory to the surface bomb 14 C concentration. That is,

Penetration depth =
$$\int_{0}^{z} \left(\Delta^{14}C - \Delta^{14}C_{1954} \right) dz / \left(\Delta^{14}C - \Delta^{14}C_{1954} \right) \Big|_{z=0}$$
(B2)

[51] Acknowledgments. We thank Danny Harvey, Kevin Mueller, and two anonymous reviewers for providing valuable comments on an earlier version of this manuscript. This research was supported in part by the Office of Science (BER), U.S. Department of Energy (DOE-DE-FG02-01ER63069), and the US National Science Foundation (ATM-0238668).

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