Impacts of atmospheric nutrient deposition on marine productivity: Roles of nitrogen, phosphorus, and iron

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[1]   Nutrients are supplied to the mixed layer of the open ocean by either atmospheric deposition or mixing from deeper waters, and these nutrients drive nitrogen and carbon fixation. To evaluate the importance of atmospheric deposition, we estimate marine nitrogen and carbon fixation from present-day simulations of atmospheric deposition of nitrogen, phosphorus, and iron. These are compared with observed rates of marine nitrogen and carbon fixation. We find that Fe deposition is more important than P deposition in supporting N fixation. Estimated rates of atmospherically supported carbon fixation are considerably lower than rates of marine carbon fixation derived from remote sensing, indicating the subsidiary role atmospheric deposition plays in total C uptake by the oceans. Nonetheless, in high-nutrient, low-chlorophyll areas, the contribution of atmospheric deposition of Fe to the surface ocean could account for about 50% of C fixation. In marine areas typically thought to be N limited, potential C fixation supported by atmospheric deposition of N is only ~1%–2% of observed rates. Although these systems are N-limited, the amount of N supplied from below appears to be much larger than that deposited from above. Atmospheric deposition of Fe has the potential to augment atmospherically supported rates of C fixation in N-limited areas. In these areas, atmospheric Fe relieves the Fe limitation of diazotrophic organisms, thus contributing to the rate of N fixation. The most important uncertainties in understanding the relative importance of different atmospheric nutrients are poorly understood speciation and solubility of Fe as well as the N:Fe ratio of diazotrophic organisms.

Citation: Okin, G. S., et al. (2011), Impacts of atmospheric nutrient deposition on marine productivity: Roles of nitrogen, phosphorus, and iron, *Global Biogeochem. Cycles*, *25*, GB2022, doi:10.1029/2010GB003858.

1. Introduction

[2] Atmospheric deposition can be an important source of nitrogen (N), phosphorus (P), and iron (Fe) to the open ocean [*Jickells et al.*, 2005; *Krishnamurthy et al.*, 2007; *Duce et al.*, 2008; *Krishnamurthy et al.*, 2009; *Mahowald et al.*, 2009; *Krishnamurthy et al.*, 2010]. Fe, deposited largely as desert dust, is thought to limit primary productivity in large areas of the surface oceans, particularly in High Nutrient Low Chlorophyll (HNLC) areas [*Martin et al.*, 1994]. On geologic time scales, the oceans are thought to be limited by P [*Falkowski et al.*, 1998], but in much of the ocean on shorter time scales, N limitation is most important [e.g., *Ryther and*

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Table 1. Estimated Rates of Nutrient Deposition to the Global Ocean and Major Ocean Basins

Basin[a]	N_{Dep} (Tg/yr)	P_{Dep} (Tg/yr)	Fe_{Dep} (Tg/yr)
Global Ocean	63	0.32	0.36
Atlantic	18	0.13	0.11
North Atlantic	7.4	0.020	0.019
North Central Atlantic	4.1	0.048	0.037
Equatorial Atlantic	4.6	0.055	0.048
South Atlantic	1.9	0.005	0.008
Indian	11	0.073	0.10
North Indian	4.1	0.043	0.040
Equatorial Indian	4.3	0.025	0.049
South Indian	2.3	0.005	0.012
Pacific	24	0.067	0.10
North Pacific	7.0	0.026	0.034
North Central Pacific	7.5	0.020	0.036
Equatorial Pacific	6.9	0.018	0.023
South Pacific	2.8	0.0039	0.007
Southern Ocean	8.2	0.032	0.039

[a]Basins as defined by *Gregg et al.* [2003]. Latitudinal boundaries between subbasins are at 30°N, 10°N, 10°S, and 30°S.

Dunstan, 1971; *Moore et al.*, 2001; *Mills et al.*, 2004; *Doney et al.*, 2007; *Krishnamurthy et al.*, 2007, 2009]. Although some marine organisms (diazotrophs) can fix molecular nitrogen into a bioavailable form, this requires the availability of Fe and P, suggesting that the flux of N to the ocean is influenced by both Fe and P availability. At the same time, atmospheric deposition of N may stimulate the growth of nondiazotrophic organisms, provided sufficient Fe and P is available to support this production. Ocean productivity is linked to carbon uptake by the ocean [*Falkowski et al.*, 1998], and thus N, P and Fe deposition to the ocean have the potential to influence the global carbon cycle.

[3] To estimate the effects of atmospheric nutrient deposition on marine productivity, their input into the oceans must be quantified. Currently there are estimates of atmospheric deposition of N, P and Fe to the oceans, based on a combination of models and observations [e.g., *Galloway et al.*, 2004; *Duce et al.*, 2008; *Mahowald et al.*, 2008, 2009]. Some studies have assessed the importance of aerosol deposition in the framework of ocean biogeochemistry models [e.g., *Krishnamurthy et al.*, 2010], however such model estimates show very different responses to atmospheric deposition [e.g., *Aumont et al.*, 2008; *Krishnamurthy et al.*, 2009]. While *Duce et al.* [2008] argues that N deposition to the ocean causes substantial impacts to ocean biogeochemistry, *Krishnamurthy et al.* [2009] uses an ocean biogeochemistry model to argue that larger changes to the ocean nitrogen budget occur due to changes in the iron deposition than through N deposition directly. *Doney et al.* [2007] considers the impact of deposition onto ocean biogeochemistry, focusing on the issue of atmospheric deposition contributions to ocean acidification.

[4] Here we compare maximum atmospherically supportable estimates of nitrogen and carbon fixation with observed rates, with the aim of establishing the extent to which these parameters are driven by atmospheric deposition. Our approach is new in that it is not based on any particular numerical physical or biological model, but on simple-to-understand, observationally based estimates of the impact of

deposition. This allows us to contrast the complicated physical model results with a simple approach to better understand the processes involved.

2. Methods

2.1. Deposition of N, P, and Fe

[5] The deposition fields of N, P and soluble Fe under current conditions used in this study (Table 1 and Figure 1) were obtained from model estimates, which have been previously compared to available observational data. Spatial estimates of inorganic and organic N deposition were obtained from *Duce et al.* [2008] from an ensemble calculation of deposition using 24 different global transport-chemistry models (Figure 1a). N sources considered were those from unmanaged terrestrial ecosystems, lightning, fossil fuel combustion, food production, the Haber-Bosch fertilization process, and marine NH_3 production. All inorganic and a large proportion of soluble organic forms in atmospheric deposition are bioavailable. The insoluble organic N fraction of atmospheric deposition [*Russell et al.*, 2003] is much less well characterized and its bioavailability is uncertain. Overall, however, the majority of atmospheric N inputs to the
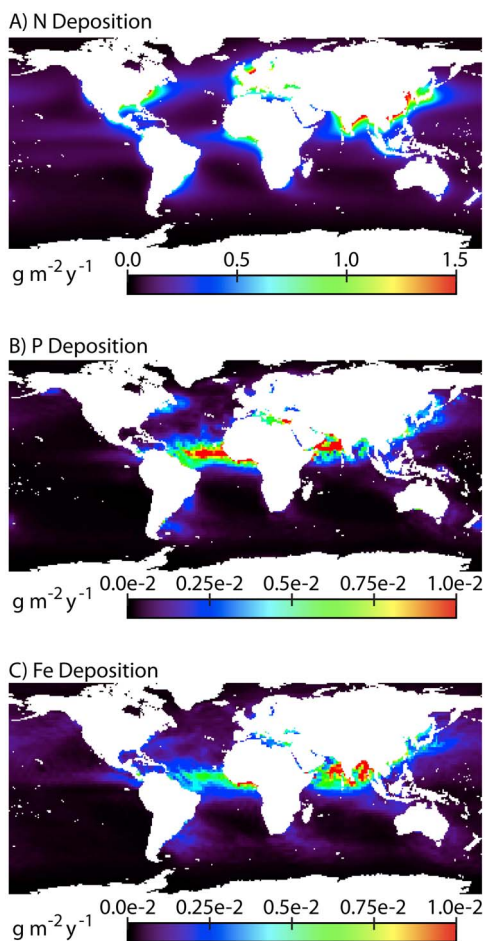

Figure 1. Estimates of atmospheric deposition of (a) N (N_{Dep}), (b) P (P_{Dep}), and (c) Fe (Fe_{Dep}) to the global ocean used in this study.

ocean appear to be both soluble and bioavailable. The global estimated rate of N deposition is 63 Tg/yr (Table 1).

[6] Estimates of P deposition were derived from estimates of desert dust deposition and deposition of phosphorus in nondust aerosols (Table 1 and Figure 1b). Total P deposition to the oceans, P_{Dep}, was calculated as

$$P_{Dep} = D_{Dep}P_{Conc} + P_{Comb} + P_{Nat}, \tag{1}$$

where D_{Dep} is the rate of dust deposition from *Luo et al.* [2008], P_{Conc} is the mass concentration of P in dust, P_{Comb} is the rate of P deposition from combustion aerosols, and P_{Nat} is the rate of P deposition from nondust natural aerosols. Here we used the same dust deposition field as in the calculation of Fe_{Dep} below. A crustal value of 700 ppm by mass (ppm(m)) was used for P_{Conc} [*Taylor and McLennan*, 1995; *Okin et al.*, 2004], though there is considerable variability in the concentration of P in mineral aerosols (e.g., reviewed by *Mahowald et al.* [2008]). While this value may be considerably higher in some regions, it is consistent with dust from North Africa, which constitutes the largest source of mineral aerosols. P_{Comb} and P_{Nat} were obtained from *Mahowald et al.* [2008], who estimate fossil fuel combustion following *Bond et al.* [2004, 2007] and biomass combustion using the monthly mean biomass burning source from *van der Werf et al.* [2003]. P_{Nat} is estimated by *Mahowald et al.* [2008] as the sum of P deposition from primary biogenic, sea salt, and volcanic aerosols. These estimates result in total P deposition to the oceans of 0.32 Tg/yr. P solubility was not considered for reasons to be explained in the discussion section. Assuming values of P_{Conc} at the high end of measured values (~1300 ppm(m)) would increase P_{Dep}, but not enough to alter our conclusions regarding the relative importance of P deposition for marine productivity.

[7] Estimates of soluble Fe deposition were derived from estimates of desert dust deposition and deposition of iron in combustion aerosols, based on the work of *Luo et al.* [2003, 2008] and *Mahowald et al.* [2009] (Table 1 and Figure 1c). Here, we assume soluble iron is the fraction of iron that is Fe(II). We used total bioavailable Fe deposition to the oceans from Scenario III of *Mahowald et al.* [2009], which was calculated as Fe_{Dep}:

$$Fe_{Dep} = Fe_{Sol} + Fe_{Comb}, \tag{2}$$

where Fe_{Sol} is the soluble Fe deposited from dust, and Fe_{Comb} is the rate of Fe deposition from combustion aerosols. Fe_{Sol} and Fe_{Comb} were obtained from *Luo et al.* [2008] and include the effect of processing of iron oxides by acids in the atmosphere. Estimated total soluble iron deposition to the oceans is 0.36 Tg/yr. All of these estimates include the dry and wet deposition of particulates, while N_{Dep} also includes dry deposition of gas-phase N, because atmospheric N contains significant gaseous as well as aerosol species.

2.2. Potential N and C Fixation From Atmospheric Input

[8] To start, we assume that all nonlimiting nutrients for N and C fixation are present in the surface in abundance and that atmospheric deposition of the limiting nutrient results in additional production. It is this additional production that we quantify here.

[9] N fixing organisms (which utilize Fe and P [e.g., *Mills et al.*, 2004] convert highly abundant molecular nitrogen gas into biologically available nitrogen compounds. We defined the likely limitation of nitrogen fixation through the surface ocean with reference to excess phosphate, $xsPO_4$ (also termed P*) [*Deutsch et al.*, 2007] concentrations:

$$xsPO_4 = PO_4 - NO_3/16. \tag{3}$$

Nitrate and phosphate concentration in surface waters were obtained from the LEVITUS94 concentration data sets [*Conkright et al.*, 1994] (see http://iridl.ldeo.columbia.edu/SOURCES/.LEVITUS94/) for the top layer (10 m). We considered that regions where $xsPO_4$ was less than 0.1 μM [*Moutin et al.*, 2008] were likely to be phosphate limited for nitrogen fixation and we assumed that in all other regions nitrogen fixation was therefore likely to be subject to Fe limitation. Other cutoff concentrations of $xsPO_4$ (0.05 and 0.2 μM) were also examined to identify the sensitivity of our results to this value. We note that these assumptions are likely to be oversimplifications. In some regions nitrogen fixation appears to be colimited by both Fe and P (e.g., the tropical northeast Atlantic [*Mills et al.*, 2004]) and the nutrient status of the oceanic waters for nitrogen fixation in general, particularly with respect to Fe, are very poorly understood. Here we estimate the maximum potential rate of marine nitrogen fixation due to atmospheric input, N_{Fix}^{Atm}, as

$$N_{Fix}^{Atm} = \begin{cases} P_{Dep}(N/P)_{diaz} & \text{where P is limiting} \\ Fe_{Dep}(N/Fe)_{diaz} & \text{where Fe is limiting.} \end{cases} \tag{4}$$

The N:P and N:Fe ratios for diazotrophic organisms ((N/P)$_{diaz}$ and (N/Fe)$_{diaz}$, respectively), were derived from values given by *Baker et al.* [2007] and based on their review of available data. Given that the reported N:Fe ratio of diazotrophic organisms spans more than an order of magnitude (from 50 to 1050 g/g) [*Berman-Frank et al.*, 2001; *Sañudo-Wilhelmy et al.*, 2001; *Kustka et al.*, 2003] and reported N:P ratios for diazotrophic organisms span only a factor of three (18 to 56 g/g), we present the estimates of N_{Fix}^{Atm} using the extreme values of the N:Fe ratio and an intermediate value of the N:P ratio (37 g N/g P). Due to temperature limitation, N_{Fix}^{Atm} at latitudes greater than 40° was considered negligible [*Carpenter and Capone*, 1992; *Langlois et al.*, 2008].

[10] At present, there are two competing views on the iron requirements for nitrogen fixation by *Trichodesmium*, due in part to the difficulty of the measurements. *Berman-Frank et al.* [2001] hold that Fe:C ratio, and hence the Fe:N ratio, is high (represented here of a value of (N/Fe)$_{diaz}$ = 1050 g/g), whereas *Kustka et al.* [2003] and *Sañudo-Wilhelmy et al.* [2001] suggest that it is lower (represented here of a value of (N/Fe)$_{diaz}$ = 50 g/g), although they suggest that it is still high relative to nondiazotrophs. There does not appear to be, at this time, a way to determine which of these two views is correct, or indeed whether a single number is appropriate everywhere.

[11] Diazotrophic organisms are generally only a small portion of the total living biomass even in ecosystems where N_2 fixation is important. This means that they are in direct competition with other microbes for any Fe or P that may be transported into the surface ocean. However, because they
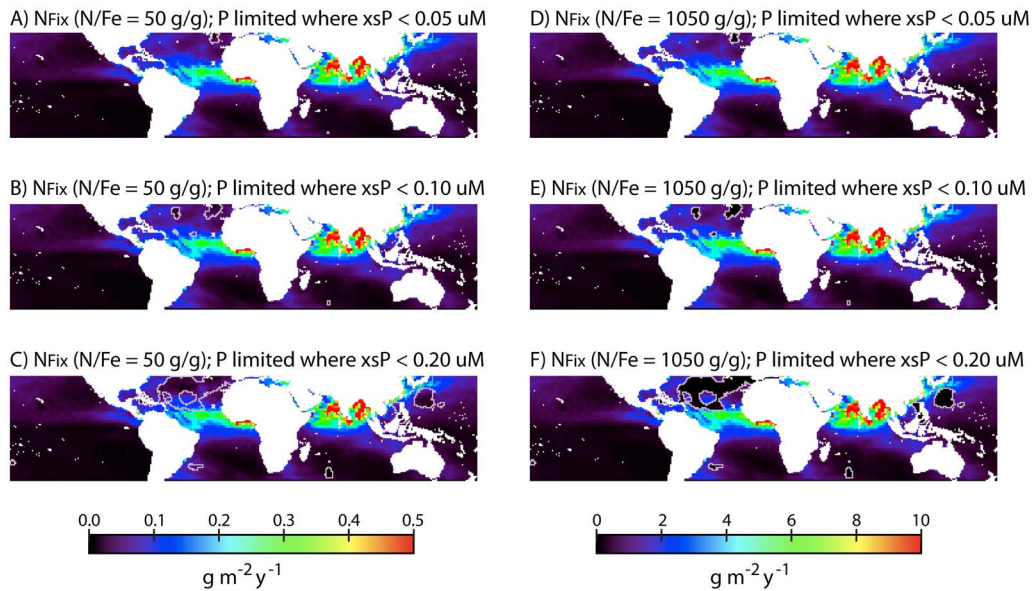
Figure 2. Estimated rates of N fixation supported by atmospheric deposition (N_{Fix}) assuming N:Fe ratio of (a–c) 50 or (d–f) 1050 g/g for diazotrophic organisms between 40°N and 40°S. Figures 2a and 2d assume P is limited where $xsPO_4$ is less than 0.05 μM. Figures 2b and 2e assume P is limited where $xsPO_4$ is less than 0.1 μM. Figures 2c and 2f assume P is limited where $xsPO_4$ is less than 0.2 μM.

are a small portion of the total biomass, we calculated direct carbon fixation due to atmospheric input of N and Fe, $C_{Fix,Fe/N\ Dep}^{Atm}$, as

$$C_{Fix,\ Fe/NDep}^{Atm} = \begin{cases} Fe_{Dep}(C/Fe) & \text{where Fe is limiting} \\ N_{Dep}(C/N)_{Redfield} & \text{elsewhere,} \end{cases} \quad (5)$$

where $(C/N)_{Redfield}$ is the Redfield mass ratio of 7.2 g/g [*Redfield*, 1934] and (C/Fe) is 1.47×10^5 g/g, which is the minimum C:Fe ratio reported by *Sunda* [2001] for iron-depleted waters. The minimum value was used in this calculation to determine the upper limit of the effect of Fe fertilization on C fixation in HNLC regions.

[12] P can limit C fixation either directly as $C_{Fix,\ P\ Dep}^{Atm} = P_{Dep}(C/P)_{Redfield}$ or indirectly, and in combination with Fe, through its impact on N_{Fix}^{Atm}. This indirect carbon fixation was calculated as $C_{Fix,\ N\ Fix}^{Atm} = N_{Fix}^{Atm}(C/N)_{Redfield}$, assuming that N fixed by diazotrophs (i.e., that calculated using the N:P and N:Fe ratios of diazotrophic organisms) is associated with C fixation among autotrophic diazotrophic organisms or can become available on a short timeframe to nondiazotrophic organisms with Redfield-like C:N ratios.

[13] Following *Duce et al.* [2008], we defined ocean areas where primary productivity was likely to be Fe limited (i.e., HNLC regions) as those having annual average NO_3^- concentration >4 μM. Nitrate concentration in surface waters was obtained from the LEVITUS94 nitrate concentration data sets [*Conkright et al.*, 1994] (see http://iridl.ldeo.columbia.edu/SOURCES/.LEVITUS94/) for the top layer (10 m).

[14] Values of atmospherically supported carbon fixation (both direct and indirect) were compared with estimates of average net primary productivity (NPP) derived from the MODerate resolution Imaging Spectrometer (MODIS) using

the method of *Behrenfeld and Falkowski* [1997] from July 2002 to June 2007 (http://www.science.oregonstate.edu/ocean.productivity/index.php) for ocean basins as defined by *Gregg et al.* [2003].

[15] Ecologists distinguish between gross primary productivity (GPP) and NPP as well as net community production (NCP). In most of the world ocean, NPP is less than about 50% of GPP whereas NCP is typically about 10–15% of GPP. This becomes important when making comparisons of the potential carbon fixation from atmospheric deposition of nutrients. The disparities in the fraction of GPP made of NPP and NCP mean that even if only a small amount of NPP is supported from atmospheric source, this could be of the same order as NCP, which, in an idealized steady state ocean, is equal to carbon export. As a result, productivity enhanced by atmospheric deposition has significant potential to impact the ocean's carbon budget.

3. Results

[16] First we consider N fixation. We find that for calculated values of $xsPO_4$, N_{Fix}^{Atm} in most of the surface oceans is Fe limited and the patterns of N_{Fix}^{Atm} are largely determined by the deposition of Fe in dust (Figure 2). The area of the surface oceans in which N_{Fix}^{Atm} is limited by P or Fe depends on the cutoff value of $xsPO_4$, though general patterns determined by the value of 0.1 μM given by *Moutin et al.* [2008] do not change considerably for cutoff values of 0.05 and 0.2 μM (Figure 2). In all cases, the northern Atlantic Ocean is most likely to be P limited with respect to N_{Fix}^{Atm}. Therefore, we do not believe that our results are particularly sensitive to the cutoff values used for $xsPO_4$, particularly given the uncertainty of values of $(N/Fe)_{diaz}$ present in the literature. Indeed, by far the largest impact on our estimates of N_{Fix}^{Atm} relate to

Table 2. Estimated Rates of Nitrogen Fixation, Carbon Fixation, and Satellite-Derived NPP for Major Ocean Basins[a]

	N_{Fix}^{Atm} [b] (Tg/yr)		$C_{Fix,Fe/N\ Dep}^{Atm}$ [c] (Pg/yr)	$C_{Fix,N\ Fix}^{Atm}$ [d] (Pg/yr)		$C_{Fix,P\ Dep}^{Atm}$ [e] (Tg/yr)	NPP[f] (Pg /yr)	$(C_{Fix,Fe/N\ Dep}^{Atm} + C_{Fix,N\ Fix}^{Atm})/NPP$ [g]	
								N/Fe = 50	N/Fe = 1050
N:Fe Ratio[h] (g/g)	50	1050	n/a	50	1050	n/a	n/a	50	1050
Global Ocean	15	323	7.5	0.085	1.8	15	42.6	16	20
Atlantic	5.0	104	0.82	0.028	0.59	6.1	9.84	6.1	13
North Atlantic	0.42	8.0	0.63	0.0024	0.046	1.0	3.68	15	17
North Central Atlantic	1.8	38	0.083	0.010	0.21	2.3	2.18	2.2	14
Equatorial Atlantic	2.4	50	0.053	0.014	0.29	2.6	2.27	2.0	16
South Atlantic	0.39	8.2	0.062	0.0021	0.044	0.24	1.71	1.7	5.1
Indian	5.1	106	0.55	0.028	0.59	3.5	5.12	2.9	12
North Indian	2.0	42	0.46	0.011	0.22	2.1	1.41	12	29
Equatorial Indian	2.4	51	0.073	0.014	0.29	1.2	1.86	2.1	16
South Indian	0.62	13	0.013	0.0036	0.075	0.26	1.84	0.8	4.4
Pacific	4.2	88	2.9	0.022	0.46	3.2	15.9	12	15
North Pacific	0.91	19	2.1	0.0046	0.10	1.2	4.58	43	45
North Central Pacific	1.8	37	0.09	0.010	0.21	1.0	3.09	1.7	8.5
Equatorial Pacific	1.1	24	0.58	0.0053	0.11	0.84	5.16	9.4	12
South Pacific	0.37	7.7	0.072	0.0020	0.041	0.19	3.06	1.7	3.4
Southern Ocean	0.77	16	3.2	0.0039	0.083	1.5	11.4	51	52

[a]Defined as in Table 1.

[b]N_{Fix}^{Atm} assuming that diazotrophic N fixation is P or Fe limited (Figures 2b and 2e) and N:Fe ratio of either 50 or 1050 g/g.

[c]C fixation from N_{Dep} and Fe_{Dep} only (Figure 4a).

[d]C fixation from N_{Fix}^{Atm} only (Figures 4c and 4d).

[e]C fixation from P_{Dep} only (Figure 4b).

[f]Average MODIS-derived NPP [*Behrenfeld and Falkowski*, 1997] from July 2002 to June 2007.

[g]See Figures 5c and 5d.

[h]Calculations of N_{Fix}^{Atm} were made using the range of ratios of N/Fe representing different estimates in the literature [*Berman-Frank et al.*, 2001; *Sañudo-Wilhelmy et al.*, 2001; *Kustka et al.*, 2003].

the assumed values of N/Fe for diazotrophic organisms. Thus, we report results using both estimates of (N/Fe)$_{diaz}$ (50 and 1050 g/g, see Table 2).

[17] In non-HNLC regions, the ratio of N:P in atmospheric deposition is always greater than the Redfield mass ratio (7.2, Log(7.2) = 0.85), suggesting that P would limit productivity if only atmospheric inputs were considered (Figure 3). However, throughout much of the non-HNLC areas, N is considered to limit primary productivity [see *Ryther and Dunstan*, 1971; *Moore et al.*, 2001; *Mills et al.*, 2004; *Doney et al.*, 2007; *Krishnamurthy et al.*, 2007; *Duce et al.*, 2008; *Krishnamurthy et al.*, 2009]. The lack of observed P limitation in surface waters has been (and still remains) widely discussed/debated [see, e.g., *Ryther and Dunstan*, 1971; *Sañudo-Wilhelmy et al.*, 2001; *Mills et al.*, 2004; *Deutsch et al.*, 2007; *Moutin et al.*, 2008]. In our results, the lack of observed P limitation in oceanic productivity results from high concentrations of xsPO$_4$ in surface waters. The amount of oceanic C fixation that can be supported by Fe and N deposition where these elements limit primary productivity ($C_{Fix,Fe/N\ Dep}^{Atm}$; Table 2 and Figure 4a) is several orders of magnitude greater than that which can be supported directly by P deposition ($C_{Fix,P\ Dep}^{Atm}$; Table 2 and Figure 4b). Due to the high C:Fe ratio we assume for phytoplankton under Fe limitation, HNLC areas experience significantly higher rates of atmospherically supported C fixation than non-HNLC (N-limited) areas. Furthermore, the indirect impact of deposition via N fixation ($C_{Fix,\ N\ Fix}^{Atm}$; Table 2 and Figures 4c and 4d), is considerably greater than the direct effect of atmospheric P deposition would be, even if C fixation in the surface oceans were P limited ($C_{Fix,\ P\ Dep}^{Atm}$; Table 2 and Figure 4b).

[18] Our estimates of $C_{Fix,Fe/N\ Dep}^{Atm}$ are somewhat sensitive to the cutoff value for nitrate below which ocean productivity is considered to be N limited (i.e., the boundary of HNLC regions; Table 3). Global values of $C_{Fix,Fe/N\ Dep}^{Atm}$ are 4.6 pg C/yr for a cutoff of 8 μM nitrate, 7.5 pg C/yr for a cutoff of 4 μM nitrate, and 13.7 pg C/yr for a cutoff of 2 μM nitrate. This nearly threefold difference in predicted $C_{Fix,Fe/N\ Dep}^{Atm}$ is not the same for all major ocean basins. For the two major basins that are dominated by HNLC conditions, the North Pacific and the Southern Ocean, the range of $C_{Fix,Fe/N\ Dep}^{Atm}$ spans a factor of 1.6–3.3. For all other basins, the differences span nearly an order of magnitude. These basins, however, contribute little to the total $C_{Fix,Fe/N\ Dep}^{Atm}$, and therefore this difference is not seen in the global oceans.

[19] In general rates of maximum potential carbon fixation sustainable by atmospheric deposition ($C_{Fix,Fe/N\ Dep}^{Atm}$ + $C_{Fix,N\ Fix}^{Atm}$) are lower than rates of NPP estimated from MODIS (Table 2 and Figure 5). However, in HNLC areas maximum potential C fixation sustainable by atmospheric depo-
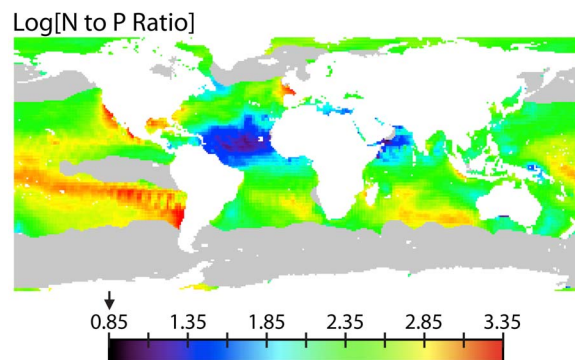

Figure 3. N to P mass ratio in atmospheric deposition. Gray areas are where annual average surface water NO$_3^-$ concentrations exceed 4 μM (assumed here to be HNLC regions). The arrow on the scale bar indicates the Redfield N:P ratio (7.2 g/g).
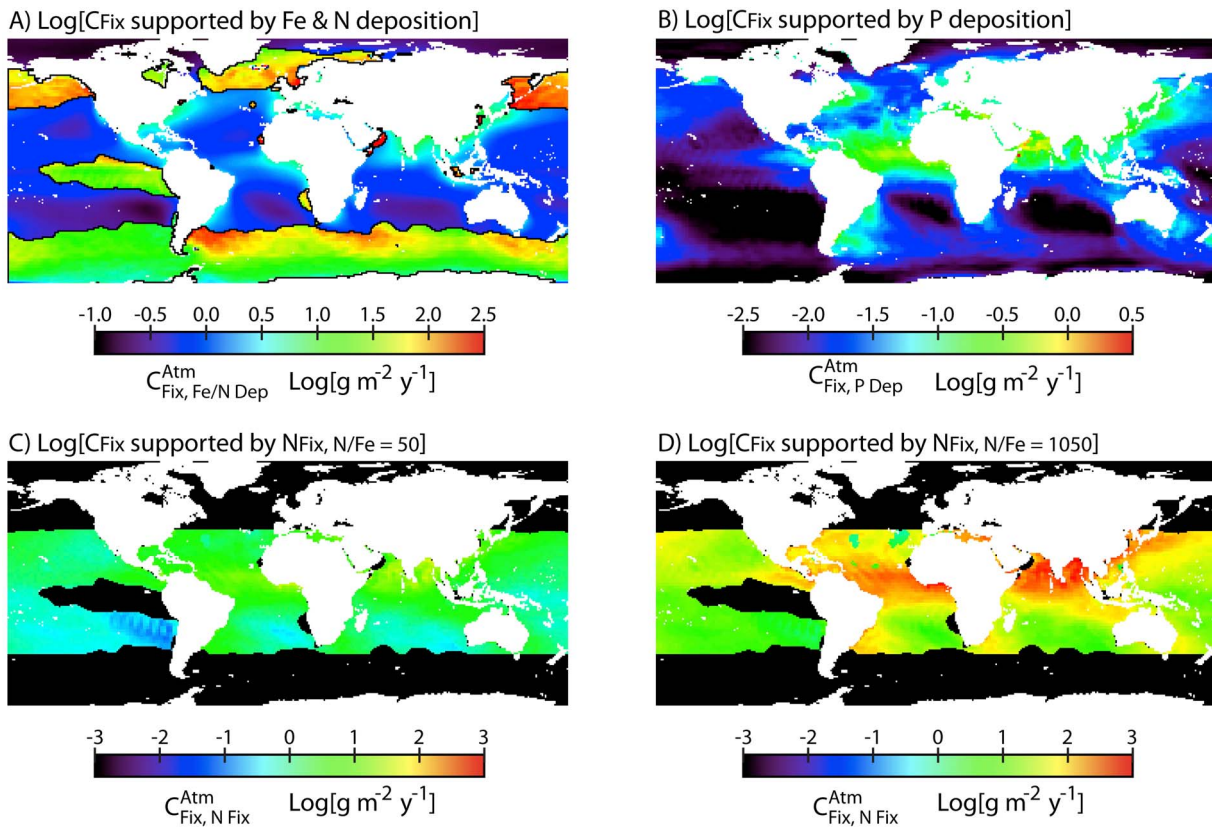
A) Log[CFix supported by Fe & N deposition]

B) Log[CFix supported by P deposition]

C) Log[CFix supported by NFix, N/Fe = 50]

D) Log[CFix supported by NFix, N/Fe = 1050]

Figure 4.   (a) Estimated rates of C fixation that can be supported by atmospheric deposition of Fe and N, $C_{Fix,Fe/N\ Dep}^{Atm}$. (b) Estimated rates of C fixation that can be supported by atmospheric deposition of P ignoring its effect on N fixation, $C_{Fix,P\ Dep}^{Atm}$. The black line shows boundaries of areas where productivity is assumed limited by availability of Fe in surface waters (see gray area in Figure 3). (c) Estimated rates of C fixation that can be supported by atmospherically supported rates of N fixation alone, $C_{Fix,N\ Fix}^{Atm}$, assuming the N:Fe of diazotrophs is 50 g/g. (d) Estimated rates of C fixation that can be supported by atmospherically supported rates of N fixation alone, $C_{Fix,N\ Fix}^{Atm}$, assuming the N:Fe of diazotrophs is 1050 g/g.

Table 3. Estimated Rates of C Fixation From Fe and N Deposition Only, Assuming Different Values for Nitrate Below Which the Ocean is N Limited[a]

	$C_{Fix,Fe/N\ Dep}^{Atm}$, 2 μM NO$_3$	$C_{Fix,Fe/N\ Dep}^{Atm}$, 4 μM NO$_3$	$C_{Fix,Fe/N\ Dep}^{Atm}$, 8 μM NO$_3$
Global	13.7	7.5	4.6
Atlantic	2.4	0.82	0.35
North Atlantic	1.1	0.63	0.28
North Central Atlantic	0.90	0.083	0.023
Equatorial Atlantic	0.25	0.053	0.026
South Atlantic	0.17	0.062	0.022
Indian	1.8	0.55	0.14
North Indian	1.2	0.46	0.098
Equatorial Indian	0.51	0.073	0.024
South Indian	0.050	0.013	0.013
Pacific	5.3	2.9	1.6
North Pacific	3.0	2.1	1.4
North Central Pacific	0.81	0.090	0.043
Equatorial Pacific	1.4	0.58	0.14
South Pacific	0.13	0.072	0.021
Southern	4.0	3.2	2.5

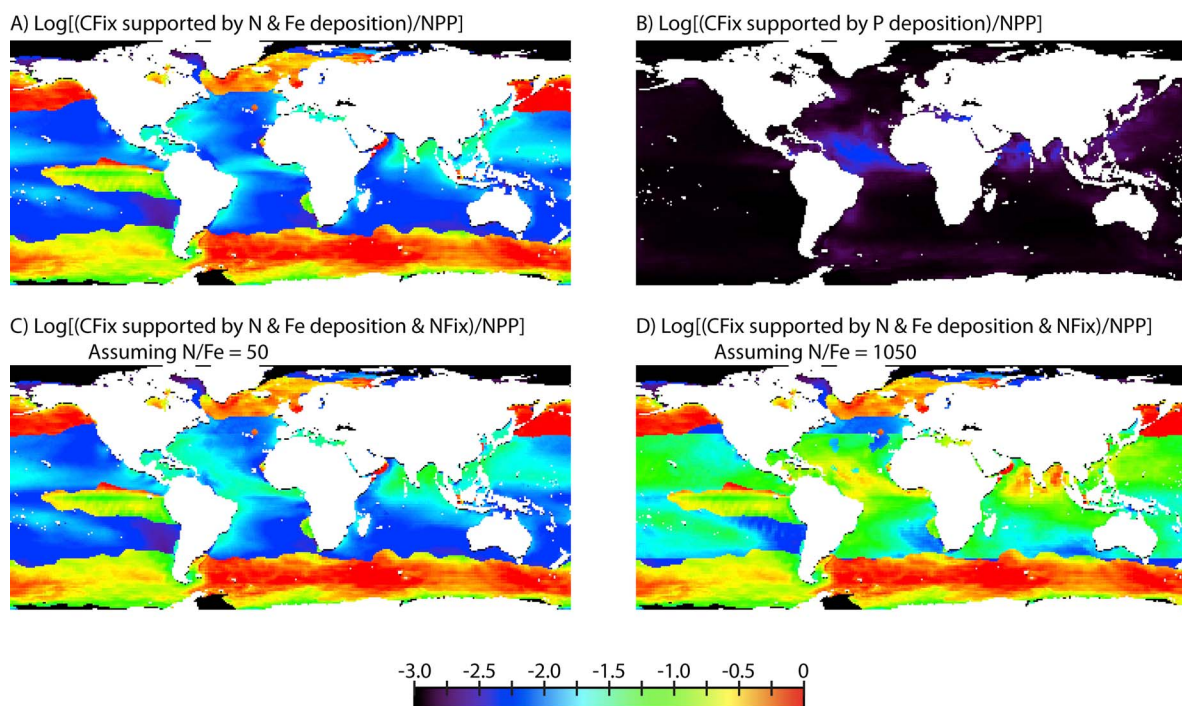[a]Basins as defined in Table 1; rates measured in petagrams of carbon per year.

Figure 5. (a) The proportion of satellite-derived NPP that can be accounted for by C fixation supported by atmospheric deposition of Fe and N, $C_{Fix,Fe/N\ Dep.}^{Atm}$. (b) The proportion of satellite-derived NPP that can be accounted for by C fixation supported by atmospheric deposition of P ignoring its effect on N fixation, $C_{Fix,P\ Dep.}^{Atm}$. (c) The proportion of satellite-derived NPP that can be accounted for by C fixation supported by atmospherically supported rates of N fixation alone, $C_{Fix,N\ Fix}^{Atm}$, assuming the N:Fe of diazotrophs is 50 g/g. (d) The proportion of satellite-derived NPP that can be accounted for by C fixation supported by atmospherically supported rates of N fixation alone, $C_{Fix,N\ Fix}^{Atm}$, assuming the N:Fe of diazotrophs is 1050 g/g.

sition may be as high as 51–52% of the observed NPP in the Southern Ocean and 43–45% in North Pacific Ocean (Table 2).

4. Discussion

[20] Deposition from the atmosphere to the ocean surface is an important but poorly understood process. Because of the limited observations of dry and wet deposition of N and dust species [e.g., *Dentener et al.*, 2006; *Mahowald et al.*, 2008, 2009; *Prospero et al.*, 2010], estimates must be based on model outputs. The long-range transport of aerosols from land sources to the ocean requires the correct simulation of source emissions and the many atmospheric processes occurring over great distances. Consequently there are often large differences among model simulations of N species [e.g., *Dentener et al.*, 2006] and dust [*Huneeus et al.*, 2010]. The dearth of observations makes it difficult to constrain the atmospheric concentrations far from the source region [e.g., *Mahowald et al.*, 2008]. Based on a comparison of mean deposition estimates from an ensemble of 23 global N models [*Dentener et al.*, 2006] with measurements made at 46 relatively remote (mostly coastal) stations, we tentatively estimate an uncertainty of +/− 25–50% in the annual N deposition on the world's oceans, the higher uncertainties being associated with the Pacific and the Southern Oceans in general. Variations between individual models and the

ensemble mean could be considerably greater. There can also be large regional differences in individual ocean basins. For example recent measurements of dry deposition over the Bay of Bengal during the dry season suggest that global models overestimate deposition by a factor of 4 to 5 [*Kumar et al.*, 2010]. In the Atlantic Ocean, *Baker et al.* [2010] recently made estimates of N deposition based on observations of aerosol and rainfall concentrations. There was good agreement between their results and recent modeling studies in the region [*Dentener et al.*, 2006], with inorganic N estimates agreeing to within <5% for the North Atlantic and to within <7% for the South Atlantic.

[21] Dust model estimates show relatively good agreement on North Atlantic deposition rates that are dominated by well-studied African dust sources. However, there are large differences in the modeled rates to other ocean basins: a factor of about three to four for the North Pacific, the South Pacific, and the Indian Ocean [*Mahowald et al.*, 2005]. Actual deposition measurements are rare. In a recent 3 year study of African dust deposition to a network nine stations in Florida [*Prospero et al.*, 2010] measurements were compared with estimates of nine dust models in the AEROCOM project [*Textor et al.*, 2007]. The mean model estimate of African dust deposition was about half of that measured; but individual model estimates ranged from 0.2 to 1.3 of measured. Model estimates to more remote ocean regions are expected to be poorer because dust sources are not as well character-

ized as those in North Africa. Overall, the uncertainty in the deposition of mineral aerosols is thought to be around an order of magnitude over ocean regions [e.g., *Mahowald et al.*, 2005, 2009]. We expect that the uncertainty in the deposition of iron and phosphorus would be at least as large as that of dust.

[22] Remarkably little is known about the nature, speciation and solubility of P aerosols that are deposited to marine ecosystems. The concentration of P in mineral dust from different natural and anthropogenically influenced sources varies from 700 to 1200 ppm(m) [*Mahowald et al.*, 2008]. P containing primary biogenic aerosol materials are poorly understood, but likely consist of small plant fragments, pollen, fern or fungal spores, or phospholipid bioaerosols. At any rate, relatively little information on organic P deposition to the oceans is available (M. Kanakidou et al., manuscript in preparation, 2010).

[23] With mineral aerosols, the lifetime of primary P bearing minerals in deposited dust in the surface layers is probably short compared to the time required for these minerals to dissolve. Soluble inorganic P, on the other hand, can probably be considered generally bioavailable to phytoplankton, whereas soluble organic forms may only be available to organisms with alkaline phosphatase enzyme activity [e.g., *Stihl et al.*, 2001; *Shaked et al.*, 2006], though this will cause biogenic aerosol P to be at least partially bioavailable [e.g., *Benitez-Nelson*, 2000; *Karl and Bjorkman*, 2002]. In the analysis here, all deposited P was assumed to be bioavailable. Thus our results represent the maximum possible contribution of atmospheric P deposition. However, we found little evidence for significant P limitation of C or N fixation, indicating that our results are not sensitive to the bioavailability of deposited P in the surface oceans.

[24] Complete understanding of the bioavailability of the atmospheric inputs of Fe is also lacking, but here there is greater information to guide us. It is apparent that only a small fraction of the total atmospheric input is soluble (e.g., of the order of magnitude of 1% in mineral dust) [*Jickells and Spokes*, 2001]. The chemistry of dissolved Fe in the oceans is extremely complex, with overall dissolution of atmospheric Fe inputs strongly modulated by the properties of seawater (e.g., the presence and nature of organic complexing ligands) [*Baker and Croot*, 2010]. There is strong evidence that the solubility of inorganic atmospheric Fe and P is dependent on the source, chemical and physical properties of the transported material [e.g., *Jickells et al.*, 2005; *Baker et al.*, 2006; *Mahowald et al.*, 2009] and that solubility appears to be altered by a variety of processes during transportation through the atmosphere [*Baker and Jickells*, 2006; *Baker and Croot*, 2010]. Also, certain organisms may be able to obtain nutrients direct from particle surfaces [*Sunda*, 2001]. Thus the approach of using the soluble Fe fraction as a proxy for bioavailable atmospheric inputs for Fe and total inputs of P should be considered a first order approximation, which will be subject to uncertainties in our understanding of the actual bioavailable fraction of these elements, of the processes that control their solubility, and of the organisms that use them.

[25] With regard to N fixation, if regions with sufficient $xsPO_4$ are omitted from consideration as possibly P limited, the bulk of the surface appears to be limited by Fe deposition. Thus, patterns of N fixation in our results are closely related to

deposition of Fe used to derive these results. This is consistent with measurements in the eastern tropical Atlantic which suggest that atmospheric P input is not sufficient to support observed N fixation, implying that upwelling P contributes significantly to N fixation [*Baker et al.*, 2007].

[26] Nonetheless, with the present uncertainty in C:Fe ratios that is apparent in the literature [see *Baker et al.*, 2007], it is difficult to derive a single estimate of atmospherically supported N and C fixation. The values derived using a N:Fe ratio of 1050 should be considered as the maximum possible rates of N and C fixation. These maximum rates of nitrogen fixation (using an N:Fe ratio of 1050) due to atmospheric Fe deposition in the tropical North Atlantic are ~4–6 g m^{-2} yr^{-1} (800–1200 μmol N m^{-2} d^{-1}). In contrast, using a N:Fe ratio of 50, the maximum rate of N fixation in this region is 0.2–0.3 g m^{-2} yr^{-1} (40–60 μmol N m^{-2} d^{-1}). In situ measurements in this region range from ~4–250 μmol N m^{-2} d^{-1}, with a median value of 25 μmol N m^{-2} d^{-1} [*Voss et al.*, 2004]. This comparison argues that the N:Fe ratio for N fixation in this region is closer to the lower value of 50 g/g used here. Nonetheless, measurements of N fixation in the Pacific of 93–137 μmol N m^{-2} d^{-1} [*Karl et al.*, 1997; *Deutsch et al.*, 2001] are inconsistent our the low inferred rates using an N:Fe ratio of 50 g/g. Instead, our estimated rates of N fixation in the Pacific are ~0.5–1 g m^{-2} yr^{-1} (100–200 μmol N m^{-2} d^{-1}) using an N:Fe ratio of 1050 g/g, suggesting that in this region the higher N:Fe value may be applicable. Alternatively, there may be other sources of Fe that may be supporting the higher rates of N fixation in this region. In particular, reducing conditions in anoxic denitrification zones could result in significant soluble Fe inputs that may subsequently be advected into regions where diazotrophy is possible.

[27] Despite the uncertainty of the stoichimetric role that Fe plays in N fixation in the surface ocean, it does seem clear that the main impact of atmospheric deposition on ocean C fixation is through the N cycle, i.e., by deposition of N or N fixation through Fe deposition, rather than through the deposition of P. The role of direct P deposition on C fixation and the role of P deposition on N fixation appear to be small. In the former case, this is due to the relatively low P concentration in dust (compared to Fe) in light of the low C:P ratio for ocean primary producers (maximum values of $C_{Fix,P\ Dep}^{Atm}$ of 1 g m^{-2} yr^{-1} are found in the equatorial tropical Atlantic and the Arabian Sea, corresponding to areas of maximum P deposition downwind of the Saharan and Arabian deserts and the biomass burning source in the Sahel). In the latter case, only small portions of the surface oceans can be P limited when the presence of $xsPO_4$ in the surface water is considered.

[28] The contribution of atmospheric deposition to productivity of the surface oceans has been a matter of considerable discussion. The results presented here support two very different regimes of atmospheric contribution to oceanic NPP. In non-HNLC regions, we predict that N deposition provides the greatest contribution to NPP and that the impact of Fe on productivity occurs through its influence on N fixation. In contrast, atmospheric deposition of Fe in HNLC regions has the potential to contribute significantly to C fixation. Rates of C fixation supported by atmospheric deposition in HNLC regions are several orders of magnitude higher than those in the non-HNLC regions. HNLC regions them-

selves contribute nearly one half of the total global NPP and about 50% of MODIS-observed NPP in the Southern Ocean, for instance, could be supported by atmospheric deposition of Fe.

5. Conclusions

[29] Using modeled fields of atmospheric deposition of N, P and Fe to the global ocean we have estimated rates of marine N and C fixation that can be supported by atmospheric supply of water column limiting nutrients. Our results are to some extent model dependent and subject to uncertainties associated with the assumptions we made about the bio-available fraction of each nutrient input. For example, very little observational data are available to help constrain our estimates of bioavailable Fe deposition to the HNLC regions of the ocean, iron solubility is strongly influenced by the properties of seawater [*Baker and Croot*, 2010] that we do not consider here, and very little is known about bioavailability of atmospherically deposited P. This is the first such global study to include the impact of organic N deposition and the deposition of nonmineral sources of P. We also address the uncertainty associated with P bioavailability by considering the potential impact of total P deposition, thus placing an upper limit on its likely influence on marine primary productivity. It is clear that there is significant variability in the contribution of the combined supply of atmospheric N, Fe and P to marine C fixation. In non-HNLC regions, atmospheric deposition of N and P can contribute just a tiny fraction of actual oceanic productivity. In HNLC regions, on the other hand, atmospheric deposition of Fe can potentially contribute considerably to rates of marine productivity. *Duce et al.* [2008] calculated that the global NPP supported by atmospheric deposition of N was ~0.38 Pg/yr. Here, we find that the global NPP potentially supported by atmospheric deposition of N and Fe is ~7.5–9.3 Pg/yr (Table 2, $C_{\text{Fix,Fe/N Dep}}^{\text{Atm}}$ and $C_{\text{Fix,N Fix}}^{\text{Atm}}$ columns), indicating clearly that Fe is likely much more important than N in supporting NPP globally, similar to the finding of *Krishnamurthy et al.* [2009]. We do not consider the impacts of aerosol deposition onto ocean acidification, as done by *Doney et al.* [2007].

[30] Globally, HNLC regions contribute significantly to the rate of carbon uptake by the surface oceans. Thus, the changes in atmospheric deposition to these regions could potentially have significant impacts on future oceanic carbon uptake. The Southern Ocean, in particular, is dominated by HNLC conditions. The major sources of Fe deposition to this basin are the arid regions of the South America, southern Africa, and Australia. Increased aeolian activity in these regions caused by increased temperatures (as predicted for southern Africa by *Thomas et al.* [2005]) and increasing atmospheric pollutants due to industrialization that may contribute to atmospheric processing of Fe bearing aerosols, thus have a significant potential to impact productivity in this area.

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