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## Impacts of increasing anthropogenic soluble iron and nitrogen deposition on ocean biogeochemistry

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[1] We present results from transient sensitivity studies with the Biogeochemical Elemental Cycling (BEC) ocean model to increasing anthropogenic atmospheric inorganic nitrogen (N) and soluble iron (Fe) deposition over the industrial era. Elevated N deposition results from fossil fuel combustion and agriculture, and elevated soluble Fe deposition results from increased atmospheric processing in the presence of anthropogenic pollutants and soluble Fe from combustion sources. Simulations with increasing Fe and increasing Fe and N inputs raised simulated marine nitrogen fixation, with the majority of the increase in the subtropical North and South Pacific, and raised primary production and export in the high-nutrient low-chlorophyll (HNLC) regions. Increasing N inputs alone elevated small phytoplankton and diatom production, resulting in increased phosphorus (P) and Fe limitation for diazotrophs, hence reducing nitrogen fixation (~6%). Globally, the simulated primary production, sinking particulate organic carbon (POC) export, and atmospheric CO<sub>2</sub> uptake were highest under combined increase in Fe and N inputs compared to preindustrial control. Our results suggest that increasing combustion iron sources and aerosol Fe solubility along with atmospheric anthropogenic nitrogen deposition are perturbing marine biogeochemical cycling and could partially explain the observed trend toward increased P limitation at station ALOHA in the subtropical North Pacific. Excess inorganic nitrogen ( $[\text{NO}_3^-] + [\text{NH}_4^+] - 16[\text{PO}_4^{3-}]$ ) distributions may offer useful insights for understanding changing ocean circulation and biogeochemistry.

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### 1. Introduction

[2] Atmospheric deposition of macronutrients and micronutrients set important controls on marine ecology and biogeochemistry. Atmospheric fluxes are changing with time because of climate and human influences, and contemporary patterns of marine biogeochemistry may reflect to some degree variations in atmospheric input of nutrients with time. Ongoing anthropogenic influences have particularly changed biogeochemical cycling of the nutrients, iron, and nitrogen. Modeling studies suggest that changes in climate and land use practices over recent decades may have altered dust fluxes and thus aeolian Fe inputs to open

ocean regions [Mahowald and Luo, 2003; IPCC, 2001]. Humans have substantially altered the global N cycle because of increased fossil fuel combustion and agriculture [Galloway *et al.*, 2004, and references therein].

[3] The past 2 decades of research in marine sciences have established the key role played by iron in driving oceanic primary production [Martin, 1990; Coale *et al.*, 1996; Falkowski *et al.*, 1998; Jickells *et al.*, 2005]. Numerous studies suggest that the micronutrient iron is responsible for limiting phytoplankton productivity in the Southern Ocean, eastern equatorial Pacific, and the subarctic Pacific; these areas are characterized as high-nutrient low-chlorophyll (HNLC) regions [Boyd *et al.*, 2000; Coale *et al.*, 1996; Boyd *et al.*, 2004]. Cassar *et al.* [2007] stated that aeolian iron additions resulted in enhanced export production in the Southern Ocean and increased gross primary production over a large area of the Southern Ocean downwind of dry continental areas. Boyd and Mackie [2008] argued that the simulated iron deposition data set used by Cassar *et al.* [2007] was primarily derived from observations in the Northern Hemisphere; hence, their correlation estimates between export production and aeolian iron inputs in the Southern Ocean may be biased. In the subtropical

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regions, diazotrophs (nitrogen fixers) may be limited by iron [Falkowski, 1997; Michaels *et al.*, 2001; Berman-Frank *et al.*, 2001; Moore *et al.*, 2004, 2006], and this iron requirement for nitrogen fixation may give the subtropics and tropics a sensitivity to atmospheric dust (iron) inputs similar to that seen in the HNLC regions [Michaels *et al.*, 2001; Gruber, 2004]. The increased biological export as a result of iron inputs to these regions leads to enhanced atmospheric CO<sub>2</sub> uptake [Moore *et al.*, 2006; Boyd *et al.*, 2007, and references therein]. This can have important implications for the global carbon cycle.

[4] In order to be utilized by marine phytoplankton, however, atmospheric iron deposited to the surface ocean must be in a bioavailable form. Although bioavailable iron may differ from soluble iron, we follow previous studies [e.g., Jickells and Spokes, 2001; Mahowald *et al.*, 2005; Luo *et al.*, 2008] and assume that soluble iron in the ferrous form is bioavailable. Much of the dissolved iron in seawater is present in colloidal forms [Moore and Braucher, 2008, and references therein]. Recent studies suggest that most of the soluble iron in surface waters derived from aerosols were bound to colloids [Wu *et al.*, 2007; Aguilar-Islas *et al.*, 2009]. In our model simulations we assumed a single “dissolved” seawater iron pool that is fully bioavailable, with no distinction between truly dissolved and colloidal forms. Solubility depends on the oxidation state of iron and mineralogy and this affects the biogeochemical processes involving iron in the ocean [Luo *et al.*, 2005]. Observational estimates of solubility of iron in desert soils found in an earlier study were  $\sim 0.4 \pm 0.3\%$  [Zhuang *et al.*, 1992]. Solubility of iron in aerosols, however, has been found to be much higher, suggesting the importance of atmospheric processing [Zhuang *et al.*, 1992; Pehkonen *et al.*, 1993; Siefert *et al.*, 1994; Spokes *et al.*, 1994; Desboeufs *et al.*, 2001; Hand *et al.*, 2004; Luo *et al.*, 2005; Sedwick *et al.*, 2005; Mahowald *et al.*, 2005; Fan *et al.*, 2006; Schroth *et al.*, 2009].

[5] A variety of chemical processes may be relevant to iron solubility. Iron solubility is defined as the labile Fe<sup>2+</sup> and Fe<sup>3+</sup> fraction of total Fe and is roughly twice as large as the Fe<sup>2+</sup> fraction [Luo *et al.*, 2005]. Here we consider the Fe<sup>2+</sup> fraction as soluble and thus bioavailable [Luo *et al.*, 2008]. We assume that 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]. Luo *et al.* [2005] concluded from atmospheric simulations that incorporating cloud processing (acidity of cloud droplets converts insoluble Fe into soluble Fe) and hematite (hematite in dust is a source of soluble iron) improved model-predicted iron solubility relative to observations. They also found that estimated aerosol iron solubility from the combined cloud and hematite processing case compared better with observations than other mixed processes. Luo *et al.* [2008] modified the rate coefficients in the cloud processing method from Luo *et al.* [2005] such that soluble iron values better matched observations, and the modified cloud processing method was combined with hematite reactions [Meskhidze *et al.*, 2005] to estimate soluble iron inputs to marine surface waters from aerosol deposition. The solubility estimates of Luo *et al.* [2008] are consistent with

observations by Siefert *et al.* [1999], Johansen *et al.* [2000], and Baker *et al.* [2006] that fine-mode (<3 μm in diameter) iron solubility is higher than coarse-mode (>3 μm in diameter) solubility. Additionally, there is some tendency for the largest solubilities to occur far from the source areas (e.g., western tropical Atlantic and midlatitude North and South Atlantic), consistent with atmospheric processing of the iron.

[6] In addition to mineral dust, combustion may also be a source of significant soluble iron input to the ocean surface [Siefert *et al.*, 1997; Chen and Siefert, 2004; Chuang *et al.*, 2005; Guieu *et al.*, 2005; Sedwick *et al.*, 2007]. Combustion sources of soluble iron can be important in regions where coal combustion from power plants and industry dominate. Biofuel and biomass burning, predominantly in the tropics, could also be an important source of new iron to the ocean [Guieu *et al.*, 2005]. Iron solubility is expected to be higher in regions dominated by combustion iron because the iron is not bound in mineral form [Schroth *et al.*, 2009].

[7] Human activities, therefore, may alter the input of bioavailable iron to the surface ocean either directly through combustion iron sources or indirectly through reactive nitrogen and sulfur emissions, which subsequently result in enhanced atmospheric acidity and cloud processing. Luo *et al.* [2008] used the limited observations of iron solubility and model simulations to argue that large increases in soluble iron input to the ocean since preindustrial times are plausible, and here we explore the impact of these predicted changes on ocean biogeochemistry.

[8] Iron solubility in many previous ocean biogeochemical simulations has been assumed to be spatially constant. Fung *et al.* [2000] modeled the global distributions of annual iron uptake by phytoplankton using constant soluble iron fractions of 1% and 10% as upper and lower bounds. Aumont *et al.* [2003] and Gregg *et al.* [2003] prescribed a surface solubility of 1% in their simulations. On the basis of sensitivities studies with a box model, Parekh *et al.* [2005] set the solubility to be 1% in their simulations. Iron solubility in the BEC model has been set to a constant 2% in past studies [Moore *et al.*, 2004, 2006]. In reality, solubility of aerosol iron over global oceans is not constant [Mahowald *et al.*, 2005, and references therein], and studying the effects of varying solubility iron on ocean ecosystems is a crucial step toward understanding its role as a limiting micronutrient in many regions.

[9] Increasing fossil fuel combustion and agriculture are substantially altering the global N cycle. Atmospheric emissions of reactive nitrogen are dominated by oxidized species of nitrogen (NO<sub>x</sub>). The net global NO<sub>x</sub> emissions have increased from an estimated preindustrial value of 12 Tg N/a [Holland *et al.*, 1999; Galloway *et al.*, 2004] to between 42 and 47 Tg N/a in 2000. Lamarque *et al.* [2005a] forecast NO<sub>x</sub> emissions to be 105 to 131 Tg N/a by 2100. There has been an increase since preindustrial times in the intensity of agricultural nitrogen cycling, the primary source of NH<sub>3</sub> emissions [Bouwman *et al.*, 2002]. Total global NH<sub>3</sub> emissions have increased from an estimated preindustrial value of 11 Tg N/a to 54 TgN/a for 2000 [Holland *et al.*, 1999; Galloway *et al.*, 2004] and are projected to increase to 116 Tg N/a by 2050 [Galloway *et al.*, 2004]. The spatial

patterns of atmospheric nitrogen loading to global oceans have also been impacted by anthropogenic activities [Duce *et al.*, 2008].

[10] Doney *et al.* [2007] estimated the impact of anthropogenic nitrogen (and sulfur) deposition on ocean acidification and the inorganic carbon system. They found that, on a global scale, oceanic uptake of CO<sub>2</sub> due to deposition of anthropogenic nitrogen resulted in an increase in the surface-dissolved inorganic carbon (DIC) and alkalinity, and a few percent increase in ocean acidification relative to that caused by anthropogenic carbon dioxide. They suggest that resulting ocean acidification could have significant effects on marine ecosystems on regional scales, especially in coastal waters downwind of industrial sources that in turn could impact human societies. Luo *et al.* [2007] used an inorganic aerosol thermodynamic equilibrium model coupled to a three-dimensional chemical transport model to understand the roles of ammonia chemistry and natural aerosols on the global distribution of NO<sub>3</sub><sup>-</sup>, NH<sub>4</sub><sup>+</sup>, SO<sub>4</sub><sup>2-</sup>, dust, and sea-salt aerosols. The thermodynamic model partitions the gas-phase precursors among simulated aerosol species on the basis of relative humidity and natural and anthropogenic aerosols present. Their study concluded that, globally, atmospheric NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup> deposition during the 1990s was double the preindustrial deposition.

[11] In an earlier study, Krishnamurthy *et al.* [2007a] modeled the effects of atmospheric nitrogen deposition on ocean biogeochemistry using simulated atmospheric inorganic N inputs (NO<sub>3</sub><sup>-</sup>, and NH<sub>4</sub><sup>+</sup>) from Luo *et al.* [2007]. Inorganic nitrogen inputs from three different periods, namely, preindustrial, current, and an IPCC-A1FI prediction for 2100, were used. 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 CO<sub>2</sub> flux. Increased atmospheric N inputs to otherwise N-limited regions stimulated primary production by small phytoplankton and diatoms, resulting in a reduction in N<sub>2</sub> fixation because diazotrophs were not able to compete as effectively for surface-dissolved P and Fe. The study suggested that new nitrogen inputs to the oceans via anthropogenic atmospheric N inputs could be partially compensated for by decreased N<sub>2</sub> fixation.

[12] In this paper we present results of transient sensitivity studies with the Biogeochemical Elemental Cycling (BEC) ocean model to increasing anthropogenic inorganic nitrogen and soluble iron from atmospheric processing of dust and combustion sources. After an initial preindustrial spin-up of 500 years, three sets of deposition data were employed as external forcings, linearly increasing over 150 years to capture the transition from the preindustrial era to current conditions. The deposition data sets included (1) variable mineral aerosol iron solubility combined with soluble iron from combustion sources; (2) increasing atmospheric inorganic nitrogen inputs; and (3) simultaneously increasing iron and nitrogen inputs. Global integrals and spatial patterns were calculated for each case for denitrification, N<sub>2</sub> fixation, primary production, sinking POC export at 103 m,

excess inorganic nitrogen ( $[\text{NO}_3^-] + [\text{NH}_4^+] - 16[\text{PO}_4^{3-}]$ ), sea-air CO<sub>2</sub> flux and atmospheric pCO<sub>2</sub>. A key goal was to examine how increasing nitrogen and soluble iron inputs since the preindustrial era may be perturbing marine biogeochemical cycles.

## 2. Methods

[13] The BEC model simulations were carried out within the National Center for Atmospheric Research's (NCAR) Community Climate System Model 3 (CCSM) Parallel Ocean Program ocean module [Collins *et al.*, 2006; Yeager *et al.*, 2006]. The horizontal resolution of the model is variable such that globally it is on average 3.6° longitude/2.0° latitude but increases to approximately 3.6° longitude/0.9° latitude at the equator. In order to allow for computations of surface ocean pCO<sub>2</sub> and air-sea CO<sub>2</sub> flux, the ecosystem component of the BEC model [Doney *et al.*, 2009] is coupled to a carbonate chemistry module based on the Ocean Carbon Model Intercomparison Project (OCMIP) [Doney *et al.*, 2003, 2004, 2006].

[14] The marine ecosystem in the BEC model includes multiple, potentially growth-limiting nutrients (nitrogen, phosphorus, iron, and silicon). The growth of four phytoplankton functional groups, namely, picoplankton, diatoms, diazotrophs, and coccolithophores, were simulated along with the biogeochemical cycling of carbon, nitrogen, phosphorus, silicon, oxygen, alkalinity, and iron. Diazotrophs have higher iron and light requirements, higher N/P ratios, and slower maximum growth rates than the other phytoplankton groups. Detailed description of the model is available in the works of Moore *et al.* [2001, 2004]. Some parameter modifications [Moore *et al.*, 2006; Krishnamurthy *et al.*, 2007b] and additional improvements to the model are described by Moore and Braucher [2008].

[15] Relevant to this study, iron sources from mineral dust and sedimentary sources were included in the model [Moore *et al.*, 2001, 2004; Moore and Braucher, 2008]. The sedimentary iron source was weighted by the actual ocean bathymetry from ETOP2 version 2.0 2-min global gridded database [U.S. Department of Commerce, 2006]. Hence, at each cell in the model, the fraction of the cell area that would consist of sediments based on the high-resolution ETOP2V2 database was calculated. This provided a more realistic distribution of sedimentary iron sources and improved the match to observed iron distributions [Moore and Braucher, 2008]. The BEC model utilized 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; Moore and Braucher, 2008].

[16] Dust deposition was from the climatology of Luo *et al.* [2003]. Atmospheric dust was assumed to contain a constant 30.8% Si and 0.105% P by weight; 7.5% of this Si and 15% of this P were treated as soluble [Duce *et al.*, 1991] and thus entered the dissolved silicate and phosphate pool upon deposition. 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]. Preindustrial deposition estimates were obtained by removing anthropogenic sources from simulations with present-day meteorology. Details on the nitrogen input data sets are given by Luo *et al.* [2007] and Krishnamurthy *et al.* [2007a]. Variable solubility iron inputs were obtained from simulations by Luo *et al.* [2008]. Their study simulated the emission, transport, and deposition of soluble iron to surface oceans from combustion sources. Soluble iron estimates were obtained by employing a combination of cloud processing (acidity of cloud droplets converts insoluble Fe into soluble Fe) [Hand *et al.*, 2004; Luo *et al.*, 2005, 2008] and hematite chemical reactions (hematite in dust is a source of soluble iron) [Meskhidze *et al.*, 2003] on modeled combustion and dust distributions.

[17] Four sets of BEC ocean model simulations were performed. A 650-year control simulation was conducted with preindustrial variable solubility iron inputs from dust and combustion sources (only biomass burning under preindustrial). After model year 500, three 150-year transient branch simulations were conducted to model the temporal evolution from preindustrial to modern conditions; anthropogenic perturbations in iron and/or nitrogen deposition (modern minus preindustrial), increasing linearly with time, are added to the preindustrial control. The first set of simulations employed linearly increasing perturbations in variable aerosol iron solubility inputs from dust and combustion sources (Scenario III from Luo *et al.* [2008], but with a 50% reduction (from 4% to 2%) in combustion Fe initial solubility, as preliminary simulations suggested excessive iron inputs to the North Pacific with the original values), hereafter referred to as case 1. The spatial patterns of the anthropogenic perturbation (modern minus preindustrial) differ from preindustrial fields, and thus the transient experiments were not just a linear increase of preindustrial fields.

[18] The cloud processing under preindustrial era was similar to that under modern era, but the hematite processing was lowered because of lower levels of atmospheric sulfur dioxide. The cloud processing case as implemented in our model [Hand *et al.*, 2004; Luo *et al.*, 2005, 2008] assumes that the clouds are always sufficiently acidic to process the iron to become more soluble. Hence, solubility of iron depends only on whether the iron is in a cloud or not. No pH parameterizations are set in the cloud processing method. Under hematite processing [Meskhidze *et al.*, 2003], the acidity of the aerosol is critical for the conversion of  $\text{Fe}^{3+}$  to  $\text{Fe}^{2+}$ , and there anthropogenic sulfate does play an important role in modulating acidity. Here the pH values are set depending on the  $\text{SO}_4$  concentration. Hence, we assumed that only the hematite processing case changes under the preindustrial conditions because of lower  $\text{SO}_4$  levels. The preindustrial case did not include biofuel emissions of iron and assumed that biomass burning was reduced by 90% from contemporary levels.

[19] The second set (case 2) of simulations employed linearly increasing, anthropogenic atmospheric inorganic nitrogen inputs (obtained from simulations by Luo *et al.* [2007]). The third set (case 3) of simulations combined the

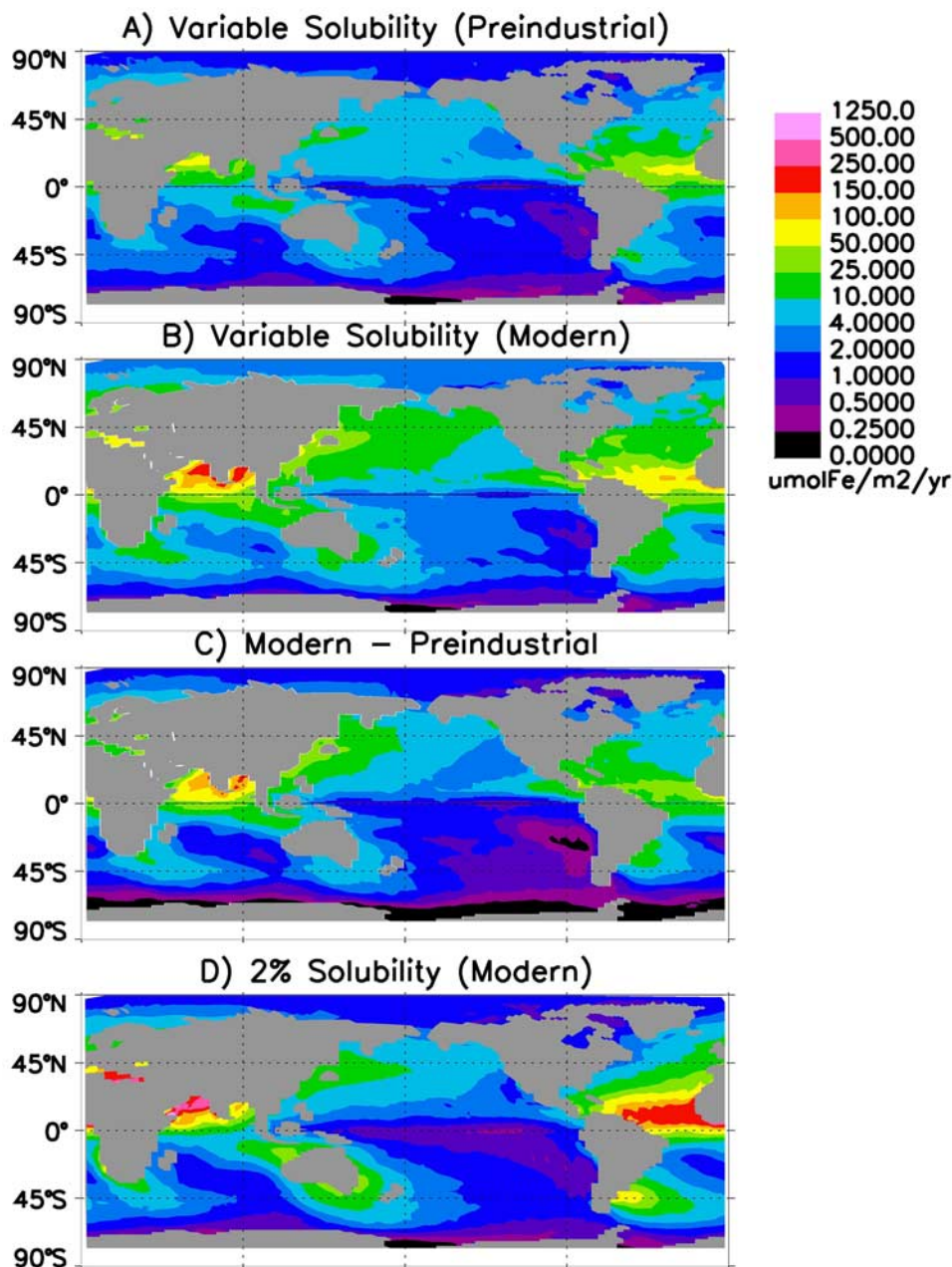
above two cases to include linearly increasing anthropogenic iron and nitrogen inputs. Total simulated inorganic N deposition to the ocean was  $\sim 22$  Tg N/a for the preindustrial and  $\sim 39$  Tg N/a for the current era [Krishnamurthy *et al.*, 2007a]. Our preindustrial estimate is on the high end of recent estimates ( $\sim 14$  Tg N/a in the work of Duce *et al.* [2008]). The response to rising N inputs to the oceans in the current era may be underestimated compared to the  $\sim 47$  Tg N/a in the work of Duce *et al.* [2008]. Total simulated soluble iron inputs to the global oceans were  $2.4 \times 10^9$  mol Fe/a for the preindustrial and  $5.5 \times 10^9$  mol Fe/a in the current era. Further details on atmospheric processing and combustion sources can be found in the work of Luo *et al.* [2008].

[20] Our approximation of a linear increase in soluble iron inputs was based in part on a recent study by Bond *et al.* [2007]. They found that black carbon emissions since the preindustrial era have increased almost linearly [Bond *et al.*, 2007, Figure 6]. In an earlier work, Bond *et al.* [2004] studied the role of combustion practices in the determination of emissions and found that 38%, 20%, and 42% of the black carbon emissions were from fossil fuel, biofuel, and open burning, respectively. Observations at Cheju, Korea, suggest a high correlation between soluble iron and black carbon coming from combustion sources [Chuang *et al.*, 2005]. Therefore, we used the linear increase in deposition of soluble iron from the preindustrial to modern values. In reality, increases in recent decades may have been somewhat more rapid than this linear trend. Also, this regional relation between soluble Fe and black carbon may vary, particularly with changes in the black carbon sources.

[21] In the preindustrial climate, mineral aerosol availability may have been different from present era conditions because of alterations in climate, hydrological cycle, and land use. For example, the intensity of cultivation in deserts would have been lower than that in modern era. Vegetation cover may also have differed because of climate and soil moisture trends and carbon dioxide fertilization effects from rising atmospheric  $\text{pCO}_2$ . We neglect for this study possible changes in mineral aerosols due to climate or land use between modern and preindustrial conditions, which could be large and perhaps dominate the anthropogenic signal, but remain largely uncertain even in terms of the sign of the anthropogenic trend [e.g., Mahowald and Luo, 2003; Tegen *et al.*, 2004; Mahowald and Dufresne, 2004; Mahowald *et al.*, 2009].

[22] A similar linear increase in anthropogenic atmospheric inorganic N inputs was assumed, but again, increases in recent decades in N inputs were likely more rapid. Atmospheric  $\text{CO}_2$  concentration was modeled as a single, well-mixed box in contact with the oceans. Climate forcings and winds were from late 20th century NCAR-NCEP climatology [Large and Yeager, 2004].

[23] Ocean iron scavenging parameters were optimized with the modern era, variable solubility atmospheric iron inputs to provide the best match to observed iron distributions [following Moore and Braucher, 2008]. The higher soluble iron inputs in the variable aerosol iron solubility case, particularly in the North Pacific, required a higher base scavenging rate to keep simulated dissolved iron concentrations reasonable, compared with the fixed 2% solubility



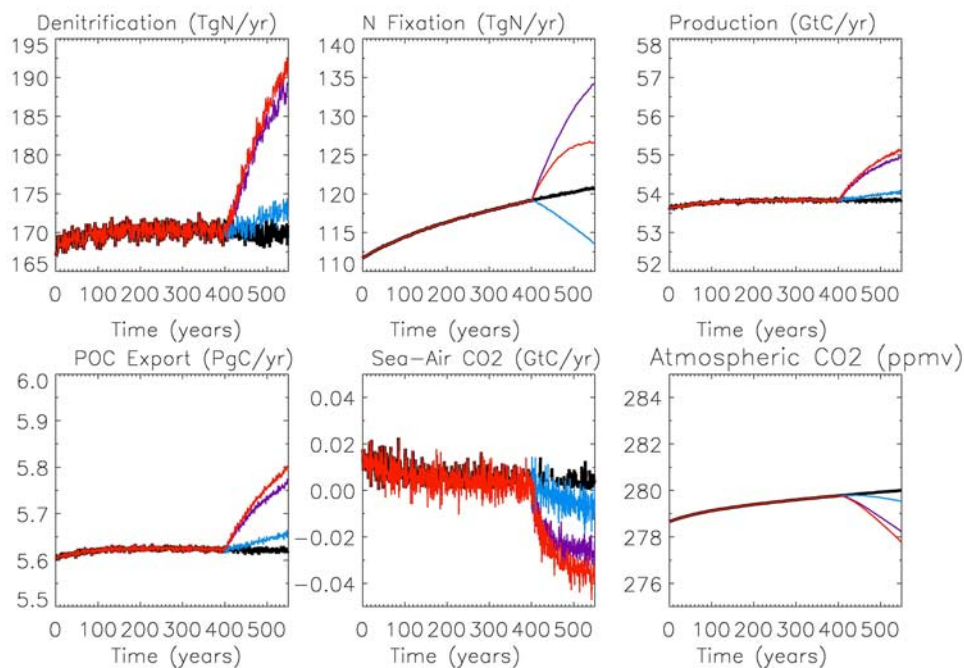
**Figure 1.** Spatial distributions of annual iron deposition under (a) preindustrial and (b) modern variable aerosol iron solubility and (c) their difference. (d) Annual iron deposition under globally constant (2%) aerosol iron solubility during the modern era.

used by *Moore and Braucher* [2008]. Conversely, lower iron inputs beneath the main dust plumes required a reduction in the scavenging coefficient that influenced scavenging at high iron concentrations ( $>0.6$  nM) to best match observations. These optimized scavenging parameter values were utilized in all simulations.

### 3. Results

[24] Globally, we estimated that surface ocean soluble iron inputs to the contemporary ocean were more than

double those during preindustrial times. The largest changes were in the northern Indian Ocean, the North Pacific, and the tropical North Atlantic (Figure 1c). Modest increases in soluble iron deposition were also observed in other remote regions compared to preindustrial. The influence of atmospheric processing in the presence of increased anthropogenic pollutants was particularly critical in remote regions away from major dust plumes (Figures 1b and 1d in the tropical and subtropical Pacific). Combustion sources of soluble iron were important near industrial and biomass burning sources where dust sources were lower (for exam-



**Figure 2.** Changes in key biogeochemical fluxes over time for 550-year simulations; the first 100 years have been excluded in the plots for clarity; hence years 101 to 650 are shown. First 400 years depicts model spin-up. Last 150 years denote the transient experiment. Black line describes the preindustrial control, and cases 1 (Fe), 2 (N), and 3 (Fe+N) are depicted by violet, blue, and red lines, respectively.

ple, off the coast of South and East Asia). Solubilities less than 2% reduced iron inputs beneath the major dust plumes (i.e., tropical Atlantic, Figures 1b and 1d). In spite of combustion iron estimated as being the source of only 5% of total global iron inputs from the atmosphere to oceans, soluble iron inputs to surface oceans from combustion sources may exceed 50% in some regions [Luo *et al.*, 2008].

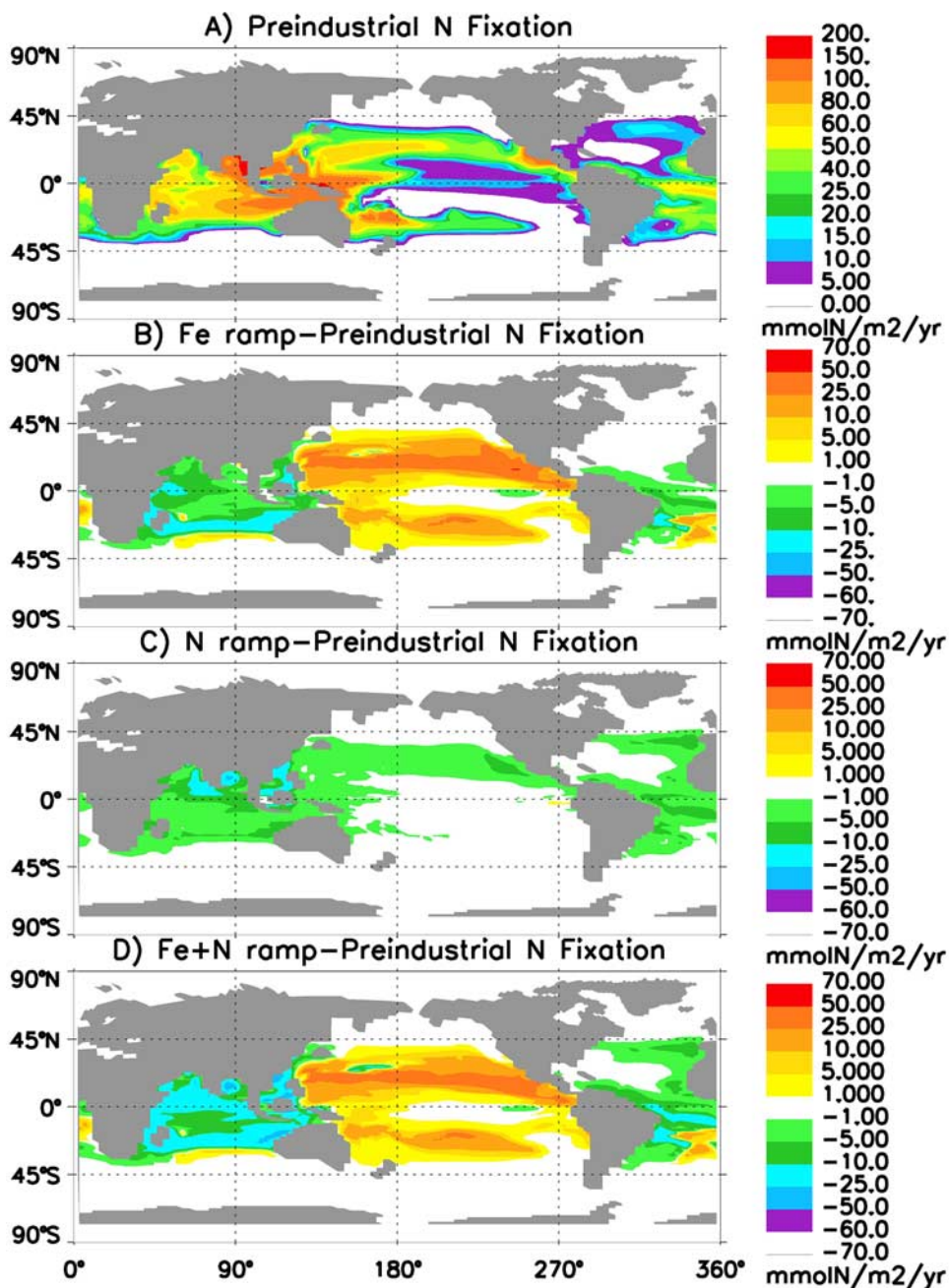
[25] Since the preindustrial era, anthropogenic inorganic nitrogen inputs to the ocean increased considerably, especially in coastal waters as well as in open ocean areas downwind of fossil-fuel-based industries and regions where intensive agriculture has resulted in increased fertilizer use [Krishnamurthy *et al.*, 2007a]. Nitrogen deposition increased significantly downwind of the coast of East and South Asia. An increase of >10 Tg N/a since preindustrial was seen off the east coast of North America and Europe. There was also a sizable increase in nitrogen deposition close to the African, Australian, and the South American continents. Compared to the preindustrial era, all the ocean basins except the Southern Ocean and the South Pacific Ocean had a substantial increase in nitrogen deposition [Krishnamurthy *et al.*, 2007a].

[26] Changes in key marine biogeochemical fluxes during the transient 150-year preindustrial to modern simulations suggest significant differences in oceanic biogeochemical cycling between the control and the three different atmospheric nutrient input regimes (Figure 2). Denitrification increased by  $\sim 20$  Tg N/a in case 1 (Fe) and case 3 (Fe+N) under increasing anthropogenic soluble iron inputs compared to the preindustrial control, with a smaller  $\sim 5$  Tg N/a increase in case 2 (N). Increasing iron (and nitrogen) inputs

fueled additional export production above the oxygen minimum zones. The largest increase in nitrogen fixation was about  $\sim 15$  Tg N/a in case 1 (Fe) followed by  $\sim 5$  Tg N/a for case 3 (Fe+N) as increased soluble iron inputs relative to the control reduced the Fe limitation in diazotrophs (N fixers). Increased diazotroph Fe and P limitation reduced nitrogen fixation by  $\sim 5$  Tg N/a in case 2 (N) as the diazotrophs were outcompeted by the diatoms and small phytoplankton under increased anthropogenic nitrogen inputs.

[27] Primary production increased from  $\sim 53.5$  Pg C/a in preindustrial control to  $\sim 55$  Pg C/a under case 3 (Fe+N), closely followed by case 1 (Fe). Case 2 (N) had only a marginal increase of  $\sim 0.5$  Pg C/a in primary production compared to the preindustrial control. Case 3 (Fe+N) had the highest sinking particulate organic carbon (POC) flux at 103 m of  $\sim 5.8$  Pg C/a closely followed by case 1 (Fe) with  $\sim 5.75$  Pg C/a, whereas it was  $\sim 5.6$  Pg C/a under preindustrial control. Sinking POC export increased only marginally ( $\sim 0.05$  Pg C/a) under case 2 (N) compared to the preindustrial control. Atmospheric  $\text{CO}_2$  declined relative to control by  $\sim 1.8$  ppm,  $\sim 0.5$  ppm, and  $\sim 2.2$  ppm at the end of the simulations for cases 1 (Fe), 2 (N), and 3 (Fe+N), respectively.

[28] Nitrogen fixation was higher in both the subtropical North and South Pacific and the South Atlantic in case 1 (Fe) compared to preindustrial control. Increased atmospheric processing resulted in higher modern soluble Fe inputs to these regions, which were away from major dust sources, partially relieving Fe limitation of diazotrophs (Figure 3b). There was a decrease in  $\text{N}_2$  fixation in the



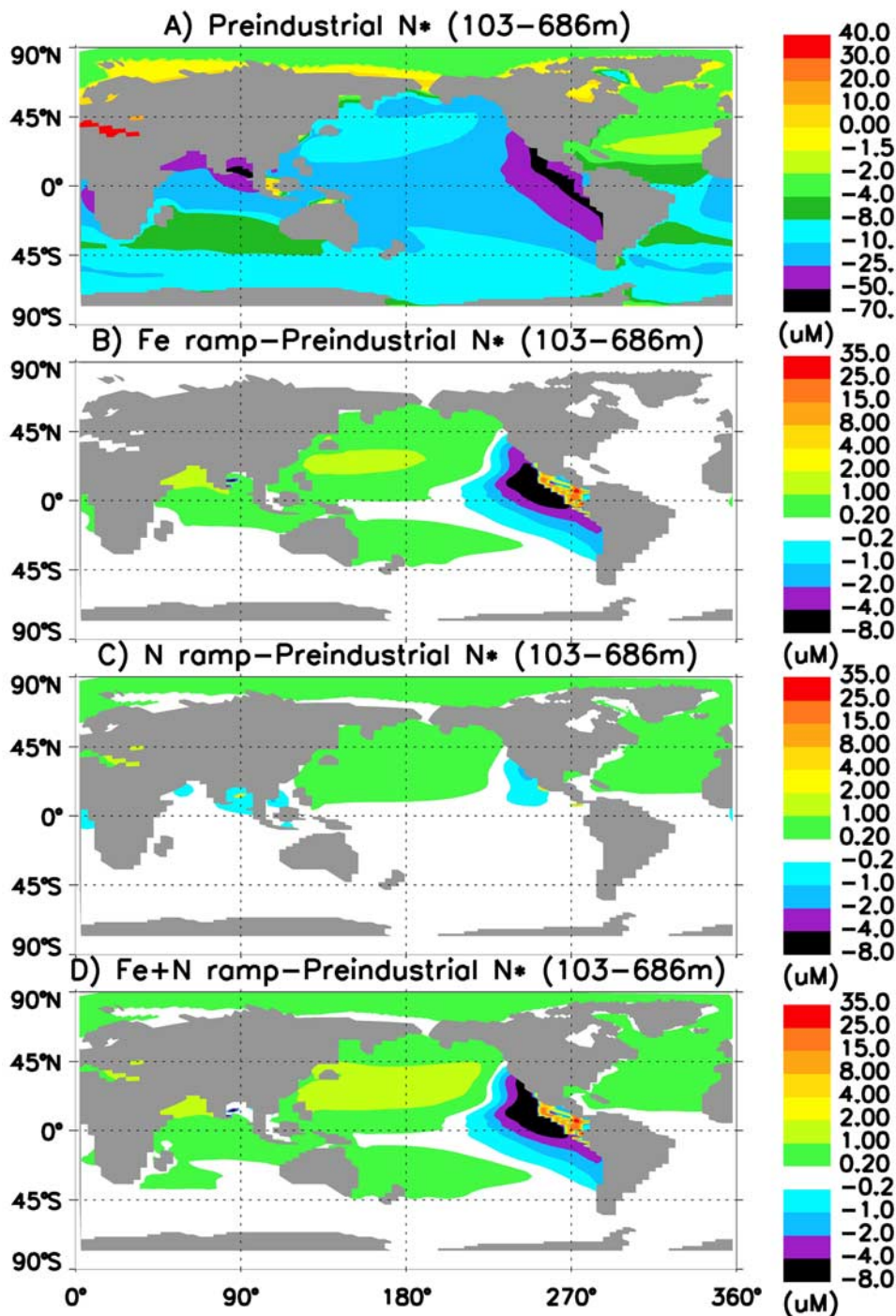
**Figure 3.** Global values of N fixation for the final year of simulations under (a) preindustrial with the difference between cases (b) 1 (Fe), (c) 2 (N), and (d) 3 (Fe+N) and preindustrial.

north Indian Ocean and equatorial Atlantic due to increased P limitation of diazotrophs, as less excess P was exported from the Pacific to the Indian Ocean and from the South Atlantic to the tropical Atlantic. Compared to preindustrial  $\text{N}_2$  fixation decreased over the subtropical North Pacific, the Indian, and the Atlantic Ocean in case 2 (N) (Figure 3c).  $\text{N}_2$  fixation increased in case 3 (Fe+N) compared to preindustrial because of increased soluble iron inputs in the subtropical North Pacific (Figure 3d). Increased Fe and P limitation of N fixers due to enhanced uptake by diatoms and small phytoplankton under higher N availability, how-

ever, resulted in a larger decrease in  $\text{N}_2$  fixation in the Arabian Sea, Bay of Bengal, and across the Atlantic Ocean. Compared to the preindustrial control, simulated global  $\text{N}_2$  fixation increased by  $\sim 11\%$  and  $\sim 5\%$  under cases 1 (Fe) and 3 (Fe+N), whereas it decreased by  $\sim 6\%$  under case 2 (N).

[29] Atmospheric N deposition was  $\sim 2.4\%$  and  $\sim 4.2\%$  of sinking particulate organic nitrogen (PON) export for the preindustrial and the 1990s, respectively [Krishnamurthy *et al.*, 2007a]. In the same study, atmospheric N supported  $\sim 18\%$  of sinking PON export in the western subtropical

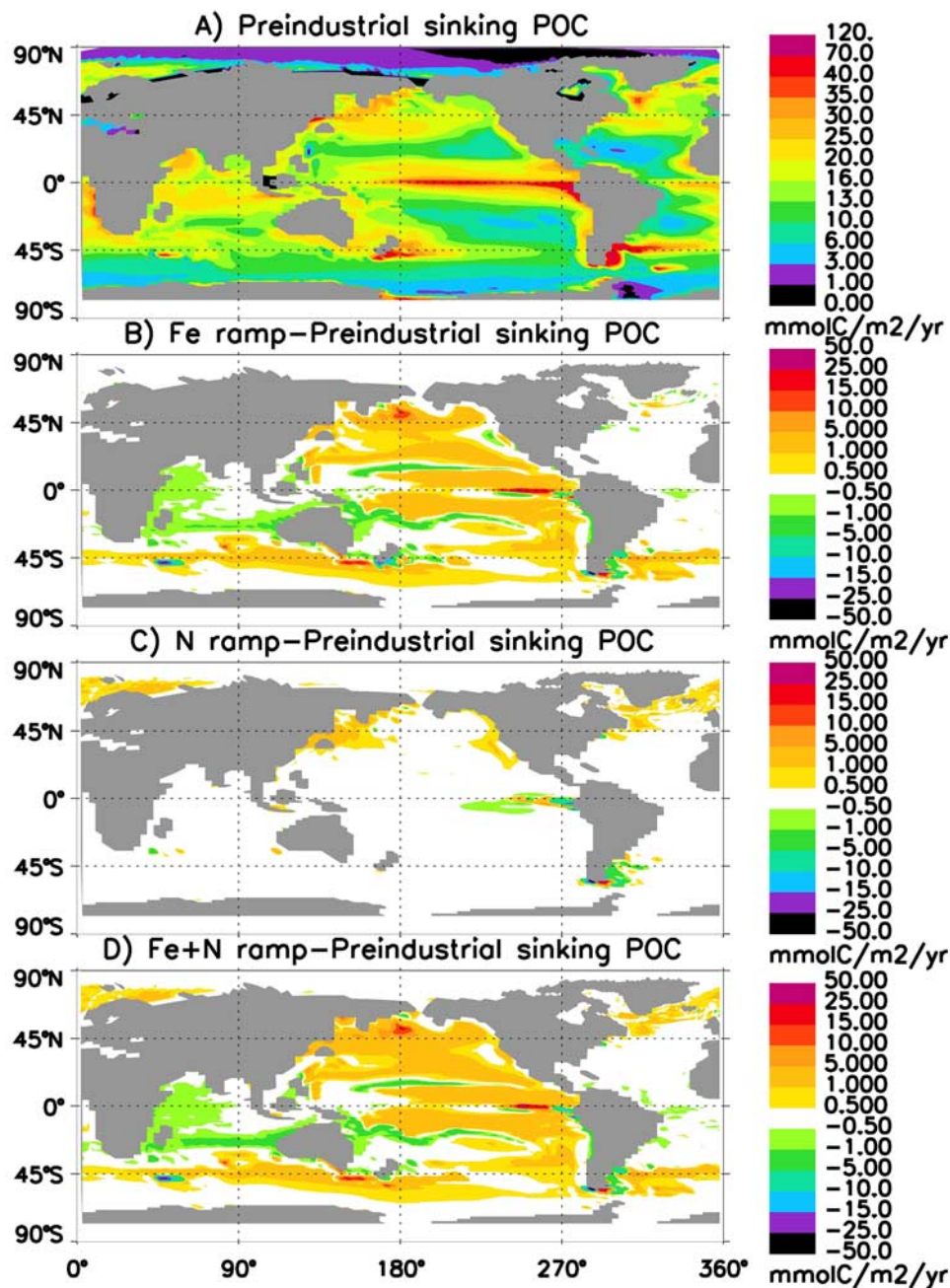




**Figure 4.** Global values of N averaged over the depths of 103 m to 686 m for the final year of simulations under (a) preindustrial with the difference between cases (b) 1 (Fe), (c) 2 (N), and (d) 3 (Fe+N) and preindustrial.

Pacific,  $\sim 6\%$  in the Arabian Sea, and  $\sim 9\%$  in the Bay of Bengal. For comparison, *Duce et al.* [2008] include both organic and inorganic nitrogen deposition over global oceans and estimated that  $\sim 3\%$  of the annual new production could be accounted for by atmospheric deposition of anthropogenic nitrogen.

[30] A number of authors since Redfield have suggested using excess inorganic nitrogen relative to inorganic phosphorus at Redfield stoichiometry as a quasi-conservative tracer that reflects the net impact of  $\text{N}_2$  fixation, denitrification, and other processes that add or remove fixed N but not P [e.g., *Michaels et al.*, 1996; *Gruber and Sarmiento*, 1997; *Gruber*, 2004]. Atmospheric deposition of Fe and N



**Figure 5.** Global values of sinking particulate organic carbon (POC) export at 103 m for the final year of simulations under (a) preindustrial with the difference between cases (b) 1 (Fe), (c) 2 (N), and (d) 3 (Fe+N) and preindustrial.

can act to increase upper ocean excess inorganic nitrogen as Fe stimulates  $N_2$  fixation and because the N/P ratios of N fixers are much higher than Redfield stoichiometries [Letelier and Karl, 1996]; conversely, Fe inputs can lower excess inorganic nitrogen if the resulting elevated export production stimulates more denitrification.

[31] We examined the depth-averaged excess inorganic nitrogen ( $[NO_3^-] + [NH_4^+] - 16[PO_4^{3-}]$ ) over the model upper thermocline (103 m to 686 m) (Figure 4). Compared to the preindustrial control (Figure 5a), cases 1 (Fe) and 3

(Fe+N) had increased nitrogen fixation under higher soluble iron inputs resulting in higher excess inorganic nitrogen values in the subtropical North Pacific. In the Arabian Sea, for cases 1 (Fe) and 3 (Fe+N), excess inorganic nitrogen increased despite lower values of  $N_2$  fixation (Figure 3), in part because of low denitrification in our model in the oxygen minimum zones in this region [Moore and Doney, 2007]. Increased  $N_2$  fixation under reduced Fe limitation resulted in increased excess inorganic nitrogen values right off the coast in the eastern equatorial Pacific (Figures 4b and 4d).

**Table 1.** Total Global Primary Production, N Fixation, Sinking POC From the Final Year of Simulation (Year 650), Atmospheric pCO<sub>2</sub> at the End of the 650 Year Simulations, Diatom, Small Phytoplankton, and Diazotroph Nutrient, and Light Limitation Factors over Global Ocean Area<sup>a</sup>

	Preindustrial	Case 1	Case 2	Case 3
Total primary production (Pg C/a)	53.65	54.75(+1.10)	53.84(+0.19)	54.95(+1.3)
Total nitrogen fixation (Tg N/a)	120.32	133.73(+13.41)	113.07(-7.25)	126.18(+5.86)
Total sinking POC (Pg C/a)	5.62	5.76(0.14)	5.65(0.03)	5.8(+0.18)
Final atmospheric PCO <sub>2</sub> (ppm/a)	280.005	278.237(-1.77)	279.531(-0.47)	277.784(-2.22)
Diatoms				
Fe-limited	34.11	28.95(-5.16)	34.74(+0.63)	29.08(-5.03)
N-limited	46.65	50.05(+3.4)	43.44(-3.21)	46.65(0)
P-limited	8.783	9.607(+0.82)	11.27(+2.48)	12.57(+3.79)
Si-limited	6.327	7.281(+0.95)	6.513(+0.19)	7.508(+1.18)
Light-limited	4.112	4.101(-0.011)	4.026(-0.09)	4.179(+0.07)
Small Phytoplankton				
Fe-limited	34.66	29.04(-5.62)	35.19(+0.53)	29.31(-5.35)
N-limited	43.99	48.15(+4.16)	41.37(-2.62)	45.28(+1.29)
P-limited	7.696	8.331(+0.64)	9.742(+2.046)	10.62(+2.92)
Light-limited	13.64	14.46(+0.82)	13.69(+0.05)	14.78(+1.14)
Diazotrophs				
Fe-limited	35.76	32.51(-3.25)	35.21(-0.55)	31.96(-3.8)
P-limited	26.74	29.8(+3.06)	27.37(+0.63)	30.47(+3.73)
Light-limited	2.332	2.525(+0.19)	2.25(-0.08)	2.401(+0.07)

<sup>a</sup>Values in parentheses are the difference of these values relative to the preindustrial control.

The increased export combined with the increased N<sub>2</sub> fixation resulted in increased denitrification in the eastern equatorial Pacific under cases 1 (Fe) and 3 (Fe+N), lower excess inorganic nitrogen compared to the preindustrial control further offshore. Under case 2 (N), compared to preindustrial control, increased nitrogen deposition resulted in higher excess inorganic nitrogen values throughout the North Atlantic and the North and South Pacific (Figure 4c).

[32] Changing atmospheric nutrient inputs modestly shifted spatial patterns of nutrient limitation. Compared to the preindustrial control, Fe-limited regions decreased by ~5% in both cases 1 (Fe) and 3 (Fe+N) and increased by ~1% under case 2 (N). N-limited regions increased by ~5% under case 1 (Fe), decreased by ~4% under case 2 (N), and remained the same under case 3 (Fe+N). P-limited regions increased by ~1%, ~3%, and ~4% under cases 1 (Fe), 2 (N), and 3 (Fe+N), respectively. Si-limited regions increased by 2% to 4% under the three cases. Exact differences in nutrient-limited and light-limited areas between control and individual cases for small phytoplankton, diatoms, and diazotrophs are summarized in Table 1.

[33] We compare spatial maps from the end of the simulations to examine how increasing anthropogenic nitrogen and soluble iron inputs may be perturbing the global carbon cycle. The mean annual sinking POC export at 103 m increased in the North and South Pacific and the Southern Ocean compared with preindustrial conditions under cases 1 and 3 (Figures 5b and 5d), because of decreased iron limitation in the HNLC areas and increased N<sub>2</sub> fixation in the subtropics. This led to a net oceanic CO<sub>2</sub> uptake at the corresponding locations (Figures 6b and 6d). The upwelling of inorganic carbon-enriched waters as a result of increased export in the subtropics resulted in some increased CO<sub>2</sub> outgassing in the equatorial Pacific.

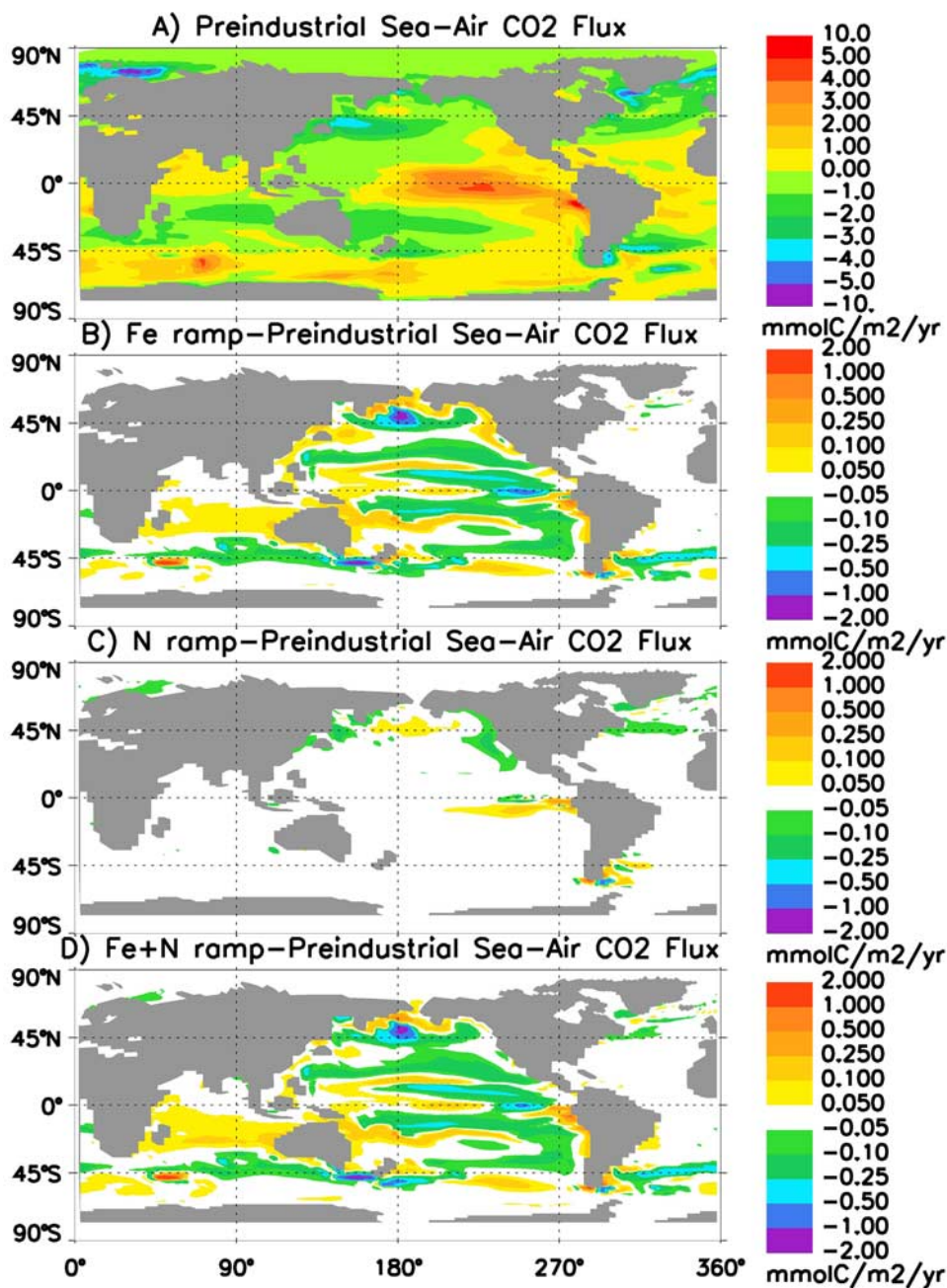
[34] There was a minimal change in sinking POC export under case 2 (N) in the North Atlantic, subarctic North

Pacific, and the equatorial Pacific resulting in a small change in the sea-air CO<sub>2</sub> flux at the corresponding locations (Figures 5b and 6b). Sinking POC export at 103 m increased globally by 0.14 Pg C/a (or ~2.5%), 0.03 Pg C/a (or ~0.5%), and 0.18 Pg C/a (or ~3.2%) since preindustrial under cases 1 (Fe), 2 (N), and 3 (Fe+N), respectively (Table 1).

[35] Overall, our simulations suggest that the influence of elevated human-induced soluble iron inputs alone as well as combined with increasing anthropogenic nitrogen inputs to global marine surface waters has a significant impact on marine biogeochemistry, particularly the nitrogen cycle and excess inorganic nitrogen distributions. Including only the increasing soluble iron, however, had a greater effect than anthropogenic nitrogen inputs alone on the sinking POC export and the air-sea CO<sub>2</sub> flux.

#### 4. Discussion and Summary

[36] Recent studies suggest that regional variations in atmospheric iron inputs may have a significant impact on the spatial patterns and rates of marine nitrogen cycling. Falcon *et al.* [2004] measured in situ rates of N<sub>2</sub> fixation in the tropical North Atlantic (summer 2001 and spring 2002) and the subtropical North Pacific (fall 2002) near station ALOHA off Hawaii. They found that the higher aeolian iron inputs to the North Atlantic resulted in higher N<sub>2</sub> fixation rates, when compared to that at the site near station ALOHA. On the basis of cruise observations collected in the North Atlantic, North Pacific, western South Pacific, and the western North Atlantic, Hynes *et al.* [2009] found that *Trichodesmium* N<sub>2</sub> fixation rates were significantly higher in the subtropical North Atlantic than in the tropical and subtropical North and South Pacific. Their study suggested that *Trichodesmium* may be more P-limited in the North Atlantic, while they inferred that *Trichodesmium*



**Figure 6.** Global values of sea-air CO<sub>2</sub> flux for the final year of simulations under (a) preindustrial with the difference between cases (b) 1 (Fe), (c) 2 (N), and (d) 3 (Fe+N) and preindustrial.

may be more iron-limited in the North Pacific. Moore *et al.* [2006] suggested that N<sub>2</sub> fixation should be sensitive to iron inputs, particularly in the Pacific. There is increasing evidence for the importance of marine nitrogen fixers beyond *Trichodesmium* including unicellular and symbiotic cyanobacteria [Church *et al.*, 2008], though there is insufficient data on basin to global scale to fully characterize the nitrogen fixation rates of these other groups. Deutsch *et al.* [2007] argued that the North Pacific has higher N<sub>2</sub> fixation rates because of high water column denitrification, despite lower rates of atmospheric iron input.

[37] In the present paper, varying soluble aerosol iron deposition influenced the regional patterns of the simulated nitrogen cycle. Annual mean N<sub>2</sub> fixation increased substantially in some regions under increasing anthropogenic soluble iron inputs, especially across the North and South Pacific, with smaller changes in the Indian and Atlantic Ocean basins (Figure 3b). In the modern era, regions away from aeolian dust sources, like station ALOHA, receive substantially higher soluble iron inputs, in a fractional sense, relative to the preindustrial period. In this study, we model the atmospheric changes on aerosol iron solubility

since preindustrial explicitly; in contrast, the model ocean iron scavenging parameters are fixed in time and space across the control and transient simulations, and time varying changes in iron scavenging are included implicitly only through changes in particle load and soluble iron concentrations. It has been suggested by *Baker and Croot* [2009, 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 post-deposition 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.

[38] Estimates of  $N_2$  fixation at station ALOHA (22.75°N, 158°W) from previous studies range between 31 and 51 mmol  $N/m^2/a$  [*Karl et al.*, 1997] to 30 and 282 mmol  $N/m^2/a$  [*Montoya et al.*, 2004].  $N_2$  fixation values for this region from our simulations were 55 mmol  $N/m^2/a$ , 80 mmol  $N/m^2/a$ , 53 mmol  $N/m^2/a$ , and 78 mmol  $N/m^2/a$  for the final year under preindustrial, case 1 (Fe), case 2 (N), and case 3 (Fe+N), respectively. At station ALOHA, the increase in  $N_2$  fixation since preindustrial was 25 mmol  $N/m^2/a$  and 23 mmol  $N/m^2/a$  under cases 1 (Fe) and 3 (Fe+N), respectively.

[39] Using observational estimates of the sinking particulate organic nitrogen data at 150 m from station ALOHA, we calculated an increasing linear trend over 1989–2006 of 0.59 mmol  $N/m^2/a$ . Using the observational sinking particulate organic C flux at 150 m, and converting it to N with the Redfield ratio, we obtained an increasing trend of 0.23 mmol  $N/m^2/a$ . This value was similar to the iron-driven increase in  $N_2$  fixation in our simulation (25 mmol  $N/m^2$  in 150 years  $\sim 0.17$  mmol  $N/m^2/a$ ). Our model results likely underestimate the increases in iron solubility in recent decades, because of the assumed linear trend. Thus, our results suggest that increasing soluble iron inputs from the atmosphere may be contributing substantially to the shift toward increasing P limitation at station ALOHA. That is, a substantial part of the observed  $N_2$  fixation increase may be due to the atmospheric deposition, rather than (or in addition to) decreases in mixed layer depth and changes in nutrient input from intermediate waters due to increased stratification as suggested previously [i.e., *Karl*, 1999].

[40] Hydrographic surveys have indicated that oceanic dissolved  $O_2$  concentrations have changed, particularly during the later part of 20th century [*Keeling and Garcia*, 2002, and references therein]. *Manning and Keeling* [2006] estimated oceanic  $O_2$  outgassing associated to global warming over 1993–2003 to be  $\sim 45$  Tmol  $O_2/a$ . In our simulations, increasing soluble iron and nitrogen inputs increased oceanic  $O_2$  outgassing by  $\sim 10$  Tmol  $O_2/a$  in the modern era compared to the preindustrial control. Thus, biogeochemical trends may be significantly influencing air-sea  $O_2$  flux, in addition to the temperature effects.

[41] Several studies have suggested a decreasing trend in the dissolved oxygen concentrations in the lower thermocline over the later part of the 20th century [*Joos et al.*, 2003] with the maximum observed change in the North

Pacific [*Emerson et al.*, 2004]. The simulated subsurface dissolved  $O_2$  distributions in the North Pacific in the present study were not significantly affected by the increases in export over the last few decades of the simulations. Our simulated changes to dissolved  $O_2$  concentrations were much less than the trends in the observations for the North Pacific during 1980 to 2000 by *Emerson et al.* [2004] as well as those described in the simulations done by *Deusch et al.* [2005, 2006]. This supports the idea that changes in physics (held constant in our simulations) and ventilation may be driving the observed changes in oxygen. The detection of long-term observational trends due to climate change and anthropogenic forcing are complicated by substantial interannual variability [*Mecking et al.*, 2008]. Recent modeling studies argue that the large trends in recent observational oxygen estimates may largely reflect natural climate variability rather than long-term secular trends [*Frölicher et al.*, 2009]. The significant changes in excess inorganic nitrogen distributions suggest that the excess inorganic nitrogen field is not static and may be a useful diagnostic of ongoing changes in ocean circulation and biogeochemistry that complements oxygen.

[42] Compared to the preindustrial era, increasing soluble iron inputs via combustion sources and atmospheric processing of mineral aerosols along with atmospheric anthropogenic nitrogen inputs resulted in a maximum global increase in primary production by  $\sim 2.4\%$  in our simulations. This increased the sinking POC export at 103 m by  $\sim 3.2\%$  leading to a reduction in atmospheric  $pCO_2$  by  $\sim 2.2$  ppm. This atmospheric  $CO_2$  reduction is small compared with the observed increase of  $\sim 100$  ppm, but would likely continue to increase in the coming decades as nutrient inputs to the oceans continue to rise (Figure 2). Extending our simulations with increasing iron and nitrogen inputs for another 100 years at the same linear rate resulted in  $\sim 4$  ppm decrease in atmospheric  $pCO_2$  compared to preindustrial control. The significant sedimentary iron source reduces the model sensitivity to variations in atmospheric iron inputs [*Moore and Braucher*, 2008].

[43] Several modeling studies have estimated changes in oceanic  $CO_2$  uptake since preindustrial conditions by applying various scenarios of atmospheric  $CO_2$  increase in the 21st century [*Sarmiento and Le Quéré*, 1996; *Sarmiento et al.*, 1998; *Joos et al.*, 1999; *Matear and Hirst*, 1999; *Greenblatt and Sarmiento*, 2004]. Compared to preindustrial conditions, these studies estimate that under the current era, there would be an increase in oceanic  $CO_2$  uptake due to biology (by  $\sim 6$ – $27\%$ ) but decreased oceanic  $CO_2$  uptake due to changes in stratification (by  $\sim 3$ – $20\%$ ) and  $CO_2$  solubility (by  $\sim 9$ – $14\%$ ) due to warming of surface waters [*Sabine and Feely*, 2007, and references therein]. Our results isolate the anthropogenic-nutrient-deposition-driven ocean biogeochemical response. This response is relatively modest at present ( $-2$  ppm  $CO_2$ ) but growing and could be important, especially when combined with the above mentioned climate-driven modifications and additional nutrient perturbations from riverine sources. Hence, these factors need to be considered when assessing the anthropogenic impacts on marine ecosystem productivity, air-sea  $CO_2$  fluxes, and the global carbon cycle.

[44] The present work highlights the importance of studying the effects of varying iron solubility on oceanic ecosystems on a global scale. Better understanding of the mechanisms by which soluble iron is delivered to the surface ocean from anthropogenic environmental changes is required. This could be critical in providing improved estimates of bioavailable iron inputs to areas remote from aeolian dust sources. Modifications to iron solubility may occur in the near future based on what energy choices humans make. Enhanced fossil fuels usage and coal-based technology in the future would influence iron solubility closer to source regions as well as impact atmospheric anthropogenic N inputs. Better understanding of the availability of atmospheric organic nitrogen inputs and the bioavailability of the atmospheric N is needed [Duce *et al.*, 2008].

[45] Ongoing changes due to increased nitrate release from land due to fertilizer use as well as nitrogen deposition from the atmosphere in highly polluted areas [De Leeuw *et al.*, 2001], combined with a decrease of dust mobilization and transport in a warmer climate [Werner *et al.*, 2002; Mahowald and Luo, 2003], will impact atmospheric iron and nitrogen availability to surface oceans. Some studies also suggest that there could be an increase in dust loads due to changes in land use [Teegen *et al.*, 2004] and in vegetation cover [Woodward *et al.*, 2005]. Increasing pollution would affect atmospheric processing of mineral dust, in turn affecting aerosol iron solubility and hence, marine ecosystem bioavailability. On the other hand, a shift toward renewable energy sources for meeting greenhouse gas emission reduction goals would likely decrease iron solubility and nitrogen inputs into the atmosphere because of changes in combustion sources and atmospheric processes. Future simulations could include variable aerosol iron solubility derived from mineral dust and combustion sources under different emission, climate, and energy use scenarios to better understand the consequences of alternative energy strategies.

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