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Impacts of atmospheric nutrient inputs on marine biogeochemistry

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[1] The primary nutrients that limit marine phytoplankton growth rates include nitrogen (N), phosphorus (P), iron (Fe), and silicon (Si). Atmospheric transport and deposition provides a source for each of these nutrients to the oceans. We utilize an ocean biogeochemical model to examine the relative importance of these atmospheric inputs for ocean biogeochemistry and export production. In the current era, simulations with the biogeochemical elemental cycling ocean model suggest that globally, atmospheric Fe inputs could support roughly 50% of the Fe exported from the euphotic zone by sinking organic and inorganic particles. Variations in atmospheric iron inputs strongly impact spatial patterns of phytoplankton growth limitation and the areal extent of the high-nutrient, low-chlorophyll regions. Atmospheric inputs of N, Si, and P have smaller impacts, potentially accounting for 5.1%, 0.21%, and 0.12% of the biogenic export of these elements from the euphotic zone, respectively. Soluble Fe input from the atmosphere is sufficient to support most of the export production in many ocean regions, whether we use a spatially variable aerosol Fe solubility, or a globally constant 2% solubility. Regionally atmospheric N inputs can have significant impacts on marine biogeochemistry, potentially supporting >25% of the export production, an impact that is increasing due to human activities. Atmospheric Si and P inputs have only minimal impacts on marine ecosystem productivity and biogeochemistry, as these inputs are typically quite small relative to the flux of these nutrients from below the euphotic zone.

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1. Introduction

[2] The later part of the twentieth century has seen an unprecedented increase in the human impacts on planet Earth. Industrialization and increased food production for sustaining the requirements of the rapidly growing world population have resulted in widespread changes in the sources and transport pathways of bioactive nutrients such as nitrogen (N), phosphorus (P), iron (Fe) and silicon (Si) [Seitzinger *et al.*, 2005; Duce *et al.*, 2008; Gruber and Galloway, 2008; Jickells *et al.*, 2005; Treguer *et al.*, 1995; Tegen and Kohfeld, 2006; Houghton *et al.*, 2001]. Extensive use of fossil fuels and N and P in fertilizer has already impacted coastal as well as open ocean marine ecosystems due to increased nutrient deposition [Rabalais, 2002; Turner *et al.*, 2003; Duce *et al.*, 2008]. Modeling studies suggest that changes in climate and land use practices over recent decades may have altered dust fluxes and thus, eolian Fe, P and Si inputs to the oceans [Mahowald and Luo, 2003; Intergovernmental Panel on Climate Change (IPCC), 2001].

[3] Anthropogenic activities have enhanced N supply to coastal and open oceans [Naqvi *et al.*, 2000; Nevison *et al.*, 2004; Luo *et al.*, 2007]. The increased N release from land due to agricultural use and fossil fuel consumption has resulted in a change in nutrient supply to the oceans [De Leeuw *et al.*, 2001]. Anthropogenic N inputs in the form of strong acid (HNO₃) and base (NH₃) dissociation products alter surface seawater alkalinity, pH, and inorganic carbon storage, increasing ocean acidification and affecting ecosystems, especially in coastal regions, which could have a significant effect on human populations [Doney *et al.*, 2007]. High rates of anthropogenic atmospheric N deposition to the North Atlantic have been reported in earlier studies [Duce *et al.*, 1991; Prospero *et al.*, 1996; Galloway *et al.*, 2004]. Recent modeling studies have estimated N and P inputs from rivers to global oceans [i.e., Seitzinger *et al.*, 2005; Dumont *et al.*, 2005]. These studies indicate that anthropogenic sources dominate the current dissolved inorganic nitrogen (DIN) inputs, whereas natural sources dominate the dissolved organic nitrogen (DON).

[4] Iron supply in particulate form is primarily from riverine sources, but most of this ends up in coastal sediments [Poulton and Raiswell, 2002]. River inputs of dissolved iron to oceans are relatively small [de Baar and de Jong, 2001], but in certain areas where large rivers discharge to the shelf, their inputs may be substantial [Tovar-Sanchez *et al.*, 2006]. Much of the external input of iron to open ocean regions is

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via eolian dust transport, from the deserts of the world [Jickells *et al.*, 2005]. Continental margin sediments are also a key source of iron [Elrod *et al.*, 2004; Moore and Braucher, 2008, and references therein]. In addition to mineral dust, direct emissions of soluble iron from combustion sources have been found to be important in regions where coal combustion from power plants and industry dominate [Siefert *et al.*, 1999; Chen and Siefert, 2004; Chuang *et al.*, 2005]. Biofuel and biomass burning areas, predominantly in the tropics could also deposit soluble iron to ocean regions close to these sources [Guieu *et al.*, 2005]. Some studies also suggest that volcanoes, anthropogenic, and extraterrestrial sources may deposit significant soluble iron [Jickells and Spokes, 2001; Johnson, 2001; Luo *et al.*, 2008].

[5] Rivers are the primary source of silicic acid to the global oceans [Treguer *et al.*, 1995]. Conley [2002] suggest that Si inputs to rivers have remained constant or even decreased as a result of dam construction over the past decades. Some Si is also transported through the atmosphere via airborne particles. Si is one of the largest constituents of soil-derived mineral aerosol [Tegen and Kohfeld, 2006]. Fly ash produced from industrial burning can also contain Si, with 5–10% dissolving at deposition [Petaloti *et al.*, 2006].

[6] The primary source of phosphorus to oceans is by rivers that bring continental, weathered P in dissolved and particulate phases [Benitez-Nelson, 2000; Compton *et al.*, 2000]. Since most of the riverine particulate P is retained within the continental shelf, this pool is largely not available for open ocean marine biota [Wollast, 1983; Seitzinger *et al.*, 2005; Beusen *et al.*, 2005]. The second significant source is eolian particulate phosphorus [Compton *et al.*, 2000]. Atmospheric deposition via eolian dust particles is particularly important in regions farther from shore, where other inputs of P are small [Benitez-Nelson, 2000]. The main external source of P to open ocean oligotrophic gyres is from dust deposition [Graham and Duce, 1982; Ridame and Guieu, 2002]. In areas remote from major deserts the primary biogenic aerosols, and combustion sources (fossil fuel, biomass burning, and biofuels) dominate P inputs [Olmez *et al.*, 1988; Smith *et al.*, 1979; Mamane *et al.*, 1986; Hildemann *et al.*, 1991; Kleeman *et al.*, 2000]. Other sources of P to marine surface waters include sea salt aerosols [Graham and Duce, 1979] that are prevalent in remote areas (e.g., Southern Ocean, but ultimately come from the oceans) and volcanic aerosols which are significant close to source regions [Frognier *et al.*, 2001]. Mahowald *et al.* [2008] suggest anthropogenic perturbations have increased by almost 25% oceanic PO₄ inputs, with larger changes over some regions.

[7] Several studies have highlighted the significance of the atmosphere as a pathway for transport of nutrients from continents to marine surface waters [Delany *et al.*, 1967; Duce *et al.*, 1991; Prospero *et al.*, 1996] and their critical role in oceanic biogeochemical cycling [Jickells *et al.*, 2005; Paerl and Whittall, 1999]. Researchers have suggested that iron delivered by dust could have stimulated oceanic productivity and may help explain the glacial-interglacial atmospheric pCO₂ transition [Martin and Gordon, 1988]. Past studies suggested that additional nitrogen fixation under increased dust loading contributed to the lower pCO₂ during the Last Glacial Maximum (LGM) [Falkowski, 1997; Broecker and Henderson, 1998; Michaels *et al.*,

2001]. Harrison [2000] proposed that increased atmospheric dust flux during glacial times increased the amount of Si in the ocean's mixed layer. This increase spurred diatoms at the expense of coccolithophores, decreasing the production of calcite and lowering the pCO₂ levels. Paytan *et al.* [2009], suggest that increased atmospheric nutrient loading via anthropogenic aerosols particularly downwind of South and East Asian continents could have toxic effects on phytoplankton, suppressing growth of some species.

[8] Previous studies have estimated regional and global impacts of atmospheric N inputs [Paerl, 1985; Spokes *et al.*, 2000; Duce *et al.*, 2008]. A few studies have examined the impacts of multiple nutrients in atmospheric deposition. Mills *et al.* [2004] suggested that the Saharan dust can be transported thousands of miles across the Atlantic and contribute large quantities of Fe, P and Si to the oligotrophic North Atlantic gyre, and that these nutrients could play a significant role in stimulating primary production by the nitrogen fixing organisms. Regional studies in the Atlantic Ocean suggest that when examining the impacts of atmospheric deposition of Fe on marine productivity, the inputs of atmospheric N and P and the stoichiometric requirements of the phytoplankton need to be considered [Baker *et al.*, 2003, 2007]. Duarte *et al.* [2006] found high rates of atmospheric nutrient inputs in the NE Atlantic and demonstrated a potentially strong impact on primary production and phytoplankton community composition. They also found relatively low inputs of P relative to inputs of C, N, Fe, and Si. Baker *et al.* [2007] examined soluble nutrient inputs along a 10°N transect in the Atlantic, and found that P and Si were supplied at very low concentrations relative to the other nutrients and the nutritional requirements of phytoplankton.

[9] In a previous study, Krishnamurthy *et al.* [2007] simulated the effects of atmospheric inorganic N inputs (NO₃⁻, and NH₄⁺) on ocean biogeochemistry using atmospheric model output from Luo *et al.* [2007] under preindustrial, current and an IPCC-A1FI prediction for 2100 emission levels. The IPCC-A1FI is a fossil fuel intensive case, which assumes a future world with extensive reliance on fossil fuels. Compared to preindustrial conditions, current era inorganic nitrogen inputs over the study period of 64 years modestly increased primary production, sinking particulate organic carbon (POC) export, and net ocean uptake from air-sea CO₂ flux. Atmospheric N inputs in the current era, can account for >20% of the export production in some regions. Increased atmospheric N inputs to otherwise N-limited regions stimulated primary production by small phytoplankton and diatoms, resulting in a reduction in N₂ fixation because diazotrophs are not able to compete as effectively for surface dissolved P and Fe. The study suggested that increasing new nitrogen inputs to the oceans via anthropogenic atmospheric N inputs could be partially compensated by decreased N fixation.

[10] We previously examined the increasing deposition of Fe and N to the oceans since the preindustrial era, including atmospheric sources of Si and PO₄ from mineral aerosols along with inorganic N deposition and a variable solubility aerosol Fe [Krishnamurthy *et al.*, 2009]. Transient simulations were performed to understand the effects of changing atmospheric nutrient inputs on ocean biogeochemistry, air-sea CO₂ exchange and their impact on global carbon cycle

from preindustrial to current era. A modest decrease in atmospheric $p\text{CO}_2$ of $\sim 2\text{ppm}$ was estimated due to the increased iron and nitrogen inputs in the current era compared to the preindustrial conditions [Krishnamurthy et al., 2009]. Inclusion of an improved sedimentary iron source reduces the biogeochemical elemental cycling (BEC) model sensitivity to variations in atmospheric iron inputs relative to earlier studies [Moore and Braucher, 2008].

[11] An overview of the various sources of N, P, Fe and Si to the oceans is presented in this paper. Here, we describe results from global-scale simulations to highlight the importance of the atmosphere as a major pathway for bringing nutrients to the oceans. We quantify for the first time at the global scale the impacts of atmospheric silicon and phosphorus inputs on marine ecosystems. We have also done some additional analysis of previously published simulations of N and Fe deposition to the oceans by Krishnamurthy et al. [2009]. This allows a comprehensive examination of nutrient deposition to the oceans. We assess the relative importance of each of the key nutrients (Fe, N, P, and Si), and estimate the fraction of ocean export production that is potentially supported by atmospheric nutrient deposition. We also examine atmospheric iron inputs under different climate states and assumed aerosol Fe solubilities to emphasize the time varying impacts of atmospheric nutrient inputs on ocean biogeochemistry.

2. Methods

[12] Duce et al. [1991] estimated the dissolved inorganic phosphorus (DIP) input to oceans to be $\sim 0.310\text{ Tg P/yr}$ assuming a crustal abundance of 0.105% by weight and a solubility of 33%. Later work suggested that the solubility of desert aerosol P was considerably lower than that from other (mainly anthropogenic) sources, perhaps as low as 8–10% [Bergametti et al., 1992; Herut et al., 1999a; Herut et al., 2002; Chen et al., 2007]. However, Herut et al. [1999b] estimated the median solubility from North Africa/Arabian dust sources was 25%, and Markaki et al. [2003] estimated the solubility of samples influenced by Saharan dust as 10–30%. Ridame and Guieu [2002] conducted a detailed study of the release of DIP from Saharan soil at different particle concentrations in both ultrapure fresh water and seawater. They found a strong influence of particle concentration on solubility, with higher solubility at lower concentrations, suggesting the concentration of dust in raindrops would strongly influence the DIP solubility. Their smallest concentration range from 5 to 20 mg/L had solubilities from 13 to 21%, with lower solubilities at higher concentrations [Ridame and Guieu, 2002]. This concentration range and smaller values accounted for 55% of their field observations of Saharan dust in the Mediterranean Sea. This proportion would increase with increasing distance from dust source regions. Due to dilution the dust input from dry deposition would always be on the low end of their concentration ranges. They estimated DIP solubility from dry deposition between 10 and 14%. Mahowald et al. [2008] simulated the global deposition of total phosphorus and phosphate. Their modeling study assumed that mineral aerosols are 10% soluble P (PO_4), while other sources (primary biogenic particles, sea salts, biomass burning, fossil fuels, biofuels)

of total P are assumed to be 50% soluble. Volcanic sources are assumed 100% soluble [Mahowald et al., 2008]. In our simulations we have assumed a global mean solubility for mineral aerosol DIP of 15% at the ocean surface and a P fraction of 0.105% by weight. Our estimates will represent a lower bound on the influence of atmospheric P deposition as we are not including the more soluble P from noncrustal sources. Our total soluble P deposition is $\sim 15\%$ lower than that estimated by Mahowald et al. [2008]. Dust deposition was obtained from climatology [Luo et al., 2008] and it was assumed that dust contained a constant 30.8% Si by weight; 7.5% of this Si was treated as soluble [Duce et al., 1991] and thus entered the dissolved silicate pool upon deposition.

[13] Global deposition data sets for variable solubility aerosol iron under preindustrial and current era were obtained from simulations by Luo et al. [2008]. Their study simulated the emission, transport and deposition of soluble iron to surface oceans from mineral dust and combustion sources. Soluble iron estimates were obtained by employing a combination of cloud processing (acidity of cloud droplets converts insoluble Fe into soluble Fe) and hematite chemical reactions (hematite in dust is a source of soluble iron) on modeled combustion and dust distributions. In addition, atmospheric deposition data sets under globally constant mineral aerosol iron solubility (2%) in the current era and during the LGM were employed as external forcings to the ocean model using the dust climatology of Luo et al. [2008] for the current era and Mahowald et al. [2005] for the LGM.

[14] Atmospheric inputs of inorganic nitrogen to oceans for the current era were obtained from University of California Irvine Chemical Transport Model (UCICTM) simulations with an embedded inorganic aerosol thermodynamic equilibrium model [Luo et al., 2007]. Details on the iron and nitrogen data sets are given by Luo et al. [2007, 2008], Krishnamurthy et al. [2007], and Krishnamurthy et al. [2009].

[15] These nutrient deposition estimates were used as inputs to the BEC ocean model. The BEC model includes an ecosystem module and a biogeochemistry model that computes the full ocean carbonate-chemistry dynamics. Four functional groups of phytoplankton (small phytoplankton, diatoms, coccolithophores, and diazotrophs) and multiple growth limiting nutrients (nitrate, ammonium, phosphate, silicate, and dissolved iron) are included in the model [Moore et al., 2004].

[16] We assume that soluble iron in the ferrous form is bioavailable as in previous studies [e.g., Jickells and Spokes, 2001; Mahowald et al., 2005; Luo et al., 2008]. All of the soluble fraction dissolves instantaneously at the surface ocean, with some further iron release though a slower dissolution/disaggregation in the water column [Moore et al., 2004]. Our model includes a single “dissolved” seawater iron pool that is fully bioavailable, with no distinction between truly dissolved and colloidal forms. Currently, there is a large uncertainty regarding bioavailability of different iron forms for different phytoplankton groups. The BEC ocean model employs a seawater iron scavenging parameterization based on simulated soluble iron concentration and number of particles available to scavenge iron; the effects of organic ligands are treated implicitly through nonlinearity in the scavenging rates, which increase at high iron concentrations [Moore et al., 2004]. It has been sug-

gested by *Baker and Croot* [2010, and references therein] that oceanic controls, namely, seawater pH, presence of strong iron-binding ligands, iron associated with nanoparticle clusters, and organic colloids may play an important role in enhancing the solubility and bioavailability of aerosol derived iron. They state that postdeposition processing of deposited aerosols, scavenging of dissolved iron by particles and speciation could be key factors in the bioavailability of iron. Other than scavenging, these processes are largely neglected here.

[17] Iron sources from mineral dust and sedimentary sources were included in the model [*Moore et al.*, 2002, 2004]. The sedimentary iron source was weighted by the actual ocean bathymetry from the ETOP2 version 2.0 2 min global gridded database (U.S. Department of Commerce, 2006). Hence, at each grid cell in our coarse resolution ocean circulation model, the fraction of that cell that would consist of sediments based on the much higher resolution ETOP2V2 database was calculated, and a source of iron based on this fraction is included. This provides a more realistic distribution of sedimentary iron source and an improved match to observed iron distributions.

[18] The BEC runs within the coarse resolution, parallel ocean program model that is part of the Community Climate System Model (CCSM 3.0) developed at the National Center for Atmospheric Research [*Collins et al.*, 2006]. The physical circulation model has 25 vertical levels, with 8 levels in the upper 103 m. The longitudinal resolution is 3.6 degrees, and the latitudinal resolution is 2 degrees at higher latitudes with finer resolution near the equator [*Collins et al.*, 2006; *Yeager et al.*, 2006].

[19] Basin-scale patterns obtained from the BEC model are comparable to observed patterns of macronutrient distributions, calcification, biogenic silica production, nitrogen fixation, primary and export production [*Moore et al.*, 2002, 2004]. Recent studies have applied the BEC model to quantify ocean biogeochemical sensitivity to variations in mineral dust deposition (iron inputs) [*Moore et al.*, 2006], the feedbacks between denitrification and nitrogen fixation [*Moore and Doney*, 2007], and the ocean biogeochemical response to atmospheric deposition of inorganic nitrogen and iron [*Krishnamurthy et al.*, 2007, 2009].

[20] The current era results presented here are from the last year of 150 year transient simulation that was conducted to model the temporal evolution of iron (obtained from mineral aerosols and combustion sources) and inorganic nitrogen atmospheric nutrient inputs from preindustrial to modern conditions [*Krishnamurthy et al.*, 2009]. This was an extension to a ~500 year preindustrial control simulation. This control was also extended another 150 years and the last year is used here for the variable Fe solubility, preindustrial era. Results using a globally constant 2% solubility under the current era and the LGM discussed here were also obtained from the last year of 650 year simulations. Only the atmospheric nutrient inputs differ between these four simulations. All other radiative and atmospheric forcings are from a NCEP/NCAR based climatology for the current era [*Large and Yeager*, 2004]. The transient simulations of Fe and N deposition have been described previously by *Krishnamurthy et al.* [2009]. Here we conduct some additional analysis of these simulations to estimate the fraction of biogenic export supported by the

atmospheric inputs and to calculate global maps of nutrient limitation. The simulation with LGM dust deposition has not been previously published.

3. Results

[21] *Mills et al.* [2004] suggest that the availability of reactive nitrogen limits primary production, the conversion of inorganic carbon to organic carbon, over much of the ocean. Reactive nitrogen enters oceans via rivers, and atmospheric deposition. Table 1 summarizes the sources of nitrogen to global oceans in the preindustrial, twentieth century and some projections for the 21st century. *Seitzinger et al.* [2006] suggest that much of the riverine inputs of reactive nitrogen is either lost to the atmosphere after conversion to N₂ by denitrification or gets buried in coastal sediments, never reaching open ocean regions. Atmospheric inputs are primarily in NO_x (NO+NO₂) and NH₃ [*Duce et al.*, 2008] forms, although recent studies suggest that atmospheric water-soluble organic nitrogen could be far more abundant than previously thought, perhaps constituting ~30% of total reactive nitrogen deposition [*Jickells*, 2006; *Cornell et al.*, 2003; *Mace et al.*, 2003; *Nakamura et al.*, 2006]. These global estimates imply that the riverine inputs have increased greatly since the preindustrial era, and that atmospheric inputs also are increasing in the present era and in future projections (Table 1). There is still considerable uncertainty in these estimates. In this study we use the 1990s estimate of *Luo et al.* [2007] as an input to the ocean biogeochemical model. Their value of 39.6 Tg N/yr is in good agreement with the estimate of 39 Tg N/yr for the early 1990s by *Galloway et al.* [2004]. Neither estimate includes a contribution from organic N.

[22] Mineral aerosols provide important nutrients, in particular the micronutrient iron to open ocean areas [*Martin*, 1990; *Swap et al.*, 1992]. Due to the limited number of direct measurements of iron deposition it is difficult to deduce iron distributions from iron deposition measurements [*Mahowald et al.*, 2009]. Over the last few decades there have been studies simulating the distribution of mineral dust [*Mahowald et al.*, 2009, and references therein]. *Zender et al.* [2004] reviewed the deposition estimates and found that dust loads varied by factor of 4 within the recent dust models (10–40 Tg/yr). In order to make better comparisons between models and measurements, *Zender et al.* [2004] recommended setting a standard size of diameter of less than 10 μm, reasoning that particles greater than 10 μm in diameter would be short-lived in the atmosphere and deposited close to source regions. Recent data have suggested that a greater proportion of iron is contained in the larger size fraction (>10 μm) and is carried longer distances than previously thought [e.g., *Neff et al.*, 2008].

[23] *Jickells et al.* [2005] suggested that atmospheric sources of iron dominate inputs to open ocean areas, as little iron from coastal erosion, glacial sediments and riverine inputs make it to the open ocean. However, *Moore and Braucher* (2008) found that the dissolved iron inputs to the open ocean from sedimentary sources and mineral dust are of similar magnitude (Table 2). They suggested that sedimentary sources from continental margins had a strong impact on open-ocean iron concentrations, particularly in the North Pacific. The Southern Ocean region also has a

Table 1. Global Distribution of Sources of N to Oceans

N Inputs	Tg N/yr	Reference
Riverine N export to Oceans		
1970	33.9	<i>Bouwman et al.</i> [2005]
1995	43.7	<i>Bouwman et al.</i> [2005]
2030	49.7	<i>Bouwman et al.</i> [2005]
Riverine fluxes to Coastal areas		
1860	27	<i>Galloway et al.</i> [2004, and references therein]
Early 1990s	47.8	<i>Galloway et al.</i> [2004, and references therein]
2050	63.2	<i>Galloway et al.</i> [2004, and references therein]
DON ^a	11.5	<i>Seitzinger et al.</i> [2005]
DIN ^b	24.8	<i>Seitzinger et al.</i> [2005]
	25	<i>Dumont et al.</i> [2005]
PN ^c	29.6	<i>Seitzinger et al.</i> [2005]
Anthropogenic sources		
Atmospheric deposition		
NO _x		
1860	6.2	<i>Galloway et al.</i> [2004]
Early 1990s	21	<i>Galloway et al.</i> [2004]
2000	23	<i>Dentener et al.</i> [2006]
2030	25	<i>Dentener et al.</i> [2006]
2050	36.3	<i>Galloway et al.</i> [2004]
preindustrial	7.38	<i>Luo et al.</i> [2007] ^d
1990s	14.8	<i>Luo et al.</i> [2007] ^e
	11.4	<i>Duce et al.</i> [1991] and <i>Galloway et al.</i> [2004]
NH _y		
1860	8	<i>Galloway et al.</i> [2004]
preindustrial	11.6	<i>Luo et al.</i> [2007] ^d
Early 1990s	18	<i>Galloway et al.</i> [2004]
1990s	24.8	<i>Luo et al.</i> [2007] ^e
2000	24	<i>Dentener et al.</i> [2006]
2030	29	<i>Dentener et al.</i> [2006]
2050	33.1	<i>Galloway et al.</i> [2004]
	26.1	<i>Adams et al.</i> [1999]
	24.8	<i>Dentener and Crutzen</i> [1994]
	13.3	<i>Duce et al.</i> [1991]
	27	<i>Rodriguez and Dabdub</i> [2004]
Organic N		
1860	6.1	<i>Galloway et al.</i> [1996]
2000	20	<i>Dentener et al.</i> [2006]
2030	23	<i>Dentener et al.</i> [2006]

^aDissolved organic nitrogen.

^bDissolved inorganic nitrogen.

^cParticulate nitrogen.

^dPreindustrial.

^eCurrent era.

strong impact from sedimentary iron sources [*Moore and Braucher*, 2008; *Tagliabue et al.*, 2009]. Much of the Fe from the sedimentary source is lost to scavenging near source regions, but even a small fraction of this large source can significantly impact open ocean iron distributions, particularly in regions where atmospheric dust is low to moderate.

[24] Total dissolved iron inputs under a globally constant aerosol iron solubility of 2% were somewhat higher than those under variable aerosol iron solubility (Table 2), but with very different spatial patterns [see *Krishnamurthy et al.*, 2009]. This is because iron inputs are overestimated under constant solubility closer to major dust sources (where solubility is often less than 2%), while they are underestimated away from these source regions due to the lack of atmospheric processing [see *Luo et al.*, 2008, and references therein]. The uncertainties in atmospheric iron inputs are larger than with N deposition, as both the total dust deposition and the soluble fraction [*Zender et al.*, 2004; *Mahowald*

et al., 2005, 2009; *Luo et al.*, 2008] are uncertain. We analyze ocean simulations with several different estimates of the iron inputs, in part, to account for this uncertainty, but also to look at how temporal changes in the atmospheric input of Fe affect the oceans.

[25] The atmosphere and river inputs are significant sources of bioavailable P to the global oceans although; riverine fluxes dominate total P inputs (Table 3). In addition to mineral dust, primary biogenic particles, biomass burning, fossil fuels, biofuels, volcanoes, and sea salts are significant sources of atmospheric P [*Mahowald et al.*, 2008]. The P inputs used in this study (coming only from mineral dust) are ~15% lower than the total dissolved inputs estimated from all sources by *Mahowald et al.* [2008]. Si inputs are dominated by river sources, but the atmosphere is also a significant source (Table 4). Both Si and P are influenced by the same uncertainties in overall mineral dust inputs to the oceans. The recent review paper by *Mahowald et al.* [2009] discusses these uncertainties and potential human perturbations of the dust cycle.

[26] Coastal, estuarine, as well as open ocean ecosystems, have been under threat over the last few decades due to human induced changes in nutrient loading. *Rabalais* [2002] suggests that changing nutrient inputs to freshwater and coastal marine ecosystems may influence the N, P and Si stoichiometric balance, which could affect the total production as well as the composition of planktonic species. In order to evaluate the relative impacts of atmospheric N, P, Fe, and Si inputs to ocean ecosystems, we estimate atmospheric nutrient deposition ratios (Figure 1). The average elemental N/P requirement for many plankton species and for bulk organic matter in the oceans has been found to be relatively constant at ~16 (mol/mol) [i.e., *Redfield*, 1934, 1958]. Atmospheric N loading is relatively far greater than atmospheric P inputs, resulting in high N/P input ratios (Figure 1). Over much of the ocean this ratio is greater than the Redfield ratio by more than 2 orders of magnitude.

[27] Past studies have suggested that aerosol N: P is universally very high, so that aerosol is always deficient in P relative to phytoplankton requirements [*Baker et al.*, 2003]. N:P values of >30 are common in the North Atlantic [*Duarte et al.*, 2006], with N:P > 1000 reported in dry deposition in the North Atlantic [*Baker et al.*, 2003] and values >500 in the Gulf of Aqaba [*Chen et al.*, 2007]. *Pahlow and Riebesell* [2000] suggested that increasing N/P in deep North Atlantic nutrients was due to increased deposition of atmospheric nitrogen from anthropogenic sources. As in the North Atlantic, the Mediterranean Sea is characterized by high nitrate to phosphate ratios (~28:1) in the deep water. N budget analyses in that region suggested that deposition from the atmosphere is the most important source of excess nitrogen [*Herut et al.*, 2002].

[28] Diatoms that build frustules from opal (amorphous, hydrated silica) have an average C/Si ratio of 8 [*Brzezinski*, 1985]. This ratio has been found to vary from 3 to 40 depending on the conditions of light, temperature, and nutrient availability [*Harrison et al.*, 1977; *Brzezinski*, 1985]. Converting this to N/Si based on Redfield ratios yields values from 0.45 to 6. Our atmospheric deposition N/Si ratio estimates are 3–4 orders of magnitude higher, strongly suggesting that N inputs are also substantially higher than Si inputs, relative to the nutritional

Table 2. Global Distribution of Sources of Fe to Oceans

Fe Inputs	Tg Fe/yr	Reference
Riverine particulate total iron	625–962	<i>Jickells et al.</i> [2005, and references therein]
Riverine dissolved iron	1.5	<i>Jickells et al.</i> [2005, and references therein]
Glacial sediments	34–211	<i>Jickells et al.</i> [2005, and references therein]
Global deposition	16	<i>Mahowald et al.</i> [1999] ^a and <i>Ginoux et al.</i> [2001] ^a
Deposition to ocean	12	<i>Luo et al.</i> [2008]
	11	<i>Zender et al.</i> [2003] ^a
	4.8	<i>Tegen and Fung</i> [1995]
	8.2	<i>Mahowald et al.</i> [1999]
	4.5–11.3	<i>Fung et al.</i> [2000]
Bioavailable iron inputs	0.36–1.63	<i>Luo et al.</i> [2008]
Coastal erosion	8	<i>Jickells et al.</i> [2005, and references therein]
Hydrothermal	14	<i>Jickells et al.</i> [2005, and references therein]
Authigenic	5	<i>Jickells et al.</i> [2005, and references therein]
Industrial combustion	0.08	<i>Luo et al.</i> [2008, and references therein]
Biomass burning	0.15	<i>Luo et al.</i> [2008, and references therein]
Bioavailable iron inputs used in this study		
Total dissolved Fe from sedimentary source	1.78	Moore and Braucher (2008)
Atmosphere 2% solubility	0.37	this study ^b
Atmosphere variable solubility	0.31	this study ^b

^aIron inputs to land and ocean averaged between the three models. *Mahowald et al.* [1999] used a single aerosol size of 2.5 μm diameter. *Ginoux et al.* [2001] included mineral dust size ranging from 0.1 to 6 μm radius. *Tegen et al.* [2004] used mineral dust size ranging from 0.5 to 50 μm radius. *Zender et al.* [2003] included mineral dust size ranging from 0.1 to 10 μm diameter.

^bIncluding some iron from subsurface (0–103 m) remineralization of dust, based on the atmospheric deposition from *Mahowald et al.* [2005] and *Luo et al.* [2008]. See *Krishnamurthy et al.* [2009].

needs of phytoplankton (Figure 1). Phytoplankton uptake of aerosol Fe depends on the cellular Fe/N quota of algal growth [*Baker et al.*, 2003]. The Fe/C ratio can range from 13 to 86 $\mu\text{mol/mol}$ depending on the Fe status of the water column [*Sunda*, 1997], which corresponds to N/Fe of

$\sim 12000\text{--}75000$ mol/mol. Global estimates of N/Fe under current-era variable aerosol iron and nitrogen inputs suggest values from 800 to 10000 over most areas, indicating excess Fe inputs relative to N (Figure 1). In the Atlantic Ocean, *Baker et al.* [2003] found that the fraction of aerosol Fe

Table 3. Global Distribution of Sources of P to Oceans

Sources	Tg P/yr	Reference
Riverine inputs		
DIP ^a	0.8–1.4	<i>Meybeck</i> [1982]
	1.09	<i>Seitzinger et al.</i> [2005]
DOP ^b	0.2	<i>Meybeck</i> [1982]
	0.67	<i>Seitzinger et al.</i> [2005]
POP ^c	0.9	<i>Meybeck</i> [1993]
PIP ^d		
Fe-bound	1.3–7.4	<i>Compton et al.</i> [2000, and references therein]
Detrital	14.5–20.5	<i>Compton et al.</i> [2000, and references therein]
	9.03	<i>Seitzinger et al.</i> [2005]
Total riverine P inputs	17.7–30.4	<i>Compton et al.</i> [2000, and references therein]
Anthropogenic riverine P inputs	18–31	<i>Paytan and McLaughlin</i> [2007, and references therein]
Bioavailable riverine P inputs	0.03–0.06	<i>Paytan and McLaughlin</i> [2007, and references therein]
Eolian inputs (from mineral dust)	1	<i>Graham and Duce</i> [1979]
	1.05	<i>Compton et al.</i> [2000]
	0.33	<i>Zender et al.</i> [2003]
Total P	1.15	<i>Mahowald et al.</i> [2008, and references therein]
Bioavailable (10% solubility)	0.115	<i>Mahowald et al.</i> [2008, and references therein]
P deposition to global oceans	0.56	<i>Mahowald et al.</i> [2008, and references therein]
Bioavailable fraction to global ocean (17.3% solubility)	0.0097	<i>Mahowald et al.</i> [2008, and references therein]
Other sources ^e	0.24	<i>Mahowald et al.</i> [2008, and references therein]
Bioavailable fraction from these sources	0.125	<i>Mahowald et al.</i> [2008, and references therein]
BEC model estimate		
Atmospheric P inputs	0.54	this study
Bioavailable P (15% solubility)	0.0083	this study
Global sinking POP at 103 m	128	this study
Percent anthropogenic		
Total P	4.8	<i>Mahowald et al.</i> [2008, and references therein]
Bioavailable P	14.3	<i>Mahowald et al.</i> [2008, and references therein]

^aDissolved inorganic phosphorus.

^bDissolved organic phosphorus.

^cParticulate organic phosphorus.

^dParticulate inorganic phosphorus.

^eOther sources include primary biogenic particles, biomass burning, fossil fuels, biofuels, volcanoes, sea salts which are assumed to be 50% soluble, with the exception of that from volcanoes, which are assumed to be 100% soluble.

Table 4. Global Distribution of Sources of Si to Oceans

Si Sources	Tmol Si/yr	Reference
River flux	5 ± 1.1	<i>Treguer et al.</i> [1995]
Eolian	0.5 ± 0.5	<i>Treguer et al.</i> [1995]
	0.08–0.1	<i>Houghton et al.</i> [2001]
	3.45	<i>Zender et al.</i> [2003] ^a
Particulate Si	0.03 to 1	<i>Treguer et al.</i> [1995] and <i>Tegen and Kohfeld</i> [2006]
Seafloor weathering	0.4 ± 0.3	<i>Treguer et al.</i> [1995]
Hydrothermal	0.2 ± 0.1	<i>Treguer et al.</i> [1995]
BEC estimate		
Atmospheric Si	5.62	
Bioavailable Si (7.5% solubility)	0.43	

^aBased on 1990s dust climatology and dust contains 30.8% Si.

input removed through aerosol N-stimulated growth was less than 20%. However, in a few instances with low Fe inputs and relatively high N inputs, this fraction exceeded 80% and the excess atmospheric Fe input was small.

[29] We also examine the relative importance of atmospheric nutrient inputs for ocean biogeochemistry by comparing soluble nutrient inputs from the atmosphere with particulate export at the base of the euphotic zone (103 m) due to the biological pump. This ratio represents the fraction of the export production potentially supported by atmospheric nutrient deposition. It is also a measure of the atmospheric nutrient source relative to total nutrient inputs to the euphotic zone. For iron we divide the atmospheric dissolved Fe inputs by the export due to biological uptake and export due to dissolved Fe scavenged onto sinking particles. In some regions atmospheric N inputs can support 30–50% of the sinking PON export (Figure 2). Globally, atmospheric N inputs can sustain $\sim 5.1\%$ of sinking PON export. These results are similar to those reported previously by us in the work of *Krishnamurthy et al.* [2007] from a different N deposition simulation. Atmospheric Fe inputs have a strong impact on ocean productivity, often supporting $>50\%$ of the sinking biogenic PFe export in the Northern Hemisphere, with a global mean of 55%. Only small amounts of export are driven by atmospheric Si and P inputs. The highest impacts of Si and P are seen in the tropical North Atlantic and Mediterranean Sea, both areas where the BEC simulated export is much too low, due to extreme P limitation. So in these regions where up to 2–3% of export is supported, our results overestimate the importance of the atmospheric inputs. Global mean percentages for atmospheric P and Si driven biological export were 0.12% and 0.21%, respectively. Thus, atmospheric P and Si inputs have minimal impacts on ocean productivity, as this source is always small relative to the flux of these nutrients from below the euphotic zone.

[30] In order to highlight the time-varying importance of atmospheric inputs of iron in driving marine biogeochemistry, we estimated the soluble atmospheric Fe inputs/sinking biogenic PFe under different input scenarios (Figure 3). In the preindustrial case with variable aerosol iron solubility, atmospheric iron inputs could support 10–50% of the export production over most ocean regions, with higher values in the North Pacific. In the modern era, with variable aerosol iron solubility, the iron driven export, away from the dust plumes, as well as closer to combustion sources

increased considerably (40–80% of export). Global mean ratios of atmospheric iron inputs/sinking biological particulate Fe flux were 0.37 and 0.55 under preindustrial and current conditions with variable aerosol iron solubility.

[31] Under constant 2% solubility for aerosol iron during the dustier LGM, atmospheric Fe deposition was high in the North and South Atlantic, the North Pacific, and parts of the Southern Ocean. In these regions atmospheric iron could support more than 100% of the simulated export (Figure 3). The global mean ratio for iron inputs and sinking biological PFe export during the LGM was 0.72. The subarctic North Pacific and most of the equatorial Pacific are shifted from Fe-limited to N-limited, because of the excess iron delivery (Figure 4). The ratio for iron inputs and sinking biological export under constant iron solubility in the modern era were high close to major dust plumes, but were lower in the HNLC regions and the subtropical North Pacific, compared with the variable Fe solubility simulation, with a global mean ratio of 0.44 (Figure 3). In each plot of Figure 3 the ratio is very low in the equatorial Pacific where dust inputs are low, and there is a large upwelling flux of Fe from below.

[32] In order to assess the impact of atmospheric iron deposition on marine ecosystems, the growth limitation factors for diatoms under the four different soluble Fe input scenarios were examined (Figure 4). In the variable aerosol Fe solubility cases during the preindustrial and modern era, diatoms were primarily N-limited across the Indian Ocean and the subtropical Pacific. In the constant solubility cases

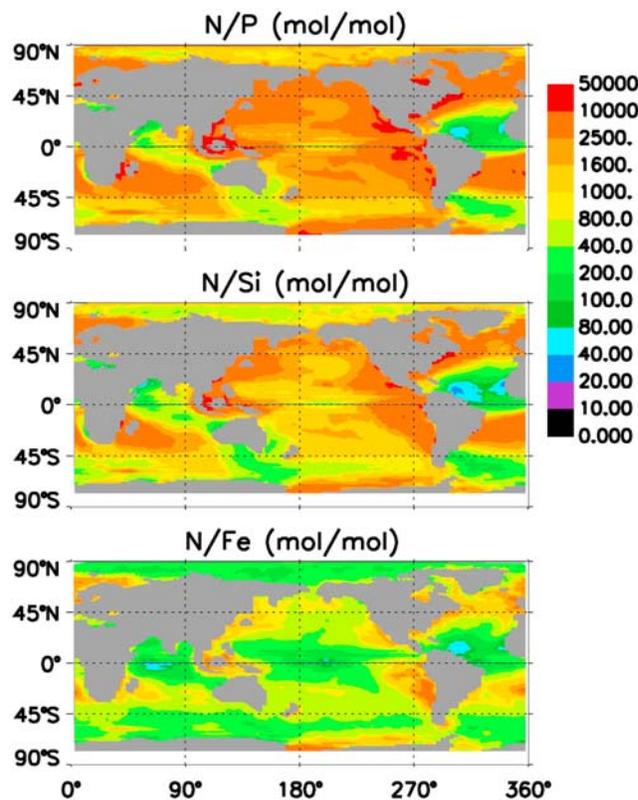


Figure 1. Aerosol derived nutrient deposition ratios for varying aerosol iron solubility, inorganic nitrogen, silicon, and phosphate inputs under the modern era climate conditions.

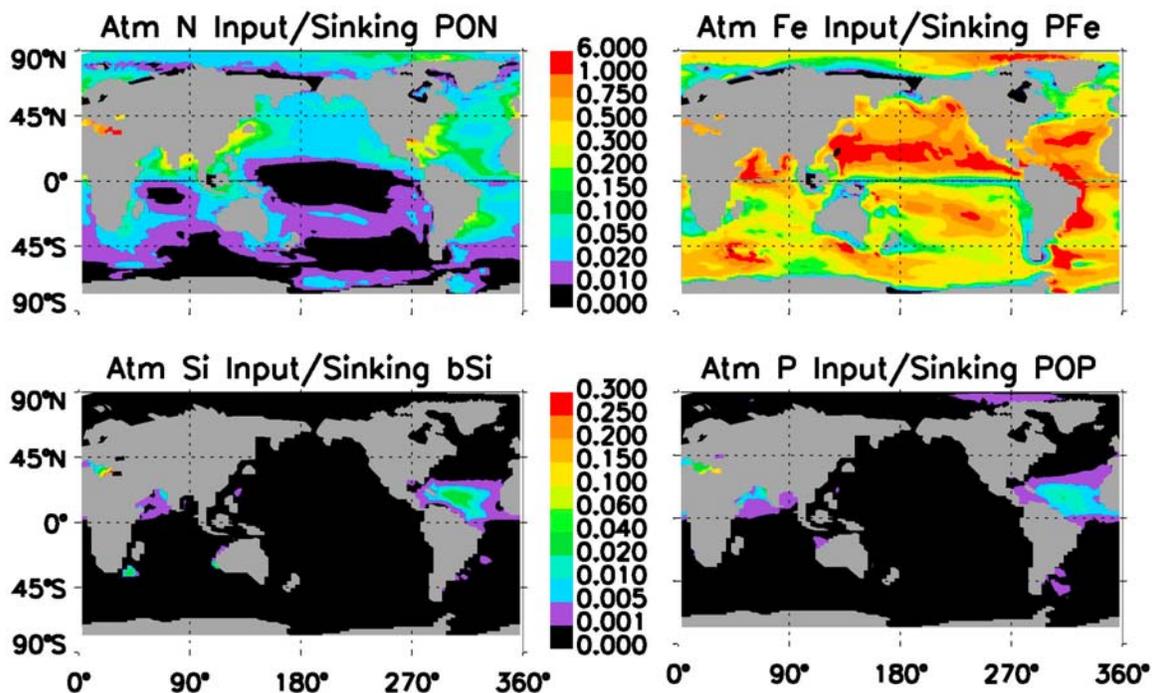


Figure 2. Ratios of atmospheric inputs of N, Fe, Si, and P with corresponding sinking particulate export under varying aerosol iron solubility, inorganic nitrogen, silicate, and phosphate inputs under the modern era climate conditions.

for the LGM and modern period, increased Fe inputs beneath major dust plumes (Arabian Sea and the low-latitude North Atlantic) lowered the Fe stress for diazotrophs, increasing N fixation. Hence, the ecosystem in the

Indian and Atlantic Oceans shifted toward P limitation. Fe limitation is considerably reduced in the Pacific Ocean for the diatoms under the higher LGM dust flux. Thus, spatial patterns of nutrient limitation and the extent of the high-

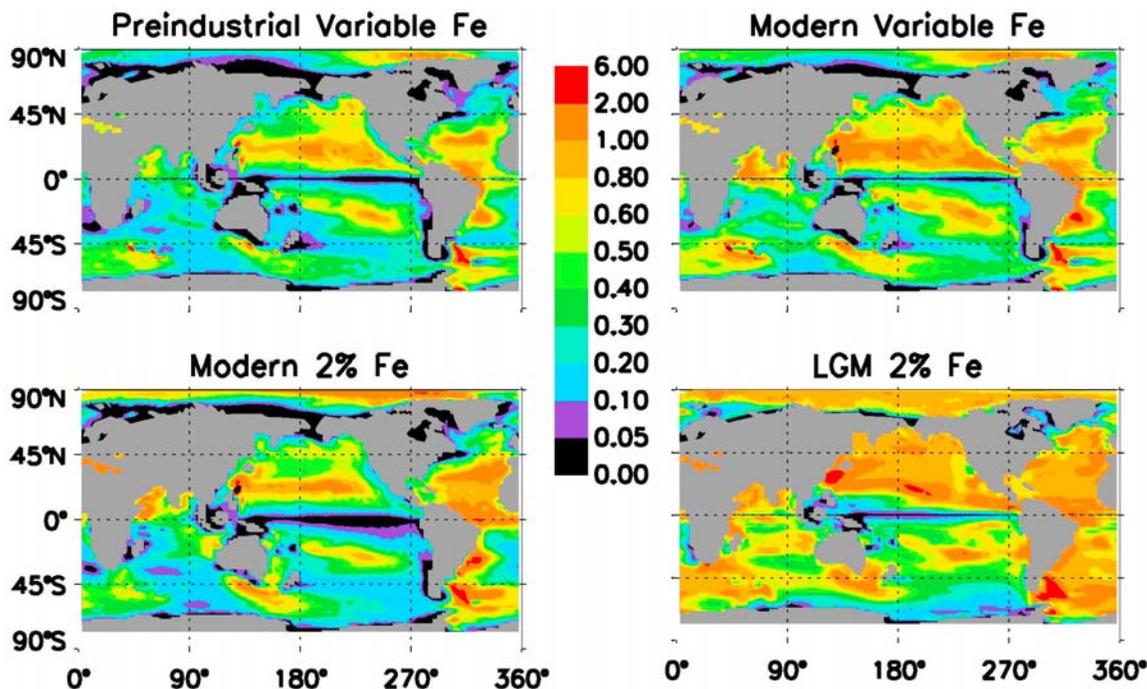


Figure 3. Ratios of atmospheric inputs of Fe over sinking particulate Fe export under varying aerosol iron solubility, during the modern and preindustrial era, and under globally constant aerosol iron solubility, during the modern era and in the LGM.

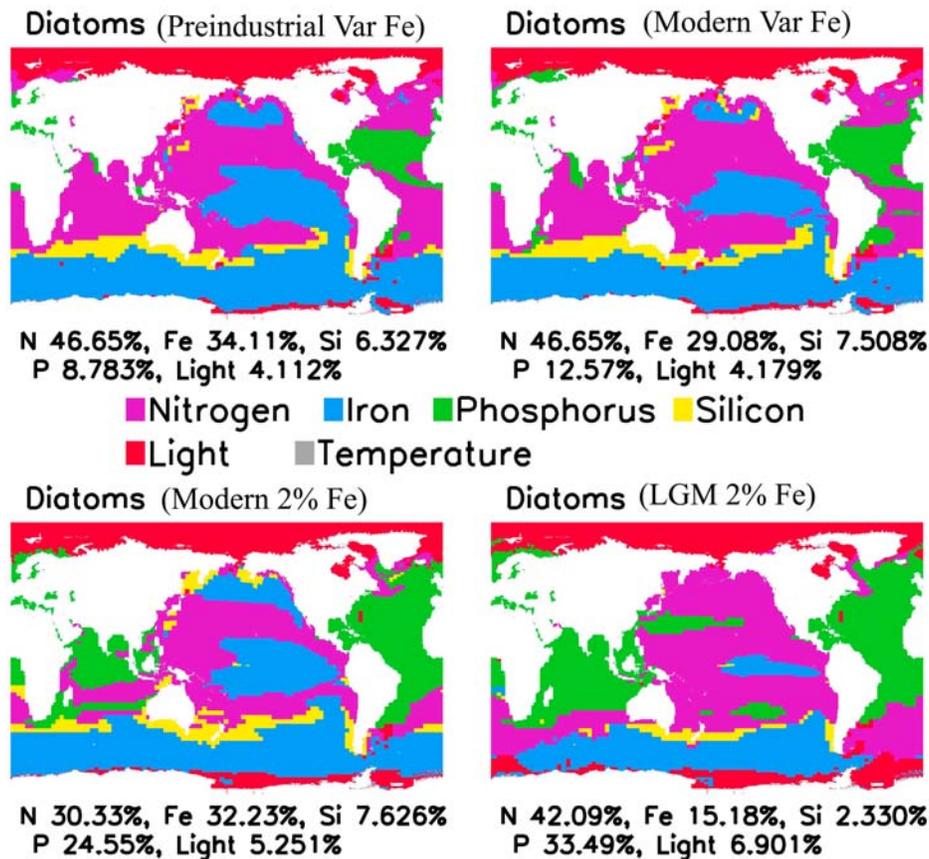


Figure 4. Factor most limiting growth rates for diatom under the four different soluble Fe input scenarios, including varying aerosol iron solubility under preindustrial and modern era conditions and constant aerosol iron solubility for modern and LGM dust fluxes.

nutrient, low-chlorophyll regions are sensitive to atmospheric Fe inputs, even when the model includes a strong sedimentary iron source.

4. Discussion

[33] Primary production in marine ecosystems occurs in the presence of nutrients that are mainly brought from below the euphotic zone by upwelling and mixing processes. Rivers and atmospheric deposition bring additional nutrients, the former being important in the coastal areas and the latter being more important in the open ocean. Global estimates of sinking export based on the BEC ocean model are on the lower end of the export estimates by other studies. Hence, our results may represent an upper bound on the relative importance of atmospheric nutrients for ocean biogeochemistry (as other models mix up more nutrients from below). This work focused on emphasizing the effects of atmospheric inputs of iron, nitrogen, silicate and phosphate on ocean biogeochemistry. Our results suggest that atmospheric iron deposition plays a significant role in driving marine primary production and biogeochemical cycling. Oceanic response to atmospheric iron inputs varies considerably, such that not only regions close to source areas are impacted, but due to atmospheric processing, enhanced in the presence of pollutants, iron solubility and

resulting impacts can increase in areas far from dust source regions.

[34] In our simulations about 50% of the sinking PFe export in a given year could be supported by atmospheric Fe inputs (Figures 2 and 3). The remaining $\sim 50\%$ of export is supported by dissolved iron coming from below the euphotic zone, which is itself partially derived from the atmospheric inputs (moved down the water column by biological uptake and export, particle scavenging of Fe, and from slow dissolution of sinking dust particles) and from iron advected from continental margins. Thus, the percentages of export production potentially supported by atmospheric Fe inputs, shown in Figures 2 and 3, underestimate the true atmospheric impacts since the subsurface iron pool is also partly dependent on this source. Thus, the atmospheric iron inputs appear to be the dominant source to open ocean regions, even though the sedimentary source along continental margins is a larger input to the oceans in absolute terms (Table 2).

[35] Atmospheric nitrogen inputs are important mainly closer to major source areas, globally accounting for 5.1% of export production. Considering some of the higher N deposition estimates in Table 1, this fraction might increase to as much as 7–9%, particularly if organic N is bioavailable and is included. Atmospheric P and Si inputs are only a very small source of external nutrient supply, with subsurface waters and rivers being a much more dominant source

of these nutrients to marine surface waters. The role of atmospheric P and Si in supporting marine ecosystem productivity would not increase substantially even with the roughly fourfold increase in atmospheric dust inputs to the oceans at the LGM (still $< 1\%$ of export production). In addition, our assumed aerosol Si solubility of 7.5% may be too high. Baker *et al.* [2006] measured much lower solubilities in Saharan dust, generally $< 1\%$. Thus, our conclusion that atmospheric inputs of Si and P have only a small impact on marine ecosystems seems robust, even given the uncertainties in dust inputs and solubilities. This is in agreement with recent field studies suggesting high N/P and Fe/P ratios in atmospheric nutrient deposition, relative to the stoichiometric requirements of phytoplankton [Duarte *et al.*, 2006; Baker *et al.*, 2007].

[36] Global estimates of marine nitrogen fixation are partly based on extrapolation of N^* -based estimates in the subtropical North Atlantic [Gruber and Sarmiento, 1997; Gruber, 2004]. N^* is a measure for nitrate concentrations in excess (or in deficit) relative to phosphate at the molar stoichiometry of 16:1 [Michaels *et al.*, 2001; Gruber and Sarmiento, 1997]. Hence, spatial distributions of N^* in the main thermocline of the world's oceans generally reflect the distribution of oceanic N_2 fixation and denitrification [Gruber, 2004]. Gruber [2004] noted that the highest N^* values are in the North Atlantic thermocline. Hansell *et al.* [2007] revisited these estimates by including recent estimates of excess N accumulated via high N/P atmospheric deposition, high N/P in dissolved organic matter (DOM), and calculated the nitrogen fixation by mass balance of the excess N field. Their study suggested that of the total excess N accumulation in the main thermocline in the North Atlantic, was $\sim 38\%$ due to atmospheric N inputs and $\sim 28\%$ from the export of high N/P DOM in the exported waters, resulting in $\sim 33\%$ from nitrogen fixation by mass balance. Yet our results indicate that the atmospheric N/P input ratio in the North Atlantic is actually lower than over most of the ocean, due to the substantial P inputs from African dust (Figure 1). Other basins do not accumulate the high excess N seen in the North Atlantic [Gruber, 2004; Moore and Doney, 2007]. It is possible that the higher atmospheric N/P input ratios in other basins are offset by denitrification, preventing a buildup of excess N. Alternatively, the role of atmospheric inputs may have been overestimated by Hansell *et al.* [2007]. In their calculations an N/P in atmospheric deposition of 100 was assumed. We estimated a somewhat lower deposition ratio for the tropical Atlantic, increasing to much higher values at higher latitudes (Figure 1).

[37] Atmospheric Fe inputs are sometimes discussed in terms of supporting N fixation in the oceans, as diazotrophs have a relatively high Fe requirement [Berman-Frank *et al.*, 2001; Kustka *et al.*, 2002]. A number of studies have suggested that N fixation rates should be sensitive to atmospheric Fe inputs to the oceans [i.e., Michaels *et al.*, 2001; Moore *et al.*, 2006]. However, it appears that atmospheric N deposition can act to suppress N fixation [Krishnamurthy *et al.*, 2007]. Thus, the ratio of N/Fe deposition has to be considered in terms of support for N fixation. High N/Fe deposition ratios could actually act to suppress N fixation by promoting depletion of P in surface waters. It appears that the N/Fe provides excess Fe in most ocean regions today,

and thus could potentially lead to increased N fixation. However, N deposition is expected to continue increasing in many regions over the next century.

[38] Human induced environmental changes over the past century have increased atmospheric inputs of Fe and N to the oceans. Increased agricultural and fossil fuel sources accompanied by massive population increase has affected the nutrient delivery by rivers to coastal waters. Fossil fuel consumption, coupled with changing climate and land use patterns have impacted atmospheric nutrient loading of nitrogen as well as iron, phosphorus and silica due to changing aridity and dust flux from the continents. Over the past decade global models capable of estimating nutrient deliveries by rivers as well as those obtained from atmospheric deposition have been developed [i.e., Seitzinger *et al.*, 2005; Zender *et al.*, 2003; Luo *et al.*, 2008]. Although, the response of increased productivity via riverine inputs to coastal ecosystems might have an immediate effect on humans due to their proximity to population centers [Howarth *et al.*, 1996; Carpenter *et al.*, 1998; Hagstrom *et al.*, 2001; Rabalais, 2002; Anderson *et al.*, 2002; Diaz *et al.*, 2003; Turner *et al.*, 2003; Townsend *et al.*, 2003], ongoing changes to atmospheric nutrient loading to the open ocean may have larger effects on biogeochemical cycles. Nutrient inputs via atmospheric deposition can be significant over large, remote areas of the oceans and, consequently important for the large-scale human impacts on marine ecosystem productivity, air-sea CO_2/O_2 flux, and the global carbon cycle.

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