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Ocean biogeochemistry exhibits contrasting responses to a large scale reduction in dust deposition

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Abstract

Dust deposition of iron is thought to be an important control on ocean biogeochemistry and air-sea CO₂ exchange. In this study, we examine the impact of a large scale, yet climatically realistic, reduction in the aeolian Fe input during a 240 year transient simulation. In contrast to previous studies, we find that the ocean biogeochemical cycles of carbon and nitrogen are relatively insensitive to a 60% reduction in Fe input from dust. Net primary productivity (NPP) is reduced in the Fe limited regions, but the excess macronutrients that result are able to fuel additional NPP elsewhere. Overall, NPP and air-sea CO₂ exchange are only reduced by around 3% between 1860 and 2100. While the nitrogen cycle is perturbed more significantly (by ~15%), reduced N₂ fixation is balanced by a concomitant decline in denitrification. Feedbacks between N₂ fixation and denitrification are controlled by variability in surface utilization of inorganic nitrogen and subsurface oxygen consumption, as well as the direct influence of Fe on N₂ fixation. Overall, there is relatively little impact of reduced aeolian Fe input (<4%) on cumulative CO₂ fluxes over 240 years. The lower sensitivity of our model to changes in dust input is primarily due to the more detailed representation of the continental shelf Fe, which was absent in previous models.

1 Introduction

The necessary nutrients for phytoplankton growth, including nitrogen (N), phosphorous (P), silica (Si), and iron (Fe), can be supplied to ocean surface waters via the aeolian deposition of mineral aerosols (Duce et al., 1991). Of these, Fe has received particular interest, due to its potential to influence the cycle of carbon (C) in climatically significant regions of the worlds oceans (Jickells et al., 2005). Total dust inputs to the ocean are on the order of 450 Tg yr⁻¹ (Ginoux et al., 2001; Mahowald and Luo, 2003; Tegen et al., 2004) and usually contain around 3.5% Fe, with a solubility that can be highly variable (e.g., Jickells and Spokes, 2001). Phytoplankton are limited by Fe in the “high

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nutrient, low chlorophyll” (HNLC) ocean regions, such as the Southern Ocean (Boyd et al., 2000), Equatorial Pacific (Coale et al., 1996) and Sub Arctic Pacific (Boyd et al., 2004) and elevated oceanic dust deposition to the Southern Ocean might drive Glacial-Inter Glacial atmospheric CO₂ variability (Martin, 1990). Oceanic dust deposition has likely declined from the pre industrial (PI) to the present day (Mahowald et al., 2006). Prediction of future deposition are model dependent and will be controlled by land use changes, as well as climate effects (Tegen et al., 2004; Mahowald et al., 2006; Mahowald, 2007¹). Processes that impact sources areas and dust loading, such as CO₂ fertilization, have the potential to drive dramatic reductions in oceanic dust deposition over the coming century (Mahowald et al., 2006).

Variability in the atmospheric deposition of Fe can significantly impact C biogeochemistry. Phytoplankton require Fe for a variety of photosynthetic and respiratory enzymes (Falkowski et al., 1998; Raven, 1990) and changes in exogenous Fe supply will therefore directly impact net primary production (NPP) and the fixation of inorganic C. Moreover, the amount of Fe that is required to fix a given quantity of C is greater under low irradiance (Sunda and Huntsman, 1997; Raven, 1990) and nitrate (NO₃) based growth (Raven, 1990), as well as for large phytoplankton functional groups (Timmermans et al., 2004). Accordingly, all HNLC mesoscale Fe addition experiments have elicited increases in large diatoms, which can efficiently export fixed C to depth, and are more efficient (per unit Fe added) in shallow mixed layers (De Baar et al., 2005; Tagliabue and Arrigo, 2006). Atmospheric pCO₂ might increase dramatically (via a decline in both NPP and air to sea CO₂ transfer) if the aeolian input of Fe were to decrease significantly over the coming decades (Moore et al., 2006; Parekh et al., 2006), although other studies show a much lower sensitivity (Bopp et al., 2003).

Aside from its direct impact on C fixation, the aeolian input of Fe can also influence the biogeochemistry of N. Nitrogenase, the enzyme responsible for fixation of dinitrogen (N₂) by phytoplankton, has a large Fe requirement (Rueter et al., 1992) and this

¹Mahowald, N. M.: Anthropocene changes in desert area: Sensitivity to climate model predictions, *Geophys. Res. Lett.*, submitted, 2007.

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has led to speculation that N fixation might be regulated by Fe and sensitive to variability in aeolian inputs (Falkowski, 1997; Moore and Doney, 2007). As such, increased N_2 fixation can elevate surface dissolved inorganic N (DIN) concentrations, potentially increasing overall NPP and the uptake of CO_2 from the atmosphere (Falkowski et al., 1997; Michaels et al., 2001; Moore and Doney, 2007). That said, it is also important to consider how changes in Fe deposition might impact processes that govern the losses of DIN. During denitrification, bacteria utilize NO_3 as an alternative electron acceptor for remineralization under low oxygen conditions. N_2 fixation and denitrification are therefore coupled by their generation and consumption of DIN and analyses of both models and observations have suggested a balanced, or homeostatic, DIN inventory (Redfield, 1934; Gruber and Sarmiento, 1997; Tyrrell, 1999; Gruber, 2004; Deutsch et al., 2007). Nevertheless, this concept remains controversial (see: Codispoti, 2007) and, with particular reference to dust, Fe has been proposed to be an agent that can unbalance the N cycle (Moore and Doney, 2007).

In this study we use the Pelagic Integration Scheme for Carbon and Ecosystem studies (PISCES) global ocean-biogeochemistry model (Aumont and Bopp, 2006) to examine the response of ocean biogeochemistry to a reduction in aeolian Fe input during a transient simulation from 1860 to 2100. We use climatically realistic projections from Mahowald et al. (2006) (rather than arbitrary changes) to investigate the biogeochemical consequences of such an extreme change. Previously, models using either simple ecosystem dynamics or performing short term simulations have shown a high sensitivity to changes in dust deposition (Dutkiewicz et al., 2005; Parekh et al., 2006; Moore et al., 2006). In contrast, we find that the biogeochemical cycles of C and N are actually relatively insensitive (globally) to a massive reduction in dust input during a transient simulation from 1860 to 2100. While N_2 fixation is more significantly impacted by reduced dust inputs, it is also accompanied by a concomitant decline in denitrification, which stabilizes ocean DIN inventories. The disparity in model results is discussed in terms of inter-model differences in the ocean Fe cycle and the timescales over which the particular study is evaluated.

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2 Methods

2.1 The PISCES model

The PISCES ocean-biogeochemistry model is extensively described in Aumont and Bopp (2006) and includes two phytoplankton functional groups (nanophytoplankton and diatoms), meso- and micro-zooplankton, 2 detrital size classes for particulate organic carbon (POC), calcium carbonate, biogenic silica, DIC, biogenic Si, dissolved organic C, NO_3 , phosphate (PO_4), Silicic acid ($\text{Si}(\text{OH})_4$), ammonium (NH_4), and Fe, as well as carbon-13 for all carbon pools. Here we describe the parameterizations relevant for this study.

2.2 Net primary production, species composition and gas exchange

In order to calculate net primary productivity (NPP) we first calculate a maximum phytoplankton specific growth rate (μ_{max}) for each phytoplankton functional type (PFT) that is a function of temperature. μ_{max} is then scaled by the product of the most limiting nutrient (assuming Michaelis Menten kinetics) and the light (using the initial slope (α) of the P-E relationship, including photoadaptation) limitation terms to produce the realized growth rate (μ), and C fixation rate of each PFT. The half saturation constant for growth ($K\mu$) is fixed for all nutrients except Fe, where it varies as a function of biomass (taken as a proxy for cell size). The uptake of NO_3 and PO_4 that accompanies NPP is calculated assuming fixed Redfield ratios ($\text{C/N} = 122/16$ and $\text{C/P} = 122/1$). On the other hand, the Fe/C ratio increases as light declines and decreases with Fe stress, while Si/C decreases as light increases and also permits lighter silicification if $\text{Si}(\text{OH})_4$ concentrations decline.

Each PFT has certain specific physiological characteristics that will govern their response to our imposed changes in aeolian Fe deposition. Diatoms are assumed to have a requirement for $\text{Si}(\text{OH})_4$ (no such requirement for nanophytoplankton) and a higher $K\mu$ (relative to nanophytoplankton) for NH_4 , PO_4 , and Fe, but a lower $K\mu$ for NO_3

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Diatoms also have a greater maximum and minimum C-specific Fe demand and $K_{\mu}\text{Fe}$ than nanophytoplankton. Finally, nanophytoplankton support a more regenerative food web, producing small particles and being preferentially grazed by microzooplankton. In contrast, mesozooplankton have a preference for diatoms and microzooplankton grazing of diatoms is zero when their biomass is greater than $0.5 \mu\text{mol C l}^{-1}$. Diatoms also produce large, rapidly sinking particles.

The air-sea exchange of $\text{CO}_{2(\text{aq})}$ follows the OCMIP protocols and is a function of the air-sea pCO_2 gradient, temperature, ice-cover, and the piston velocity. We represent the wind speed dependence of the piston velocity via a quadratic function of the wind speed at 10 m (Wanninkhof, 1992).

The Fe cycle is represented in a relatively simple fashion and dissolved Fe is assumed to be made up of free inorganic and organically complexed Fe, both of which are bioavailable. The concentration of organically complexed Fe is computed using a chemical equilibrium constant and a fixed ligand concentration of 0.6 nM. The impact of abiotic Fe chemistry (e.g., Tagliabue and Arrigo, 2006) on Fe speciation is not included. Free inorganic Fe can be converted to particulate species by scavenging onto particles. Atmospheric deposition of Fe assumes that Fe is 3.5% of dust, with a solubility of 0.5% and includes subsurface dissolution of dust particles. Additional sources of Fe are sediment resuspension, which includes a representation of continental shelf supply that is usually ignored by global models (Aumont and Bopp, 2006), as well as riverine supply (hydrothermal sources are not represented in PISCES).

2.3 Nitrogen cycling in PISCES

The uptake of DIN by phytoplankton and subsequent remineralization of organic matter to DIN is described in Aumont and Bopp (2006), here we concentrate on describing N_2 fixation and denitrification.

Marine N_2 fixation is carried out by numerous planktonic organisms, including the oft mentioned *Trichodesmium* sp., as well as a variety of unicellular diazotrophes, crocospheara and diatom endosymbionts (see: Mahaffey et al., 2005 and references therein).

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Given this taxonomic diversity and the associated physiological variability, we decided not to prognostically simulate a N_2 fixing plankton functional type (PFT) within PISCES. Instead, we chose to include N_2 fixation as a process that produces NH_4 under certain conditions (Aumont and Bopp, 2006). Specifically, N_2 fixation is a function of the growth rate of the nanophytoplankton function group and only occurs in surface waters that have a temperature warmer than, or equal to, $20^\circ C$. This maximum potential rate of N_2 fixation is then scaled by the limitation term for Fe and one minus the limitation term for DIN (NO_3 and NH_4). Therefore, the realized rate of N_2 fixation increases with decreasing DIN and declines as Fe becomes more limiting.

Denitrification occurs in suboxic waters and results in NO_3 instead of O_2 being consumed during the remineralization of organic matter. In PISCES, we restrict denitrification to waters whose O_2 concentration is below $8 \mu M$ (Lipshultz et al. 2000) and set the N/C stoichiometric ratio for denitrification to be 0.8 mol : mol (Middelburg et al., 1996; Soetaert et al., 2000). Finally, we also include the bacterial conversion of NH_4 to NO_3 via nitrification, which occurs in oxic waters and is inhibited by light.

Despite our relatively simple parameterization scheme, denitrification and N_2 fixation are implicitly coupled in PISCES. Although we do not include a diazotrophic PFT in PISCES, we do include the main role of N_2 fixation in supplying NH_4 under certain conditions. The absence of an explicit PFT might be important if diazotrophes are significantly different in their sinking and remineralization characteristics, or food web structure; all of which are currently difficult to generalize for use in a global model. Variability in denitrification will alter DIN concentrations which will, in turn, impact the rate of N_2 fixation. While on the other hand, changes in export production (mediated by N_2 fixation and/or Fe and other nutrients) will modify subsurface O_2 concentrations and thus denitrification rates. This approach also allows the long spin up and simulation periods to be conducted with a relatively low computational cost.

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2.4 Physical model

The physical model coupled to PISCES is based on the ORCA2 global configuration of OPA version 8.2 (Madec et al., 1998), including a dynamic-thermodynamic sea ice model (Timmermann et al., 2003). The mean horizontal resolution is approximately 2° by 2° cos latitude and the meridional resolution is enhanced to 0.5° at the equator. The model has 30 vertical levels, with an increment that increases from 10 m at the surface to 500 m at depth (12 levels are located in the first 125 m). Our standard physical model employs climatological atmospheric forcing from various datasets. These include NCEP/NCAR 2 m atmospheric temperature (averaged between 1948 and 2003) and relative humidity, ISCCP total cloudiness (averaged between 1983 and 2001), CMAP precipitation (averaged between 1979 and 2001), weekly wind stress based on ERS and TAO observations and creates a representation of ocean circulation/mixing that is constrained by observations. Please see Aumont and Bopp (2006) for more details and the associated references.

2.5 Experimental design

After spinning up the model for over 3000 years under preindustrial conditions, we ran PISCES for a 240 year period from 1860 to 2100 with a specified atmospheric pCO₂ that is constrained by observations between 1860 and 2000 (Keeling et al., 2001) and the A2 scenario from 2000 to 2100 (Nakicenovic and Swart, 2000). A recent study by Mahowald et al. (2006) used the National Center for Atmospheric Research's Community Climate System Model (NCAR-CCSM) to investigate the change in mineral aerosols in response to climate and predict pre industrial, current, and future dust sources, as well as oceanic deposition. We linearly interpolated the aeolian Fe supply to the ocean between the pre industrial (1860), current (taken to be 2000) and doubled CO₂ (taken as 2100) fields of Mahowald et al. (2006), and assume Fe to be 3.5% of dust, with a solubility of 0.5%. We also ran a control experiment from 1860 to 2100 that was forced by the annually changing atmospheric pCO₂ concentrations, but constant

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PI dust deposition. Finally, we use the year 2000 as a reference, as it represents 'modern' dust deposition.

3 Results and discussion

3.1 Changes in aeolian Fe input in 2100

5 The predicted reduction in aeolian Fe input in 2100 (relative to 2000) is spatially heterogeneous and results from changes in sources area and dust loading (see: Mahowald et al., 2006). The largest percentage reductions in aeolian Fe input are found throughout the sub-Antarctic Southern Ocean, specifically to the south of Australia, Patagonia and, to a lesser degree, South Africa (Fig. 1). In the remaining HNLC regions (the sub-
10 Arctic and Equatorial Pacific) there are slightly lesser, but still significant, reductions in Fe deposition (Fig. 1). It is noteworthy that the locations where a proportionally high reduction in dust is predicted are all HNLC regions of relatively high NPP. Aside from that, there are also large absolute reductions in Fe deposition in the Northern Sub-tropical Atlantic Ocean (Fig. 1). The decline in dust deposition is due to the predicted
15 decline in future dust loading that results from climate induced changes in vegetation, as well as CO₂ fertilization (Mahowald et al., 2006; Mahowald, 2007¹) and can therefore be thought to be climatically realistic (rather than arbitrary changes in model forcings). Between the pre-industrial and modern periods; the decline in dust deposition is controlled by changes in source area, mostly at southerly latitudes (Mahowald et al., 2006). Quantitatively, this represents a reduction in total aeolian Fe deposition to ocean surface waters of 38% and 60% by 2000 and 2100, respectively (relative to
20 1860).

3.2 Impact on NPP

25 Despite an overall reduction of 60% in aeolian Fe input, we find little impact upon global NPP by 2100. At the beginning of our experiments (1860), annual NPP is

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35.8 Pg C yr⁻¹ and declines to 35.3 and 34.6 Pg C yr⁻¹ by 2000 and 2100, respectively (Table 1). This represents a decline of only 3.3% (1.2 Pg C) or 1.8% (0.7 Pg C) between 1860 and 2100 or 2000 and 2100, respectively (Table 1). As expected, NPP is reduced in the HNLC regions that are impacted by the reduced aeolian Fe input, such as the Atlantic sector of the Southern Ocean (ATL-SO, by 15 to 30 gC m⁻²), the Sub Arctic Pacific (SAPac, by 5 to 15 gC m⁻²) and the Equatorial Pacific (EqPac, by 5 to 30 gC m⁻²) (Fig. 1, Fig. 2a, Fig. 3a, between 2000 and 2100). Nevertheless, the overall reductions in NPP are only slight (especially in the Sub Arctic Pacific and the Antarctic sector of the Southern Ocean) and suggest that aeolian Fe input is only important in regions directly adjacent to dust sources. This is probably because the continental shelf is the principal source of Fe to the mixed layer in the large majority of HNLC waters (Tyrrell et al., 2005; Blain et al., 2007; Aumont et al., 2007²).

The spatial re-organization in NPP that follows the reduction in aeolian Fe input is responsible for the small change in global NPP that results. The decline in ATL-SO NPP (Fig. 3a) reduces the utilization of macronutrients (such as NO₃) and an excess therefore remains in surface waters (up to 2 μM between 2000 and 2100, Fig. 4a). These excess nutrients are now able to enter mode waters (especially in the ATL-SO, Fig. 4d) and fuel additional NPP in the macronutrient limited low latitudes (Fig. 3a) (Aumont et al., 2003; Sarmiento et al., 2004a; Aumont and Bopp, 2006). A similar mechanism also occurs in the EqPac, where the decline in eastern tropical Pacific (ETP) NPP and NO₃ utilization is balanced by elevated NPP in the western tropical Pacific (WTP) (Fig. 3a) as the unused upwelled nutrients flow westward (Dutkiewicz et al., 2005). This highlights the dynamic nature of the global ocean system, whereby changes in local nutrient utilization can have far field ramifications (Sarmiento et al., 2004a; Dutkiewicz et al., 2005). Overall, the net effect of this re-organization of NPP is a slight decline in globally integrated NPP (Table 1), despite the greatly reduced dust inputs.

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3.3 Species composition and nutrients

Despite a small effect on global NPP, reduced aeolian Fe input does cause a decline in diatom abundance in high latitude HNLC waters. In the Southern Ocean, for example, lower aeolian Fe inputs reduce the competitive advantage of diatoms and drive a floristic shift to the smaller nanophytoplankton functional group (by as much as 30%, Fig. 4d). This is due to the higher affinity for dissolved Fe that is associated with nanophytoplankton, relative to diatoms (Aumont and Bopp, 2006). Overall, diatom Fe limitation increases by between 10 and over 60% between 2000 and 2100, compared to around 5 to 25% for the nanophytoplankton PFT. As a consequence, the abundance of diatom grazing mesozooplankton also declines, resulting in a more regenerative marine food web. Overall, the Fe-mediated shift in food web structure results in a larger relative decline in C export, relative to NPP, by 2100 (−5.1 and −3.3% for carbon export and NPP, respectively, Table 1).

Only the concentrations of non-limiting nutrients will change greatly by 2100. As mentioned previously, NO_3 concentrations increase in the Fe-limited HNLC regions where aeolian Fe input declines (Fig. 1, Fig. 4a). While high residual NO_3 concentrations unsurprisingly persist in the Southern Ocean, EqPac, and SAPac, we also predict excess NO_3 in North Atlantic under low aeolian Fe deposition conditions (Fig. 4a). This would suggest a switch from a predominantly N limited system to one limited by Fe, thus highlighting the potential importance of atmospheric supply as a source of Fe to this particular region. In the N-STP and tropical Atlantic, N remains the limiting nutrient and prevents any accumulation of residual NO_3 (Fig. 4a) despite the increased transport of NO_3 from adjacent regions of reduced NPP (Fig. 3a). Similarly, it is only in regions that are not Fe limited that we see any change in Fe concentrations by 2100 (Fig. 4b). Although there is less Fe being delivered to the Southern Ocean and the EqPac, in particular, by 2100 (Fig. 1), phytoplankton utilize all available Fe and therefore do not change surface concentrations. That said, there are notable reductions in Fe concentrations in the northern subtropical Atlantic Ocean, as well as the Indian

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Ocean; regions of particularly high atmospheric deposition rates (Jickells et al., 2005; Mahowald et al., 2006).

The diagnostic tracer Si^* ($(\text{Si}(\text{OH})_4) - (\text{NO}_3)$) was originally proposed by Sarmiento et al. (2004a) and declines with increasing Fe stress, due to the elevated Si/N uptake ratios of Fe limited diatoms (e.g., Takeda, 1998). However, we find that although Fe stress does increase in 2100 (see above), the replacement of diatoms by nanophytoplankton (which have no $\text{Si}(\text{OH})_4$ requirement) actually increases Si^* (not shown). Therefore, Fe-mediated changes in phytoplankton species composition, with particular regard to any taxon-specific Si requirements, should be considered when evaluating spatio-temporal changes in Si^* . For example, the exhaustion of $\text{Si}(\text{OH})_4$ at more southerly latitudes than NO_3 in the Southern Ocean (causing negative Si^*) (Franck et al., 2000; Sarmiento et al., 2004a), might arise from local aeolian inputs of Fe, or the advection of high-Fe subsurface waters from the Patagonian shelf. Greater Fe supply would allow diatoms to make up a greater proportion of phytoplankton biomass near dust deposition sites, therefore increasing $\text{Si}(\text{OH})_4$ uptake and reducing Si^* (i.e. the inverse of our results).

3.4 Nitrogen biogeochemistry

3.4.1 Integrated rates and spatial distributions

Annual global N_2 fixation is 75 Tg N yr^{-1} (in 2000, Table 1), which is towards the lower end of the estimate of $120 \pm 50 \text{ Tg N yr}^{-1}$ proposed by Gruber (2004) in a recent synthesis. Regarding specific ocean basins, Mahaffey et al. (2005) compiled estimates of Atlantic N_2 fixation that are between 15 and 77 Tg N yr^{-1} , while Pacific N_2 fixation rates were 21 to 59 Tg N yr^{-1} (Mahaffey et al., 2005 and references therein). We find area-integrated rates are 24.2 and $31.3 \text{ Tg N yr}^{-1}$ for the Atlantic and Pacific Oceans, respectively during this study (in 2000, Table 2). More recently, a model-based study (Deutsch et al., 2007) concluded that N_2 fixation rates in the Pacific may be as much as twice as high as those in the Atlantic and proposed one of the highest global N_2 fix

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ation rates (137 Tg N yr^{-1}). In our model, N_2 fixation is of roughly the same order in the Atlantic and the Pacific and is distributed throughout the tropical oceans, apart from the high NO_3 EqPac (Fig. 2c). This is a region where Deutsch et al. (2007) predict very high rates of N_2 fixation (greater than $200 \text{ mmol N m}^{-2} \text{ yr}^{-1}$), despite high concentrations of DIN, and may go somewhat towards explaining the disparity between our estimates. In the Indian Ocean we obtain 20 Tg N yr^{-1} of N_2 fixation, but while both geochemical evidence (Gruber and Sarmiento, 1997) and the few measurements available (Capone et al., 1997) would support high rates of N_2 fixation, no basin scale estimate exists.

Global denitrification is $36.1 \text{ Tg of N yr}^{-1}$ (in 2000, Table 1) and is dominated by the ETP region (Table 2, Fig. 2d). This is somewhat below the $65 \pm 20 \text{ Tg N yr}^{-1}$ proposed by Gruber (2004) for pelagic denitrification and well beneath estimates of up to 150 Tg N yr^{-1} (Codispoti et al., 2001). Unfortunately, while we are able to capture the major features of ETP denitrification (Fig. 2d, Table 2), we evidently underestimate Indian Ocean denitrification severely (Table 2). As a result of the high rates of Antarctic bottom water formation near the Antarctic shelf, as well as the relatively high resolution simulations that are needed to accurately capture the complex physics, Indian Ocean deep waters contain too much oxygen in PISCES. Therefore, denitrification rates are retarded in a region that has been estimated to consume over 20 Tg N per year (Howell et al., 1997), a factor of 5 greater than we find (Table 2). In addition, our model also does not include any sedimentary denitrification, which can be between 180 and 300 Tg N yr^{-1} (Codispoti et al., 2001; Gruber, 2004).

Overall, our value for global annual pelagic N_2 fixation of 75.7 Tg N (for 2000, Table 1) is very similar (also in terms of spatial distributions, Fig. 2a) to estimates of 58 to 75 Tg N yr^{-1} that are produced by models that contain a prognostic N_2 fixing PFT (Moore et al., 2006; Moore and Doney, 2007) and well within the geochemistry-based range of $120 \pm 50 \text{ Tg N yr}^{-1}$ (Gruber, 2004). As regards denitrification, our model results are low relative to those produced by Moore and Doney (2007) (65 Tg N yr^{-1}), but within the range of 45 to 85 Tg N yr^{-1} proposed by Gruber (2004) (especially considering our aforementioned underestimation in the Indian Ocean). Reconciling model

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results with the high end estimates of water column denitrification of up to 150 Tg N yr⁻¹ (Codispoti et al., 2001; Galloway et al., 2004) remains a challenge for global ocean-biogeochemistry models.

3.4.2 Changes in N₂ fixation with dust

5 N₂ fixation responds much more dramatically to changes in aeolian Fe deposition than NPP. Globally, N₂ fixation declines by 17% (or 14 Tg N) to approximately 67 Tg N yr⁻¹ by the end of our 240 year simulation (in 2100, Table 1). The majority of this reduction is due to changes in the Pacific Ocean basin, wherein N₂ fixation declines by almost 25% (Table 2). In contrast, N₂ fixation declined by slightly more than 10% in the Atlantic and Indian Oceans. Overall, changes in aeolian Fe input have a far greater impact on
10 N₂ fixation (by as much as 25%) than NPP or C export (3 to 5%, Table 1).

N₂ fixation reacts both directly and indirectly to the reduced aeolian Fe inputs. The largest absolute decline in N₂ fixation occurs in the southern subtropical Atlantic (Fig. 3c) and is a result of both reduced aeolian input of Fe (Fig. 1), as well as additional NO₃ that results from the Fe-mediated decline in ATL-SO NPP (Figs. 3a, 4a).
15 Moreover, the large area of reduced absolute N₂ fixation in the N-STP gyre (Fig. 3c) is also a consequence of both the additional NO₃ that is transported from the region of reduced NPP in the EqPac (Figs. 3a, 4a), as well as reduced Fe input (Fig. 1). Indeed, N₂ fixation in the N-STP gyre will be highly sensitive to any Fe-mediated changes in
20 DIN utilization in the directly adjacent HNLC regions such as the EqPac and SAPac (Figs. 3a and 4a), as well as local changes in Fe inputs.

Moore et al. (2006) suggest that annual N₂ fixation would decline by almost 40 Tg N under an identical reduction in dust, far greater than our reduction of <10 Tg N (Table 1). This is because Moore et al. (2006) conducted 40 year simulations using the future
25 dust deposition field, wherein the impact of the reduced dust input will be exaggerated, relative to our incremental decline in dust deposition from 1860 to 2100. Indeed, the reduction in annual N₂ fixation of 10 to 20 Tg N from the first 2 years of Moore et

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al. (2006) is more in line with our results.

3.4.3 Changes in denitrification with dust

Denitrification also exhibits a strong response to the reduced atmospheric Fe inputs simulated in this study. This is due to the Fe mediated changes in C export, which modify the amount of O₂ consumed during remineralization and consequentially, the amount of denitrification that occurs. In the Pacific Ocean, the reduction in ETP NPP (Fig. 3a), results in greater subsurface O₂ concentrations and denitrification declines accordingly (by 24%, Table 2). On the other hand, the increase in low latitude NPP is responsible for the very slight absolute increase in Atlantic Ocean denitrification (Figs. 3a and d, Table 2). The decline in ETP denitrification results in additional subsurface NO₃ (Fig. 4d), which will eventually enter surface waters and will reduce the area over which N₂ fixation is selected for. While N₂ fixation can be directly controlled by the degree of Fe limitation, denitrification responds to the change in export production (and thus O₂ consumption) that results from the impact of Fe (or any other resource) on NPP (processes “I”, “J”, “K”, and “F” in Fig. 5).

3.4.4 The interplay between N₂ fixation and denitrification

At the beginning of our experiments the imbalance between pelagic N₂ fixation and denitrification amounts to excess of 42.3 Tg N. As mentioned previously, this most likely results from our underestimation of Indian Ocean denitrification and the absence of any benthic DIN consumption. For comparison, the recent synthesis of Gruber (2004) includes an imbalance of 65 Tg N between N₂ fixation and denitrification. Over the course of our experiment, the degree of imbalance declines by around 9 Tg N (to 33.4 Tg N, Table 1) and results from the rapid response of N₂ fixation to the surface water consequences of the reduction in aeolian Fe input (on both Fe and DIN concentrations).

Denitrification remains a relatively constant fraction of N₂ fixation by the end of our simulations (Table 2), in accord with hypotheses of a homeostatic DIN inventory. This

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is especially so in the Pacific Ocean, where N_2 fixation and denitrification are tightly coupled over the duration of our experiment (240 years, Table 2). This is probably due to the geographic proximity of regions of N_2 fixation and denitrification in the Pacific, as well as the increased deep-water ventilation over ETP denitrification sites (Deutsch et al., 2007). In contrast, Moore and Doney (2007) used an alternative experimental design (varying nitrogen cycle parameters) to suggest N_2 fixation and denitrification feedbacks were ‘muted’ in the Pacific Ocean. For example, simulated changes in N_2 fixation yielded a lesser change in denitrification (and vice-versa) than was seen in other ocean basins (Moore and Doney, 2007). In our study, the high rates of denitrification in the ETP are mostly supported by the high rates of NPP (e.g., Fig. 2a), and thus C export, that arise from the local upwelling of nutrients. We find that denitrification reacts to the impact of Fe on ETP NPP, rather than N_2 fixation, and that N_2 fixation responds to changes in DIN utilization in the ETP, as well as to Fe.

The impact of N_2 fixation and denitrification on DIN concentrations are segregated with respect to depth (Fig. 5). Denitrification responds to the subsurface consequences of the reduction in aeolian Fe input on C export and O_2 consumption (processes “K” and “F” in Fig. 5), by either stimulating N_2 fixation or NPP (processes ‘B’ and ‘I’ in Fig. 5). Therefore, the degree of deep-water ventilation will control the efficiency with which denitrification can negatively feedback onto N_2 fixation (via DIN concentrations, processes “G”, “L”, and “C” in Fig. 5). Reduced ventilation will also reduce vertical nutrient supply (process “L” in Fig. 5), as well as reducing subsurface O_2 concentrations, which may further reduce vertical DIN supply (via greater consumption during denitrification (processes “F” and “G” in Fig. 5). Nevertheless, any decline in vertical nutrient supply would reduce surface NPP, C export and subsurface O_2 consumption, thereby resulting in reduced denitrification (Fig. 5). Overall, the inter play between Fe fuelled NPP and N_2 fixation and the subsequent impact on DIN, C export, as well as remineralization and denitrification in subsurface waters becomes much more complex when changes in exogenous nutrient inputs are accompanied by changes in physical mixing (Fig. 5). Additional field measurements of the temporal changes in the processes and

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pools represented in Fig. 5, as well the degree of ocean ventilation, would greatly assist future investigations.

Overall, our results suggest that it is unlikely that Fe can significantly unbalance the oceanic N cycle (Moore and Doney, 2007), especially over long timescales. While the decline in aeolian Fe deposition does reduce N₂ fixation, it is accompanied by an oxygen mediated decline in the amount of denitrification (Tables 1 and 2). Moreover, besides the direct requirement of N₂ fixation for Fe, it is also important to consider that Fe will influence the amount of NPP, and thus DIN utilization, that occurs in the HNLC regions (Figs. 4a and 5). In this sense, anything that changes the degree of EqPac DIN utilization (not only Fe) has the potential to feedback onto both surface N₂ fixation and subsurface denitrification (processes “D” and “H”, as well as “D”, “J”, “K”, “F” and “G” in Fig. 5). This suggests that the ocean DIN inventory would be relatively insensitive to any increases in aeolian Fe input that may have occurred during the geologic past (e.g., during the last glaciation, LGM). That said, if increased dust deposition was also accompanied by a decline in ETP ocean ventilation (i.e. altering the strength of processes ‘L’ in Fig. 5), then the efficiency of the denitrification-N₂ fixation feedback can potentially become more complex (see above).

3.5 Air-sea CO₂ exchange

Ocean uptake of atmospheric CO₂ (FCO₂) is 1.9 Pg C yr⁻¹ in 2000 (Table 1, Fig. 2b) and compares well with current estimates of approximately 2 Pg C yr⁻¹ (Le Quéré et al., 2003). The 38% decline in dust deposition between the PI period and present day reduces FCO₂ by 5.5% (or 0.1 Pg C), relative to the impact of the 80 μatm rise in atmospheric pCO₂ (Table 1). However, by 2100 the 470 μatm increase in atmospheric pCO₂ is predominantly responsible for the change in the global ocean C sink (to 6.2 Pg C yr⁻¹) and dust decreases ocean uptake by only 3.4% (or 0.22 Pg C, Table 1). Cumulatively, the ocean takes up 22.8 Pg less C between 1860 and 2100, despite an 60% reduction in aeolian Fe input (or 4% of the 529.8 Pg of C taken up by the ocean assuming PI dust for 240 years).

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As seen for NPP, the change in dust deposition results in a spatial reorganization in FCO_2 (Fig. 3b). The degree of uptake of atmospheric CO_2 in the ATL-SO (Fig. 2b) is retarded and shifted to lower latitudes (Fig. 3b). Since higher temperatures and lower wind speeds characterize the tropical south Atlantic, gas exchange is reduced and FCO_2 declines more than NPP (-5.5% and -1.5% for FCO_2 and NPP, respectively, between the PI and present, Table 1). This disparity is all but eliminated by 2100, by which time the rise in atmospheric pCO_2 is the principal control on FCO_2 and the reduction in FCO_2 is only 0.1% greater than for NPP (Table 1). The decrease in EqPac FCO_2 by 2100 is due to an increase in the degree of outgassing (as NPP declines, Fig. 3a) and is partly balanced by increased ocean uptake in the WTP (Figs. 2b and 3b).

Recent studies have suggested a large reduction in the air-sea CO_2 flux (Moore et al., 2006) and a dramatic increase in atmospheric pCO_2 concentrations (Parekh et al., 2006) might result from a marked decline in oceanic dust deposition. In contrast, our results indicate that the impact of a massive decline in dust on air-sea CO_2 fluxes is much lower and is, at most, on the order of 3 to 5% (Table 1). Moore et al. (2006) found that the ocean uptake of CO_2 declined by 0.5 Pg C yr^{-1} under a future dust scenario, but kept atmospheric pCO_2 at PI levels ($\sim 280 \mu\text{atm}$). Given that atmospheric pCO_2 is projected to increase by as much as $470 \mu\text{atm}$ by 2100, it is difficult to compare to the results of our study, which includes the observed and predicted change in atmospheric pCO_2 between 1860 and 2100 (see Methods). Although we did not compute the impact of ocean CO_2 fluxes on atmospheric pCO_2 (instead this was used as a model forcing), it is clear that the impact of the reduced dust deposition will be far lower than the increase of up to $180 \mu\text{atm}$ from Parekh et al. (2006) and would be closer to that of $3 \mu\text{atm}$ from Bopp et al. (2003). In a more recent study, phytoplankton biomass in PISCES was found to be highly insensitive to variability in atmospheric deposition of Fe (Aumont et al., 2007²), mostly because of the explicit representation (Aumont and

²Aumont, O., Bopp, L., and Schulz, M.: What does temporal variability in aeolian dust deposition contribute to sea-surface iron and chlorophyll distributions?, *Geophys. Res. Lett.*,

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Bopp, 2006) of the high Fe flux that arises from continental shelves (Elrod et al., 2004; Tyrrell et al., 2005; Lam et al., 2006; Blain et al., 2007). Coarse resolution ocean-biogeochemistry models are often unable to represent the continental shelves and the absence of any sediment Fe source (Dutkiewicz et al., 2005; Parekh et al., 2006) will obviously increase the sensitivity of such models to any changes in aeolian input.

4 Perspectives

Our study only addresses the impacts of a change in dust deposition and atmospheric pCO₂ on ocean biogeochemistry. In reality, it is highly likely that changes in ocean circulation, such as increased surface stratification (e.g., Sarmiento et al., 2004b), will accompany any possible change in dust deposition. Greater stratification will impact the vertical supply of nutrients to surface waters, which might hamper the growth of large PFTs, such as diatoms (Bopp et al., 2005). Changing the degree of ocean ventilation also has the potential to change subsurface O₂ concentrations, which would impact denitrification rates. Moreover, reduced ventilation might also reduce the efficiency of the denitrification – N₂ fixation negative feedback (process “L” in Fig. 5), especially in the ETP. To that end, longer simulations would be required to adequately address the impact of changes in deep nutrient inventories (driven by export production and denitrification) on surface biogeochemistry.

Recent research has demonstrated an enhancement of N₂ fixation rates under higher CO₂ concentrations (Hutchins et al., 2007; Barcelos e Ramos et al., 2007), which might offset any dust-mediated decline in N₂ fixation. As an initial calculation, we use our modeled spatial increase in CO₂(aq) between 2000 and 2100, alongside a Michaelis-Menten fit to the proportional increase in N₂ fixation per unit change in pCO₂ from Hutchins et al. (2007), to suggest that global N₂ fixation could be enhanced by 22 Tg N (or 30%) by 2100 (relative to 2000); more than compensating for the dust-mediated decline of 8 Tg N (Table 1). However, this upper bound estimate assumes

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that N₂ fixation will simply increase with CO₂(aq), while in reality, concentrations of DIN, Fe, and PO₄ also exert a strong control. Therefore, any CO₂ enhancement would be unlikely to realize the complete 22 Tg N (or 30%) by 2100. Nevertheless, the CO₂ effect on N₂ fixation merits further investigation, especially as anthropogenic CO₂ is accumulating in the poorly ventilated subtropical gyres (Sabine et al., 2004) that are frequented by N₂ fixing phytoplankton (e.g., Mahaffey et al., 2005).

The sensitivity of individual global ocean biogeochemistry models to changes in aeolian iron input will be dependent on the model specific importance of atmospheric deposition, relative to other sources of Fe. The significance of aeolian Fe inputs to global ocean biogeochemistry depends on whether a given model also represents continental shelf inputs of Fe (low sensitivity to dust), or relies solely on dust as an external source (high sensitivity to dust). Although current global models do not address the observed spatial variability in the solubility of dust-bound Fe, including (or increasing the importance of) the continental shelf Fe inputs does permit a more accurate representation of the elevated phytoplankton biomass and Fe concentrations typical of these regions (Aumont and Bopp, 2006). Notwithstanding, observational estimates of sedimentary Fe fluxes, as well as their geographic variability and the bioavailability of the Fe supplied, remain sparse (but see: Elrod et al., 2004; Lam et al., 2006, Blain et al., 2007) and require further investigation.

5 Conclusions

Despite enforcing a climatically realistic dramatic reduction in the ocean deposition of aerosol Fe, we find that global NPP and FCO₂ only decline by 3–5%. This is due to the spatial redistribution of NPP that accompanies a decline in aeolian Fe input, as well as the lesser importance of aeolian Fe sources relative to continental shelf supply in PISCES. Locally, dust deposition does have a role in sustaining diatom biomass in the ATL-SO and could therefore be important in controlling the relative utilization of NO₃ and Si(OH)₄ and the associated marine food web. N₂ fixation declines by 17%

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in response to both the reduction in Fe supply, as well as the increased DIN concentrations that result from reduced nutrient utilization in dust impacted HNLC regions. Nevertheless, concomitant declines in denitrification stabilize the ocean DIN inventory (especially in the Pacific Ocean) and the excess NO_3 that arises from reduced bacterial consumption can act as a negative feedback on N_2 fixation. The denitrification – N_2 fixation feedback is controlled by changes in nutrients and NPP, and its efficiency depends on the degree and timescales of deep-water ventilation above denitrification sites (summarized in Fig. 5). The change in air-sea CO_2 exchange between 1860 and 2100 is predominantly controlled by the change in atmospheric pCO_2 and the decline in dust deposition reduces cumulative uptake (over the 240 year study period) by only 22.8 Pg C, or 4%. The impact of future changes in dust deposition, as well as other changes in climate, on ocean biogeochemical cycles should utilize a variety of global ocean biogeochemical models in order to minimize model-specific conclusions.

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Table 1. The impact of the simulated change in aeolian Fe deposition (Fe Dep, Tg Fe yr⁻¹) on globally integrated NPP (NPP, Pg C yr⁻¹), N₂ fixation (N₂Fix, Tg N yr⁻¹), denitrification (Denitr, Tg N yr⁻¹), the air-sea CO₂ exchange (FCO₂, Pg C yr⁻¹), and export production (Ex P, Pg C yr⁻¹) at three time points. The number in parenthesis is the percentage change relative to a control run, at the same point in time, which includes the change in atmospheric pCO₂, but assumes pre industrial (i.e. 1860) dust deposition.

| Year | Fe Dep | NPP | N ₂ Fix | Denitr | FCO ₂ * | Ex P [^] |
|------|------------|-------------|--------------------|--------------|--------------------|-------------------|
| 1860 | 0.25 | 35.8 | 81.7 | 39.4 | -0.1 | 8.3 |
| 2000 | 0.16 (-38) | 35.3 (-1.5) | 75.7 (-7.4) | 36.1 (-8.5) | 1.9 (-5.5) | 8.1 (-2.1) |
| 2100 | 0.06 (-60) | 34.6 (-3.3) | 67.8 (-17.0) | 34.4 (-12.7) | 6.2 (-3.4) | 7.8 (-5.1) |

*For FCO₂, a positive value denotes ocean uptake of atmospheric CO₂ and a negative percentage change therefore represents a reduction in ocean uptake.

[^]Export production is defined as the amount of POC passing the 100 m depth horizon.

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Table 2. Integrated annual rates (Tg N yr^{-1}) of N_2 fixation and denitrification, as a function of year and ocean basin. The number in parenthesis is the percentage change relative to a control run, at the same point in time, which includes the change in atmospheric pCO_2 , but assumes pre industrial (i.e. 1860) dust deposition. See Table 1 for the globally integrated rates.

| | N_2 Fixation (Tg N yr^{-1}) | | | Denitrification (Tg N yr^{-1}) | | |
|------|---|---------|---------|---|---------|---------|
| | Atlantic | Pacific | Indian | Atlantic | Pacific | Indian |
| 1860 | 25.2 | 35.2 | 21.0 | 2.0 | 33.4 | 4.2 |
| 2000 | 24.2 | 31.3 | 20.0 | 2.1 | 29.9 | 4.1 |
| | (-4.1) | (-11.2) | (-5.1) | (+7.9) | (-10.5) | (-2.3) |
| 2100 | 22.2 | 26.8 | 18.6 | 2.2 | 28.5 | 3.8 |
| | (-12.1) | (-23.9) | (-11.6) | (+13.1) | (-14.7) | (-10.7) |

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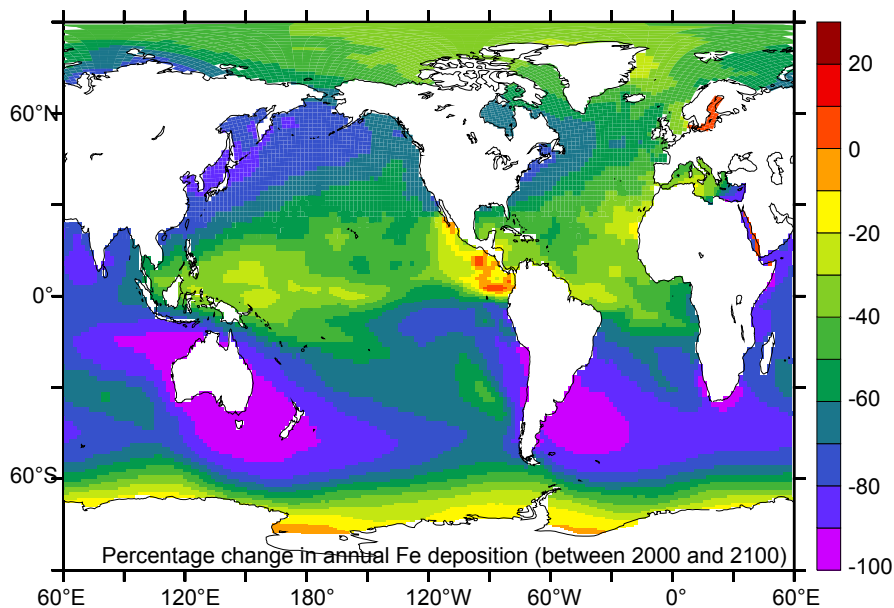


Fig. 1. The percentage change in the oceanic Fe deposition from the atmosphere between 2000 and 2100.

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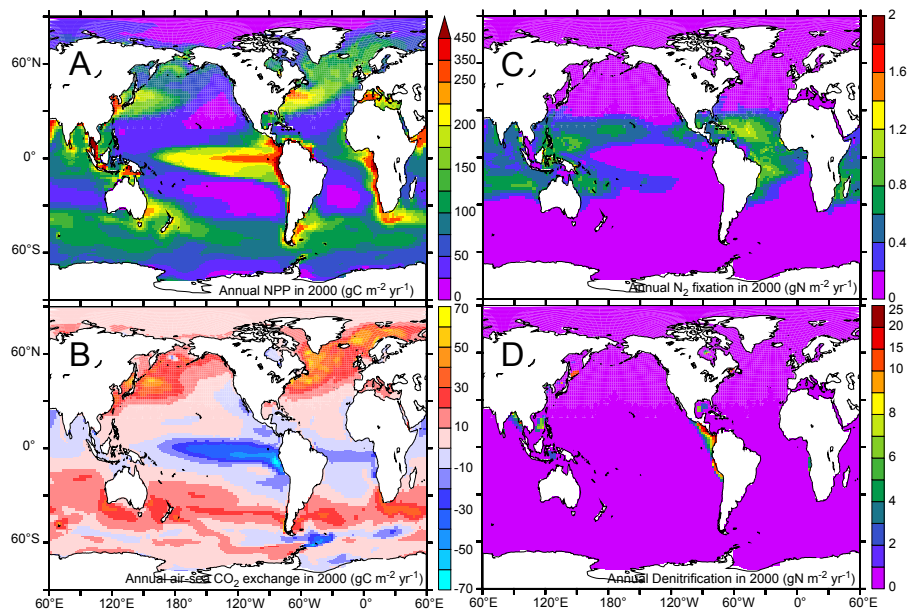


Fig. 2. The annual rate of **(a)** NPP ($\text{gC m}^{-2} \text{yr}^{-1}$), **(b)** air-sea CO_2 exchange ($\text{gC m}^{-2} \text{yr}^{-1}$), **(c)** N_2 fixation ($\text{gN m}^{-2} \text{yr}^{-1}$), and **(d)** denitrification ($\text{gN m}^{-2} \text{yr}^{-1}$) in 2000.

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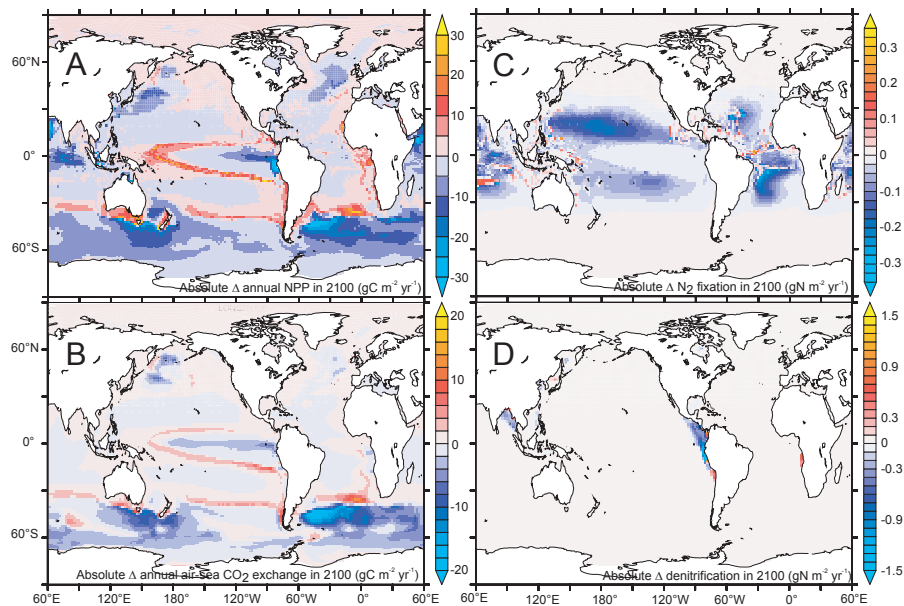


Fig. 3. The absolute change in (a) NPP ($\text{gC m}^{-2} \text{yr}^{-1}$), (b) air-sea CO_2 exchange ($\text{gC m}^{-2} \text{yr}^{-1}$), (c) N_2 fixation ($\text{gN m}^{-2} \text{yr}^{-1}$), and (d) denitrification ($\text{gN m}^{-2} \text{yr}^{-1}$), between 2000 and 2100.

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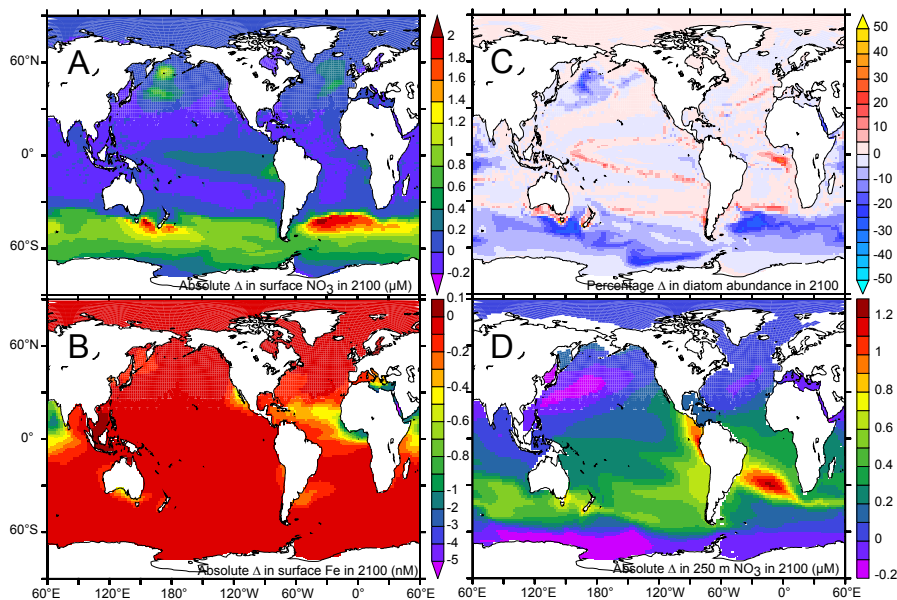


Fig. 4. The change in the annually averaged (a) NO_3 at the surface (absolute, μM), (b) Fe at the surface (absolute, nM), (c) the abundance of diatoms (percentage change in the proportion of total phytoplankton C associated with diatoms), and (d) NO_3 at 250 m (absolute, μM), between 2000 and 2100.

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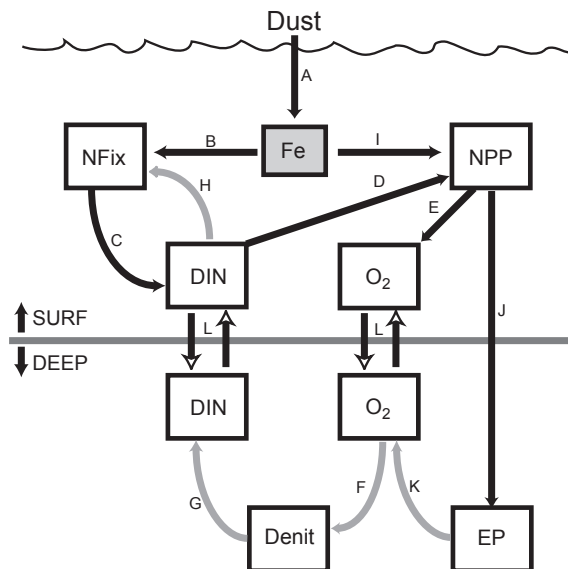


Fig. 5. A schematic to illustrate the impact of a change in Fe deposition on, and the feedbacks between, NPP, export production (EP), N_2 fixation (Nfix), and denitrification (Denit). Positive and negative feedbacks are represented by black and gray arrows, respectively. The deposition of dust alters the dissolved Fe concentration (process A), which then increases N_2 fixation (B) and, by consequence, surface DIN concentrations (assumed here to be $NO_3 + NH_4$) (C). Elevated DIN concentrations can increase NPP (D) and oxygen concentrations (E). Once transported to the subsurface (L), greater oxygen concentrations will retard denitrification (F) and therefore increase subsurface DIN (G). When this DIN is mixed into surface waters (L), it will reduce N_2 fixation (H). Alternatively, additional Fe can also stimulate NPP (I), increasing export production (J), which will also reduce subsurface oxygen via remineralization (K), which will stimulate denitrification (F).

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