Atmospheric carbon dioxide in a less dusty world

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[1] The availability of iron exerts a significant control on primary production and the export of organic matter over large areas of the ocean, especially those far from land sources. We explore the regulation of the global soft tissue pump of carbon and atmospheric CO_2 by the atmospheric delivery of iron in a three-dimensional ocean circulation and biogeochemistry model. There is only a small change in atmospheric CO_2 when the aeolian iron source is increased several fold but a significant increase in response to a reduction in the aeolian iron source. This strong asymmetry suggests a positive feedback, amplifying an increase in atmospheric CO_2 if a warmer world is also less dusty.
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1. Introduction

[2] Carbon is sequestered in the deep ocean, reducing atmospheric CO_2, in part due to the photosynthetic formation of organic material in the surface ocean which is transported to depth, much as gravitationally sinking detritus, where most is remineralized. This process is termed the soft tissue pump of carbon. Iron enrichment experiments in the field clearly demonstrate the enhancement of primary production by the addition of iron in the high nutrient, low chlorophyll (HNLC) regions [*Martin et al.*, 1994; *Coale et al.*, 1996; *Boyd et al.*, 2004]; the Southern Ocean, Equatorial Pacific and sub-Arctic Pacific. Recent evidence also indicates an associated enhancement of the flux of sinking, organic particles [*Boyd et al.*, 2004; *Buesseler et al.*, 2005].

[3] Data from ice cores [*Petit et al.*, 1999] and sediments [*Rea*, 1994] suggest that the aeolian iron supply during the Last Glacial Maximum (LGM) was significantly higher than the present day. *Martin* [1990] hypothesized that this increased iron supply could have fertilized the oceans and increased the efficiency of the soft-tissue biological pump. Idealized box models of the ocean-atmosphere carbon cycle [*Knox and McElroy*, 1984; *Sarmiento and Toggweiler*, 1984; *Toggweiler*, 1999] indicated that modulation of the soft tissue pump by iron limitation or other mechanisms could drive significant variations in atmospheric CO_2. They have suggested that elimination of the soft-tissue pump could increase pCO_2 by almost 200 ppmv while its enhancement might reduce pCO_2 by up to 100 ppmv, sufficient to explain the recorded glacial drawdown. More recent, three-dimensional models [*Bopp et al.*, 2003; *Archer et al.*, 2000] however, find that the enhancement of the soft-tissue pump by iron fertilization is considerably less effective, reducing atmospheric CO_2 on the order of 10 ppmv. Sedimentary proxies of ocean productivity do not provide a simple picture of globally increased export production at the LGM but indicate more complex and regional changes in productivity and nutrient utilization [*Bopp et al.*, 2003; *Sigman and Boyle*, 2000] which may also be driven by other factors, including changes to ocean circulation or ice cover [e.g., *Toggweiler*, 1999].

[4] Discussions of the role of iron have centered around fertilization of the HNLC regions in a windier, drier and dustier glacial climate. There have, however, been periods in geological history when climate has been warmer, probably less windy and with a stronger hydrological cycle. Such conditions would favor reduced dust transport and lower iron supply to the oceans. Near-future climate change scenarios also suggest a warmer, wetter climate [*Intergovernmental Panel on Climate Change* (*IPCC*), 2001]. Studies of atmospheric dust loading in such a future climate suggest a response which ranges from an increase of 20% [*Tegen et al.*, 2004] to a decrease of 60% [*Mahowald and Luo*, 2003] relative to present-day, depending on assumptions about anthropogenic effects, land use change and CO_2 fertilization of terrestrial plants.

[5] Here we do not attempt to recreate past or future climates. Instead we use a highly idealized framework, including an ocean with constant circulation, to examine the sensitivity of atmospheric pCO_2 and the biological storage of carbon in the ocean to both increased and decreased atmospheric supply of iron-rich dust.

2. Biogeochemical-Circulation Model Results

[6] We employ a global, coarse resolution configuration of the MIT ocean circulation and biogeochemistry model with explicit representation of the cycles of iron, phosphorus and carbon, coupled to a well mixed atmospheric reservoir of CO_2. The physical model is forced with observed, seasonal climatologies of heat, freshwater and momentum fluxes at the sea surface. We use a simplified parameterization of the export of organic matter from the sunlit surface ocean where production can be limited by the availability of Fe, PO_4 and light [*Parekh et al.*, 2005; *Dutkiewicz et al.*, 2006]. We explicitly model the oceanic iron cycle following *Parekh et al.* [2004, 2005]. The scavenging of iron onto sinking particles in the ocean is guided by the scavenging rates observed for isotopes of thorium [*Bacon and Anderson*, 1982]. We assume equilibrium partitioning between free and complexed iron assuming a fixed, uniform ligand concentration (here 1 nM) and conditional stability coefficient guided by laboratory and

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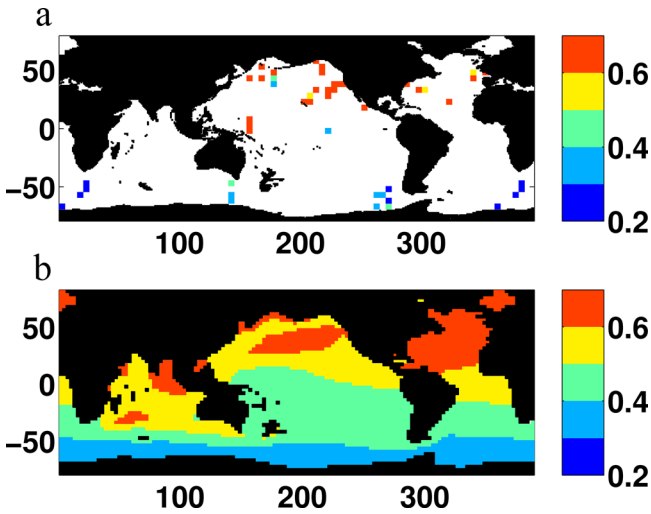

Figure 1. Total dissolved iron concentration at 1000 m (*nM*): (a) Observations are compiled from numerous published sources [*Parekh et al.*, 2005], (b) modelled. The model captures the basin scale deep-water horizontal gradients (high in the northern basins, and low in the Southern Ocean) which regulates the delivery of iron to the remote southern ocean.

field observations [*Rue and Bruland*, 1997]. In the modern, control configuration, integrated for 3500 years, the aeolian iron source is prescribed from modeled estimates of atmospheric dust transport [*Luo et al.*, 2003] assuming that iron is 3.5% by weight of dust and that 1% of that iron becomes bio-available in the surface ocean [*Jickells and Spokes*, 2001]. Modelled phosphate and iron distributions capture the observed, large scale horizontal and vertical gradients [see also *Parekh et al.*, 2005]. Surface phosphate is high in the upwelling regions, particularly in the HNLC regimes where iron is the limiting nutrient. Iron is replete over much of the surface Atlantic and Northern Indian Oceans, where there is significant aeolian deposition, and lowest over the Equatorial Pacific, South Pacific and Southern Oceans. The horizontal gradients of iron in the deep waters (Figure 1) are controlled by the balance between regeneration of organic matter, scavenging and complexation. The concentration of iron in the upwelling deep waters is important, especially in regions far from land dust sources [*Le Fèvre and Watson*, 1999; *Aumont et al.*, 2003; *Parekh et al.*, 2005].

[7] Sensitivity studies (Table 1) with the model are initialized with the modern, control state and the amplitude of the aeolian dust flux to the ocean is modulated by a fixed factor (0.0 "no dust"; 0.1; 0.5; 1.0 "modern control"; 1.5; and 2.0) and integrated for a further 1000 years. In one further case we impose the aeolian dust deposition estimated for the LGM with an atmospheric model [*Mahowald et al.*, 1999] in which both pattern and magnitude are altered. In that case the globally integrated aeolian iron source is increased more than five-fold. In all cases the modeled ocean circulation is the same as in the modern control. Most of these experiments are close to a new equilibrium after 1000 years; the no dust case approaches, but is not yet at equilibrium.

Table 1. Summary of Model Sensitivity Studies

Experiment	Dust Flux	Δ pCO_2 (After 1000 Years)
EXP0	modern	0
EXP1	No dust	181
EXP2	0.1 × modern	95
EXP3	0.5 × modern	14
EXP4	1.5 × modern	−2
EXP5	1.9 × modern	−3.5
EXP6	2.0 × modern	−4
EXP7	LGM	−8

[8] Increasing the aeolian dust source in our model drives only a small drawdown of atmospheric CO_2, concurring with other recent model studies [*Archer et al.*, 2000; *Bopp et al.*, 2003]. LGM dust deposition leads to a decrease of only 8 ppmv (Figure 2). The focus on understanding glacial drawdown of CO_2 means that little attention has been paid to the scenario of a decreased aeolian source. Here we find that a corresponding five-fold reduction drives an increase in atmospheric CO_2 of about 75 ppmv (inferred from Figure 2); almost an order of magnitude greater sensitivity in this direction. Why is the response so asymmetric?

2.1. Increased Aeolian Iron Flux

[9] The impact of an increased iron source is damped by a combination of factors: For instance, the upwelling, iron-limited, HNLC zones respond with increased productivity, driving down local surface phosphate concentrations and reducing the lateral transfer of phosphate to the neighboring regions where productivity therefore decreases [*Dutkiewicz et al.*, 2005]. A more important damping factor, though, is the regulation that the complexing ligand plays in the cycling of the iron, by protecting it from scavenging. The amount of ligand controls the concentration of bio-available iron, particularly in upwelling water which is the major source of iron to the surface ocean in remote regions [*Le Fèvre and Watson*, 1999; *Aumont et al.*, 2003; *Parekh et al.*, 2005]. As the aeolian source of iron increases, so too does the deep ocean concentration and the upwelling source to the HNLC regions. If the ligand saturates, the upwelling iron concentration no longer increases significantly in response to higher iron inputs. In this limit, ocean productivity and the biological pump become considerably less sensitive to further increases in dust deposition. In this model we have imposed the total ligand concentration at 1 nM. Little is known about the sources, sinks and cycling of the organic ligands but it is possible that they are side-

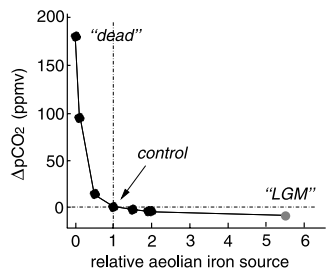

Figure 2. Sensitivity of modelled pCO_2 to the dust-borne, aeolian iron source. Δ pCO_2 (ppmv) is the change in atmospheric pCO_2 relative to the modern control after 1000 years with modified dust source. The response is very asymmetric with low sensitivity to increasing iron supply and a strong response to a decrease.

rophores, the production of which may increase as a function of increased dust flux [*Rue and Bruland*, 1997]. In an additional LGM experiment with a concurrent increase in total ligand concentration to 2 nM, reduction of atmospheric pCO_2 is enhanced by an additional 6 ppmv.

[10] However, even in the LGM case, surface Southern Ocean iron concentrations approach 0.5nM, yet phosphate is not fully utilized. What is preventing further drawdown of both phosphate and iron? High latitude productivity is also significantly regulated by seasonal mixing and light dynamics. In widespread regions of the Southern Ocean, where there is a significant upwelling of macro-nutrients, there is a very short growing season, during which persistently deep mixed layers can limit productivity [*Mitchell et al.*, 1991; *van Oijen et al.*, 2004]. In this model, productivity and macro-nutrient utilization [*Sigman and Boyle*, 2000] over wide areas of the high latitude North Pacific and Southern Oceans are ultimately limited by light and mixing dynamics [see also *Dutkiewicz et al.*, 2006].

2.2. Decreased Aeolian Iron Flux

[11] In strong contrast, reducing the aeolian source of iron relative to the modern control results in a significant increase in atmospheric pCO_2. There are no mechanisms limiting the response in this direction. As the atmospheric source is reduced, iron stress becomes more and more significant on the global scale, reducing export production, nutrient utilization efficiency, and the soft tissue pump of carbon. Over hundreds to thousands of years the deep ocean is depleted of iron. Carbon that was previously sequestered in the deep ocean by the soft tissue pump is slowly expelled to the atmosphere as this water is brought to the surface. Similarly much of the phosphate upwelling to the surface will not be utilized and lateral transfer to other regions of the ocean will lead to a more homogenized surface phosphate distribution [*Dutkiewicz et al.*, 2005] and higher preformed phosphate. The limit of no external iron source leads to an ocean carbon cycle that is driven purely by the solubility pump.

[12] In this "dead ocean" limit, the increase in atmospheric pCO_2 in the three-dimensional model is broadly consistent with expectations from box models [*Knox and McElroy*, 1984; *Sarmiento and Toggweiler*, 1984; *Toggweiler*, 1999]. This result, however, contrasts with the study of *Bopp et al.* [2003] who found only a 3 ppmv increase after eliminating the atmospheric dust source. *Bopp et al.* [2003] employ the model of *Johnson et al.* [1997], in which the loss to scavenging is represented as a Newtonian damping of dissolved iron concentration toward a uniform, specified concentration: scavenging = $-k_{sc}([Fe] - [Fe_{sol}])$, where k_{sc} is a scavenging rate, and $[Fe_{sol}] = 0.6\ nM$. $[Fe_{sol}]$ could be considered as the concentration of a saturated organic ligand. In this parameterization, when $[Fe] < [Fe_{sol}]$ scavenging ceases with the consequence that $[Fe]$ at depth never drops below 0.6 nM, even when the external aeolian iron supply is completely shut down. We parameterize scavenging as a function of the free iron concentration, Fe', and assume thermodynamic equilibrium between free and complexed forms of iron [*Parekh et al.*, 2004], therefore allowing horizontal gradients of iron in the deep ocean (Figure 1). This form allows scavenging to continue even at very low dissolved iron concentrations since a fraction, albeit very small, of dissolved iron is always in the free iron pool which may be scavenged. We argue that the dynamic representation of complexation and scavenging employed here is more appropriate for this scenario.

3. Discussion

[13] Our results suggest that while an increase in the aeolian iron source seems likely to have had only a small impact on atmospheric CO_2 in glacial periods, there may be a very significant response if the atmospheric source were to be reduced. The response of the oceanic storage of CO_2 to increased dust flux is tempered by compensating regional changes in productivity, modulated by macro-nutrient transport, and by the availability of organic ligands which complex iron and protect it from scavenging. However most importantly, light rather than iron, appears to be the major control on biological productivity and CO_2 draw-down in the high latitude regions [*Dutkiewicz et al.*, 2006] which control, to a large extent, the mean preformed nutrient concentration of the deep ocean. The response to decreased dust flux does not have similar strong controls – the limit is an abiotic ocean where all the biogenic carbon is lost to the atmosphere.

[14] The paradigm for a glacial climate is a cooler atmosphere, with stronger meridional temperature gradient, stronger winds, and a more arid and dustier world. Conversely we expect warmer, or equable, climates to have a moister atmosphere, with weaker meridional temperature gradient and weaker zonal winds [e.g., *Farrell*, 1990]. These factors could reduce the transport of dust to the ocean regions far from land, driving up atmospheric carbon dioxide and leading to a positive feedback between climate, carbon and iron cycles during the transition to a warmer climate. Such a feedback may have contributed to the shift to equable climates in earth history. More significantly, recent model studies [*Mahowald and Luo*, 2003] indicate that atmospheric dust loading could decrease in the forthcoming decades (though *Tegen et al.* [2004] suggest a possible increase). If the present warming trend continues over the next decades [*IPCC*, 2001], and if dust delivery is reduced, aggravated iron stress could amplify the anthropogenic increase in atmospheric CO_2.

[15] The significantly asymmetric response to changes in the aeolian iron source in our ocean model suggests a significant potential carbon-climate-iron feedback as climate warms relative to today. It is interesting to note that our modern "control" falls at the transition between low and high sensitivities (Figure 2). Is this an accident of our parameter choices or does this reflect the nature of the real modern ocean? We cannot yet answer this question. These results must be viewed in context of the limitations and simplifications of the model based on current understanding of the oceanic iron cycle and fixed ocean circulation. We have not addressed important issues such as other sources of iron (e.g., continental margins), the regulation of nitrogen fixation by iron availability, and modifications of the soft tissue pump by a changing circulation. However, this study has highlighted the need to better understand the nature of the organic ligands which regulate the availability of iron in the oceans (are they siderophores? [*Macrellis et al.*, 2001]), and the importance of the co-limitation of productivity by iron and light in high latitude oceans. These may be particularly important questions to address in understanding

the relationship between aeolian iron supply, carbon and climate.

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