

# Effects of patchy ocean fertilization on atmospheric carbon dioxide and biological production

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[1] Increasing oceanic productivity by fertilizing nutrient-rich regions with iron has been proposed as a mechanism to offset anthropogenic emissions of carbon dioxide. Earlier studies examined the impact of large-scale fertilization of vast reaches of the ocean for long periods of time. We use an ocean general circulation model to consider more realistic scenarios involving fertilizing small regions (a few hundred kilometers on a side) for limited periods of time (of order 1 month). A century after such a fertilization event, the reduction of atmospheric carbon dioxide is between 2% and 44% of the initial pulse of organic carbon export to the abyssal ocean. The fraction depends on how rapidly the surface nutrient and carbon fields recover from the fertilization event. The modeled recovery is very sensitive to the representation of biological productivity and remineralization. Direct verification of the uptake would be nearly impossible since changes in the air-sea flux due to fertilization would be much smaller than those resulting from natural spatial variability. Because of the sensitivity of the uptake to the long-term fate of the iron and organic matter, indirect verification by measurement of the organic matter flux would require high vertical resolution and long-term monitoring. Finally, the downward displacement of the nutrient profile resulting from an iron-induced productivity spurt may paradoxically lead to a long-term reduction in biological productivity. In the worst-case scenario, removing 1 ton of carbon from the atmosphere for a century is associated with a 30-ton reduction in biological export of carbon.

**INDEX TERMS:** 4806 Oceanography: Biological and Chemical: Carbon cycling; 4842 Oceanography: Biological and Chemical: Modeling; 4845 Oceanography: Biological and Chemical: Nutrients and nutrient cycling; 4875 Oceanography: Biological and Chemical: Trace elements; **KEYWORDS:** carbon sequestration, iron fertilization, general circulation model, fisheries impacts

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## 1. Introduction

[2] In recent years, the idea of fertilizing the ocean with iron as a means of offsetting anthropogenic carbon dioxide emissions has been proposed in a number of forums [Martin *et al.*, 1991; U.S. Department of Energy (DOE), 1999]. Iron is an important limiting micronutrient for phytoplankton [Martin *et al.*, 1991; Raven and Falkowski, 1999], which use it in a large number of important physiological processes. Iron limitation appears to be especially important in explaining why large diatoms do not grow in regions such as the tropical Pacific and Southern Ocean where concentrations of surface macronutrients such as phosphate and

nitrate are high [Coale *et al.*, 1996; Boyd *et al.*, 2000]. Small phytoplankton are easily limited by small grazers which have generation times at least as short as those of the phytoplankton and recycle nutrients very efficiently. By contrast, large plankton are grazed by larger grazers with longer generation times. When these large plankton are given the iron they need to grow, they can escape grazing control and take up large amounts of macronutrients and carbon. It is presumed that over time this carbon is exported to depth. If the magnitude of this process could be increased, the result would be to remove carbon from the surface ocean, and thus from the atmosphere. This could offset some portion of anthropogenic emissions.

[3] Fertilization can take two forms. As generally envisioned, micronutrient fertilization involves adding iron to regions where macronutrients are already abundant, result-

ing in macronutrient depletion. It is important to stress that macronutrient depletion is itself important, not just the increase in the rate of biological cycling within the mixed layer. *Sarmiento and Toggweiler* [1984], *Knox and McElroy* [1984] and *Siegenthaler and Wenk* [1984] argued that macronutrient depletion in high latitudes could play an important role in explaining why atmospheric carbon dioxide concentrations have been lower during recent glacial periods. *Sarmiento and Orr* [1991] used an ocean general circulation model (OGCM) to estimate the maximum possible drawdown of atmospheric carbon dioxide resulting from macronutrient depletion driven by micronutrient fertilization. In work by *Sarmiento and Orr* [1991], surface phosphate concentrations were forced toward zero for 100 years in four regions: the Southern Ocean (90°S–30°S), the tropics (18°S–18°N), the North Atlantic (poleward of 30°N) and the North Pacific (also poleward of 30°N). The largest effect (75 ppmv reduction of atmospheric carbon dioxide after a century) came from fertilizing the Southern Ocean. By contrast, macronutrient depletion in the tropics resulted in an extremely weak drawdown of atmospheric carbon dioxide (1 ppmv) after a century. Recent simulations [*Archer et al.*, 2000; *I. Marinov et al.*, What sets the response of the ocean to fertilization, manuscript in preparation, 2003] suggest that these numbers may underestimate the relative importance of tropical and Southern Ocean fertilization but that the basic picture is accurate.

[4] An alternative form of fertilization would involve addition of macronutrients such as nitrate and phosphate as well as micronutrients [*Orr and Sarmiento*, 1992; *Jones*, 1996]. We will term this strategy *macronutrient addition* to distinguish its effect on the surface dissolved nutrient pool from the macronutrient depletion associated with iron fertilization alone. *Orr and Sarmiento* [1992] investigated the potential of long-term and large-scale increases in tropical biological productivity to alter atmospheric carbon dioxide. They found that if long-term (100 year) enhancement of biological productivity occurred over the entire tropics, with the required macronutrients being supplied from the already-present dissolved macronutrient pool (resulting in macronutrient depletion), only 20% of the resulting additional exported carbon came from the atmosphere over a century. If macronutrients were also added, however, they found that a larger fraction of the additional exported carbon came from the atmosphere.

[5] Long-term and complete depletion of macronutrients may serve as a limiting scenario for biogeochemical changes driven by climate change. However, anthropogenic fertilization is likely to be much more modest, both in its spatial and temporal scale. The present set of runs were undertaken to evaluate the efficiency of such a scenario, one in which fertilization is applied to a single grid box of our general circulation model, (a patch of ocean with an area of 200,000 km<sup>2</sup>) for a limited time (1 month). Given the spatial dimensions of the patch, reducing surface phosphate by 0.1 μmol/l over a depth range of 100 m would correspond to a removal of 2.8 Mt C from the surface ocean. This is a factor of 10 larger than the largest operational carbon sequestration project, that underway in the Sleipner West gas field [*U.S. DOE*, 1999]. It should be noted that the size of the patch

being fertilized is also significantly larger than those fertilized during the IRONEX experiments in the equatorial Pacific and the SOIREE experiment in the Southern Ocean.

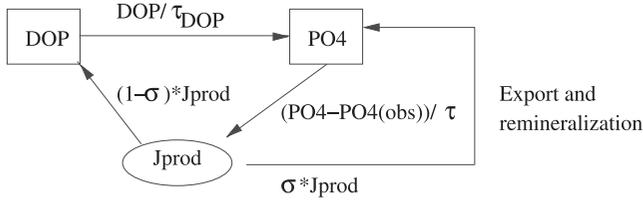
[6] Fertilization as a means of carbon sequestration has been controversial since it was first proposed in the late 1980s. The American Society of Limnology and Oceanography passed a resolution in 1991 discouraging the use of iron fertilization as a policy option. Recently, *Chisholm et al.* [2001] argued against the claim that “ocean fertilization is an easily controlled, verifiable process that mimics nature; and that it is an environmentally benign long-term solution to atmospheric CO<sub>2</sub> accumulation.”

[7] This paper addresses a number of the issues discussed by *Chisholm et al.* [2001], in particular the efficiency of fertilization, the verifiability of sequestration resulting from fertilization and the long-term consequences of fertilization. Although our simulations assume that localized fertilization can carry large amounts of carbon from the surface ocean to the deep ocean, the degree to which this process removes carbon dioxide from the atmosphere is quite variable. In simulations where macronutrients are depleted, much of the carbon comes from the ocean rather than the atmosphere. In simulations where macronutrients are added to the surface ocean, the fraction of carbon supplied by the atmosphere is much larger. However, there is compensatory outgassing that occurs on a global scale. Because the efficiency of fertilization is so variable, it will prove challenging to verify the amount of carbon sequestered by fertilization. Moreover, we find that the impacts on the export flux of organic matter can greatly exceed the impact on ocean carbon uptake. This implies that long-term, nonlocal impacts on marine biology, including fisheries, could dominate the environmental impact of sequestration.

## 2. Model Description

### 2.1. Physical Model

[8] The physical model used in this study was developed for the Ocean Carbon Model Intercomparison Project (OCMIP). The horizontal resolution is the same coarse 3.75 × 4.5 resolution used in previous papers published by our group [*Najjar et al.*, 1992; *Anderson and Sarmiento*, 1995; *Gnanadesikan*, 1999; *Murnane et al.*, 1999; *Gnanadesikan et al.*, 2002]. Surface temperatures and salinities are restored toward observations with a 1 month time constant. This procedure may be thought of as an assimilation procedure whereby the relatively well known salinity and temperature data are used to estimate the much more poorly known surface fluxes of heat and freshwater. Additional surface fluxes of heat and freshwater (taken from *da Silva et al.*, 1994) are imposed so that the total surface fluxes are a blend of purely observational estimates and dynamically constrained estimates. Surface winds are given by *Hellerman and Rosenstein* [1983] as in previous versions of the model. The model produces an average temperature and salinity structure which is relatively realistic [*Gnanadesikan et al.*, 2002]. Although, as in all coarse-resolution ocean models, there are significant problems with the detailed representation of the equatorial zone, *Gnanadesikan et al.* [2002] show that the overall transports of water in the



Boxes denote tracer fields which are advected and diffused.  
 Ellipse denotes a source/sink term.

**Figure 1.** Schematic of the OCMIP 2 biogeochemical model.

Equatorial Undercurrent and Ekman layers are in line with those produced by much higher-resolution models. The model has too little convection within the Southern Ocean and so may overestimate the efficiency with which carbon is stored in the deep ocean on long timescales. However, over the century-scale timescales considered in this paper we do not anticipate that this effect will be of great importance.

## 2.2. Biogeochemical Model

[9] The biogeochemical model used in this paper is the same as that used in the OCMIP 2 program (R. G. Najjar and J. C. Orr, Design of OCMIP 2 simulations of chlorofluorocarbons, the solubility pump and common biogeochemistry, unpublished manuscript, 1998 (available at [www.ipsl.jussieu.fr/OCMIP/](http://www.ipsl.jussieu.fr/OCMIP/))). The model is shown schematically in Figure 1. There are four state variables: phosphate ( $PO_4$ ), dissolved organic phosphate (DOP), dissolved inorganic carbon (DIC) and alkalinity (ALK). The equations for these variables, the key source and flux terms involving them, and their relation to the DIC concentration are shown below.

$$\frac{d}{dt}PO_4 - \nabla D \nabla PO_4 = -J_{prod} + \frac{DOP}{\tau_{DOP}} + \frac{\partial}{\partial z} F_{POP}, \quad (1a)$$

$$\frac{d}{dt}DOP - \nabla D \nabla DOP = (1 - \sigma)J_{prod} - \frac{DOP}{\tau_{DOP}}, \quad (1b)$$

$$J_{prod} = (PO_4 - PO_4(obs))/\tau \quad PO_4 > PO_4(obs), z > -z_c, \quad (1c)$$

$$F_{POP} = 0 \quad z > -z_c$$

$$F_{POP} = (z/z_c)^\beta \sigma * \int_{-z_c}^0 J_{prod}(x, y, z, t) dz \quad z < -z_c, \quad (1d)$$

$$\frac{d}{dt}DIC - \nabla D \nabla DIC = R_{C:P} \cdot \left( -J_{prod} + \frac{DOP}{\tau_{DOP}} + \frac{\partial}{\partial z} F_{POP} \right) + \frac{\partial}{\partial z} F_{Ca}, \quad (1e)$$

$$\frac{d}{dt}ALK - \nabla D \nabla ALK = R_{Cinorg:Corg}(- (1 - \sigma)J_{prod}) + \frac{\partial}{\partial z} F_{Ca}, \quad (1f)$$

$$F_{Ca} = 0 \quad z > -z_c$$

$$F_{Ca} = R_{Cinorg:Corg} \sigma \exp(z/L_{ca}) \int_{-z_c}^0 J_{prod}(x, y, z, t) dz \quad z < -z_c, \quad (1g)$$

$$Flux_C = k_g(pCO_2(atm) - pCO_2(ocean))$$

$$= K_v \frac{\partial DIC}{\partial z}(z=0) \frac{\partial DIC}{\partial pCO_2}. \quad (1h)$$

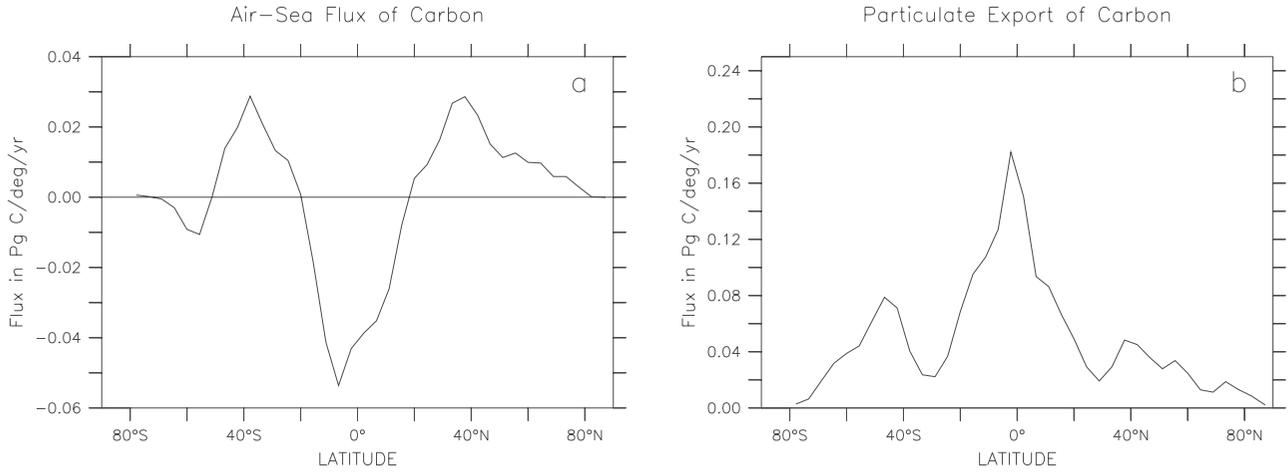
In the equations above,  $d/dt$  is the total derivative,  $D$  is the diffusion tensor,  $J_{prod}$  is the total production of organic matter,  $F_{POP}$  is the vertical flux of particulate organic phosphate below the compensation depth  $z_c = 75$  m,  $F_{ca}$  is the vertical flux of calcium carbonate,  $\tau_{DOP}$  is a remineralization timescale for dissolved organic matter (set to 6 months),  $\sigma$  is the fraction of production exported as particulate organic matter (set to 1/3),  $PO_4(obs)$  is the observed phosphate concentration from *Louanchi and Najjar* [2000] and  $\tau = 1$  month.  $R_{C:P} = 117$ , following *Anderson and Sarmiento* [1994]. Note that  $R_{C:P}$  is taken to be the same for dissolved and particulate matter. The remineralization parameters have values of  $\beta = -0.9$ ,  $R_{Cinorg:Corg} = 0.07$  and  $L_{ca} = 2$  km following *Yamanaka and Tajika* [1996]. Gas exchange (equation 1h) is computed using a piston velocity  $k_g$  adapted from *Wanninkhof* [1992]. Total carbon inventory is conserved in the simulations, so that a net flux of carbon into the ocean reduces the amount of carbon in the atmosphere.

[10] Figure 2 shows a summary of the annually and zonally averaged carbon cycle as simulated by the model. As seen in Figure 2a, the ocean loses carbon dioxide to the atmosphere in the tropics and takes it up in the subtropics. There is a weak outgassing flux from the Southern Ocean as well. The export of particulate organic carbon is largest in the tropics, drops off in the subtropics and rises again in the subpolar zones of both the Northern and Southern Hemispheres.

[11] In a manner similar to *Sarmiento and Orr* [1991], we simulate iron fertilization implicitly, by looking at the effect of drawing down surface macronutrients. We do this in the model by restoring phosphate to zero, rather than to observations. Locally, this increases the uptake and particulate export of phosphate and carbon, and lowers the  $pCO_2$ . The low- $CO_2$ , low-phosphate water at the surface is then advected through the model, so that the effects of fertilization on both air-sea uptake and new production are felt at a distance. In contrast to *Sarmiento and Orr* [1991], however, we apply this change in forcing for a limited time (1 month) over limited spatial regions.

[12] An extensive set of studies was carried out to evaluate the impact of fertilization both on atmospheric carbon dioxide and on biological productivity. The scenarios are listed below, along with the questions they were intended to answer, and summarized in Table 1.

[13] • How does the impact of fertilization depend on how fertilization changes surface macronutrient concentrations? Three kinds of fertilization were considered. In the first,  $PO_4(obs)$  in equation (1c) was replaced by zero for the month of September, so chosen because the maximum nutrients at the fertilization site are found during this month.



**Figure 2.** Key diagnostics of the carbon cycle in a general circulation model at equilibrium. (a) Air-sea flux of carbon dioxide. Note that carbon dioxide tends to invade the ocean at midlatitudes and to escape from the ocean at low latitudes. (b) Export flux of carbon across the base of the euphotic zone.

This produces a partial depletion in macronutrients over the fertilization time period (as shown by the dashed line in Figure 3) and is denoted by PART. In a second set of runs,  $PO_4(obs)$  was set to zero and  $\tau$  was reduced from 30 days to 3 days. This produces almost complete removal of macronutrients (dotted line in Figure 3) and runs made with this parameterization of fertilization are denoted as FULL.

[14] Finally, a set of runs were made in which  $PO_4(obs)$  was set to zero and  $\tau$  to three days, but phosphate was added at each timestep so as to maintain the surface concentrations at the same level as in the control run (the solid line in Figure 3). These runs were denoted as ADD. They are intended to separate the effect of surface macronutrient depletion from the carbon drawdown associated with fertilization. Implicitly, the ADD scenario corresponds to adding a “supernutrient” which is always rapidly exported to depth whenever it comes to the surface.

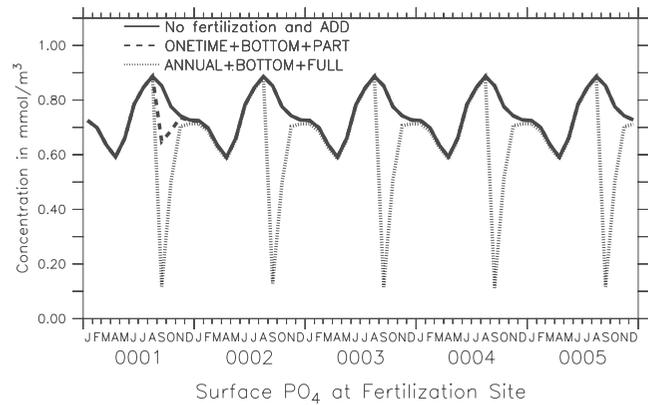
[15] • Can the results from a single fertilization study be used to predict the impact of future fertilization studies? This question was attacked by changing the number of times that fertilization was done. In one set of cases

(denoted by ONETIME) the fertilization was only applied for the month of September during the first year of the run. In a second set of cases (denoted by ANNUAL) the fertilization was applied each September. In contrast to *Sarmiento and Orr* [1991], we did not change the total carbon in the system according to a prescribed emissions scenario. This removes changes in the impact of fertilization due to changes in the buffer factor.

[16] • How does the impact of fertilization depend on what happens to particulate matter once it leaves the surface layer? We investigated this question by varying the remineralization profile. In one set of runs, the remineralization of particulate organic carbon produced by fertilization was parameterized to occur at shallow depths following the function in equation (1d) (denoted by EXP). In a second set of runs (denoted by BOTTOM), designed to provide an

**Table 1.** Types of Runs Made to Evaluate Sensitivity of Atmospheric  $CO_2$  to Spot Fertilization

Parameter	Type of Experiment	Acronym
Frequency of fertilization	single event (1 month)	ONETIME
	1 month each year	ANNUAL
Nutrient change	partial depletion	PART
	full depletion	FULL
	nutrient addition	ADD
Remineralization profile	same as for normal export	EXP
	export at site to bottom	BOTTOM
<i>Runs made with ONETIME + (FULL, ADD) + BOTTOM</i>		
Size of fertilization patch	one grid box	(default)
	nine contiguous boxes	9BOXREG
	nine scattered boxes	9BOXSCAT
	equatorial Pacific	EQPAC
Circulation scheme	low vertical, lateral mixing	(default)
	high vertical, lateral mixing	HIMIX



**Figure 3.** Surface phosphate concentration at the fertilization site, demonstrating the effect of changing  $PO_4$  and  $T$  in equation (9). Concentrations are monthly averages, instantaneous concentrations during micronutrient fertilization at the fertilization site will be slightly lower (since the depletion in nutrients takes some time to establish itself).

upper limit on the impact of remineralization, the entire excess particulate export went all the way to the bottom before remineralizing. In all models the DOP produced by fertilization (2/3 of the additional production) is transported and remineralized in a manner identical to that in the baseline model. *Sarmiento and Orr* [1991] found that for climatological fertilization, the impact of changing the remineralization length scale was relatively minor. We do not.

[17] Simulations were carried out for each combination of these three parameters. Each simulation is denoted by number of repetitions + type of fertilization + remineralization scheme so that ONETIME + ADD + BOTTOM is a simulation in which the fertilization was carried out once, nutrients were added, and the remineralization at the fertilization site occurred at the ocean bottom.

[18] In addition, several sensitivity studies, as described below, were carried out to evaluate the robustness of these results. These simulations were only run with the ONETIME + ADD + BOTTOM and ONETIME + FULL + BOTTOM settings.

[19] • To what extent can the results for a single grid box be extrapolated to larger regions? We performed a set of simulations in which the area over which fertilization was applied varied. We ran one set of simulations in which the region of fertilization was expanded from one grid box to a square of nine grid boxes with the same central point. These simulations are denoted by 9BOXREG. In another set of simulations (denoted by EQPAC) a massive fertilization event was applied over the entire equatorial Pacific east of the date line between 9°S and 9°N.

[20] • How sensitive are the results to model physics? A set of runs, denoted by HIMIX, was carried out in which the lateral mixing was doubled and diapycnal mixing was increased by a factor of 4 within the pycnocline. This run has much more tropical upwelling and a level of biological production that is on average double that of the baseline run. More details about this run are provided by *Gnanadesikan et al.* [2002] (where it is denoted as AIHIGH + KVHIGH).

[21] The biological cycling model employed here embodies a number of key assumptions. It is important to state at the outset that not all of these assumptions necessarily describe the real ocean.

[22] • The first key assumption that is made here is that fertilization actually results in an increase in particulate export. This is still controversial. *Nodder et al.* [2001] did not find an increase in particulate organic carbon export during the SOIREE experiment, and *Landry et al.* [2000] argue that changes in the food web acted to keep the carbon export ratio low during IronEx III. By contrast, *Boyd et al.* [1998] argue that iron-mediated diatom blooms are the best explanation for episodic increases in particulate organic flux in the North Pacific.

[23] • A second key assumption is that biological productivity actually depends on whether macronutrient concentrations exceed the climatological mean in the control experiments (or 0 during the fertilization experiments) and will decrease as the excess macronutrient concentration decreases. Constraining production to shut off when the concentration drops below climatological values (as we

do) is particularly severe. Other approaches to modeling biological production (such as in the HAMOCC model of *Maier-Reimer* [1993]) use a Michaelis-Menten approach to growth, assuming that growth becomes small (but not zero) when the nutrient concentration drops. As will be shown in the discussion section, this assumption has profound consequences for the macronutrient depletion scenarios.

[24] • A final set of assumptions about the role of iron is built into both the macronutrient depletion and macronutrient addition cases. In the depletion cases, nutrients remineralized at depth as a result of fertilization are not treated differently when they return to the surface. This implicitly assumes that any added iron has been lost to the system. By contrast, in the macronutrient addition cases, any nutrients exported to depth are rapidly taken up when they return to the surface. This corresponds to assuming that any iron added along with the macronutrients remains associated with these nutrients over time. In reality, iron interacts strongly with sinking particles and so will not be retained for very long time periods. However, because some iron is bound by biologically secreted chelating compounds, it may not be lost to the system as quickly as in the depletion experiments.

[25] In combination, the last two sets of assumptions mean that the macronutrient depletion experiments could be lower bounds for the efficiency of fertilization, while the macronutrient addition experiments are upper bounds. If the first assumption does not hold, all the experiments here are likely to be upper bounds.

### 3. Results

#### 3.1. Relationship Between Phosphate Depletion and Atmospheric CO<sub>2</sub> Drawdown

[26] An implicit assumption in many discussions of ocean fertilization is that depletion of surface nutrients leads to additional export of organic carbon from the surface ocean to the deep ocean, which in turn leads to a roughly equivalent drawdown in atmospheric carbon dioxide. A way of comparing runs at different spatial scales is to normalize them by the amount of phosphate initially present within the euphotic zone. This gives a “potential POC export”  $\Delta C_{POC}^{pot}$ ,

$$\Delta C_{POC}^{pot} = R_{C:P} \int_x \int_y \int_{z=-D}^0 PO_4(\text{control}) dx dy dz, \quad (2)$$

(shown in the second column of Table 2).  $\Delta C_{POC}^{pot}$  is the amount of carbon that could be exported if all of the available phosphate in the mixed layer were converted to POP and exported, carrying particulate organic carbon with it, and if no additional phosphate were added either from below or laterally. We would not, a priori, expect to realize the full amount of this POC export, as two thirds of the additional production resulting from nutrient depletion is converted to DOP. Approximately one sixth of the DOP is then converted back to PO<sub>4</sub> during the monthlong fertilization, and one third of this PO<sub>4</sub> is in turn exported in particulate matter (little DOP is exported to any depth in our model). In the absence of horizontal and vertical supply,

**Table 2.** Summary of Carbon Dynamics Changes Associated With Onetime Macronutrient Depletion Runs<sup>a</sup>

Experiment	$\Delta C_{POC}^{pot}$	$\Delta C_{POC}^{fert}$	$\Delta C_{global}^{uptake}$ (1 yr)	$\Delta C_{global}^{uptake}$ (100 yr)	$Eff_{depl}$
ONETIME + PART + BOTTOM	24.34	4.15	0.76	0.48	0.115
ONETIME + FULL + BOTTOM	24.34	15.3	2.78	1.48	0.097
ONETIME + FULL + EXP	24.34	16.7	2.43	0.34	0.020
9BOXSCAT	143.9	104.5	19.6	7.43	0.074
9BOXREG	201.0	93.62	14.7	8.42	0.090
EQPAC	1814	734.8	151.6	72.7	0.099

<sup>a</sup> $\Delta C_{POC}^{pot}$  (equation (2)) is the potential export of POC if all  $PO_4$  in the mixed layer at the at the fertilization site were converted to POC.  $\Delta C_{POC}^{fert}$  is the actual additional POC export over the time macronutrients are being depleted.  $\Delta C_{global}^{uptake}$  (1 yr, 100 yr) are the oceanic drawdown of atmospheric  $CO_2$  after 1 and 100 years, respectively, for the entire ocean.  $Eff_{depl} = \Delta C_{global}^{uptake}(100 \text{ yr})/\Delta C_{POC}^{fert}$  is the fraction of POC initially exported that ends up coming out of the atmosphere. An implicit assumption of some studies of macronutrient depletion is that  $\Delta C_{POC}^{fert} = \Delta C_{global}^{uptake}$ . In our simulations this is not the case.

with a small value for  $\tau$ , we would thus expect that approximately 37% of the potential POC export would in fact be exported from the euphotic zone during a 1-month fertilization event.

[27] We define the cumulative additional POC export  $\Delta C_{POC}$  as

$$\Delta C_{POC} = \int_x \int_y \int_{t=0}^T (F_{POC}(fert) - F_{POC}(control)) dx dy dt, \quad (3)$$

where  $F_{POC}(fert)$  and  $F_{POC}(control)$  are the particulate fluxes to the abyss in the fertilization and control runs. Let  $\Delta C_{POC}^{fert}(T_f)$  be  $\Delta C_{POC}$  when the spatial integration is performed over the fertilization site and  $T_f$  is the end of the fertilization time.  $\Delta C_{POC}^{fert}(T_f)$  (shown in the third column of Table 2), is the actual additional flux of particulate organic carbon to the deep ocean at the fertilization site.

[28] We can also define the oceanic uptake of atmospheric carbon dioxide over some region at time  $T$  following the initiation of fertilization as

$$\Delta C_A^{uptake}(T) = \int_x \int_y \int_{t=0}^T (F_{CO_2}(fert) - F_{CO_2}(control)) dx dy dt, \quad (4)$$

where  $F_{CO_2}$  is the air-sea flux of atmospheric carbon dioxide, and  $A$  represents the area over which the air-sea flux difference is integrated. Finally, we can define the efficiency of fertilization for the macronutrient depletion runs as

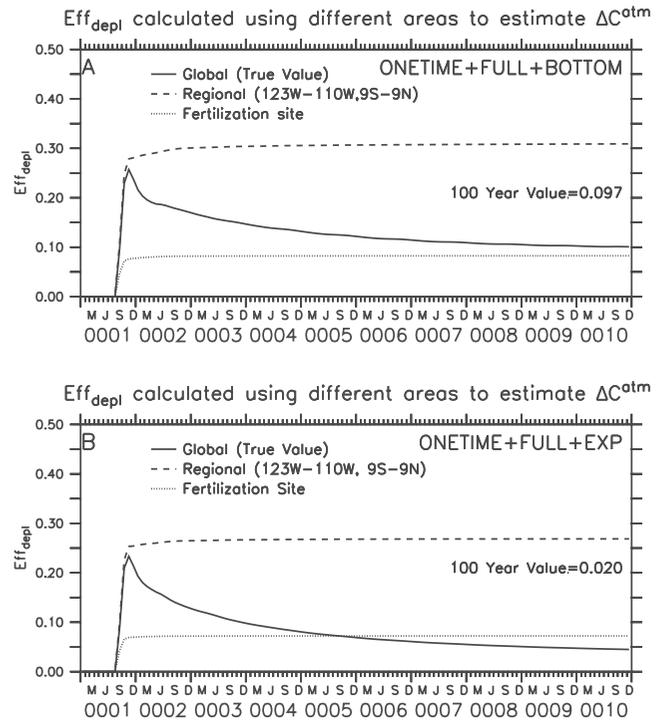
$$Eff_{depl} = \Delta C_{global}^{uptake}(T) / \Delta C_{POC}^{fert}(T_f). \quad (5)$$

$Eff_{depl}$  measures the fraction of particulate organic carbon exported to the deep ocean as a result of fertilization which eventually comes out of the atmosphere. If the implicit assumption that phosphorus depletion leads to atmospheric carbon dioxide drawdown is true, then at some time after fertilization it might be expected that  $\Delta C_{global}^{uptake}(T)$ , integrated globally, should approach  $\Delta C_{POC}^{fert}(T_f)$ . The efficiency would then approach a value of 1.0.

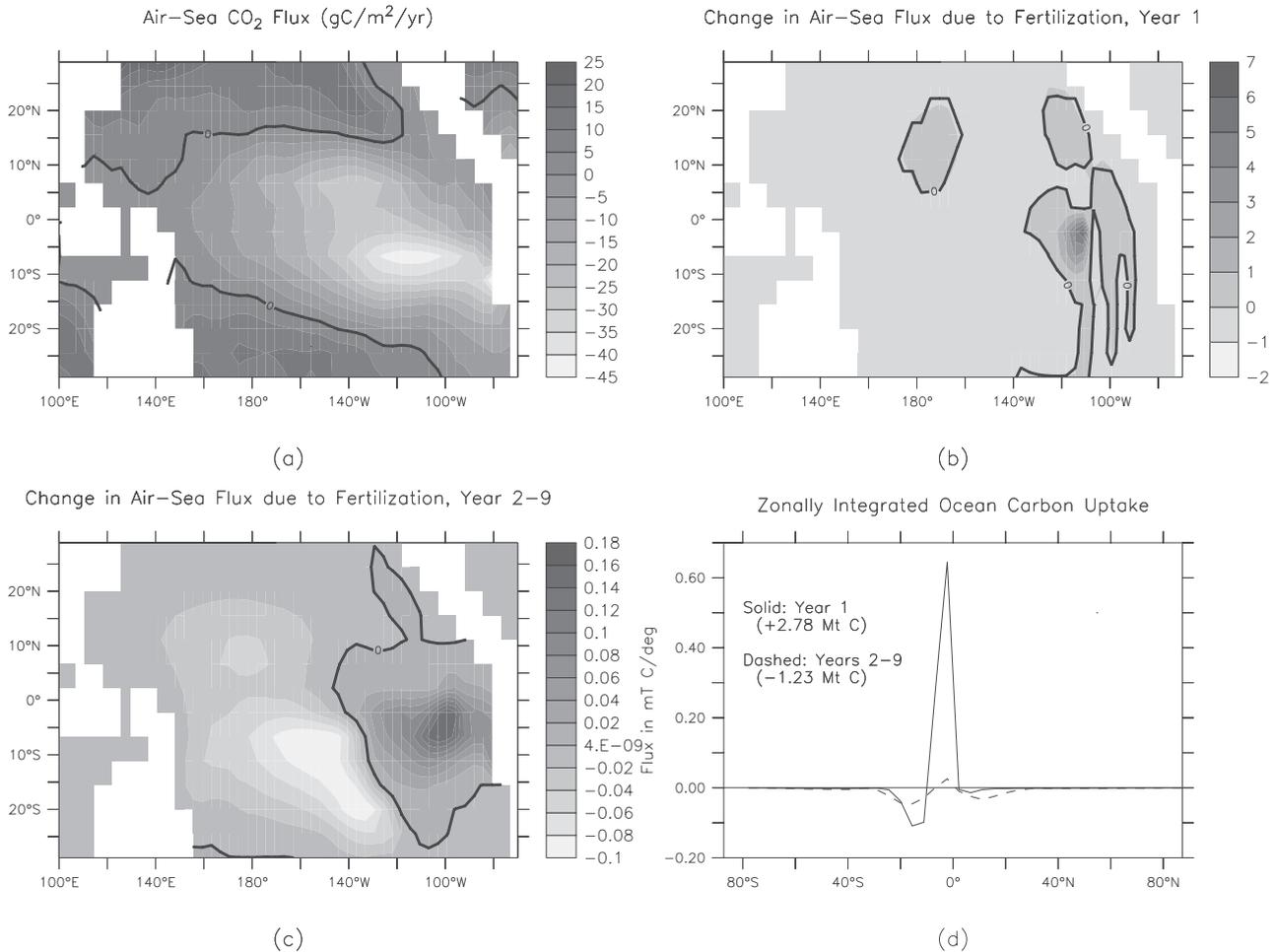
[29] Results from a number of the nutrient depletion runs are summarized in Table 2.  $Eff_{depl}$  for ONETIME + FULL + BOTTOM and ONETIME + FULL + EXP (calculated using different areas for the integration of  $\Delta C_{global}^{uptake}(T)$ ) is

shown in Figure 4. A number of important conclusions can be drawn from these results

[30] 1. For isolated fertilization events where nutrients are fully depleted at the fertilization site  $\Delta C_{POC}^{fert}(T_f) > 0.37 \Delta C_{POC}^{pot}$  (compare columns 2 and 3 of Table 2), but as more contiguous points are fertilized, the additional POC export drops toward what might be expected from a simple analysis of the biogeochemical model. This is because diffusion and advection supply nutrients to the fertilization



**Figure 4.**  $Eff_{depl}$  for runs (a) ONETIME + FULL + BOTTOM and (b) ONETIME + FULL + EXP using different surface areas to estimate  $\Delta C_{global}^{uptake}$ . The solid lines show the true value, the dashed lines the value if the air-sea flux is only measured at the fertilization site, and the dotted lines show the value if a region around the fertilization is used. A value of 1 would mean that all the sinking POC was eventually taken out of the atmosphere. Note that neither the estimate using air-sea gas flux at the site nor that made regionally captures the temporal dependence of  $Eff_{depl}$ .



**Figure 5.** Change in air-sea CO<sub>2</sub> flux due to fertilization. (a) Background air-sea flux in gC/m<sup>2</sup>/yr. (b) Change in air-sea flux integrated over the first year subsequent to fertilization (simulation shown is ONETIME + FULL + BOTTOM). (c) Change in air-sea flux integrated over years 2–9 following fertilization. (d) Zonally integrated cumulative changes in air-sea flux.

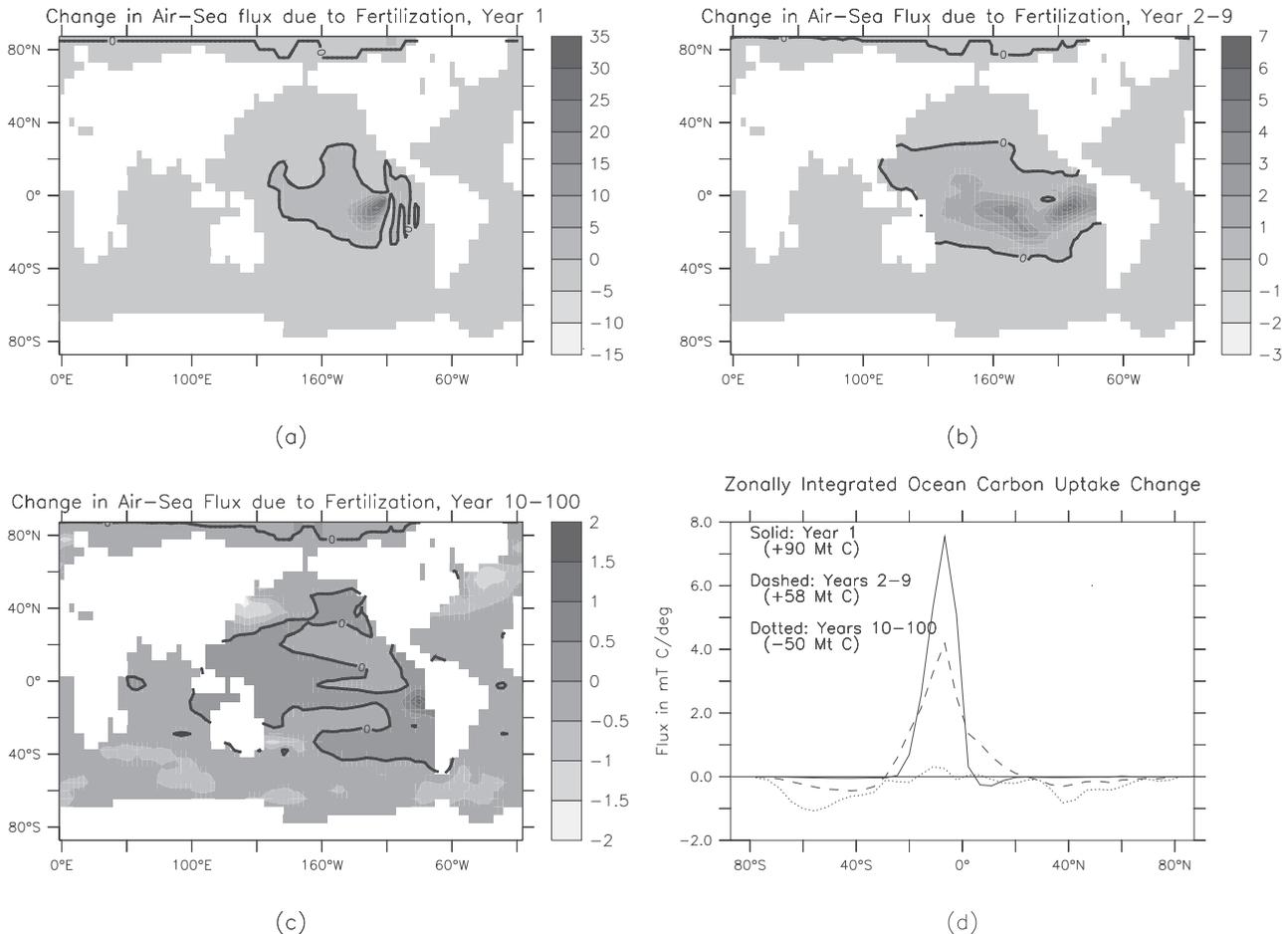
site(s) during a fertilization event. As a result  $\Delta C_{POC}^{pot}$  is underestimated if only the site itself is considered. When a larger contiguous region is fertilized, lateral supply of nutrients becomes much less important. This has a number of implications. On the one hand, it demonstrates that fertilizing small patches would result in drawing down nutrients over a broader region, enhancing the “effective size” of a patch. On the other hand, it means that large-scale fertilization would not necessarily be as efficient at removing carbon from the atmosphere as one might conclude from an experiment involving a small patch.

[31] 2. The efficiency of fertilization ( $Eff_{depl}$ ) is much less than 1.0. After 1 year it is around 20%, and after 100 years it varies between 2 and 11.5%. Moreover, the efficiency depends quite strongly on the length scale of the remineralization. When the additional particulate export remineralizes at shallow depths, it is rapidly returned to the surface ocean and the atmosphere, and efficiencies are very low. Such a strong dependence on the location of remineralization represents a key difference between these results, and those of *Sarmiento and Orr [1991]*. A more complete

discussion of why efficiencies are so low in these runs is presented in section 4.

[32] 3.  $\Delta C_{global}^{uptake}(T)$  cannot be determined simply by measuring fluxes at the fertilization site itself or even over a region surrounding the fertilization site.  $Eff_{depl}$  varies significantly depending on whether  $\Delta C_{uptake}(T)$  is measured at the site itself, in a region surrounding the fertilization site, or globally. The air-sea fluxes of carbon dioxide that contribute to  $\Delta C_{uptake}(T)$  have complicated spatial and temporal dependence. Figure 5 shows the background air-sea CO<sub>2</sub> flux, and the change in CO<sub>2</sub> flux integrated over the 1 year after fertilization and 2–9 years after fertilization. Initially, a large flux is seen into the ocean at the fertilization site, but over time a large-scale pattern develops in which additional flux into the ocean to the east of the fertilization site is balanced by additional flux out of the ocean to the west of the fertilization site. This pattern of fluxes results in a significant net outgassing that cancels 40% of the initial CO<sub>2</sub> drawdown from the atmosphere. The amplitude of the fluxes associated with this pattern are small in comparison with the background air-sea CO<sub>2</sub> fluxes. This means that the





**Figure 8.** Change in air-sea carbon flux due to fertilization. (a) Integrated change in air-sea flux in 1st year following fertilization (ONETIME + ADD + BOTTOM). (b) Integrated change in air-sea flux in the first year over years 2–9 following fertilization event. (c) Integrated change in air-sea flux over years 10–100 of simulation (10–99 years following fertilization). (d) Zonally integrated changes in air-sea flux.

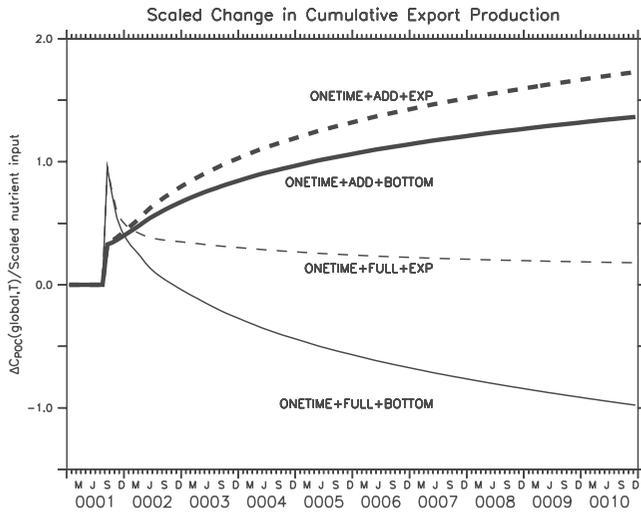
dependence on the remineralization profile. When the simulation is run out for 100 years, the fraction removed from the atmosphere drops to 42–44%. Figure 8 shows the spatial distribution of the outgassing that cancels the initial drawdown of carbon dioxide. In contrast to the macronutrient depletion runs, where most of the compensation occurred within the tropics, in the macronutrient addition runs the compensation largely occurs in the high latitudes. As deep water whose carbon content was set by being in contact with the prefertilization atmosphere with higher carbon dioxide concentrations is brought to the surface, it tends to restore the atmosphere toward its prefertilization value.

[35] These results are similar to those of *Orr and Sarmiento* [1992]. They are also similar to those in a recent study by *Matear and Elliot* [2003] who use a similar ocean circulation model to examine the effects of macronutrient fertilization. *Matear and Elliot* [2003] find efficiencies of about 65% when examining regions which are phosphate-limited. However, they do not simulate the same decreases in atmospheric  $\text{CO}_2$  drawdown due to outgassing on century

timescales, largely because they prescribe the concentration of atmospheric carbon dioxide. In our model, removal of carbon dioxide from the atmosphere as a result of sequestration results in lower atmospheric carbon dioxide and thus outgassing. In *Matear and Elliot's* model, more carbon is added to the atmosphere-ocean system to compensate for that taken up by the oceans as a result of fertilization. As a result, they do not simulate the outgassing that we obtain.

### 3.2. Effects on Biological Productivity

[36] In addition to changing the carbon cycle, fertilization alters the cycling of nutrients. Such secondary impacts of fertilization should also be considered in evaluating its overall impact. Figure 9 and Table 3 show the effect of fertilization on the cumulative POC export flux in a number of runs. We focus on POC because particulate export is associated with large plankton and higher trophic levels and may be particularly important when considering impacts on fisheries. We note that the effect on the total production is likely to be significantly larger than the effect on particulate export. Several key results emerge from these simulations



**Figure 9.** Scaled change in globally integrated POC export resulting from fertilization. For depletion runs, the ratio shown is the change in globally integrated POC flux over the change in POC flux at the fertilization site during the month of fertilization. This ratio is initially close to 1, but then drops as nutrient removal from the surface ocean results in a decrease in production in surrounding areas. For additional runs, the ratio is the globally integrated export production over the added phosphate  $\ast R_{C,P}$  the stoichiometric ratio of phosphate to carbon. This ratio is initially 1/3, (as only one third of the added phosphate is initially exported as POC) but then increases as the added nutrients are recycled in and below the mixed layer.

[37] 1. The net impact of macronutrient depletion on export production can be negative. In the nutrient depletion runs with BOTTOM remineralization, there is a significant drop in global export production over 100 years. The cumulative drop in global export production exceeds the local increase by a factor of 3 and exceeds  $\Delta C^{uptake}(T=100\text{ yr})$  by a factor of 30. In the FULL + EXP simulation the export production increases during fertilization and then drops in succeeding years, so that after 100 years the cumulative change in export production is essentially identical in the fertilized and nonfertilized cases. In other words, the simulation that is the most efficient at removing CO<sub>2</sub> from the atmosphere is also the one that causes the largest reduction in global export production and vice versa.

[38] 2. The net impact of macronutrient addition on export production can be positive over long times. In both of the macronutrient addition runs, about half of the additional production over the first 100 years occurs after year 10 of the experiment.

[39] 3. The bulk of the changes in production do not occur at the fertilization site or in the immediate vicinity but on larger spatial scales. Figure 10 illustrates this for the ONETIME + FULL + BOTTOM, ONETIME + ADD + BOTTOM and the ONETIME + ADD + EXP simulations. It might be thought that the principal effect of changing the nutrient burden of the upper water column would be felt on the edges of the equatorial high production zone. If production were modeled with Michaelis-Menten kinetics, and

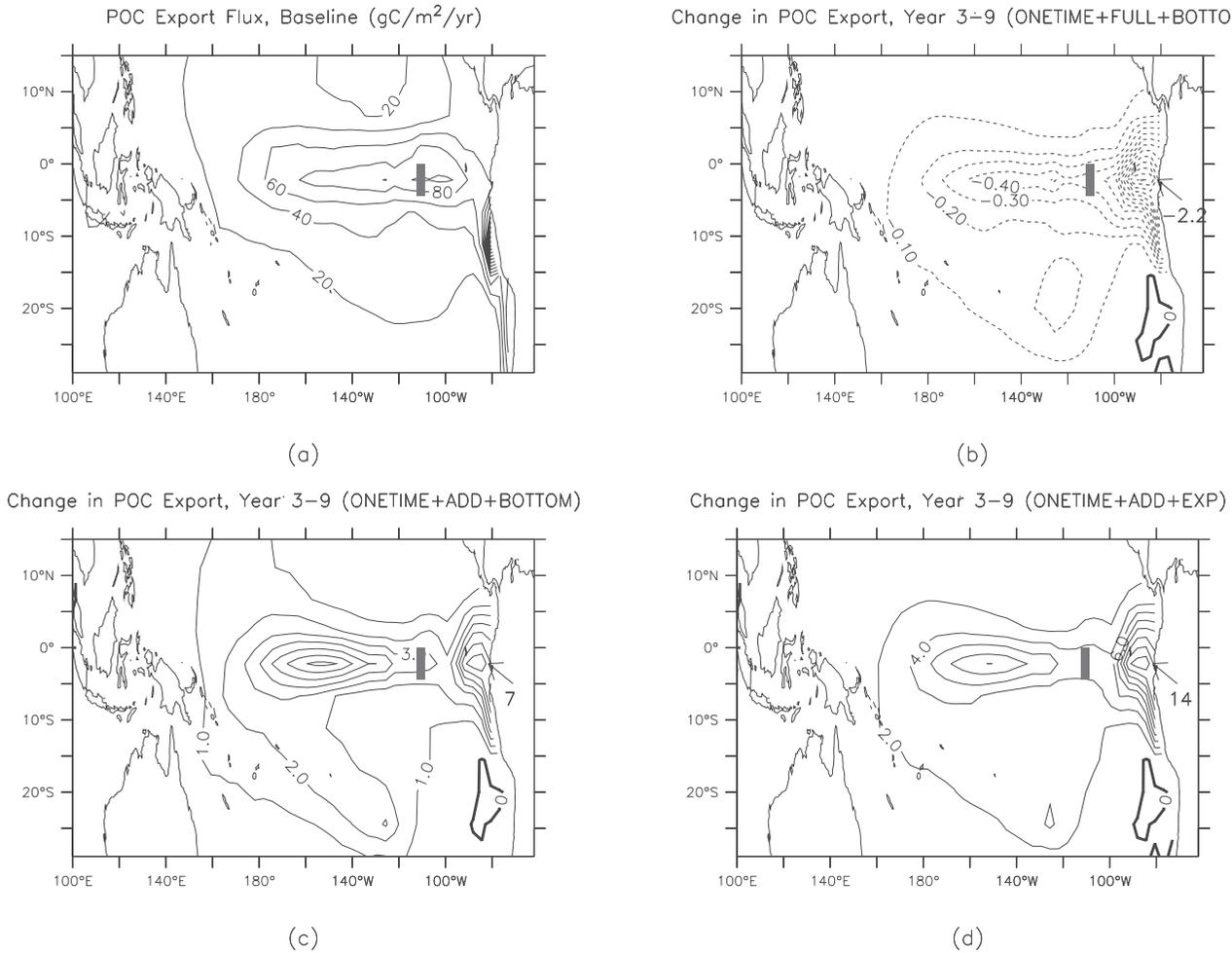
the growth rates were saturated in the upwelling zone, this would likely be true. However, since our models represent production by restoring nutrients to observations, the effect of changing nutrient concentrations in upwelled waters is felt most strongly near the upwelling itself.

[40] An order-of-magnitude estimate of the potential cost of changes in export production for the worst case (FULL + BOTTOM) scenarios can be made as follows. The southeast Pacific region, where a significant portion of the impacts are found in the present study, accounts for almost 20% of global fisheries landings [FAO Marine Resources Service, 1997], with some 15–20 million tons of fish caught each year and a first sale value in the \$10–20 billion range. Subsequent processing would raise the value of this catch substantially. The total export flux predicted by the GCM in this region is round 2 GtC/yr. Suppose we make the (undoubtedly oversimplified) assumption that fisheries landings are directly proportional to export flux. Given that much of the export flux in this region is in fact the result of fecal pellet formation from higher trophic levels, this is not an unreasonable first guess. Then the “value” of one ton of export flux is at a minimum \$10 billion/2 billion tons = \$5/ton. If the ONETIME + FULL + BOTTOM presented here were to accurately describe the response of oceanic biology to changes in nutrient supply, the cost to fisheries over 100 years of sequestering a ton of carbon through micronutrient fertilization of the tropical ocean would be 30 times this or around \$150/ton C sequestered. While it should be recognized that there are huge uncertainties surrounding this estimate, it is important to note that this cost is significant. The cost of separating and capturing

**Table 3.** Change in Particulate Export<sup>a</sup>

Time	$\Delta C_{POC}$ (Site, MtC)	$\Delta C_{POC}$ (Regional, MtC)	$\Delta C_{POC}$ (Global, MtC)	$\Delta C_{POC}$ Over $\Delta C^{uptake}$
<i>ONETIME + FULL + BOTTOM</i>				
1 mo	15.3	14.3	14.6	10.1
1 yr	11.9	7.4	1.1	0.4
10 yr	11.8	6.0	-15.6	-10.1
100 yr	11.6	3.9	-45.9	-31.0
<i>ONETIME + FULL + EXP</i>				
1 mo	16.7	15.7	16.0	10.9
1 yr	13.8	9.1	6.1	2.5
10 yr	13.7	8.6	2.9	4.0
100 yr	13.7	8.4	0.4	1.2
<i>ONETIME + ADD + BOTTOM</i>				
1 mo	71.9	72.1	72.1	20.6
1 yr	77.1	83.2	129.0	1.4
10 yr	77.7	93.2	308.2	2.1
100 yr	79.9	114.3	622.2	6.3
<i>ONETIME + ADD + EXP</i>				
1 mo	85.4	85.6	85.6	24.4
1 yr	97.7	103.7	175.3	1.8
10 yr	99.0	120.1	455.5	2.7
100 yr	102.7	155.1	970.3	8.9

<sup>a</sup>First three columns are  $\Delta C_{POC} = \int_x \int_y \int_t F_{POC}(fert) - F_{POC}(control) dx dy dt$ , where  $F_{POC}$  is particulate export flux. First column shows  $\Delta C_{POC}$  integrated over the fertilization site, second is  $\Delta C_{POC}$  integrated regionally (9°S–9°N, 123°W–108°W) and the third is for  $\Delta C_{POC}$  integrated globally. The right-hand column shows the ratio between  $\Delta C_{POC}(global)$  and  $\Delta C^{uptake}$ , also integrated globally.



**Figure 10.** Change in export flux of POC due to fertilization. Fertilization region is shown by the hatched box at 110W, 2.2 S. All simulations are shown integrated from 3–9 years after the fertilization event. (a) Baseline export flux of POC. (b) Change in export flux of POC, ONETIME + FULL + BOTTOM. (c) Change in export flux of POC, ONETIME + ADD + BOTTOM. (d) Change in export flux of POC, ONETIME + ADD + EXP.

carbon dioxide from flue gases using currently available technology ranges from \$35–\$264/ton C [U.S. DOE, 1999] so that it will be important to consider potential impacts of fertilization on tropical fisheries when evaluating the costs and benefits of fertilization.

### 3.3. Sensitivity to Temporal Scale of Fertilization

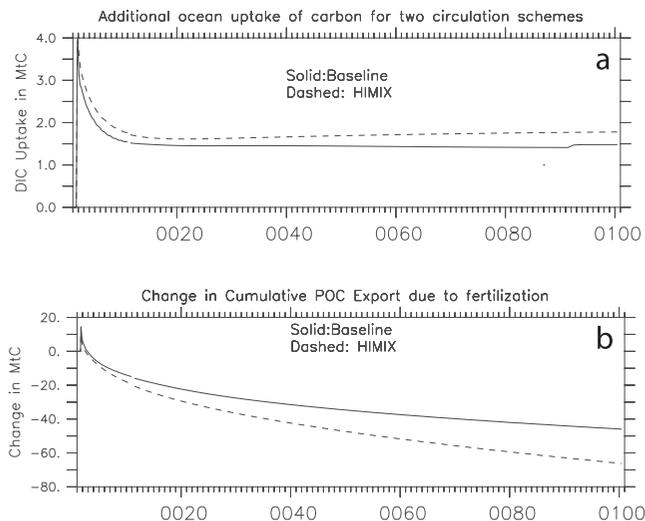
[41] In order to apply these results more widely, it is important to know how they can be scaled up in space and time. We have already considered the spatial scaling of these runs, and shown that it may not be strictly linear. What about scaling in time? Can a ONETIME run be considered a Green’s function calculation that can then be used to characterize results for fertilization in subsequent years, or does fertilizing in one year affect the results in subsequent years? Comparison of the ONETIME and ANNUAL runs (Table 4) reveals that the ANNUAL runs respond in a linear fashion in most cases, with the cumulative additional POC export at the fertilization site, and oceanic  $\text{CO}_2$  uptake almost exactly 100 times the ONETIME case in all the

cases shown (the exception being the  $\text{CO}_2$  uptake in the FULL+EXP simulations). This result might be expected from Figure 4 since the bulk of the transient response of air-sea flux to fertilization occurs in the first 10 years. In

**Table 4.** Cumulative Change in POC Export at the Fertilization Site and  $\Delta C_{\text{global}}^{\text{uptake}}$  (100 yr) in ONETIME and ANNUAL Simulations After 100 Years<sup>a</sup>

Experiment	$\Delta C_{\text{POC}}$ (100 yr, Site)	$\Delta C^{\text{uptake}}$ (100 yr)
ONETIME + PART + BOTTOM	2.58	0.48
ANNUAL + PART + BOTTOM	268.3	50.2
ONETIME + FULL + BOTTOM	11.61	1.49
ANNUAL + FULL + BOTTOM	1158	150
ONETIME + FULL + EXP	13.68	0.34
ANNUAL + FULL + EXP	1353	52.5

<sup>a</sup>Major differences in the fraction of POC flux supplied from the atmosphere occur as a result of changing remineralization and depletion. Differences between this result and Table 1 are because cumulative change in POC export over 100 years is considered, rather than just the change in POC export after 1 month.



**Figure 11.** Sensitivity of the results to changes in circulation scheme for case ONETIME + FULL + BOTTOM. (a) Cumulative change in air-sea CO<sub>2</sub> flux ( $\Delta C_{\text{uptake}}$ ). (b) Cumulative additional export flux of POC ( $\Delta C_{\text{POC}}(T)$ ).

general, knowing the response of the ONETIME simulations allows us to predict a number of key features of the ANNUAL simulations (in particular the amount of additional POC export and the fraction of this export that comes from the atmosphere).

### 3.4. Sensitivity to Physical Circulation

[42] A final question to consider is the importance of the detailed physical representation of the large-scale overturning to the results presented here. As shown in Figure 11a, changes in the circulation scheme make relatively little difference to the amount of carbon sequestered when the exported material goes all the way to the bottom. Despite the fact that run HIMIX has a much more direct connection between the deep ocean and the surface of the tropical Pacific, it still does not allow carbon that goes all the way to the bottom to escape. However, there are larger differences when the changes in biological production are considered (Figure 11b). In the high-mixing simulation, nutrients circulate more rapidly through the upper ocean and tropical production is about twice that in the low mixing case [Gnanadesikan *et al.*, 2002]. When nutrients are removed from the upper ocean, the resulting impact on export production is thus larger than in the low-mixing case.

## 4. Discussion

### 4.1. Sequestration and the Preformed and Remineralized Nutrient Pools

[43] Several interesting questions emerge from the results presented above. Perhaps the most important is why is the response to macronutrient depletion so different from the response to macronutrient addition? The key to the answer, as will be demonstrated below, lies in the different responses of biological cycling to the two perturbations.

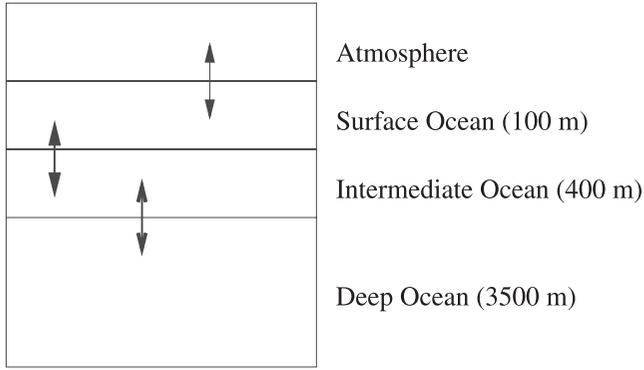
These different responses result in different impacts on the preformed and remineralized nutrient pools.

[44] By definition, the preformed concentration of a substance is the concentration in water leaving the euphotic zone. In a steady unperturbed state, the surface DIC concentration of the ocean is in equilibrium with the pCO<sub>2</sub> of the atmosphere, thus the preformed concentration is effectively a measure of the atmospheric CO<sub>2</sub> inventory. By contrast, remineralized DIC remains isolated from the atmosphere so long as biological productivity continues to remove any remineralized DIC (along with its associated pool of remineralized nutrients) that is transported into the surface ocean and thus into potential contact with the atmosphere. In this paradigm of how the ocean DIC cycle functions, fertilization will only be effective to the extent that it succeeds in shifting DIC from the preformed pool to the remineralized pool, which is what both macronutrient depletion and macronutrient addition do. The difference between these two scenarios is that macronutrient depletion, as simulated in our models, results in a temporary shift of carbon from the preformed to the remineralized pool, while macronutrient addition results in a permanent addition of carbon to the remineralized pool.

[45] Consider first the macronutrient depletion scenario. Here, the temporary monthlong reduction of surface phosphate from its climatological mean to a concentration of near zero (induced by the addition of micronutrients) results in a conversion of phosphate from the preformed to the remineralized pool, along with its associated DIC. However, the instant that the fertilization ceases, the surface concentration of phosphate begins to increase back toward its prefertilization concentration. This occurs because the surface biological production is determined by forcing the model-predicted phosphate concentration back toward its climatological mean, so that there is a drop in POP export in the months succeeding a fertilization experiment. Thus the temporary macronutrient depletion increases the remineralized pool near the bottom of the ocean, but there is a quick compensatory conversion of DIC from the remineralized to the preformed pool in the upper ocean near the fertilization site.

[46] In the macronutrient addition case, the total pool of nutrients in the ocean is “permanently” increased, but the preformed pool is kept constant because of the way that we force model predicted nutrients back to their climatological mean at the surface of the ocean. The additional nutrients thus become a permanent part of the remineralized pool. The additional nutrients in the remineralized pool lead to a permanent shift of DIC from the preformed to the remineralized pool, and thus to a reduction in atmospheric carbon dioxide.

[47] One thing that the foregoing analysis should make clear is that the parameterization of biological processes will have a considerable influence on how the ocean responds to perturbations such as those described in this paper. Our method of forcing nutrients back to their climatological mean immediately after macronutrient depletion ensures that the recovery will be rapid. If, in fact, ocean ecosystems behave in a such a way as to slow the recovery by maintaining the reduced surface nutrient concentrations



**Figure 12.** Schematic of four-box model.

over an extremely long period of time, then the atmospheric  $\text{CO}_2$  uptake will be larger. Conversely, if in the macronutrient addition case, the biological production is not able to consume all the additional nutrients (either because of loss of the micronutrients added at the same time or because of changes in the ocean biology), then some portion of the added macronutrients will shift from the remineralized to the preformed pool, and the efficiency of the fertilization will drop.

#### 4.2. A Simple Box Model

[48] This connection between long-term biological impacts and sequestration can be illustrated in a very simple model, shown in Figure 12, which consists of three oceanic boxes and one atmospheric box. We prescribe a gas exchange coefficient between the surface box and the atmosphere, and fix the temperature of all the ocean boxes to  $10^\circ\text{C}$ , the alkalinity in the ocean to  $2291 \mu\text{mol/kg}$ , the atmospheric  $p\text{CO}_2$  to  $280 \mu\text{atm}$ , the exchange between the surface and intermediate boxes to  $150 \text{ Sv}$  and the exchange between the intermediate and deep boxes to  $30 \text{ Sv}$ . Biological production is allowed to remove carbon and phosphate from the surface ocean and export it to depth. For normal production, 90% of the exported carbon and phosphate are deposited in the intermediate layer with the remainder deposited in the deep layer.

[49] The expected behavior of this model is as follows. The change in the carbon content of the surface ocean due to gas exchange is

$$\begin{aligned} \frac{\partial}{\partial t} \text{DIC} &= k_g * A * (p\text{CO}_2^{\text{atm}} - p\text{CO}_2^{\text{oc}}) * \frac{\partial \text{DIC}}{\partial p\text{CO}_2} \\ &= k_g * A * \Delta p\text{CO}_2 * B, \end{aligned} \quad (8)$$

where  $k_g$  is the gas exchange coefficient,  $A$  is the surface area of the ocean,  $p\text{CO}_2^{\text{atm}}$  and  $p\text{CO}_2^{\text{oc}}$  are the partial pressures of  $\text{CO}_2$  in the atmosphere and ocean respectively, and  $B$  is a buffer factor. Suppose that some change in carbon content is introduced into the system. At equilibrium, the change in  $p\text{CO}_2$  resulting from this perturbation in the carbon content  $\delta p\text{CO}_2$  must be the same in the atmosphere and the ocean. Thus for a net removal of  $\delta C$  from the combined atmosphere and ocean system,

$$\delta p\text{CO}_2 = \delta C * (2.123 \times 10^6 \text{ Pg/atm} + B * V)^{-1}, \quad (9)$$

where  $V$  is the volume of that portion of the ocean which has come into equilibrium with the atmosphere. Equilibration happens quickly for the surface ocean, but more slowly for the deep ocean. Thus on short timescales,  $B * V$  is small, and the majority of a perturbation in the carbon content of the surface ocean and atmosphere would be expected to be reflected in the atmosphere. On longer timescales, the effect of intermediate and deep waters would be expected to come into play, and the effect on atmospheric concentrations would drop.

[50] We ran four experiments with this model.

[51] 1. The first was direct injection: We removed 1 Gt of carbon from the atmosphere and injected it into the deep ocean.

[52] 2. The second was macronutrient depletion with no impact on production: We removed 1 Gt C and a proportional amount of phosphate (in the same stoichiometric ratio as in the GCM) from the surface layer and injected it into the deep layer, but kept the biological production constant at all times other than the single sequestration event.

[53] 3. The third was macronutrient depletion with an impact on production: The same transfer of phosphate and carbon is made as in the second case, but the production is made proportional to the nutrient content of the surface ocean- analogous to the way in which production is parameterized in the GCM.

[54] 4. The fourth was macronutrient addition: 1 Gt C is removed from the surface waters, 0.9 Gt C is added to the intermediate waters, 0.1 Gt C is added to the deep waters, and phosphate is added to the intermediate and deep waters but not removed from the surface water (simulating the effect of macronutrient addition). In this experiment, surface production of organic matter is left proportional to the nutrient concentration in the surface water.

[55] Figure 13 shows the results of these box model simulations. Results are shown on a logarithmic time axis, relative to the fertilization event. Several important points emerge from this plot:

[56] • If macronutrient depletion at one time has no effect on the productivity at subsequent times, its efficiency over long times is essentially identical to that associated with direct injection of  $\text{CO}_2$  into the deep ocean.

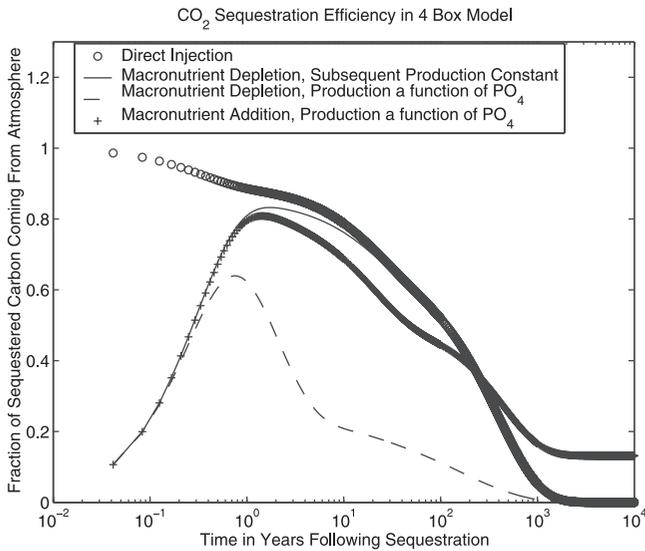
[57] • If macronutrient depletion at one time does cause a reduction in productivity at subsequent times, the fraction of POC export coming from the atmosphere is much smaller.

[58] • Macronutrient addition has an efficiency that is similar to that associated with deep injection at short timescales. At long timescales, its efficiency is much greater, as carbon is permanently added to the remineralized pool in the deep ocean.

#### 4.3. Biological Compensation With a GCM

[59] Why is the exact formulation of the biological cycling scheme of such importance? Examination of the details of one GCM run gives us some insight. The change in the carbon balance of the surface ocean between the fertilization and control runs can be described by the following terms:

$$\frac{\partial}{\partial t} \Delta \text{DIC} = \Delta \text{Biology} + \Delta \text{Transport} + \Delta \text{Gas Exchange}. \quad (10)$$



**Figure 13.** Results of four experiments with a four-box model involving shifts of 1 GtC. Vertical axis shows the fraction of carbon coming from the atmosphere. Lines are as follows. Circles denote direct injection. Initially all carbon comes from atmosphere, but over a few years the ocean outgasses carbon to bring the atmosphere into equilibrium with the surface ocean. Over 100 years, as more intermediate water is brought to the surface, the fraction drops yet further. Over a few thousand years the initial equilibrium is restored. The solid line denotes macronutrient depletion, which has no subsequent effect on production. Once equilibration with the atmosphere occurs, this case is essentially identical to the direct injection case. The dashed line denotes macronutrient depletion with subsequent production dependent on  $\text{PO}_4$  concentration. This case has a much lower efficiency and is directly analogous to the macronutrient depletion case. Pluses denote macronutrient addition with subsequent production a function of  $\text{PO}_4$  concentration. This case is associated with a much higher efficiency, close to direct injection. Unlike direct injection, it results in permanent storage of carbon in the deep ocean.

Figure 14 shows these terms, integrated over the mixed layer globally, for three time periods in run ONETIME + FULL + BOTTOM, during which three separate regimes emerge: the first month of fertilization, the succeeding 3 months, and the remainder of the first 5 years. During the fertilization event itself, there is a reduction of DIC in the surface layer ( $-25.6 \text{ MtC}$ ), driven by the biological drawdown of carbon ( $-41.9 \text{ MtC}$ ). This drawdown is compensated to a certain extent by vertical transport of carbon and nutrients ( $+14.4 \text{ MtC}$ ). The total effect of gas exchange ( $+1.47 \text{ MtC}$ ) is quite small in contrast. Over the succeeding three months, which may be referred to as a “rebound” period, there is a significant rise in the DIC content of the surface ocean ( $+25.7 \text{ MtC}$ ), which is essentially due to the biological terms ( $+26.0 \text{ MtC}$ ). This change in the biological term in the carbon balance reflects a reduction in primary production ( $-20 \text{ MtC}$ ). In the remainder of the first 5 years, a regime is seen in which the

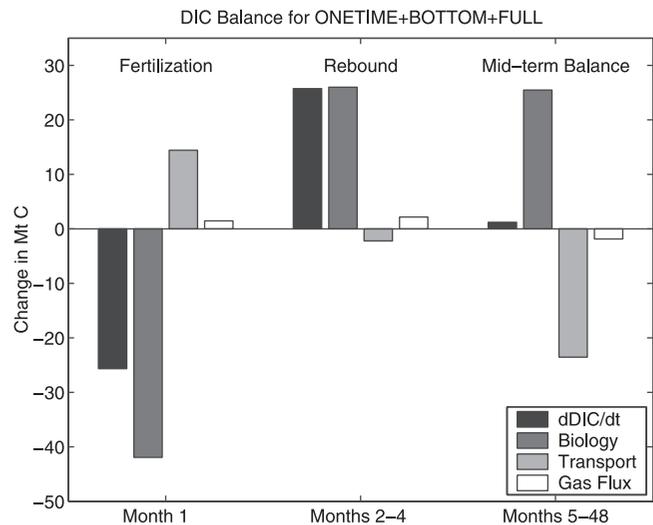
primary balance is between an increase in the biological term (corresponding to a decrease in primary production) and a decrease in the vertical transport of carbon (resulting from the export of this carbon to the deep ocean).

[60] In combination, the box models and term balances show that the formulation of biological cycling plays a key role in evaluating both the efficiency and impacts of ocean fertilization on carbon cycling. Insofar as reducing the supply of nutrients to the surface ocean would be expected to reduce the productivity of the surface ocean, macronutrient depletion with deep remineralization will not be particularly efficient at sequestering atmospheric carbon dioxide and subsequent impacts on fisheries might be large.

#### 4.4. Carbon Fertilization and Verification of the Oceanic Carbon Uptake

[61] Our results have important implications for how the atmospheric  $\text{CO}_2$  drawdown resulting from fertilization might be verified. In particular

[62] 1. Direct measurements of  $\Delta C^{\text{uptake}}(T)$  by measuring changes in the air-sea flux are not likely to work. While it might be possible to track the initial atmospheric  $\text{CO}_2$  drawdown over the first year following nutrient depletion, it would be very difficult to track the large-scale compensation that occurs in succeeding years over large spatial



**Figure 14.** Globally integrated carbon balance for the ONETIME + DEEP + FULL model. Integrations are made over the top 85 m. The bar plot shows that there are three periods associated with this fertilization event. During the event (first set of bars), the reduction in DIC is largely driven by an increase in the biological source minus sink term, offset in part by vertical transport. During the following 3 months, there is a rapid increase in upper ocean carbon driven by the source minus sink terms. As in the box model, the fact that biological productivity depends on nutrient concentration leads to a reduction in the biological uptake of carbon in the months following fertilization. In the remainder of the first 5 years after the fertilization event, a balance is established where reduced upwelling of nutrients and carbon is balanced by reduced production and export of nutrients and carbon.

scales. As seen in Figure 5 the drop in  $\Delta C^{\text{uptake}}(T)$  resulting from macronutrient depletion is associated with a pattern of fluxes covering the entire tropical Pacific. In Figure 8, the drop in  $\Delta C^{\text{uptake}}(T)$  between  $T = 10$  years and  $T = 100$  years results from a pattern of outgassing covering the entire globe, with substantial compensation occurring poleward of  $40^\circ\text{S}$ . The amplitude of the fluxes involved is very small (compare the range of  $0.3 \text{ mol/m}^2/\text{yr}$  in Figure 5c with the full range of  $60 \text{ mol/m}^2/\text{yr}$  in Figure 5a). Temporal variability and the trend due to continued anthropogenic  $\text{CO}_2$  additions would dwarf the magnitude of this signal.

[63] 2. Indirect estimates of carbon sequestration based on particle export from the euphotic zone will also be insufficient to estimate  $\Delta C^{\text{uptake}}(T)$ . Although particle export from the mixed layer can be estimated with sediment traps and by using the concentration of thorium in the upper ocean [Buesseler, 1991] such measurements are not in and of themselves sufficient to establish how much carbon will be removed from the atmosphere. As can be seen in Table 2, there is a huge difference in the atmospheric  $\text{CO}_2$  drawdown associated with nutrient depletion if the remineralization of POC occurs at shallow depths or deep in the ocean. Although the macronutrient addition scenarios (Figure 7) show little sensitivity to remineralization profile, these simulations implicitly assume that any iron added with the macronutrients to produce additional production remains associated with these nutrients for all time. This assumption is unlikely to be true (if macronutrients and iron were tightly coupled to each other, HNLC regions would not be iron-limited).

[64] A further complication involves the role of dissolved organic matter. In the present simulations, the assumption has been made that the lifetime of DOM is short (6 months). As a result, the export of DOC to the deep ocean is relatively limited. It is unclear that this is the case in the real world. For example, measurements of DOM made by Doval and Hansell [2000] indicate that DOC oxidation at depth in the Pacific can be quite significant. This in turn implies that some fraction of DOC produced in the surface layer could have a fairly long lifetime. Moreover, it is well known that certain species of DOC are very refractory and can persist for millennia [Hansell et al., 1997]. Any production of such long-lived DOC would be functionally equivalent to exporting carbon to the ocean bottom, since it would be thousands of years before the carbon thus sequestered could come into contact with the atmosphere. While it might be concluded that models might be used to extrapolate from export flux to carbon uptake, such a conclusion would be premature given the gross uncertainties as to the details of how remineralization and DOC production would actually function were the real ocean to be fertilized.

## 5. Discussion and Conclusions

[65] The central point we wish to emphasize is that ocean fertilization cannot be thought of as a simple story that ends when the products of a bloom induced by fertilization sink out of the surface layer. Several points are particularly important.

[66] 1. The efficiency of tropical micronutrient fertilization that results in the depletion of surface nutrients can be

very low if the nutrient depletion reduces production at other places and times. Even under the best of circumstances (assuming all POC to go all the way to the ocean bottom), only about 10% of the initial POC export eventually comes out of the atmosphere after 100 years.

[67] 2. Much of the initial drawdown of atmospheric  $\text{CO}_2$  takes place not at the fertilization site, but in the surrounding area. There are also significant compensating fluxes that reduce this drawdown over timescales of years or decades on spatial scales of thousands of kilometers. Direct measurement of these compensating fluxes is not likely to be achievable.

[68] 3. The efficiency of patchy nutrient depletion depends very sensitively on the profile of remineralization. If remineralization occurs at shallow depths, very little (2%) of the POC initially exported ends up coming out of the atmosphere after 100 years. This also poses a challenge to verifying sequestration due to nutrient depletion.

[69] 4. Micronutrient fertilization resulting in nutrient depletion can reduce biological productivity over the long term. If it is effective at producing atmospheric  $\text{CO}_2$  drawdown by exporting POC to great depths, it also exports phosphate to great depth. This means less phosphate is available for tropical production. Over century timescales, the reduction in export production can be up to 30 times the atmospheric  $\text{CO}_2$  drawdown resulting from fertilization.

[70] 5. Addition of both macronutrients and micronutrients can have a distinctly different behavior from macronutrient depletion. The efficiency of fertilization is much higher, while the pattern of air-sea  $\text{CO}_2$  flux and the sensitivity to remineralization profile are substantially different. The results, however, depend sensitively on the assumption that once macronutrient is added to the system, it remains active along with its associated micronutrients.

[71] 6. The location at which changes in export production occur may be a long way from the fertilization site. Large-scale, long-term consideration of the effects of fertilization is vital if the true environmental impact is to be estimated.

[72] 7. Not all aspects of small-scale micronutrient fertilization experiments are likely to scale up in space. At small scales, lateral diffusion can supply nutrients to a patch in a way that will not be realistic at larger scales.

[73] Taken together, these results raise serious questions about the usefulness of micronutrient fertilization for reducing atmospheric carbon dioxide. Atmospheric  $\text{CO}_2$  drawdown associated with micronutrient fertilization is likely to be relatively inefficient, highly dependent on the remineralization profile, and may have disproportionate long-term impacts on export production. Moreover, verifying the amount of carbon sequestered is likely to be very difficult (if not impossible) because of the large space and timescales involved.

[74] While idealized macronutrient addition does appear to have a higher efficiency within our simulations, it also shares some of the same problems as macronutrient depletion. In particular, the large space and timescales associated with compensatory air-sea  $\text{CO}_2$  fluxes make verification of sequestration very difficult. Moreover, we have presented a best-case scenario for macronutrient addition in which the

micronutrients which are implicitly added at the same time as the macronutrient remained associated with the macronutrients for all time. In the real ocean it is unlikely that macronutrients added at one point in time will remain active (that is to say associated with sufficient iron to drive production) while being repeatedly cycled through the tropical pycnocline. The very fact that the tropics are iron-limited despite considerable recycling of nutrients suggests that iron is preferentially removed from the system relative to macronutrients. Additionally, as noted by *Orr and Sarmiento* [1992], adding macronutrients might be expected to increase ocean anoxia, with resultant increases in denitrification. Increased denitrification would in turn be expected to result in increased production of nitrous oxide (a powerful greenhouse gas) and nitrogen limitation in upwelling regions. Neither of these effects was considered in this paper.

[75] Understanding the net impact of fertilization on air-sea carbon flux and biological cycling of carbon within the ocean will require focused research into the following questions:

[76] • What are the nonlocal effects of fertilization? Will, for example, iron fertilization result in a decrease in production at and around the fertilization site in succeeding months? One route to answer this question will be more sophisticated modeling studies. For example, *Barber and Chai* [2001] find that they are able to reproduce to first order the chlorophyll increase and carbon drawdown in a more sophisticated biological model involving two classes of phytoplankton (small plankton and diatoms), two classes of zooplankton and two nutrient currencies (silicon and nitrogen). In their model, the effect of iron fertilization is parameterized in terms of observed changes on diatom growth rate. It should be possible to build on this model to evaluate long-term, nonlocal effects of iron fertilization. Additional data will also be required to evaluate the extent to which mechanisms in models such as Barber and Chai's are realistic, and to realistically include a cycle of iron as well as silicon and nitrogen.

[77] • How does fertilization affect the location of remineralization? *Armstrong et al.* [2001] have suggested that export to the deep ocean is controlled by the production of ballast materials such as silicate and carbonate tests. *Hutchins and Bruland* [1998] and *Takeda* [1998] have demonstrated that iron fertilization can lead to a shift in the C:Si ratio in diatoms, resulting in a lower production of ballast per unit carbon. Such an effect might offset some portion of the additional fixation of carbon associated with fertilization by changing the profile of remineralization. In order to estimate whether such an effect is likely to be important in the real world, fundamental measurements to better characterize the processes involved in remineralization will be necessary.

[78] • What is the fate of fertilizing agents (in particular iron) away from the surface layer? To what extent does iron behave like a nutrient (as its vertical profile in the Equatorial Pacific would suggest) and to what extent do internal sinks such as adsorption onto sinking particles make it behave like a highly absorptive metal? What are the natural time and length scales for such behavior? In order to answer this

question, fundamental studies to understand remineralization in the upper ocean need to be conducted.

[79] • What is the role of dissolved organic matter in carbon export? The simulations presented here stipulate that all organic matter produced by fertilization has a short life time, essentially limiting the possibility that it can be exported. More simulations considering the effects of different dissolved organic matter cycles need to be carried out and comparisons made with data in order to better estimate this effect.

[80] Two recently proposed programs offer opportunities to address key aspects of these gaps in understanding. The proposed OCTET program ([Alpha.msrc.sunysb.edu/octet](http://alpha.msrc.sunysb.edu/octet)), with its proposed focus on remineralization has the potential to address the final three gaps. The proposed Ecological Dimensions of Climate Change program (EDOCC, <http://picasso.oce.orst.edu/ORSOO/EDOCC>), with its emphasis on how ecosystems change as forcing and routes of nutrient supply vary, could play a critical role in answering the first set of questions. In order to do this, however, these programs will need to do three things. First, the role of iron cycling will need to be incorporated into the design of these programs from the beginning, so that collection of iron data on the same spatial scales as the WOCE hydrographic program can be included as part of future hydrographic work. Second, studies will need to examine the vertical structure of nutrient cycles. Finally, measurements of interannual variability in ecosystems that tie biological variability to changes in iron supply could be very important for estimating the impact of fertilization on a range of ecosystems.

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