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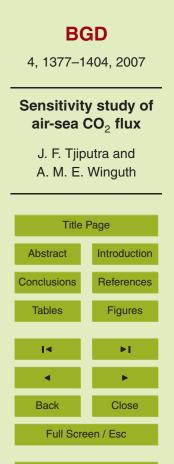
# Adjoint sensitivity of the air-sea CO<sub>2</sub> flux to ecosystem parameterization in a three-dimensional global ocean carbon cycle model

#### J. F. Tjiputra and A. M. E. Winguth

Department of Atmospheric Oceanic and Space Sciences, University of Wisconsin, Madison, Wisconsin, USA

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Correspondence to: J. F. Tjiputra (jftjiputra@wisc.edu)



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#### Abstract

The regional sensitivity of air-sea  $CO_2$  flux to ecosystem components and parameters in a three-dimensional ocean carbon cycle model is estimated using an adjoint model. Adjoint sensitivities to the global air-sea  $CO_2$  flux reveal that the biological component

- <sup>5</sup> of the model is significant in the high latitudes of both hemispheres and in the Equatorial Pacific. More detailed analysis indicates that zooplankton grazing activity plays a major role in the carbon exchange in the above regions. The herbivores' ingestion parameter in the model regulates the flux of remineralized (i.e. regenerated) biogenic nutrients; thus, substantially controls the biological production and the concentration of
- <sup>10</sup> dissolved inorganic carbon (*DIC*) in the euphotic zone. Over a 10-year period, reducing the herbivores' ingestion parameter in the model by 25% could increase the global uptake of atmospheric carbon by 6 Pg C. Thus, climate induced changes in the marine ecosystem structure are of importance for the future uptake of atmospheric CO<sub>2</sub>.

#### 1 Introduction

- <sup>15</sup> Carbon dioxide is an important anthropogenic greenhouse gas and its concentration in the atmosphere increased from about 280 ppm in the pre-industrial era to 379 ppm in 2005 (Intergovernmental Panel of Climate Change (IPCC), Summary for Policymakers, 2007, http://www.ipcc.ch). This number exceeds the natural ranges over the past 650 000 years and is expected to keep increasing in the future due to continuous fossil
- <sup>20</sup> fuel burning and land use changes, such as deforestation. Atmospheric carbon dioxide will eventually be distributed into three main carbon reservoirs: the atmosphere, the ocean, and the terrestrial biosphere, with the ocean being the largest of the three. Sabine et al. (2004) have estimated that nearly half of the emitted anthropogenic  $CO_2$ has ended up in the ocean. Within the euphotic zone (i.e. the ocean surface layer with
- <sup>25</sup> sufficient light), carbon exchange between the atmosphere and ocean is determined by physical solubility of CO<sub>2</sub> and by marine photosynthesis. These mechanisms, first in-

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troduced by Volk and Hoffert (1985), are known as the solubility and biological pumps. Seawater solubility of  $CO_2$  increases with decreasing sea surface temperatures (Weiss, 1974). The biological pump is divided into soft tissue and calcium carbonate pumps, which are controlled by the vertical flux of particulate organic carbon and inorganic cal-

- <sup>5</sup> careous shell material into the deep sea, respectively. The soft tissue pump transfers carbon formed by photosynthesis from the surface to the deep sea, which decreases the surface  $pCO_2$  and increases uptake of  $CO_2$  from the overlying atmosphere. On the other hand, the calcium carbonate pump transfers carbonate ions from the surface to the deep sea, which decreases the alkalinity and increases the  $pCO_2$  at the surface.
- Many model studies have shown the importance of marine biology in lowering atmospheric CO<sub>2</sub> (e.g. Najjar et al., 1992; Siegenthaler and Sarmiento, 1993; Maier-Reimer et al., 1996; Sarmiento et al., 1998; Archer et al., 2000; Plattner et al., 2001; Takahashi et al., 2002; Bopp et al., 2003; Gloor et al., 2003; Cameron et al., 2005; Le Quéré et al., 2005; Wetzel et al., 2005; Winguth et al., 2005), but relatively few studies have been
   conducted to analyze in detail the sensitivity of air-sea CO<sub>2</sub> flux to perturbations of different ecosystem components or processes (Sarmiento and Orr, 1991; Gnanadesikan et al., 2003; Zeebe and Archer, 2005; Dutkiewicz et al., 2006; Parekh et al., 2006). Of the above studies, only Dutkiewicz et al., (2006) applied the adjoint model to estimate
- the above studies, only Dutkiewicz et al. (2006) applied the adjoint model to estimate such sensitivity.
  20 Here, we will attempt to estimate and examine the sensitivity of air-sea CO<sub>2</sub> flux to
- perturbations of ecosystem components and parameters in a three-dimensional global ocean carbon cycle model using the adjoint approach. The adjoint model is applied to efficiently produce a global map of sensitivity of atmospheric  $CO_2$  uptake with respect to changes of regional ecosystem parameters and components. The sensitivity map
- will be applied to analyze the role of different ecosystem processes (e.g. productivity, grazing, remineralization, etc.) in regulating atmospheric  $CO_2$  concentrations. The manuscript is designed as follow: Sect. 2 briefly describes the forward and adjoint models used in this study. Section 3 explains the model interpolation and sensitivity experiment results. The paper concludes in Sect. 4.

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#### 2 Forward and adjoint model description

This study employs the Hamburg Ocean Carbon Cycle (HAMOCC5) model, which uses monthly climatology forcing and is based on studies by Maier-Reimer (1993), Six and Maier-Reimer (1996), and Aumont et al. (2003). The model has a 3.5°×3.5° horizontal resolution and 22 vertical layers with thickness varying from 50 m at the surface to 700 m in the deepest layer. The geochemical tracers in the model are advected with a time step of one month, while a faster time step of three days is used in the euphotic zone (i.e. the upper two layers of the model) to resolve ecosystem and air-sea gas exchange processes. The marine productivity is controlled by macronutrients (phosphate, nitrate, and silicate) and micronutrients (dissolved iron). The biological pump is comprised of organic and inorganic export out of the two uppermost model layers, which correspond to a mean euphotic zone with a depth of 107 m. The model is able to simulate the main characteristics of ocean biogeochemistry and resolve some of the High Nutrient-Low Chlorophyll (HNLC) regions in the world by implementing iron lim-

- itation productivity. For example, it is capable to simulate the distribution of the main biochemical tracers in the ocean including phosphate, nitrate, iron, dissolved inorganic carbon, alkalinity, and radiocarbon. Compared to the more computationally expensive, high-resolution ocean model, this model allows us to efficiently apply the adjoint technique in order to study the large-scale pattern of the biogeochemical state in the entire
   water column. Due to the relatively short integration time scale (ten years) of the ex-
- periments, the sediment module is simplified by introducing a sink-only sediment layer. A more detailed description of the model can be found in Howard et al. (2006).

In this experiment, the model run is started in the year 1995 and integrated through 2005, while maintaining averaged atmospheric CO<sub>2</sub> concentration at 375 ppm. The seasonal cycle of air-sea CO<sub>2</sub> flux in the high and low latitudes at different ocean locations (e.g. Bermuda Atlantic Time-Series Study (BATS), Hawaii Ocean Time-Series (HOTS) and Southern Ocean, 56° N, 275° W) simulated by the model fit reasonably well with the observation compiled by Takahashi et al. (2002) as shown in Fig. 1. However,

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a phase lag exist in the Southern Ocean, which may be associated to interannual variations, deficiencies in the model, and the sparse data in this location. The average annual air-sea  $CO_2$  flux for this period is also shown in Fig. 1 together with interpolated averaged observations from Takahashi et al. (2002), which is based on 940 000 pCO<sub>2</sub>

- <sup>5</sup> measurements in surface mixed-layer and corrected to the wind speeds at 10 m (http: //www.ldeo.columbia.edu/res/pi/CO2/). In general, the model agrees reasonably well with the data with common features including efflux in the equatorial oceans due to upwelling of cold, CO<sub>2</sub>-rich deep water and uptake in the subtropical gyres and high latitude oceans.
- The adjoint model allows us to efficiently estimate the sensitivity of model output (i.e. air-sea CO<sub>2</sub> flux) with respect to small perturbations of model input or control parameters. In contrast to finite perturbation method, adjoint sensitivity are equivalent to thousands of forward perturbation runs with only a fraction of the computational expense by running one forward and one adjoint model integration. Here, the adjoint codes have been generated by the TAMC (Tangent linear and Adjoint Model Compiler) (Giering and Kaminski, 1998; www.fastopt.com) and improved manually to be com-
- putationally efficient. The accuracy of the adjoint code has been tested using a finite difference approximation. The cost function ( $J_{FCO2}$ ) is defined as the global air-sea  $CO_2$  flux ( $F_{CO2}$ ), and is based on the Wanninkhof (1992) formulation integrated over all ocean surface area for a period of ten years (from 1995–2005):

$$J_{FCO2} = \iint F_{CO2} dA dt \tag{1}$$

$$F_{\rm CO2} = k(p\rm CO_{2(air)} - p\rm CO_{2(sea)})$$
<sup>(2)</sup>

where *k* is the gas transfer velocity, and  $pCO_{2(air)}$  and  $pCO_{2(sea)}$  represent the partial pressure of  $CO_2$  in the atmosphere and in the surface ocean, respectively. *A* represents the area of the specific model grid and *t* represents time integration. The sensitivity map in this study is produced by using the adjoint model to estimate the partial derivative of the cost function to a small perturbation of a specific control variable,

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#### 3 Sensitivity experiments

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#### 3.1 Seasonal variability of pCO<sub>2</sub>

The change of the partial pressure of CO<sub>2</sub> in the surface ocean depends on the *DIC* concentration, temperature, alkalinity, and the salinity of the water (Le Quéré et al., 2000; Wetzel et al., 2005).

$$\frac{dpCO_2}{dt} = \frac{\partial pCO_2}{\partial D/C} \frac{dD/C}{dt} + \frac{\partial pCO_2}{\partial T} \frac{dT}{dt} + \frac{\partial pCO_2}{\partial TALK} \frac{dTALK}{dt} + \frac{\partial pCO_2}{\partial S} \frac{dS}{dt}$$
(3)

Here, the seasonal cycle of surface pCO<sub>2</sub> due to each component in Eq. (3) is calculated separately. The adjoint model is applied to calculate the monthly partial derivative term in the equation (i.e.  $\frac{\partial pCO_2}{\partial DIC}$ ,  $\frac{\partial pCO_2}{\partial T}$ ,  $\frac{\partial pCO_2}{\partial TALK}$ ,  $\frac{\partial pCO_2}{\partial S}$ ). Figure 2 shows the seasonal cycle of each component for different regions of world oceans according to Takahashi et al. (2002). The variability of the sum of all components has been confirmed using the differential of monthly output of the forward model.

In most regions, the seasonal variability of pCO<sub>2</sub> is controlled by the temperature and *DIC* concentration variability at the surface. There is a relatively small variability of pCO<sub>2</sub> caused by the alkalinity and salinity terms in all regions due to their small seasonal variability (Weiss et al., 1982). As expected, in the tropical regions (14° N–14° S), the seasonal variability is relatively small, except in the Indian Ocean, which may be a result of seasonal monsoon events. The temperature variability is more apparent in the low latitudes. The *DIC* term in the tropical regions is relatively small because the biol-

ogy and advection processes, which control the *DIC* variability in these regions, almost cancel each other out (see below). In the subtropical regions, where warm tropical water meets colder high latitude water, the temperature and *DIC* terms are of the same magnitude. In the high latitude (north of 14° N and south of 14° S) the pCO<sub>2</sub> variability

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is dominated by the *DIC* term, especially in the Southern Ocean. This analysis is consistent with regional (North Atlantic) studies by McKinley et al. (2004) and Takahashi et al. (2002). The prescribed surface temperatures in the model consider factors from advection, shortwave and longwave radiation, and latent and sensible heat.

#### 5 3.2 Seasonal variability of *DIC*

The change of *DIC* concentration in the model is proportional to the sources and sinks combination of advection (i.e. advection and diffusion), biological production, fresh water fluxes, air-sea exchange, and convective mixing.

$$\frac{dDIC}{dt} = J_{\text{add}} + J_{\text{bio}} + J_{fW} + J_{FCO2} + J_{\text{con}}$$
(4)

<sup>10</sup> The biological production term in the model is further comprised of:

$$J_{\text{bio}} = -r(T, L) \cdot P \cdot \frac{N}{N + N_o} + P(\text{CaCO}_3) + g \cdot Z \cdot \varepsilon_{\text{her}} \cdot (1 - \text{zinges}) \cdot$$

$$\frac{(P - P_{\min})}{P + P_o} + d_o \cdot \frac{N}{N + k_{\text{doc}}} \cdot \text{DOC} + l_o \cdot \text{POC}$$

$$r(T, L) = \frac{f(T) \cdot f(L)}{\sqrt{f(T)^2 + f(L)^2}} \cdot \frac{\text{Fe}}{(\text{Fe} + k_{\text{Fe}})}$$
(6)

$$P(CaCO_3) = A(P_{orgC} - 0.5 \cdot P_{BSi})$$
(7)

<sup>15</sup> All the notations in Eqs. (5), (6), and (7) are described in Table 1. The terms in Eq. (5) represent primary production, calcium carbonate production, and remineralization of the following: non-assimilated zooplankton (Z) grazing, dissolved organic carbon (DOC), dead zooplankton, and particulate organic carbon (POC). Primary production depends on regionally varying growth rates and nutrient concentration

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(5)

 $N = \min(PO_4, NO_3)$ . The growth rate of phytoplankton (P) is a function of light, temperature and dissolved iron as shown in Eq. (6), and calcium carbonate production is a function of organic production and silicate concentration in the euphotic layer. A more complete description of the ecosystem processes in the model is available at Tjiputra <sup>5</sup> et al. (2007).

Figure 3 shows the seasonal cycle of each component in Eq. (4) for different ocean regions. In almost all regions, the biology term has a stronger seasonal amplitude than the other terms. The convective mixing term occurs seasonally in the mid and high latitude regions with stronger mixing during winter periods. Fresh water flux has
<sup>10</sup> little effect on the seasonal *DIC* variability. In the tropical regions, the advection term, which is dominated by continuous upwelling, is as strong as the biological term. In the mid latitude regions, the *DIC* variability is dominated by the biological and convective mixing terms. In the high latitudes, the variability of *DIC* is dominated by the biology term, consistent to study by Wetzel (2004).

<sup>15</sup> 3.3 Regional sensitivity of air-sea CO<sub>2</sub> flux with respect to the ecosystem components in the model

To investigate the biological contribution to the air-sea CO<sub>2</sub> flux in more detail, the adjoint model is applied to estimate the regional sensitivity map of global air-sea CO<sub>2</sub> flux to perturbations of the nutrient  $\partial J_{FCO2}/\partial N_{(i,j)}$ , phytoplankton  $\partial J_{FCO2}/\partial P_{(i,j)}$ , and zooplankton  $\partial J_{FCO2}/\partial Z_{(i,j)}$  components of the model. Both forward and the adjoint models are integrated for ten years with a monthly perturbation of nitrate (0.16  $\mu$ mol N/L), phosphate (0.01  $\mu$ mol P/L), zooplankton (1.27  $\mu$ mol C/L), phytoplankton (1.27  $\mu$ mol C/L), and iron (0.033 nmol Fe/L) concentration for each experiment. The sensitivities (or gradient) of the air-sea CO<sub>2</sub> flux to the perturbation of these components are plotted in Fig. 4.

The sensitivity runs show that, in general, the cost function is more sensitive to change of phosphate than nitrate by a factor of two (i.e. based on Redfield ratio P:N of 1:16). In most regions of the world, certain type of organism (e.g. cyanobacteria)



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can fix nitrate from atmospheric dinitrogen ( $N_2$ ) (Tyrrell, 1999). Nitrate concentration is crucial in the Eastern Equatorial Pacific and Northeast Pacific. The sensitivity map shows that phosphate is the dominant limiting nutrient in the tropical Atlantic regions. For example, over ten years period, the sensitivity map indicates high sensitivity of the

air-sea CO<sub>2</sub> flux to changes in phosphate in the eastern subtropical North Atlantic. This high sensitivity is especially due to high concentration of dust input in this region, which allow more efficient macronutrient uptake for phytoplankton growth. The atmospheric carbon uptake is more sensitive to phosphate perturbation in the tropical and subtropical regions than in the high latitude regions. This is due to the seasonal requirement
 of phosphate in the high latitudes whereas a year long supply of phosphate is needed

 of phosphate in the high latitudes whereas a year long supply of phosphate in the equatorial regions.
 In general, zeenlankton, grazing limits the growth of phytoplankton, thus

In general, zooplankton grazing limits the growth of phytoplankton, thus limits the uptake of atmospheric  $CO_2$  by photosynthesis. Figure 4c illustrates that reducing the zooplankton concentration in most ocean regions would increase the uptake of  $CO_2$  by

- <sup>15</sup> the ocean, with the strongest region being the Western Equatorial Pacific Ocean. This region has sufficient light and temperature for phytoplankton growth, but relatively low nutrient concentration. The consequence of reduced zooplankton concentration is the increase in average phytoplankton lifetime, as there are fewer predators, thus allowing more photosynthesis and uptake of surface *DIC*. Additionally, reducing the zooplankton is the increase in average of surface *DIC*.
- ton concentration also increases the carbon flux from phytoplankton to the POC and DOC components of the model and increases the concentration of regenerated nutrient flux. More detail role of the zooplankton activity in CO<sub>2</sub> uptake is further explored in the next section.

The air-sea CO<sub>2</sub> flux is sensitive toward phytoplankton addition in the tropical and high latitude regions. In the equatorial regions, adding more phytoplankton results in increase of *DIC* concentration, hence surface pCO<sub>2</sub>. To better analyze the process occurring in the high latitude, we run the similar sensitivity experiments toward phytoplankton, with perturbation only during JFM (January February March) and JAS (July August September) periods. The results are shown in Fig. 5 together with convec-

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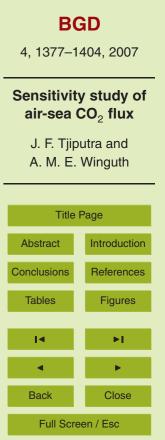


tive mixing event simulated in the model during the respective periods. In both hemisphere's winter season, the global uptake of CO<sub>2</sub> increase with the addition of phytoplankton concentration, especially in regions with high convective overturning processes, which upwelled additional nutrient to fuel more biological production. During summer seasons, adding phytoplankton concentration increase outgassing of carbon, similar to the tropic regions.

The micronutrient iron is found to be the most sensitive in the Equatorial Pacific, followed by the Southern Ocean and the North Pacific regions. This is expected since there is a constant supply of macronutrients in the Equatorial Pacific but a very limited source of iron. In contrast, the Atlantic Ocean has a large flux of aeolian dust input

- source of iron. In contrast, the Atlantic Ocean has a large flux of aeolian dust input from the Sahara desert (Mahowald et al., 2005), making it insensitive to iron input. The Southern Ocean and the North Pacific are also known for iron limited regions, but since the uptake of CO<sub>2</sub> in these regions occurs seasonally, the magnitude of sensitivity is relatively weaker than that of the Equatorial Pacific. The iron sensitivity is relatively weaker than that of the Equatorial Pacific.
- <sup>15</sup> map confirms an earlier sensitivity study by Dutkiewicz et al. (2006). However, their study indicates much stronger sensitivity in the Equatorial Pacific than our results. This may be contributed by the significantly low iron concentration in the Equatorial Pacific Ocean simulated by their model. The models discrepancy is difficult to validate due to the very limited and sparse observation of dissolved iron currently available (Parekh et al., 2005).
  - 3.4 Regional sensitivity of air-sea CO<sub>2</sub> flux with respect to ecosystem parameters in the model

In order to explore in more detail the role of plankton activity in the air-sea carbon exchange, the regional sensitivity of air-sea CO<sub>2</sub> flux toward ecosystem parameters <sup>25</sup> are explored as well. Earlier global and regional adjoint sensitivity studies (Zhao et al., 2005; Tjiputra et al., 2007) indicate that phytoplankton maximum growth, phytoplankton exudation, ingestion by herbivores, zooplankton grazing, and zooplankton excretion parameters are sensitive and hence will be tested for the regional scale. To



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simplify the analysis, the parameter units are uniformed by scaling the parameters to Pi = [P(new)/P(first guess)] (e.g. Giering, 1989). The adjoint model is applied to calculate  $\partial J_{FCO2}/\partial Pi_{(i,j)}$  for each parameter. Overall, the global air-sea CO<sub>2</sub> flux is sensitive toward these ecosystem parameters in regions with large annual productivity, including the Equatorial Pacific, the North Pacific (Western Gyre), the North Atlantic, and the

Southern Ocean.

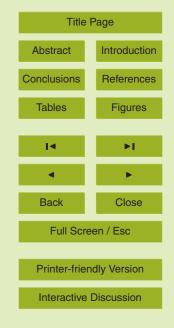
With respect to phytoplankton-associated parameters in the model, the air-sea  $CO_2$  flux is sensitive toward growth rate and exudation rate parameters. Both Figs. 6a and b indicate that carbon uptake is sensitive to these parameters, especially in the Equatorial Pacific, North Pacific, and North Atlantic regions. Notable sensitivities are also

- torial Pacific, North Pacific, and North Atlantic regions. Notable sensitivities are also revealed in parts of Southern Ocean and Indian Ocean. The sensitivity maps indicate that increasing the growth rate and reducing the exudation rate in these regions would increase the global uptake of CO<sub>2</sub>. The change of the respected parameters would result in more phytoplankton mass remain in the surface, thus increasing the primary
- <sup>15</sup> production and reducing the *DIC* concentration in the euphotic zone. In contrast, the uptake of CO<sub>2</sub> in regions with low nutrient concentrations (e.g. subtropical Pacific), are reduced if the phytoplankton growth rate is increased. These oligotrophic regions have low biological production all year long (Behrenfeld et al., 2005) and are considered to be atmospheric carbon sinks (see Fig. 1). However, due to the low nutrient concen-<sup>20</sup> tration in these regions, increasing the exudation rate may provide additional dissolved
- organic carbon (DOC), which could be remineralized as regenerated nutrients.

Both in the ocean and lake ecosystems, zooplankton play an important role in controlling the nutrient uptake (i.e. primary production) by phytoplankton (Carpenter et al., 2001; Moore et al., 2002; Pasquer et al., 2005; Schindler et al., 1997). For exam-<sup>25</sup> ple, lakes with high zooplankton biomass tend to uptake atmospheric CO<sub>2</sub>, whereas lakes with low zooplankton tend to do the opposite. In this study, we show that in regions where the air-sea CO<sub>2</sub> flux is controlled by the biological component, the surface carbon fluxes are sensitive to herbivore ingestion and grazing. The sensitivity map (Fig. 6c) illustrates that in regions with strong upwelling and convective mixing. 4, 1377–1404, 2007

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reducing the ingestion by herbivores would substantially increase the uptake of atmospheric  $CO_2$ . Having a low ingestion rate (i.e. high egestion rate) reduces the amount of carbon stored in the zooplankton biomass, allowing more carbon to be stored in POC and DOC. Therefore, there is more available carbon that can be recycled imme-

- <sup>5</sup> diately into nutrients in the euphotic zone. Previous sensitivity studies using only a onedimensional forward model demonstrate that the model output (e.g. the sea-to-air  $CO_2$ flux and surface  $pCO_2$ ) is sensitive toward grazing parameters in the Equatorial Pacific region (Chai et al., 2002; Dugdale et al., 2002). Here, the adjoint sensitivity identifies three main regions where the air-sea  $CO_2$  flux is sensitive toward grazing parame-
- ters: the Equatorial Pacific, North Pacific, and Southern Ocean. Figure 6d also shows strong sensitivity toward grazing parameters in the Bermuda Atlantic Time-series Study (BATS), which is consistent with in situ observations. This indicates that zooplankton grazing consumes most of the production at this location (Lessard and Murrell, 1998). In these regions, a reduction in the grazing rate increases phytoplankton mass in the
   euphotic zone, thus increasing the nutrient and *DIC* uptake by phytoplankton. The sen-
- sitivity of the zooplankton excretion parameter further emphasizes that the ecosystem dynamics are highly sensitive to zooplankton biomass.

In order to quantitatively test the sensitivity toward ecosystem parameters, the forward model is integrated for 10-years with different parameter perturbations. The anomalies of global air-sea CO<sub>2</sub> flux to these perturbation are plotted in Fig. 7. The control run has an average annual carbon uptake of 1.6 Pg C yr<sup>-1</sup>. This value is close to the corrected Takahashi et al. (2002) estimate of 1.64 Pg C yr<sup>-1</sup> for the year 1995 (http://www.ldeo.columbia.edu/CO2/) and within limits of Sabine et al. (2004) estimate.

- Over the 10-year period, significant atmospheric carbon sinks occur in the Equatorial Pacific, Western North Pacific Gyre, North Atlantic, and Southern Ocean regions. Essentially, the largest drawdown occurs when the herbivore ingestion parameter is
- reduced by 25%. The total uptake of atmospheric carbon yielded by perturbing the phytoplankton growth rate, exudation rate, herbivores ingestion, grazing rate, and excretion rate are 1.7, 1.7, 2.2, 2.0, and 1.9 Pg C yr<sup>-1</sup>, respectively, over 10-year period.

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Figure 7 also provides a first order estimate of strong biological pump regions. Southern Ocean, Equatorial Pacific, and North Pacific are three of the strongest ocean region in which the biological production contributes significantly to the sinks of the *DIC* concentration in the surface. Furthermore, these regions, which are also known as HighNutrient-Low-Chlorophyll (HNLC) regions, have not reach their maximum production due to limited iron supply in these regions (Martin et al., 1994; de Baar et al., 1995). In addition, our adjoint sensitivity study indicates that the air-sea gas exchanges in these regions are not only sensitive toward iron fertilization but also very sensitive toward zooplankton-associated parameters (i.e. grazing and ingestion parameters). These results are also supported by previous field studies, which indicate the significant role of zooplankton in suppressing the phytoplankton growth, thus biological production in

HNLC regions (Landry et al., 2003; Price et al., 1994; Saito et al., 2005).

#### 4 Conclusions

This study highlights the importance of the marine ecosystem structure in controlling
global air-sea CO<sub>2</sub> flux. Here, we demonstrate with an adjoint three-dimensional carbon cycle model that, outside of the tropic regions, convective mixing, biological production, and seasonal temperature variability mainly control the seasonal variability of pCO<sub>2</sub>. In the subpolar gyres, the biology activity is essentially responsible for the variability. These results are consistent with the observations (Takahashi et al., 2002;
Watson and Orr, 2003) and a study by Pasquer et al. (2005) who claimed that the Southern Ocean would be a source rather than a sink of atmospheric CO<sub>2</sub> without biological activity.

The analysis with the adjoint carbon cycle model has provided valuable information on which processes in the marine ecosystem significantly control the air-sea CO<sub>2</sub> gas <sup>25</sup> exchange. Along equatorial upwelling regions, the primary production is fueled sufficiently by a continuous supply of upwelled nutrients, which decrease the *DIC* concentration and pCO<sub>2</sub> in the surface layer. However, this effect is more than compensated

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by the upwelled *DIC*-rich watermass. At high latitudes, however, photosynthesis peaks during the spring and summer months, and drops significantly during the fall and winter months. The sensitivity experiments demonstrate that the uptake of atmospheric  $CO_2$  is sensitive to biological processes at both the high latitudes (e.g. North Pacific, McKin-

 Iey et al., 2006, Antarctic Ocean, Metzl et al., 1999) and the nutrient-rich low latitudes (i.e. Equatorial Pacific).

In regions where nutrients are the limiting factor for primary production, nutrient fertilization should increase carbon fixation and the uptake of atmospheric  $CO_2$ . However, this study shows that the amplitude of the outcome crucially depends on the structure within the regional ecosystem. The adjoint sensitivity maps illustrate that phosphate is an essential limiting factor for regions with sufficient light and temperature for growth. Regions with strong iron input, such as the Equatorial Atlantic, are very limited by Phosphate concentration. In nearly all parts of the ocean, atmospheric  $CO_2$  uptake increases with a reduction of zooplankton concentration, which yields faster recycling of nutrients in the euphotic zone. Moreover, lowering the zooplankton concentration

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<sup>15</sup> of nutrients in the euphotic zone. Moreover, lowering the zooplankton concentration results in extended phytoplankton life span, which prolongs high photosynthesis production periods.

The sensitivity map of air-sea CO<sub>2</sub> flux toward ecosystem parameters also indicates that plankton activity and the food web structure can significantly alter the flux of CO<sub>2</sub> between the atmosphere and ocean. For example, the air-sea carbon exchange in the high-nutrient low-chlorophyll (HNLC) regions, such as the Equatorial Pacific, the Subarctic North Pacific, and the Southern Ocean are sensitive not only toward iron, but also toward zooplankton ingestion, grazing, and excretion parameters. The role of zooplankton in these regions is mainly to suppress phytoplankton growth and slow down

<sup>25</sup> the process of nutrient remineralization. In the oligotrophic sub-tropical regions, the biological production can be amplified by increasing the flux of regenerated nutrients in the euphotic zone; for example, this can be achieved by increasing the exudation rate of phytoplankton.

Despite the effectiveness of using an adjoint model to estimate the sensitivity, the

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outcome essentially depends on the performance of the ecosystem model. For example, the adjoint sensitivity also depends on the initial condition of the ecosystem component and the first guess of the parameters. These parameters are determined based on observations and laboratory studies (e.g. Eppley, 1972; Amon and Benner,

- <sup>5</sup> 1994), and therefore do not necessarily represent the global marine ecosystem. Optimization techniques, such as the 4-DVAR method, can provide more insight on how to better constrain these parameters (Tjiputra et al., 2007; Zhao et al., 2005). Furthermore, a study by Doney et al. (2004) demonstrates that physical properties (e.g. surface forcing, convective mixing parameterization, vertical and horizontal resolutions,
- etc.) of many biogeochemical models can significantly impact the carbon cycle forecast. Thus, improving the parameterization of the physical processes in the model is also necessary in the near future. In summary, the adjoint model provides valuable information in identifying processes that could significantly influence the air-sea CO<sub>2</sub> flux estimates of the model. Having a more detailed and accurate parameterization
   of the sensitive processes can substantially reduce the model-data discrepancies and improve the model forecast. Future field experiments to test the results of this study

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are undoubtedly necessary.

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 Table 1. Description of ecosystem parameters.

Symbol	Description
T	Temperature
L	Light
P	Phytoplankton
Ν	Nutrient
No	Nutrient half saturation constant
G	Zooplankton grazing rate
Ζ	Zooplankton
${m arepsilon}_{ m her}$	Herbivores ingestion parameter
Zinges	Zooplankton assimilation efficiency
$P_{\min}$	Minimum concentration of phytoplankton
$P_o$	Phytoplankton half saturation constant
$d_o$	Remineralization rate of DOC
k <sub>doc</sub>	DOC parameter
I <sub>o</sub>	POC remineralization rate
Fe	Dissolved iron
k <sub>Fe</sub>	Iron half saturation constant
A	Calcium carbonate production parameter
P <sub>orgC</sub>	Organic carbon production
P <sub>BSi</sub>	Biogenic silica parameter

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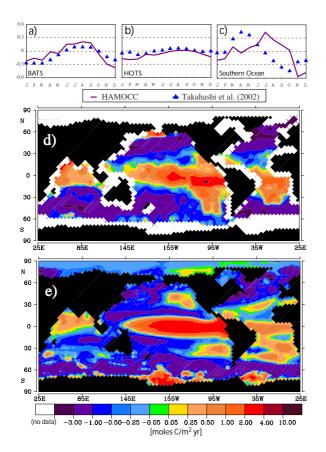
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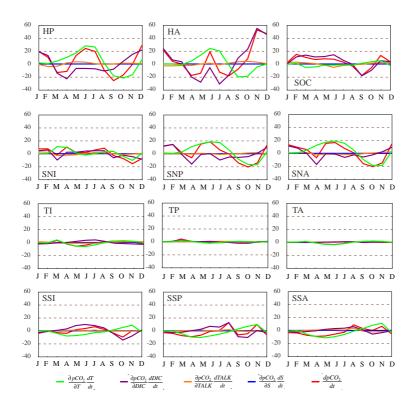
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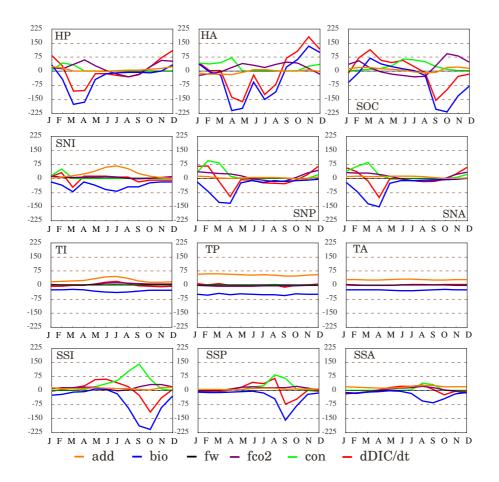
**Fig. 1.** HAMOCC5 model simulated seasonal cycle of air-sea  $CO_2$  flux [in moles  $CO_2 \text{ m}^{-2}\text{yr}^{-1}$ ] at **(a)** Bermuda Atlantic Time-Series Study (BATS), **(b)** Hawaii Ocean Time-Series (HOTS), and **(c)** Southern Ocean (56° S, 275° W) with observation estimates from Takahashi et al. (2002). Averaged air-sea  $CO_2$  flux for the year 1995 estimated from **(d)** observations by Takahashi et al. (2002) and **(e)** from HAMOCC5 model simulation. Positive values represent outgassing to atmosphere, whereas negative values indicate uptake of  $CO_2$  from the atmosphere.

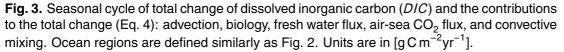
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**Fig. 2.** Seasonal cycle of the total change of  $pCO_2$  and the contributions to the total change (Eq. 3) for different ocean regions: sea surface temperature, dissolved inorganic carbon, alkalinity, and salinity. Ocean regions are defined as follow: High latitude Pacific (HP, N of 50° N), High latitude Atlantic (HA, N of 50° N), Southern Ocean (SOC, S of 50° S), Subtropical North Indian (SNI, 14° N–50° N), Subtropical North Pacific (SNP, 14° N–50° N), Subtropical North Atlantic (SNA, 14° N–50° N), Tropical Indian (TI, 14° N–14° S), Tropical Pacific (TP, 14° N– 14° S), Tropical Atlantic (TA, 14° N–14° S) Subtropical South Indian (SSI, 14° S–50° S), Subtropical South Pacific (SSP, 14° S–50° S), and Subtropical South Atlantic (SSA, 14° S–50° S). Units are in [ppm month<sup>-1</sup>]. 1399

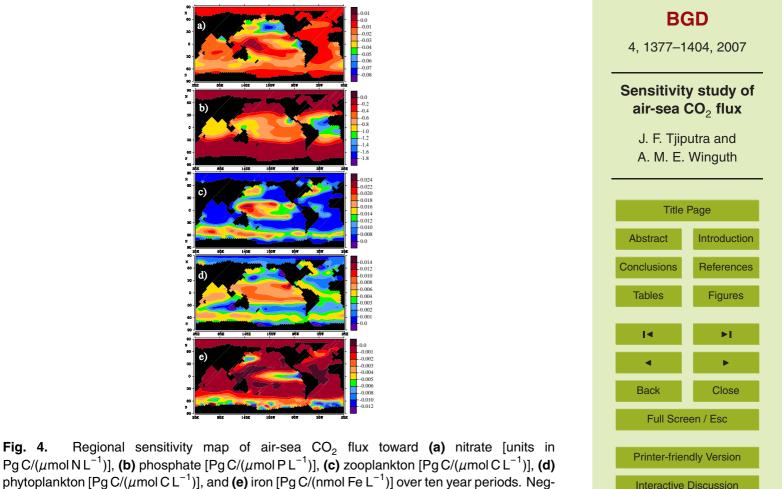
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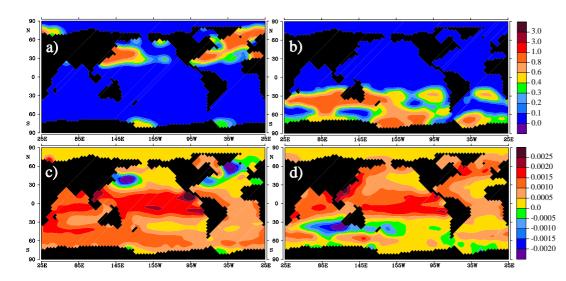


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phytoplankton [Pg C/( $\mu$ mol C L<sup>-1</sup>)], and (e) iron [Pg C/(nmol Fe L<sup>-1</sup>)] over ten year periods. Negative values indicate that an increase in the correspondent variable will increase the uptake of CO<sub>2</sub> from the atmosphere. Positive values indicate otherwise.

Fig. 4.



**Fig. 5.** Convective mixing event for the **(a)** JFM and **(b)** JAS in the HAMOCC5. Larger values represent stronger mixing due to convective overturning. Ten-year regional sensitivity map of air-sea  $CO_2$  flux toward phytoplankton for perturbation on **(c)** JFM and **(d)** JAS periods [Pg  $C/(\mu \text{mol C L}^{-1})$ ]. Negative values indicate that an increase in the correspondent variable will increase the uptake of  $CO_2$  from the atmosphere.

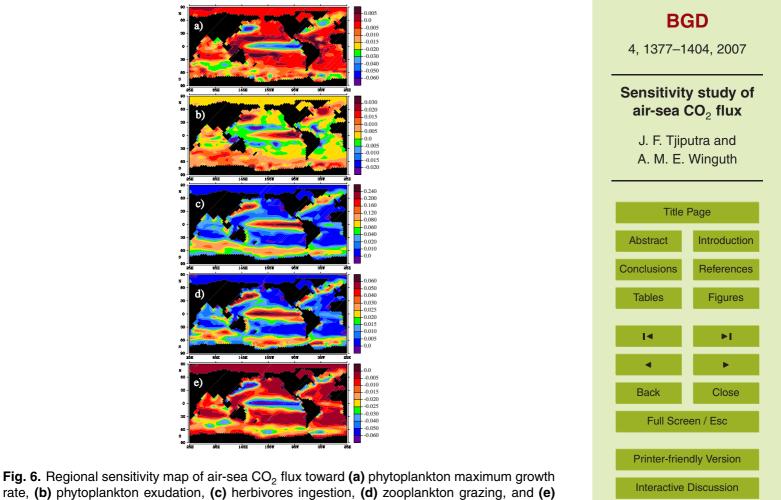
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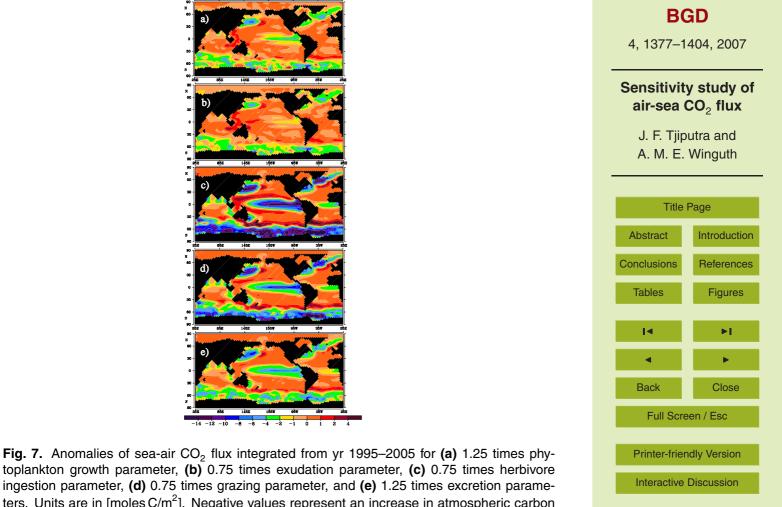
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zooplankton excretion parameters over ten year periods. Negative values indicate that an increase in the correspondent variable will increase the uptake of  $CO_2$  from the atmosphere. Units are in [Pg C]. 1403



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toplankton growth parameter, (b) 0.75 times exudation parameter, (c) 0.75 times herbivore ingestion parameter, (d) 0.75 times grazing parameter, and (e) 1.25 times excretion parameters. Units are in [moles C/m<sup>2</sup>]. Negative values represent an increase in atmospheric carbon uptake, whereas positive values represent increase in outgassing.