# MIT Joint Program on the Science and Policy of Global Change



# Uncertainty in Atmospheric CO<sub>2</sub> Predictions from a Parametric Uncertainty Analysis of a Global Ocean Carbon Cycle Model

Gary Holian, Andrei P. Sokolov and Ronald G. Prinn

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#### Abstract

Key uncertainties in the global carbon cycle are explored with a 2-D model for the oceanic carbon sink. This model has many enhancements over simple 1-D box-diffusion models, including mixed-layer inorganic carbon chemistry, climate-dependent air-sea exchange rates, and mixing of dissolved inorganic carbon into the deep ocean that is parameterized by 2-D eddy diffusion. At the same time it is much more computationally efficient than 3-D models which makes it applicable to a comprehensive parametric uncertainty analysis. By calibrating the key parameters of this ocean carbon sink model to widely referenced values, it produces an average oceanic carbon sink during the 1980s of 1.94 Pg yr<sup>-1</sup>, consistent with the range estimated by the IPCC of 2.0 Pg yr<sup>-1</sup>  $\pm$  0.8 Pg yr<sup>-1</sup> (1994). The uncertainty range cited in the IPCC study and widely reported elsewhere is principally the product of the structural uncertainty derived from the results of several ocean carbon sink models of varying degrees of complexity. This range does not directly take into account the parametric uncertainty inherent in these models or how those uncertainties will impact on forecasts of future atmospheric CO<sub>2</sub> concentrations.

A sensitivity analysis of the parameter values used as inputs to the 2-D ocean carbon sink model developed for this study suggests that the IPCC's range for the oceanic carbon sink of 1.2 to 2.8 Pg yr<sup>-1</sup> during the 1980s may be too conservative. By applying the Probabilistic Collocation Method (Tatang *et al.*, 1997) to this simple ocean carbon sink model, the uncertainty in the size of the oceanic sink for carbon and hence future atmospheric CO<sub>2</sub> concentrations is quantitatively examined. This uncertainty is found to be larger than that implied by the structural differences examined in the IPCC study alone. An average 1980s oceanic carbon sink of  $2.06 \pm 0.9$  Pg yr<sup>-1</sup> (with 67% confidence) is estimated. This uncertainty is found to be dominated the uncertainty in by the rate of vertical mixing of dissolved carbon from the surface into the deep ocean which is parameterized in this study by vertical diffusion. A contribution of the uncertainty in vertical diffusion even increases with time from 83% in the 80s to about 97% in 2100. In contrast a contribution of an uncertainty in the rate of air-sea CO<sub>2</sub> exchange decreases from 15% to less than 1% during the same period.

It is observed that a wide range of parameter values can be used to balance the contemporary carbon cycle due to the large uncertainties in the total oceanic and terrestrial sinks. These parameter values yield correspondingly large differences in the range of future atmospheric CO<sub>2</sub> concentrations when driven by forecasts of anthropogenic CO<sub>2</sub> emissions scenarios over the next century. For a reference set of emissions similar to the IS92a scenario of the IPCC (1992), the uncertainty in the atmospheric CO<sub>2</sub> concentration in 2100 is found to be 659 ppm  $\pm$  35 ppm (with 67% confidence). This uncertainty is solely due to uncertainties identified in the "solubility pump" mechanism of the oceanic sink, which is only one of the many large uncertainties lacking a quantitative examination in the global carbon cycle. Such uncertainties have implications for the predictability of atmospheric CO<sub>2</sub> levels, a necessity for gauging the impact of different rates of anthropogenic CO<sub>2</sub> emissions on climate and for policy-making purposes. Because of the negative feedback between the natural carbon uptake by the terrestrial ecosystem and atmospheric CO<sub>2</sub> concentration, taking changes in the former into account leads to a smaller uncertainty in the latter compared to that in the case with the fixed terrestrial uptake.

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#### **1. INTRODUCTION**

Recent concerns about increasing anthropogenic emissions of greenhouse gases over the course of the last century have given impetus to studies of the global carbon cycle, focusing particularly on the existence and magnitude of natural sinks for atmospheric  $CO_2$ . Ice core and atmospheric data show that the  $CO_2$  mole fraction has increased from an average value of 275 ppm in preindustrial times to approximately 366 ppm (Keeling *et al.* 1999). Future emissions of  $CO_2$ , chiefly due to the combustion of fossil fuels are predicted to double or even quadruple the atmospheric concentration of  $CO_2$  over the 21<sup>st</sup> century with potentially adverse consequences for global climate. However, the ability to accurately predict future atmospheric  $CO_2$  levels is predicated on the ability to successfully model the natural carbon cycle and its response to continuing emissions of  $CO_2$  and perturbations of global climate.

To predict  $CO_2$  levels over the 21<sup>st</sup> century, emissions forecasts for anthropogenic  $CO_2$  are combined with models of the surface sinks for carbon. The various processes which determine the size of the natural sinks for  $CO_2$  in the terrestrial biosphere and the ocean are not fully understood. Of these two primary surface sinks for carbon, the ocean has been widely studied for its potential to be the dominant sink for carbon owing to its large capacity to take up  $CO_2$  through dissolution. These studies have resulted in models of the contemporary oceanic carbon sink that vary in complexity from simple box models to complete global biogeochemical models that include full dynamical simulations of the ocean general circulation (Oeschger *et al.*, 1975; Maier-Reimer and Hasselman, 1987; Sarmiento *et al.*, 1992). These models are important scientific tools for understanding the behavior of the global carbon cycle. But when they are used to forecast atmospheric  $CO_2$  concentrations, it is important to assess the uncertainties in these calculations resulting from model imperfections. Relatively small uncertainties now may lead to large uncertainties in future predictions.

Differences in carbon sink estimates produced by different models are, to a large extent, the results of structural differences between those models. These include the use of different parameterizations of physical processes, different model resolutions, and so on. The impact of such structural uncertainty on the forecast of atmospheric CO<sub>2</sub> concentration is difficult to evaluate in an ordered way. The two-dimensional (2-D) Ocean Carbon Model (OCM) used in this study has a fixed set of parameterizations of the transfer of CO<sub>2</sub> gas across the air-sea interface, chemical interactions with dissolved inorganic carbon in the ocean, and transport of dissolved carbon into the thermocline and deep waters by assumed Fickian diffusive mixing processes. However, the range of values for its parameters are chosen so as to cover the range of structural uncertainty exhibited by a range of models. This allows us to treat structural uncertainty as parametric uncertainty. Such an approach was used by Webster and Sokolov (2000) for studying uncertainty in climate change projections by the MIT 2D Climate Model. Because these parameters have some measured or otherwise quantifiable uncertainty, it is possible to apply an analytical method of uncertainty analysis to the model, namely the Probabilistic Collocation Method (Tatang et al., 1997), also known as the Deterministically Equivalent Modeling Method, to determine the impact of parametric uncertainly on the uncertainty of the size of the oceanic carbon sink. The parameters that are most important in contributing to the variance of the carbon sink can also be ranked in order of importance using this method.

Projections of atmospheric concentrations of  $CO_2$  can then be calculated as a function of this uncertain oceanic carbon sink, after considering a contemporary carbon budget consistent with current observations. Instead of projecting a single concentration path for atmospheric  $CO_2$  for a given anthropogenic emissions scenario, it is possible to produce probability distributions for future atmospheric  $CO_2$  concentrations as a function of time due to the quantifiable uncertainties in the oceanic carbon sink.

# 2. THE OCEAN CARBON MODEL (OCM)

#### 2.1 Model Description

The OCM is a component of the MIT Joint Program on the Science and Policy of Climate Change's 2-D Land-Ocean resolving (2D-LO) Climate-Chemistry model (Prinn *et al.*, 1999; Sokolov *et al.*, 1998; Wang, Prinn and Sokolov, 1998) and therefore shares that model's simplified oceanic structure. The OCM is a latitudinally-extended box-diffusion model composed of 24 zones centered 7.826 degrees apart. Oceanic mixed layer depths are kept constant at the observed average annual values. The mixed layer is attached to an eddy-diffusive deep ocean with 10 vertical layers of increasing depth. The model has many similarities to existing box-diffusion models (Oeschger *et al.*, 1975; Siegenthaler and Joos, 1991) and zonally-averaged models of the ocean (Stocker *et al.*, 1994).

The OCM uses the real world distribution of oceanic surface area as a function of latitude, but treats all basins as a zonal average. Open ocean, for the purposes of air-to-sea transfer of gaseous  $CO_2$  in each latitude, is the total open ocean area minus the amount covered in sea-ice. The two southernmost latitude zones contain no ocean and are composed perpetually of the Antarctic continent, while the northernmost zone is covered by ice. Bottom ocean topography is not considered and depth is assumed to extend to a constant 3000 meters. The OCM is designed to be run either interactively with the 2-D Climate-Chemistry model, or alone (obtaining its required chemical and climatic inputs exogenously). The basic 2-D structure of the model is depicted in **Figure 1**.



Figure 1. Structure of the OCM

In the OCM, the calculated air-to-sea flux of carbon is proportional to the CO<sub>2</sub> partial pressure  $(pCO_2)$  gradient between the atmosphere and the oceanic surface layer multiplied by a calculated piston (*i.e.* transfer) velocity, V<sub>p</sub>:

$$CO_2 Flux = V_p \left( p C O_2^{air} - p C O_2^{sea} \right)$$
(1)

The piston velocity is calculated as a function of the surface wind speed at each latitude (Tans *et al.*, 1990). Partial pressure of  $CO_2$  at the ocean surface p $CO_2$  is calculated from dissolved  $CO_2$  via Henry's law.

In mixed-layer of the ocean dissolved  $CO_2$  is assumed to be in chemical equilibrium with dissolved carbonate and bicarbonate ions in the surface ocean, with this equilibrium being dependent on temperature, alkalinity, and the concentrations of boric, silicic, phosphoric and other acids. Together, total dissolved inorganic carbon (DIC) in the ocean is defined as the sum of the concentrations of three carbon species:

$$DIC = [CO_2^*] + [HCO_3^-] + [CO_3^{2-}]$$
(2)

Less than 1% of total DIC in the oceans (averaging about 2.05 mol m<sup>-3</sup>) is actually dissolved  $CO_2$  gas. Over 89% is present as  $HCO_3^-$  and 10% as  $CO_3^{2-}$  and while total DIC is preserved for changes in temperature and pressure, the relative proportions of these three species are not, affecting the concentration of dissolved  $CO_2$ . The equilibrium reactions therefore play a direct role in determining the size of the pCO<sub>2</sub> gradient between the atmosphere and sea. The temperature dependence of these reactions is primarily responsible for the natural seasonal cycle and distribution of pCO<sub>2</sub> in the surface ocean. In the OCM, this translates into a latitudinal gradient in the surface partial pressure of  $CO_2$  and a net flux that depends on latitude.

The specific formulation of the carbonate chemistry used in the OCM is based primarily on that of Peng *et al.* (1987), which includes the effects of temperature, dilute acids and tritation alkalinity on inorganic carbonate chemistry. The first and second apparent dissociation constants for carbonic acid,  $K_1$  and  $K_2$ , are strongly dependent on the temperature, alkalinity, and salinity of the ocean (Weiss, 1974; Mehrbach *et al.*, 1973). By exogenously supplying known observed quantities for each of the latitude zones of the model for titration alkalinity, DIC, total borate, total silicate, and total phosphorus, the seven equations which determine the system are reduced into a single higher order equation in  $[H^+]$ , DIC, and carbonate alkalinity ( $Alk_{carb}$ ).

$$[H^{+}] = \frac{K_{1}}{2 \cdot Alk_{carb}} \{ (DIC - Alk_{carb}) + ((DIC - Alk_{carb})^{2} + 4(Alk_{carb} \cdot \frac{K_{2}}{K_{1}})(2 \cdot DIC - Alk_{carb}))^{\frac{1}{2}} \}$$
(3)

Equation (3) cannot be solved analytically, since  $Alk_{carb}$  depends on  $[H^+]$ , so an iterative method is used to solve for a consistent value of  $[H^+]$ . Once this value is known, the  $pCO_2^{sea}$  and all other concentrations consistent with that  $[H^+]$  are determined in the model for that latitude and time step. The resultant  $pCO_2^{sea}$  is used in equation (1) to compute the carbon flux into the ocean.

The mixed layer DIC concentration depends on the surface flux from air-to-sea and the amount exported to the deep ocean by diffusion in each latitude zone in the model. This flux to the deep ocean has the effect of increasing the air-to-sea gradient which allows more carbon to enter the ocean than gets in by buffering alone. Vertical and horizontal transports DIC are parameterized by diffusion (Sokolov *et al.*, 1998) according to the equation:

$$\frac{dDIC}{dt} = K_V \frac{d^2 DIC}{dz^2} + K_H \frac{d^2 DIC}{dy^2}$$
(4)

The baseline values of vertical diffusion coefficients ( $K_v$ ) determined from tritium observations (Hansen *et al.*, 1984) are given in **Figure 2**. These values allow simulation of the strong sinking motion at high latitudes compared to equatorial waters due to the thermohaline circulation and other mixing. When averaged and weighted by area, the global mean value of the vertical diffusion coefficients is 2.5 cm<sup>2</sup>s<sup>-1</sup>. K<sub>H</sub> values are prescribed as a function of depth (decreasing with the latter) but do not depend on latitude. The values of K<sub>H</sub> used in this study (Stocker *et al.*, 1994) are close to those used in Sokolov *et al.* (1998) in the upper part of the ocean (2.8 x 10<sup>4</sup> compared to 2.4 x 10<sup>4</sup> in the mixed layer), but larger in the deep ocean.



**Figure 2**. Vertical Diffusion Coefficients,  $K_V$  (in 101 cm<sup>2</sup>sec<sup>-1</sup>) as a Function of Latitude

#### 2.2 Pre-industrial Steady State Spin-up

The OCM is initially spun-up to a pre-industrial steady-state, characterized by a fixed preindustrial atmospheric  $CO_2$  concentration of 280 ppm and zero net annual exchange between all the major carbon reservoirs. Climate data, in the form of zonal mean sea surface temperatures and surface wind speeds, was obtained from a run of the climate model of Sokolov and Stone (1998) under equilibrium pre-industrial conditions. Due to the extremely long time it takes carbon to reach the deepest layers of the ocean, this spin-up required 10,000 years starting from an ocean devoid of carbon. Since by assumption, the land biosphere was also in steady-state at this time, no model or assumption for terrestrial exchange is required.

#### 2.3 Transient Spin-up for 1765–1989

In order to spin up the 2-D OCM from the steady-state established above (which must be reestablished for any changes of the chemical parameters or diffusion coefficients), it is driven from steady-state to the present day using the historical atmospheric  $CO_2$  record which has been estimated from ice-cores and post-1957 observations at the South Pole and Mauna Loa. The oceanic carbon sink model is also driven with zonally-averaged climatic data, including sea surface temperatures and surface wind speeds predicted over the same period in the MIT 2D-LO Climate Model to spin it up to the present.

Using reference values for all the parameters of the OCM, a net carbon flux into the ocean that grows with time between 1765 and 1990 is predicted as shown in **Figure 3**. The average annual



**Figure 3**. Annual Carbon Flux (Pg yr<sup>-1</sup>) into the Ocean between 1765-1990

global flux in the OCM rises from zero in the steady-state at 1765 to over 2.0 Pg yr<sup>-1</sup> in the late 1980s. Most of the increase in the oceanic carbon sink comes in the last 90 years of the run, when atmospheric  $CO_2$  has been rising the fastest. An examination of the change in the distribution of DIC in the ocean from the steady-state in 1765 to 1990 indicates specifically where the carbon is being taken up by the ocean. The distribution of additional DIC added to the ocean as a function of latitude and depth for the top 2000 meters of the ocean is plotted in **Figure 4**. DIC additions represent about 1% of the surface concentration of background DIC. Most of the carbon taken up by the model is confined to the top 500 meters of the ocean.

As expected, the carbon is being taken up primarily at high latitudes, where lower temperatures, larger mixed layer depths, and faster vertical diffusion rates favor a larger sink. A comparison of the output of the oceanic carbon sink model in this study with a group of the most commonly referenced models in the literature (IPCC, 1994) and the Ocean Carbon Model Intercomparison Project (OCMIP) reveals that the reference response of the OCM that is spun up to the present with the historical  $CO_2$  record falls within the range of other models of greater and lesser complexity. Average concentrations of dissolved inorganic carbon in the current ocean are also in good



**Figure 4**. Distribution of Additional Total dissolved inorganic carbon (mol DIC m<sup>-3</sup>) in the Ocean, between 1765-1990

agreement with globally observed values of 2.06 mol m<sup>-3</sup>. A north-to-south gradient is observed in this current state, consistent with inorganic carbonate chemistry favoring higher DIC (lower pCO<sub>2</sub>) concentrations at lower temperatures and lower DIC (higher pCO<sub>2</sub>) at higher temperatures. However, the vertical gradient in DIC is weaker than that of the observed quantity in the oceans, due to the lack of a marine biological cycle in the model which tends to deplete the surface of carbon with respect to the deep ocean. However, this is generally considered a "fly-wheel" effect, since it does not increase the net transfer of carbon to depth because of nutrient limitations that cause the marine biological cycle to be relatively insensitive to rising DIC on the century time-frames addressed here. The model is constructed to simulate the changes in uptake of CO<sub>2</sub> due to CO<sub>2</sub> perturbations in the atmosphere, and not the observed distribution of carbon in the oceans upon for which the imposition of an additional cycling of carbon by marine biota in the oceans is required.

# 3. FORECASTING ATMOSPHERIC CO<sub>2</sub> CONCENTRATIONS

# 3.1 Closing the Carbon Cycle

In the industrial era, increases in the global average atmospheric concentration of  $CO_2([CO_2]^{atm})$  can be expressed by

$$\frac{d[CO_2]^{adm}}{dt} = F_t - O_t + L_t - B_t$$
(5)

where  $F_t$  is the rate of fossil fuel emission,  $O_t$  is the oceanic carbon sink,  $L_t$  is the anthropogenic carbon emission from land-use, and  $B_t$  is the natural terrestrial biospheric sink for carbon The net biospheric sink for carbon is defined as  $NB_t = L_t - B_t$ . Estimates of fossil fuel emissions and the historical oceanic sink from model simulations indicate that the biosphere as a whole has probably been acting as a net sink for carbon over the last 50 years. This sink is currently believed to be comparable in size to the oceanic carbon sink (within the uncertainty of both; see *e.g.*, Tans *et al.*, 1990).

Both the anthropogenic fossil fuel emission scenario and carbon emission from land-use used to forecast atmospheric  $CO_2$  concentrations over the next century come from a recent reference run of the MIT Integrated Global System Model (Reilly *et al.*, 1999). This emissions prediction is consistent with current observations of atmospheric  $CO_2$  concentrations through 1998 and forecasts fossil fuel emissions growth as a function of economic activity through 2100 (**Figure 5**). It represents a frame of reference from which to compare the behavior of the OCM with other models that are also driven by similar carbon emissions scenario.



**Figure 5**. *Reference Fossil Fuel Emissions (Pg yr<sup>-1</sup>) for 1990-2100* 

Uncertainty in *anthropogenic carbon* emission is not considered in this paper, since this study is limited to the physical processes that produce or feedback on carbon sinks in the ocean and their contribution to uncertainty in atmospheric  $CO_2$  concentrations. This is not a study of the total uncertainty in future atmospheric  $CO_2$  concentrations, which must necessarily include economic and policy considerations (*e.g.*, Prinn *et al.*, 1999).

To model the terrestrial sink of carbon we use a simple parameterization of the Terrestrial Ecosystems Model (TEM; McGuire *et al.*, 1997; Melillo *et al.*, 1993; Xiao *et al.*, 1995, 1997) which is a process-oriented model that can simulate either the equilibrium or transient response of surface land biota and soils to changes in CO<sub>2</sub> levels in the atmosphere as well as to changes in climate. The TEM model includes 18 distinct ecosystems, called "biomes," which are defined on a grid of the world,  $\frac{1}{2}^{\circ}$  by  $\frac{1}{2}^{\circ}$  in size that contains 62,483 land grid cells (about 25% of the globe), including 3,059 ice grid cells, and 1,525 wetland grid cells, ranging between 83°N and 56°S. The TEM is computationally very demanding so a parameterization for the net ecosystems productivity calculated by the model between 1990 and 2100 and driven by atmospheric CO<sub>2</sub> concentrations and changing climate is constructed. A constant term which accounts for additional sinks (*e.g.*, due to nitrate deposition and Northern Hemisphere reforestation) that are not as yet incorporated into the TEM is also added to close the carbon budget in the 1980s. The following equation for the natural biospheric sink for carbon is assumed:

$$B_t = B_t^{TEM} + \overline{B}$$
(6)

The constant  $\overline{B}$  is determined as the residual flux needed to satisfy equation (5) in the 1980s, using the oceanic carbon sink computed by the OCM and the parameterized carbon sink from the TEM:

$$B_t^{TEM} = \frac{[CO_2]_t - 277}{T_t^B}$$
(7)

where  $T_t^B$  is the time-dependent atmospheric lifetime of CO<sub>2</sub> due to the natural biospheric uptake estimated from runs of the TEM driven by different forecasts of atmospheric CO<sub>2</sub> increase and concomitant climate change as predicted in the IGSM (Prinn *et al.*, 1999). Simulations in which the TEM is driven with predicted atmospheric CO<sub>2</sub> concentrations between 1990-2100 cause it to generate a carbon sink as a function of time that is approximated by equation (7). Included in the simulations are the effects of increasing temperature that acts as a negative feedback to the terrestrial carbon sink and CO<sub>2</sub> fertilization effect that acts as a positive feedback (Prinn *et al.*, 1999). From these estimates,  $T_t^B$  is found to increase with time primarily due to a "saturation effect" where higher CO<sub>2</sub> levels, changes in precipitation, and higher temperature decrease the effectiveness of the terrestrial sink for carbon as a function of time. This parameterization for the CO<sub>2</sub> fertilization sink was shown to approximate the TEM output for atmospheric CO<sub>2</sub> concentrations increasing between the range 500-900 ppm (well within the range of uncertainty that is estimated for the atmospheric CO<sub>2</sub> later in this paper). Parametric uncertainty in the terrestrial sink is not considered in this study, focusing instead on the uncertainty in the oceanic sink.

# 3.2 Reference Atmospheric CO<sub>2</sub> Forecast

Taking the above fossil fuel emissions scenario, terrestrial source and sink assumptions, and the OCM to compute  $O_{i}$ , atmospheric  $CO_{2}$  concentrations forecast using equation (5) for the period, 1990-2100 is shown in **Figure 6**, together with observations until 1998.



Figure 6. Reference Atmospheric CO<sub>2</sub> Mole Fractions (ppm) Forecast for 1990-2100

Cabon dioxide concentrations under the reference scenario increase from 353 ppm in 1990 to 661 ppm in 2100. The reference forecast calculated by the IPCC (1995) using the HILDA Model of Seigenthaler and Joos (1992) under the IS92a emissions scenario, forecasted atmospheric  $CO_2$  concentrations to rise to just over 700 ppm. The range of results for all of the models tested in the IPCC survey was about  $\pm$  20 ppm. The atmospheric  $CO_2$  concentration forecast in this study falls below that range in part because of the reduced emissions assumed in the reference anthropogenic  $CO_2$  forecast of the EPPA model (Reilly *et al.*, 1999).

Beyond differences in structure and terrestrial carbon sink assumptions assumed in the above referenced oceanic carbon models, the results for models rely on assumptions of parameter values which are themselves not known with certainty. Numerous models which also purport to do a reasonable job of forecasting atmospheric  $CO_2$  concentrations ignore the degree to which assumptions made in the parameter choices of the model inherently affect their accuracy. The sensitivity of the carbon sink to uncertainty in its parameters should therefore be explored to gauge the robustness of prediction of the ocean sink.

# 4. APPLICATION OF UNCERTAINTY ANALYSIS TO THE OCM

### 4.1 Probabilistic Collocation Method

One of the most commonly used methods for addressing parametric uncertainty in complex models is the Monte Carlo procedure. The Monte Carlo procedure, however, is impractical for studies of models requiring significant computational effort because the number of runs necessary to complete the analysis can typically run into the thousands. For long-term climate change studies, where even simple representations of the systems involved can require large amounts of computational time, the Monte Carlo method is not feasible.

The Probabilistic Collocation Method (PCM, also called the Deterministically Equivalent Modeling Method or DEMM), which is described fully in Tatang *et al.* (1997), provides an efficient method for examining parametric uncertainty in large complex models. The PCM approximates the response of the uncertain outputs of the model under examination by a polynomial chaos expansion. The resultant expansions, constitute a reduced-form version of the original model to which traditional uncertainty methods such as the Monte Carlo method can then be applied with great computational efficiency.

Applying the PCM requires specifying the probability distribution functions (PDFs) of the input parameters and using them to calculate the orthogonal polynomials to be used in the expansion of the model. The roots of these polynomials are then used to define the representative parameter values that are to be used as inputs to runs of the original model to obtain coefficients of the polynomials in these expansions (polynomial power should be chosen in advance). Because these parameter values (called "collocation points") are weighted toward the high probability region of each input parameter, the approximation of the outputs is particularly good within this region. The error of the approximation is estimated by choosing another set of inputs (e.g., from the roots of the polynomials of next order or randomly chosen values), running the original model and comparing its output with the approximation. By iteratively increasing the order of the polynomial chaos expansions, the accuracy of the PCM approximation can be tested until the error of the approximation is sufficiently small to consider it a successful reduced-form model of the original model. The more uncertain inputs and higher the order of the approximation, the more runs of the complex parent model are required to perform the analysis. Since an approximation for every output of the model can be constructed from the same set of model runs the number of outputs chosen is not a computational issue. The outputs selected for approximation by the PCM in this study are the oceanic carbon sink at five year intervals between 1765 and 2100 and atmospheric CO<sub>2</sub> concentrations forecast between 1990 and 2100 by the ocean carbon sink model under a fixed emissions scenario and constrained by the observed carbon budget described in Section 3.

Since the steady-state simulated by the oceanic carbon model differ for any changes in the input parameters, the OCM needs to be driven to a new pre-industrial equilibrium with an atmospheric  $CO_2$  concentration of 280 ppm and spun-up to the present state with the historical  $CO_2$  record for each set of parameters values before a new atmospheric  $CO_2$  forecast can be calculated.

#### 4.2 Uncertain Parameters in the OCM

The first step required to perform the uncertainty analysis using PCM is to choose the uncertain input parameters to which the model's outputs are most sensitive. From Section 2, it is recalled that the major transport mechanisms for the removal of atmospheric  $CO_2$  by the oceans are: 1) the air-to-sea transfer across the surface, 2) the buffering by the oceanic mixed-layer, and 3) the mixing of dissolved carbon into the deep sea. The OCM addresses these three "solubility pump"  $CO_2$  sink processes in the following way: 1) a wind-speed dependent piston velocity that drives the air-sea exchange over the interface; 2) explicit inorganic carbon chemistry in the oceanic mixed layer that converts additional dissolved  $CO_2$  into dissolved inorganic carbon; and 3) the diffusion of dissolved inorganic carbon from the mixed layer to depth.

The six parameters discussed below are needed to quantify these processes and from previous sensitivity studies are found to directly impact on the size of the oceanic sink for carbon. Because these six parameters have not up until now been well defined, exact estimates of their probability distributions are not available; their PDFs must be qualitatively specified. Such specification is made based on a combination of the current state of knowledge and expert judgment. The principle chosen to guide the specification of these PDFs is that they be broad and capture as much of the uncertainty supported by theory or observations so that the maximum reasonable parametric uncertainty can be examined. The PDFs of the six uncertain parameters are specified below.

# a) $K_v$ — The Coefficient of Vertical Diffusion

As a standard probability distribution for  $K_v$  we use that constructed by Webster and Sokolov (2000) based on the values needed to fit the transient responses of different coupled atmosphereocean GCMs to increasing CO<sub>2</sub> concentration (Sokolov and Stone, 1998). The PDF for  $K_v$  is a beta distribution with parameters  $\alpha = 2.72$  and  $\beta = 12.2$ . This distribution (see Figure 8) has a median of 2.5 cm<sup>2</sup>s<sup>-1</sup> and 95% fractiles of 0.5 and 12.5 cm<sup>2</sup>s<sup>-1</sup>. Since, as discussed later in the paper, uncertainty in the vertical mixing plays a dominant role in defining an overall uncertainty in an oceanic carbon uptake, we will also present results obtained with an alternative distribution.

# b) $K_{H}$ — The Coefficient of Horizontal Diffusion

The values for the horizontal diffusion coefficient in the upper ocean were obtained by Stocker *et al.* (1994) "assuming a circulation of 40 Sv in a latitude band between 20° and 40° latitude over 60° angular width and 500 m depth." This assumption leads to  $K_H \approx 3 \times 10^4 \text{ m}^2 \text{s}^{-1}$ . As mentioned above this value is close to that obtained under assumptions that inter-hemispheric recirculation time for the top layer of the ocean is equal to 25 years (Sokolov *et al.*, 1998). There are, however, significant uncertainties in the strength of the ocean circulation. The probability density function for horizontal diffusion coefficient used in this study is a beta distribution with  $\alpha = 8.3$  and  $\beta = 41.5$ . It has mean of 42 Sv and a standard deviation of 13 Sv, and the 2.5% and 97.5% fractiles are 20 and 70 Sv respectively. Such a distribution provides a reasonable representation of uncertainty in the strength of ocean circulation.

# c) $V_p$ — The Piston Velocity

The chosen piston velocities  $(V_p)$  should be consistent with the current measured global average carbon uptake rate of  $20 \pm 3$  moles of CO<sub>2</sub> per m<sup>2</sup>yr<sup>-1</sup>. Tans *et al.* (1990) (TTF) point out that the factor calculated by Liss and Merlivat (1986) (LM) to calculate the transfer velocity for a given difference in partial pressure of CO<sub>2</sub> between the atmosphere and ocean is easily uncertain to within a factor of two. Further works by Wanninkhof (1992) and Wanninkhof and McGillis (1999) (WM) demonstrate a significantly stronger dependence at higher wind speed (cubic) than LM relationships, leading to higher uptake rates (see **Figure 7**).

For this uncertainty calculation we use the estimate of Tans *et al.* (1990) (which is intermediate between LM and WM) as a reference value, a factor 2 and 0.5 times the reference is assumed to contain about 67% of the probability. Because the  $V_p$  cannot be negative, the beta distribution is



Figure 7. Transfer Velocity Dependence on Wind Speed

also chosen and constructed assuming a mode of 1 (the reference value) as a multiplier on the value of  $V_p$ . The asymmetry assumed above indicates that the probability that the standard piston velocity multiplier is less than 1 is  $\frac{1}{3}$  and greater than 1 is  $\frac{2}{3}$ . The multiplier is assumed to range between 0 and 5 with a 95% probability that it is less than 3.

### d) $K_0$ — The Henry's Law Coefficient

The Henry's law coefficient is highly temperature dependent and the normal distribution is chosen to characterize its PDF. Reference values in this PDF are taken from Weiss (1974), which at 20° C and 35 % salinity gives a value of pK<sub>0</sub> (-log10[K<sub>0</sub>]) of approximately 3.2. A standard deviation of ± 0.1 pK<sub>0</sub> is assumed normally distributed from this reference value.

e) K<sub>1</sub> and K<sub>2</sub> — The First and Second Disassociation Constants of Carbonic Acid

 $K_1$  and  $K_2$  are strongly temperature dependent, as well as dependent on salinity and pressure. Thevalues for these parameters have been measured empirically in a handful of studies (Mehrbach, 1973; Hansson, 1979; Goyet and Poisson, 1989; Roy *et al.*, 1993; Lee and Millero, 1995). A study of the uncertainty in these values performed by Lee and Millero (1995) show that most of these measured  $K_1$  and  $K_2$  values do a reasonable job of fitting the observations of quantities such as dissolved inorganic carbon, alkalinity, and pH with standard deviations in the range of 0.01 to 0.04 pK units. The values of these coefficients in this study are assumed normally distributed. The parameters are assumed to have a mean in the middle of the measured values of  $pK_1 = 5.89$ ,  $pK_2 = 9.04$  at 20° C and 35‰ with standard deviations of  $\pm 0.05$  pK units (larger than the above standard deviation estimates that focused on data from the North Atlantic to account for the accumulation of errors when zonally-averaged temperature values are used to determine the coefficients).

#### 4.3 Uncertain Response of the OCM

The PDFs for the six uncertain parameters (**Figure 8**) are used to generate the polynomials and collocation points. A third-order fit with cross-terms in all six of the parameters is chosen to approximate the outputs of the Ocean Carbon Model, requiring a total of 74 runs of the OCM. The PCM approximations were constructed for five-year averages of the oceanic carbon sink and the atmospheric  $CO_2$  concentration for the length of the simulation.



Figure 8. Probability Distributions of the Uncertain Parameters in the OCM

A third order approximation with cross-terms appears to do a very good job of fitting the output of the OCM. There are quantitative means of checking the performance of the reduced model. One such variable is the index of agreement, d (Wilmott, 1982) which measures the quality of the approximation by summarizing normally calculated indexes such as the root mean square error, the mean absolute error, the intercept and the slope of the least-squares regression among others. Specifically:

$$d = 1 - \left(\frac{\sum_{i=1}^{N} (y_{M}^{i} - y_{A}^{i})^{2}}{\sum_{i=1}^{N} (|y_{A}^{i} - \overline{y}_{M}| + |y_{M}^{i} - \overline{y}_{M}|)^{2}}\right)$$
(8)

where *N* is the number of runs used to check the error,  $y_M$  is the true model result and  $y_A$  is the approximated model result. The closer to unity that *d* is, the better the approximation. Here *d* is calculated for each of the output variables using 50 runs of the parent and PCM model using randomly chosen input parameters from the PDFs in Figure 8. We find *d* ranges between 0.92 and 0.99 for the atmospheric CO<sub>2</sub> concentration between 1990 and 2100 and between 0.91 and 0.99 for

the oceanic carbon sink between 1860 and 2100. Such a good agreement indicates that the PCM-based reduced-form models do a very effective job of capturing the variability of the real parent model. **Figure 9** shows the accuracy of the fit of atmospheric  $CO_2$  concentrations in the year 2000 by plotting the results obtained from runs of the OCM and the PCM approximation for the same 50 randomly chosen sets of parametric inputs and indeed they show that the PCM approximations and the actual OCM model are in very good agreement. Looking beyond the year 2000, knowing that the uncertainty in the outputs is increasing as a function of time, the same experiment is conducted in the years 2050 and 2100 and the comparisons are plotted in **Figure 10**.



**Figure 9**. Accuracy of the PCM Forecasts of Atmospheric CO<sub>2</sub> Concentrations in 2000 Relative to the OCM Forecasts



Figure 10. Accuracy of the Forecast of Atmospheric CO<sub>2</sub> Concentrations in 2050 and 2100

The reduced-form model still does a reasonably good job in approximating the output of the real model as a function of time after 2000. However, it is evident in Figures 9 and 10 that agreement is better in the regions of high probability for the uncertain input parameters (and hence high probability for the outputs) than further out in the tails of the distributions. As pointed out in Section 4.1, this is expected from the way the PCM approximation is constructed as it is meant to be most accurate in the high probability regions of the input parameters.

The results of the approximation between 1860 and 1990 are displayed in **Figure 11**. Plotted are the mean and confidence intervals that represent two standard deviations of the ocean carbon sink calculated by the PCM approximation as a function of time.

The trend in the mean ocean carbon sink determined by the PCM approximation is qualitatively similar to that obtained when the parent model was spun up with the reference parameter values. However, the mean oceanic carbon sink in the 1980s estimated by the PCM approximation is



**Figure 11**. Mean (solid-line) and Two Standard Deviation (dotted-line) of the PCM Approximation of the Historical Oceanic Carbon Sink (Pg yr<sup>-1</sup>)

2.06 Pg yr<sup>-1</sup>, which is 6.2% higher than in the reference run noted in **Table 1** of 1.94 Pg yr<sup>-1</sup>. The standard deviation of the PCM estimates which grows with time, averages about 0.92 Pg yr<sup>-1</sup> in the 1980s, giving a range of values for the oceanic carbon sink of 1.14 to 2.98 Pg yr<sup>-1</sup> (with 67% confidence). The IPCC (1994) range for the 1980s oceanic carbon sink from intermodel comparisons is significantly smaller and is estimated at 1.4 to 2.6 Pg yr<sup>-1</sup> with a subjective estimate of confidence of 90% (Siegenthaler and Sarmiento, 1993).

A Monte Carlo simulation based on 10,000 runs with the reduced-form model was performed to generate a discrete representation of the PDF of the oceanic sink for the 1980s (**Figure 12**). This PDF covers a broad range of values with over 90% of the probability contained between 0.0

Model	OCM (2-D)	Box-Diffusion (1-D)	Princeton OGCM	HILDA (2-D)	Hamburg HAMOCC-3	Stocker 2-D OGCM
Author(s)	(This Paper)	Oeschger et al. (1975)	Sarmiento et al. (1996)	Siegenthaler & Joos (1992)	Maier-Reimer et al. (1996)	Stocker et. al. (1994)
Mean Ocean Carbon Sink for 1980s (Pg yr <sup>-1</sup> )	1.94	2.32	2.2	2.15	1.6	2.10
Cumulative Carbon Sink 1770-1980 (Pg)	99.4	123	N/A	107	N/A	100

Table 1. Comparison of the OCM Carbon Uptake to Other Models

and 4.0 Pg yr<sup>-1</sup>, indicating that the uncertainty in the carbon sink derived from the uncertainty in the input parameters for this model has exceeded that of the IPCC (1994) range. However, we emphasize that balancing the carbon cycle in the 1980s requires that the sum of the oceanic and terrestrial sinks total about 4 Pg yr<sup>-1</sup>; a low estimate of the oceanic sink demands a high estimate of the terrestrial sink and vice versa.

# 4.4 Forecasting Atmospheric CO<sub>2</sub> Concentrations Under Uncertainty



**Figure 12**. *Histogram of 10,000 Monte Carlo Runs for the Approximation for the mean 1980s Oceanic Carbon Sink (in Pg yr<sup>-1</sup>)* 

Using the same emissions (fossil fuel +

deforestation) scenario as for the reference forecast in Section 3.2, the PCM is used to approximate the atmospheric  $CO_2$  concentration at 5 year intervals between 1990 and 2100. The mean value of the approximation and intervals containing two standard deviations are plotted in **Figure 13**.



**Figure 13**. Mean (solid-line) and 95% Confidence Intervals (dotted-lines) of the *PCM Approximation of the Atmospheric CO*<sub>2</sub> Forecast: 1990-2100

The mean atmospheric  $CO_2$  forecast using the PCM predicts slightly lower atmospheric  $CO_2$  concentrations than the reference run, though the latter clearly falls well within the uncertainty of the  $CO_2$  forecast using the PCM. The atmospheric  $CO_2$  concentration forecast in 2100 is 659 ppm compared to 661 ppm when the model is run with reference values. As one might expect, the uncertainty in the atmospheric  $CO_2$  concentration forecast by the model grows with time. The standard deviation is 35.4 ppm by 2100 which indicates an uncertain range of over 70 ppm (95% confidence) in the forecast atmospheric  $CO_2$  concentration in 2100 from the solubility sink in the ocean alone (assuming a normal distribution).

Looking specifically at the forecasts of atmospheric  $CO_2$  for 2000, 2050, and 2100 a Monte Carlo simulation of 10,000 runs was carried out using the PCM reduced-form model to produce discrete PDFs for  $CO_2$  mole fractions (**Figure 14**). As expected, the discrete PDFs broaden as a



**Figure 14**. Histograms of 10,000 Monte Carlo Runs of the Approximation of Atmospheric CO<sub>2</sub> Concentrations in 2000, 2050, and 2100

function of time due to an accumulation of uncertainty. The extent of the uncertainty in atmospheric  $CO_2$  concentrations is dependent on the assumed uncertainty in the input parameters. For the uncertainty assumptions in Section 4.1, the range covered by two standard deviations in the atmospheric  $CO_2$  concentration by 2100 is almost 150 ppm.

The range of the uncertainty in atmospheric  $CO_2$  concentrations is actually smaller than it might have been absent the assumption of an atmospheric  $CO_2$ -dependent terrestrial sink (described in Section 3). Looking at the uncertainty in the variable component of the  $B_t$  term (NEP) in the carbon budget (from equations 6 and 7) which depends on the atmospheric  $CO_2$  concentration, the PCM estimates of the  $CO_2$  mole fraction uncertainty can be used to estimate the resultant uncertainty in the terrestrial ecosystems sink calculated by the TEM. By 2100, the mean terrestrial carbon sink grows to over 3.9 Pg yr<sup>-1</sup> and the variable component (about 2.15 Pg yr<sup>-1</sup>) has an uncertainty of 0.39 Pg yr<sup>-1</sup> (with 95% confidence) due to the uncertainty of the  $CO_2$  predictions (**Figure 15**), which are in turn due to the uncertainty in the oceanic sink for carbon. Because the size of the terrestrial carbon sink increases with increasing atmospheric  $CO_2$  concentrations it acts as a negative feedback to the growth in atmospheric  $CO_2$  concentrations. When atmospheric  $CO_2$ 



**Figure 15**. Uncertainty in the Terrestrial Sink (Pg yr<sup>-1</sup>) Implied by Uncertainty in the OCM CO<sub>2</sub> Mole Fraction Predictions

concentrations rise faster (because of a smaller oceanic sink driven in our study by changes in OCM parameters) the terrestrial sink increases faster which slows the increase in atmospheric  $CO_2$  concentrations. Conversely, when atmospheric  $CO_2$  concentrations rise slowly because of a fast growing oceanic sink, the terrestrial sink must rise more slowly, increasing atmospheric  $CO_2$  concentrations. This represents a decrease in the uncertainty in atmospheric  $CO_2$  concentrations relative to an assumption of a fixed terrestrial sink for carbon because of interactions between the two sinks.

### 4.5 Relative Importance of the Uncertain Parameters

The prescribed uncertainty of the six parameters chosen for this study is responsible for the uncertainty calculated in the oceanic carbon sink and hence in the atmospheric  $CO_2$  concentration. However, not all of the chosen parameters contribute to this uncertainty equally and their relative impacts change as a function of time. The PCM method used in this study provides not only the mean and standard deviation (assuming a normal distribution) of the approximated output variables, but also the relative contribution of each parameter to the variance in that output variable. For example, looking at the value of the mean oceanic carbon sink for the 1980s and 2100, the relative contribution of each parameter to the total variance is given in **Table 2**.

Of the six parameters, the coefficient of vertical diffusion clearly dominates all of the others in terms of its contribution to the variance in the output variable. This is due to the fact that the surface mixed layer of the ocean is usually close to equilibrium with the atmosphere so that the major control on whether more carbon can be taken up by the ocean is the amount exported to depth from the mixed-layer. The only other parameter with a significant contribution is the piston velocity, which controls the air-to-sea exchange rate. For positive gradients into the ocean, this factor determines the size of the oceanic carbon sink that drives the ocean back to a steady-state with the atmosphere. The effects of uncertainty in the three chemical parameters are small due in large part to the relatively high accuracy with which these parameters are known. The importance of the coefficient of vertical diffusion increases from 83 to 97% of the total variance from the 1980s to 2100 while the relative importance of the piston velocity collapses from just over 15% in the 1980s to less than 1% of the total variance by 2100. This is a result of the large gradient between the atmosphere and ocean which dominates the carbon flux in the future relative to the surface exchange rate which is not limiting. The importance of all three of the chemical parameters,  $K_0$ ,  $K_1$ , and  $K_2$  steadily increases as a function of time, but they remain small even in 2100. This is due in part to the effect of a changing effective "Revelle Factor" as the carbon sink becomes increasingly sensitive to the chemistry of the surface water. More dissolved carbon is held as

Parameter	1980s	2100
Kv	83.4	96.7
Kh	00.2	00.4
Vp	15.2	00.7
K0	00.0	00.0
K1	00.1	01.5
K2	00.0	00.7

Table 2. Relative Contributions of the Uncertain Parameters to the Variance in the Carbon Sink

dissolved  $CO_2$  rather than as  $HCO_3^2$  and  $CO_3^2^2$ . But uncertainty in these chemical parameters is small and does not appear to significantly contribute to uncertainty in future atmospheric  $CO_2$  concentrations on time-scales of decades to centuries.

#### 4.6 Sensitivity of the Results to Parametric Specification

Because the accuracy of the PCM-based approximation to the actual model is very good (as demonstrated in Section 4.3) our confidence in the uncertainty analysis for an oceanic carbon sink model is not so much dependent on the possible deficiency of the probabilistic collocation method as on the robustness of the input parameter probability distributions used to perform the analysis. As noted in Section 4.1, the true probability distributions of some of inputs chosen in this study are not very well known. We must therefore test for the possibility that the results are strongly sensitive to our assumptions about the prescribed uncertainty in our chosen parameters.

Because the uncertainty in the coefficient of vertical diffusion,  $K_v$ , dominates the total uncertainty in the output of the model, we focus on how sensitive the results are to our assumptions about the relative certainty of this parameter. Increase in the mean of the PDF, holding the standard deviation fixed, increases  $CO_2$  uptake and reduces atmospheric concentrations and vice versa. A 25% decrease in the mean of the PDF of  $K_v$  decreases the mean oceanic sink calculated in the 1980s from  $2.06 \pm 1.81$  Pg yr<sup>-1</sup> (95% confidence) to  $1.93 \pm 1.71$  Pg yr<sup>-1</sup>. Conversely, a 25% decrease in the mean of the distribution of  $K_v$  increases the mean oceanic sink in the 1980s to  $2.20 \pm 1.82$  Pg yr<sup>-1</sup>. While the mean uptake of carbon changes, the relative uncertainty is as expected almost unaffected for modest changes in the mean of the parameter.

As an additional test we constructed an alternative distribution for the vertical diffusion coefficient with the same mean, but one-half the standard deviation of the reference distribution, thereby changing its overall variance. The PDFs for the alternate the reference distributions are compared in **Figure 16**. Holding the uncertainty in all of the other parameters at their reference values, the PCM analysis is performed again and fits are made to the desired outputs: the oceanic sink and the atmospheric CO<sub>2</sub> concentration. The reference fit and the fit with the alternate distribution are plotted in **Figure 17**. The atmospheric CO<sub>2</sub> estimated in 2100 (at 95% confidence)



**Figure 16**. Alternative PDF (dotted-line) and Reference PDF (solid-line) for the Vertical Diffusion Parameter,  $K_v$ 



**Figure 17**. Comparing the Uncertainty in Future Atmospheric CO<sub>2</sub> Concentrations between the Alternative (red) and the Reference (black) PDFs

is  $657 \pm 43$  ppm compared with  $659 \pm 70$  ppm in the reference case. As expected, there is an overall reduction (38.6 %) in the uncertainty of the forecast due to the increased certainty in the vertical diffusion parameter.

By changing the PDF of  $K_v$ , an impact is made upon the relative importance of the six uncertain parameters chosen for the analysis, particularly of course on the coefficient of vertical diffusion,  $K_v$ , which decreases from nearly 80% to almost 50% in 1990 as shown in **Figure 18**. The reduction in uncertainty in the coefficient of vertical diffusion has resulted in an increase in the relative importance of the air-sea transfer coefficient,  $V_p$  in 1990 from 10% to 30% in determining the uncertainty of the oceanic sink which is  $2.06 \pm 1.81$  Pg yr<sup>-1</sup> (95% confidence) for the 1980s in the reference case and  $2.08 \pm 1.16$  Pg yr<sup>-1</sup> in the alternative one. But this increased importance is not maintained over the length of the calculation and the importance of the piston velocity once again becomes negligible by 2100. The importance of the vertical diffusion coefficient increases as before to dominate the other parameters. This indicates that while specific parameter choices can be important for calibrating the model during the period of observation in the 1980s, the mechanism most important in determining the uncertainty in future oceanic uptake is the mixing into the deep ocean, here parameterized by vertical diffusion.



**Figure 18**. Relative Importance of the Vertical Diffusion Parameter  $(K_v)$  and the Exchange Coefficient  $(V_v)$  between the reference (solid-line) and alternative (dotted-line) distributions

#### 5. CONCLUSIONS

In this paper a parameterized reduced-form model of the oceanic sink for carbon has been constructed for use in uncertainty studies of the contemporary global carbon budget and for forecasting future atmospheric CO<sub>2</sub> concentrations in climate change simulations. The Ocean Carbon Model (OCM) determines the global sink for CO<sub>2</sub> in the ocean by means of the so-called "solubility pump" mechanism including air-to-sea exchange, dissolution in the surface mixed-layer, and diffusive mixing of dissolved carbon to sequestration at depth. The marine organic carbon cycle is not included in this simple model. Using reference parameter values, the model is initialized by being driven to a steady-state with a pre-industrial atmospheric CO<sub>2</sub> record to generate a current oceanic carbon sink of 1.9 Pg yr<sup>-1</sup>, consistent with the results of other oceanic carbon sink models. The model is used to forecast atmospheric CO<sub>2</sub> concentrations into the next century by closing the current global carbon budget with the addition of an atmospheric CO<sub>2</sub> concentration of the land sink for carbon estimated from the Terrestrial Ecosystems Model (Xiao *et al.*, 1997) and driving it into the future with an anthropogenic emissions prediction from a global economic model (Prinn *et al.*, 1999).

The two-dimensional OCM has been developed with an acknowledgment of the parametric uncertainty inherent in such models. This study explores that uncertainty quantitatively by examining the uncertainty in the outputs resulting from specified uncertainty in the input parameters through the application of the Probabilistic Collocation Method (PCM; Tatang *et al.*, 1997). The PCM makes it possible to efficiently quantify the uncertainty in the model's most important outputs: the global oceanic carbon sink and future atmospheric  $CO_2$  concentrations. The method requires specifying the probability distributions of the input parameters. For the OCM, the air-to-sea piston velocity, the Henry's Law coefficient, the first and second dissociation constants for carbonic acid, and the vertical and horizontal diffusion rates are the parameters chosen to represent the uncertainty in the model's primary carbon sink mechanisms.

For a reasonable range of uncertainty in these six parameters, the mean oceanic carbon sink for the 1980s is calculated to be  $2.06 \pm 1.8$  Pg yr<sup>-1</sup> (with 95% confidence). When compared to the IPCC range for the uncertainty of the oceanic carbon sink of  $2.0 \pm 0.8$  Pg yr<sup>-1</sup> (1992, 1994, 1995), it is demonstrated that the parametric uncertainty in this oceanic carbon sink model spans almost twice this range. The IPCC range is due primarily to an undefined combination of parametric and structural uncertainties underlying differences between various oceanic carbon sink models, while the uncertainty in the output of the OCM is strictly parametric. The uncertainty in the oceanic sink and hence the atmospheric CO<sub>2</sub> concentration is shown to increase with time. For anthropogenic CO<sub>2</sub> emissions similar to the IS92a scenario of the IPCC (1992), the uncertainty in atmospheric CO<sub>2</sub> concentrations is found to be  $659 \pm 71$  ppm in 2100 (with 95% confidence). Like in the case of the oceanic carbon sink for the 1980s, the range of atmospheric CO<sub>2</sub> concentrations of 588 to 730 ppm by 2100 is much larger than that implied by the uncertainty presented in the IPCC study (1994, 1995). As discussed in Section 4, taking into account changes in the natural terrestrial carbon sink leads to a decrease in the uncertainty in the projected atmospheric CO<sub>2</sub> concentration.

Because it has been well established that atmospheric  $CO_2$  levels are a strong driver of radiative forcing in the atmosphere, this result has implications for quantifying uncertainty in studies of

future climate change. A range of atmospheric  $CO_2$  concentrations of nearly 150 ppm in 2100 is expected to produce discernible differences in global surface temperatures that may dwarf the impacts of certain modest policies to reduce or stabilize  $CO_2$  concentrations. The ability to produce probability distributions for atmospheric  $CO_2$  concentrations as a function of time from quantifiable uncertainties in the oceanic carbon sink allows for the testing of anthropogenic carbon emissions paths and scenarios for the stabilization of atmospheric  $CO_2$  concentration with some estimate of the likelihood that they can be achieved. Distributions of future atmospheric  $CO_2$  concentrations can then be run through global climate models in order to propagate the uncertainty in atmospheric  $CO_2$  concentrations through the climatic system to quantify impacts on key outputs of interest such as global temperature, precipitation, and sea-level rise. However, the relative quality of such results depends on the quality of the estimates of uncertainty in the input parameters, since the uncertainty in the oceanic carbon sink is directly keyed to the assumptions of uncertainty in the parametric inputs as is noted in Section 4.

Improving the quality of the probability distributions of the uncertain input parameters in the OCM is therefore important for decreasing the uncertainty in the size of the predicted oceanic carbon sink. The "solubility pump" is a well-recognized process in the ocean, although it has been difficult to quantify by observation since net fluxes are small compared to gross exchanges. By examining the contribution of the uncertainty in the parameters of the model in this study to the variance in the outputs, it has been possible to rank their relative importance. The three chemical parameters and the coefficient of horizontal diffusion appear to have relatively small impacts on the uncertainty of the oceanic carbon sink relative to the coefficient of vertical diffusion which accounts for 50% or more of the uncertainty. That the rate of vertical mixing into the deep ocean is determined to have the greatest effect on the uncertainty of the oceanic carbon sink, agrees with the observation that the surface ocean and atmosphere remain reasonably close to equilibrium and that the rate of export of dissolved carbon to the deep ocean should there fore control the size of the sink. This study also indicates that the importance of such transport increases with time. Much of the recent work into inferring the vertical mixing rate from tracer distributions such as bombproduced radiocarbon appears to be well justified (Broecker et al., 1985, 1995) since reductions in the uncertainty of the mixing rate will have the greatest impact on the total uncertainty of the oceanic carbon sink. The strength of the oceanic carbon sink can be expected to be strongly sensitive to the strength and nature of the mixing assumed in a particular oceanic carbon sink model, which might be parameterized as in this model, or dynamical as in various 3-D Ocean GCMs. The strong sensitivity of the carbon sink computed by our OCM to the choice of the vertical diffusion parameter points up a major problem with the reasonableness of using a simple fixed diffusive (or fixed advective-diffusive) ocean carbon sink model to predict future carbon concentrations since this approach is calibrated only to reproduce the current rate of mixing into the ocean. These uncertainties suggest caution when interpreting the results of individual models of the oceanic carbon sink as they exist now, since critical gaps in knowledge and a disparity in the degree of complication make the effectiveness of such models difficult to compare without doing so in conjunction with an analysis of the parametric uncertainty of each model.

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