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Modeling dust and soluble iron deposition to the South Atlantic Ocean

Matthew S. Johnson,¹ Nicholas Meskhidze,¹ Fabien Solmon,² Santiago Gassó,³ Patrick Y. Chuang,⁴ Diego M. Gaiero,⁵ Robert M. Yantosca,⁶ Shiliang Wu,⁷ Yuxuan Wang,⁸ and Claire Carouge⁶

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[1] The global chemical transport model GEOS-Chem, implemented with a dust-iron dissolution scheme, was used to analyze the magnitude and spatial distribution of mineral dust and soluble-iron (sol-Fe) deposition to the South Atlantic Ocean (SAO). The comparison of model results with remotely sensed data shows that GEOS-Chem can capture dust source regions in Patagonia and characterize the temporal variability of dust outflow. For a year-long model simulation, 22 Tg of mineral dust and 4 Gg of sol-Fe were deposited to the surface waters of the entire SAO region, with roughly 30% of this dust and sol-Fe predicted to be deposited to possible high nitrate low chlorophyll oceanic regions. Model-predicted dissolved iron fraction of mineral dust over the SAO was small, on average only accounting for 0.57% of total iron. Simulations suggest that the primary reason for such a small fraction of sol-Fe is the low ambient concentrations of acidic trace gases available for mixing with dust plumes. Overall, the amount of acid added to the deliquesced aerosol solution was not enough to overcome the alkalinity buffer of Patagonian dust and initiate considerable acid dissolution of mineral-iron. Sensitivity studies show that the amount of sol-Fe deposited to the SAO was largely controlled by the initial amount of sol-Fe at the source region, with limited contribution from the spatial variability of Patagonian-desert topsoil mineralogy and natural sources of acidic trace gases. Simulations suggest that Patagonian dust should have a minor effect on biological productivity in the SAO.

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1. Introduction

[2] Aeolian dust deposition has proven to be a critical source of iron (Fe) to high nitrate low chlorophyll (HNLC) oceanic regions [Martin and Fitzwater, 1988; Coale *et al.*, 1996; Boyd *et al.*, 2000; Fung *et al.*, 2000]. HNLC waters

comprise ~30% of the world oceans; thus the supply of Fe to the surface waters of these regions may play a key role in regulating biological productivity, atmospheric CO₂ concentration, and climate [Martin, 1990; Zhuang *et al.*, 1992a; Jickells *et al.*, 2005]. Out of the three main HNLC oceanic regions (subarctic north Pacific, equatorial east Pacific, and the Southern Ocean) the Southern Ocean (SO) is suggested to be the largest region where marine productivity is limited by the micronutrient Fe [Martin, 1990; Watson *et al.*, 2000; Boyd *et al.*, 2000]. The importance of this region is reflected in a hypothesis that the glacial-interglacial changes in atmospheric CO₂ can be attributed to fluctuations in the atmospheric dust-Fe supply to the SO [Martin and Fitzwater, 1988]. However, recent studies have also pointed out the importance of marine processes for the biogeochemical cycling of Fe in the surface waters of the SO [Meskhidze *et al.*, 2007; Blain *et al.*, 2007; Wagoner *et al.*, 2008]. Despite the potentially important role of Fe-laden dust for primary productivity in the polar and sub-polar waters of the SO, few studies exist that can help constrain Fe-laden dust fluxes to the surface waters of the South Atlantic Ocean (SAO) and the possible HNLC regions of the SAO. Throughout this study we define the SAO as the part of the Atlantic Ocean

¹Department of Marine, Earth, and Atmospheric Sciences, North Carolina State University, Raleigh, North Carolina, USA.

²Laboratoire d'Aérodynamique, UPS, CNRS, Toulouse, France.

³Goddard Earth Sciences and Technology Center, University of Maryland Baltimore County, Baltimore, Maryland, USA.

⁴Earth and Planetary Sciences Department, University of California, Santa Cruz, California, USA.

⁵CIGeS, FCFEYN, Universidad Nacional de Córdoba, Córdoba, Argentina.

⁶School of Engineering and Applied Sciences, Harvard University, Cambridge, Massachusetts, USA.

⁷Atmospheric Science Program, Department of Geological and Mining Engineering and Sciences and Department of Civil and Environmental Engineering, Michigan Technological University, Houghton, Michigan, USA.

⁸Department of Environmental Science and Engineering, Tsinghua University, Beijing, China.

between the equator and the Antarctic coastline (from north to south) and from the 70°W to 20°E, and the possible HNLC region as the portion of SAO south of the Antarctic Circumpolar Current (ACC) (~42°S) [Boyd *et al.*, 2007]. Patagonia has been estimated to supply the majority (up to 90%) of aeolian-Fe deposited to the SAO [Gabric *et al.*, 2002; Gaiero *et al.*, 2003; Erickson *et al.*, 2003]. Therefore, there is a great need for improved quantification of Fe-laden dust emissions from Patagonia, transport pathways, and deposition to the surface waters of the SAO.

[3] The role of aeolian Fe deposition in marine ecosystems is further complicated by the fact that oceanic primary productivity is influenced not by the total amount of mineral-Fe, but the portion of Fe that is water soluble (or bioavailable) [e.g., Jickells *et al.*, 2005]. Despite Fe being the fourth most abundant element in the earth's crust (~3.5% of mineral dust weight [Taylor and McLennan, 1985; Duce and Tindale, 1991]) and can be delivered in large quantities (~0.2–1.0 Tg yr⁻¹) to the surface waters of the SAO through atmospheric pathways [Li *et al.*, 2008; Gaiero *et al.*, 2003], this oceanic environment contains regions that are, biologically speaking, in short supply of Fe. Sampling studies of Patagonian-desert topsoils are limited, and currently no data exists for the Fe speciation in Patagonian soils. From a global perspective, Fe is primarily found in a form of (oxyhydr)oxides such as hematite (α -Fe₂O₃) and goethite (α -FeO(OH)), and to a lesser extent as ferric Fe (Fe(III)) that is substituted into aluminosilicate minerals [Dedik and Hoffmann, 1992; Hoffmann *et al.*, 1996; Arimoto *et al.*, 2002]. These forms of Fe are highly insoluble in oceanic waters [Stumm and Morgan, 1996; Fung *et al.*, 2000; Bonnet and Guieu, 2004; Jickells *et al.*, 2005; Journet *et al.*, 2008].

[4] The acidification of mineral dust aerosols during atmospheric transport has been proposed as the primary mechanism for the production of water-soluble forms of iron (sol-Fe) in mineral dust [Duce and Tindale, 1991; Zhu *et al.*, 1992; Zhuang *et al.*, 1992a, 1992b; Zhu *et al.*, 1993, 1997; Meskhidze *et al.*, 2003, 2005]. A number of natural (via biological production of dimethyl sulfide (DMS) and volcanic emission [Zhuang *et al.*, 1992b; Bay *et al.*, 2004]) and anthropogenic (via the mixing of dust plumes with acidic trace gases such as SO₂ and HNO₃ [Meskhidze *et al.*, 2003; Fan *et al.*, 2004; Solmon *et al.*, 2009]) pathways for the acid mobilization of mineral dust have been proposed. Modeling studies incorporating the acid mobilization of Fe, resulting from the mixing of mineral dust with anthropogenic acidic trace gases, have predicted considerable enhancements of sol-Fe as a result of this mixing process [Meskhidze *et al.*, 2005; Fan *et al.*, 2004; Yang and Gao, 2007; Solmon *et al.*, 2009]. However, Patagonia, the postulated source region of Fe-laden dust for the SAO, is sparsely populated and considered to be one of the most pristine areas of the globe. Therefore, the amount of anthropogenic acidic trace gases available for mixing with mineral dust plumes are unlikely to be high enough to sufficiently acidify dust and initiate large scale mineral-Fe dissolution. Under such conditions, natural sources of SO₂ are likely to play a key role for enhancing Fe dissolution in mineral aerosols and increasing bioavailable Fe fluxes to the surface ocean [Zhuang *et al.*, 1992b].

[5] In this work we use a global chemical transport model implemented with a prognostic dust-Fe dissolution scheme (GEOS-Chem/DFeS model) [Solmon *et al.*, 2009] to estimate dust aerosol mobilization in Patagonia, as well as transport, and deposition of dust and sol-Fe to the surface waters of the SAO. Our model simulations are focused on the SAO domain and consider three main issues: 1) Patagonian dust mobilization, transport, and deposition to the SAO; 2) mineral Fe dissolution within advecting Patagonian dust and subsequent deposition of sol-Fe to the surface waters of SAO; 3) sensitivity simulations to estimate the fluxes of sol-Fe associated with variations in natural sources of SO₂, different mineralogical compositions of Patagonian soil, and possible relative humidity (RH) dependent SO₂ uptake. Natural sources of SO₂ considered here include volcanic emissions and biological oceanic sources (i.e., DMS). The model results for mineral dust advection, transport, and deposition are compared to past research studies, local dust reports, and aerosol optical depth (AOD) values derived from the Aerosol Robotic Network (AERONET), the MultiAngle Imaging Spectroradiometer (MISR), and the Moderate Resolution Imaging Spectroradiometer (MODIS). A year-long model simulation was conducted to capture two contrasting dust advection seasons: high dust mobilization during the austral summer (DJF) and the low dust mobilization of winter. The time period between October 2006 and September 2007 was selected to take advantage of the most up-to-date meteorological fields available for the model and the limited dust/visibility reports available from Patagonia.

2. Methods

2.1. Model Description

[6] The global chemical transport model GEOS-Chem (v8-01-01) was applied to the Patagonia/SAO domain to quantify mineral dust mobilization at the source regions, and atmospheric transport and deposition to the SAO. GEOS-Chem is driven by assimilated meteorological fields from the Goddard Earth Observing System (GEOS) of the NASA Global Modeling Assimilation Office [Bey *et al.*, 2001; Park *et al.*, 2004]. The model uses GEOS-5 meteorological fields at a 2° × 2.5° (latitude-longitude) grid resolution and 47 vertical levels. In its full chemistry configuration, GEOS-Chem includes H₂SO₄-HNO₃-NH₃ aerosol thermodynamics coupled to an O₃-NO_x-hydrocarbon-aerosol chemical mechanism [Bey *et al.*, 2001; Park *et al.*, 2004]. Sulfur compounds, carbonaceous aerosols, and sea-salt emission and chemistry are accounted for and described by Park *et al.* [2004], Heald *et al.* [2004], and Alexander *et al.* [2005]. The mineral dust module in GEOS-Chem treats the mobilization of dust from the Earth's surface using the Dust Entrainment and Deposition (DEAD) scheme [Zender *et al.*, 2003], gravitational settling, and wet and dry deposition [Fairlie *et al.*, 2007]. The dust is distributed in four-size bins with diameters 0.2–2.0, 2.0–3.6, 3.6–6.0 and 6.0–12.0 μm [Fairlie *et al.*, 2007]. GEOS-Chem was previously shown to capture much of the amplitude and seasonal cycle in dust climatology at the surface sites of the North Pacific Ocean [Fairlie *et al.*, 2007], while this study is the first

Table 1. Mineralogical Composition of Patagonian Dust Prescribed to GEOS-Chem/DFeS

Mineral	Baseline (wt%) ^a	Clay Dissolution Sensitivity (wt%)	Reduced CaCO ₃ Sensitivity (wt%)
Hematite	5	2.5	5
Albite	20 (~7–54)	20	20
Microcline	7 (~0–45)	7	7
Illite	17 (~0–40)	17	17
Smectite	11 (~0–25)	11	11
Kaolinite	7 (~0–50)	7	7
CaCO ₃	9 (~3–25)	9	3
Gypsum	3 (~0–10)	3	3
Quartz ^b	21 (~11–75)	23.5	27

^aValues within the parentheses display the upper and lower limits of individual minerals (wt%) within Patagonian topsoils [Ramsperger et al., 1998; Claquin et al., 1999; Smith et al., 2003; Gaiero et al., 2004].

^bQuartz is assumed to play no role in Fe dissolution calculations and therefore was used to balance chemical composition of Patagonian soil.

application of GEOS-Chem with the focus on South American dust source regions.

2.2. Fe Dissolution Scheme

[7] A prognostic, physically based dust-Fe dissolution module (DFeS) of Meskhidze et al. [2005] has previously been implemented into GEOS-Chem (v7-03-06) [Solmon et al., 2009]. The DFeS module explicitly considers mineralogical composition of windblown dust and uses aqueous phase equilibrium and dissolution/precipitation reactions for the following minerals contained in dust: calcite (CaCO₃), albite, microcline, illite, smectite, gypsum, and hematite [Meskhidze et al., 2005; Solmon et al., 2009]. In agreement with most field measurement studies [e.g., Sullivan et al., 2007, and references therein], the atmospheric aging and uptake of acidic species will increase mineral dust hygroscopicity allowing for the aqueous reactions to proceed. The dissolution/precipitation of each mineral is then estimated based on solution pH, temperature, dust mineralogy, and the specific surface area of the individual minerals. With the prescribed mineralogical composition of dust and specified Fe content of minerals, the DFeS module can estimate the amount of sol-Fe produced during atmospheric transport and transformation of mineral aerosols. GEOS-Chem/DFeS-predicted fluxes of mineral dust and sol-Fe to the subarctic North Pacific Ocean have been shown to be in reasonable agreement with available observational data [Solmon et al., 2009].

2.3. Dust Mineral Composition

[8] The mineralogical composition of aerosols is one of the key factors influencing Fe solubility [e.g., Spokes et al., 1994; Meskhidze et al., 2005; Sedwick et al., 2007; Cwiertny et al., 2008; Journet et al., 2008]. The initial dust mineralogy was developed to represent the transportable fraction (i.e., the fraction of the particles within desert topsoils that can be uplifted and transported over large distances by wind) of South American/Patagonian-desert topsoils. Nine key minerals that can influence the concentration of sol-Fe formed during the atmospheric transport of dust particles were chosen for the model simulations. The limited number of Patagonian-desert topsoil samples have shown

that clay minerals, quartz, and plagioclase are the main components likely available for uplift [Ramsperger et al., 1998; Claquin et al., 1999; Smith et al., 2003; Gaiero et al., 2004]. In order to reduce the number of transported tracers, we define a “dust mode” in which all minerals (including Fe containing species) can be emitted, undergo chemical and physical transformation, and be removed by wet and dry processes [Solmon et al., 2009]. The mineralogical composition for this study (summarized in Table 1) was chosen to be representative of the transportable fraction of Patagonian-desert topsoil [Ramsperger et al., 1998; Claquin et al., 1999; Smith et al., 2003; Gaiero et al., 2004] and was developed following the methodology of Meskhidze et al. [2005].

[9] Past studies have commonly assumed that “free” Fe [Sumner, 1963; Anderson and Jenne, 1970] within Fe (oxyhydr)oxides such as goethite and hematite was the major form of Fe suitable for solubilization in mineral dust plumes [Claquin et al., 1999; Meskhidze et al., 2003; Luo et al., 2005; Fan et al., 2004; Solmon et al., 2009]. However, in their recent papers Journet et al. [2008] and Cwiertny et al. [2008] suggest that in addition to Fe oxides, sizable fractions of sol-Fe can be released from clay minerals. To account for the fraction of sol-Fe from clay minerals, the initial Fe fractions of smectite and illite were prescribed following Journet et al. [2008]. Mineral dissolution equations of Meskhidze et al. [2005] were modified to calculate the clay mineral contribution to the total amount of sol-Fe, formerly determined exclusively by the initial content of sol-Fe within hematite (see below) and sol-Fe produced during mineral dissolution [Solmon et al., 2009].

[10] Currently no data exists for the Fe content of aluminosilicates (clays) in Patagonian-desert soils. Therefore, all baseline simulations were carried out assuming clays contain no mineral Fe. To consider the effect of aluminosilicates on atmospheric fluxes of sol-Fe, model sensitivities were conducted (in section 4) where the average Fe content of smectite and illite was prescribed to be ~1.2% and ~0.6% w/w, respectively [Journet et al., 2008]. To keep the initial Fe content of dust at 3.5% [Taylor and McLennan, 1985; Duce and Tindale, 1991], we reduced the weight percent of hematite in mineral dust to 2.5% (see Table 1). No additional tracers were implemented in the standard GEOS-Chem/DFeS code and it was assumed that the fraction of sol-Fe from clay minerals does not influence the mineral aerosol pH.

[11] Previous modeling studies have shown that the production of sol-Fe varies temporally and spatially depending on mineral dust composition and the amount of acid added to the aerosol during its atmospheric transport [e.g., Meskhidze et al., 2005]. The limited number of studies on the mineralogy of Patagonian-desert topsoils show that CaCO₃ concentrations have large spatial variability, ranging (by weight) from 3% to 24%, with an average concentration of 9% [Claquin et al., 1999; Gaiero et al., 2004]. Since the prognostic dust-Fe dissolution scheme is sensitive to the alkalinity buffer produced by mineral dust (e.g., CaCO₃ concentration in dust particles), a sensitivity study was conducted in which CaCO₃ within Patagonian dust plumes was reduced to 3% by weight which is the lowest value reported in previous literature (see Table 1). The reduced initial CaCO₃ concentration sets a high-limit on the amount of sol-Fe that can be formed within Patagonian-dust through

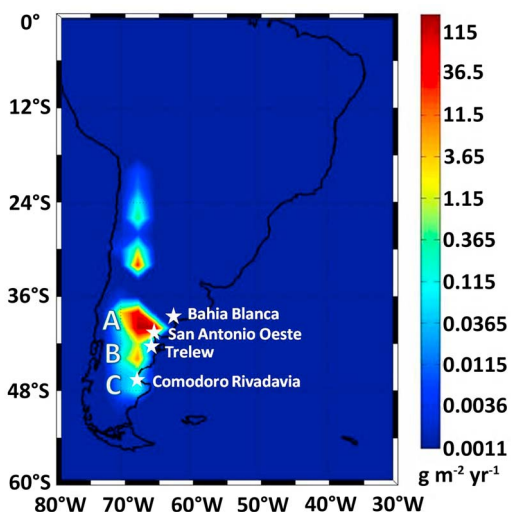


Figure 1. Annually averaged emission strength of dust ($\text{g m}^{-2} \text{yr}^{-1}$) between October 2006 and September 2007. Letters A, B, and C denote model-predicted locations of the three largest dust emission sources in Patagonia (36°S – 48°S). The stars indicate the surface stations at Bahia Blanca (38.7°S , 62.2°W), San Antonio Oeste (40.8°S , 65.1°W), Trelew (43.2°S , 65.3°W), and Comodoro Rivadavia (45.8°S , 67.5°W) with the local visibility reports.

acid mobilization. As the default concentration of CaCO_3 (9%) already yields minor dissolution of mineral Fe (see below), the simulations with increased CaCO_3 concentrations are not reported.

2.4. Initial Fe Solubility

[12] A major source of uncertainty in modeling mineral-Fe dissolution and deposition to the surface ocean is the initial fraction of sol-Fe, i.e., the fraction of Fe within mineral dust that is assumed to be water soluble at the source region. Due to the lack of information regarding the initial dissolved iron fraction (DIF) of sol-Fe in Patagonian-desert soils, an initial Fe solubility of 0.45% is prescribed in this study based on the synthesis of data on water soluble fraction or reactive Fe nano-particles in global desert soils [Guieu and Thomas, 1996; Spokes and Jickells, 1996; Zhuang et al., 1990; Bonnet and Guieu, 2004; Ooki et al., 2009; Solmon et al., 2009; Shi et al., 2009].

2.5. Satellite Data

[13] To evaluate the GEOS-Chem-predicted aerosol abundances in the vicinity of the dust source regions, model-predicted AOD values (550 nm) were compared to remotely sensed data obtained from Terra MISR (555 nm) [Martonchik et al., 1998, 2002] and Terra and Aqua MODIS retrievals (550 nm) [Kaufman et al., 1997; Tanré et al., 1997; Remer et al., 2005]. Previous studies show, that both MISR [Martonchik et al., 2004; Kahn et al., 2005] and MODIS [Remer et al., 2005; Hsu et al., 2006; Levy et al., 2007a, 2007b] satellites can accurately retrieve AOD over oceanic and land regions. In this study we use the MISR Collection 4 Level-3 global daily gridded AOD data sets

averaged at $0.5^{\circ} \times 0.5^{\circ}$ resolution and MODIS Collection 5 Level-3 global daily gridded AOD values averaged at $1^{\circ} \times 1^{\circ}$ resolution.

3. Results

3.1. Dust Mobilization From Patagonia

3.1.1. Dust Source Regions

[14] Figure 1 shows GEOS-Chem-predicted dust source locations and emission rates in South America. According to Figure 1, between October 2006 and September 2007 GEOS-Chem produced three distinct sources in South America. With an estimated annual emission flux of $\sim 30 \text{ Tg yr}^{-1}$, Patagonia is expected to contribute over 95% of South American mineral dust, followed by 1.0 Tg yr^{-1} from two separate source regions in Western Argentina. Model-predicted South American dust source locations and annual emission rates are in agreement with the analysis of dust sources conducted using Total Ozone Mapping Spectrometer (TOMS) satellite retrievals [Prospero et al., 2002] and previous model simulations [Li et al., 2008; Wagener et al., 2008]. The only dust source region in South America that was not distinctly captured by the model is a relatively weak source located in the salt flats (salars) of the Bolivian Altiplano (centered at 20°S and 67°W) [Prospero et al., 2002]. Comparison of GEOS-Chem-predicted dust source regions with remotely sensed data from Patagonia is difficult, as the dust plumes are traveling at low altitudes and are generally accompanied by large amounts of cloud cover [Gassó and Stein, 2007]. To assess the model's ability to characterize dust source regions in Patagonia, GEOS-Chem simulated dust emission plots were overlaid on Google Earth topography maps. Visual analysis of the three major dust source regions predicted by the model in Patagonia (indicated by the letters A, B, and C in Figure 1) show that the model-predicted maximum mineral dust emission areas correlate well with dry lake/river beds and low lying regions with little vegetative cover (see auxiliary material Figure S1) that have been suggested to be predominant sources of windblown dust from Patagonia [e.g., Prospero et al., 2002; Gassó and Stein, 2007].¹

[15] Since Patagonia is the major source of model-predicted mineral dust advected from South America and deposited to the SAO domain, for the model calculation of mineral dust and sol-Fe fluxes to the ocean we will only consider Patagonian dust sources located between 36°S and 48°S (see Figure 1).

3.1.2. Dust Events

[16] Large dust outbreaks generally occur only during a few days throughout the year, yet they may contribute the majority of dust deposited annually to the world's oceans [Lojé-Pilot et al., 1986]. To evaluate the model's ability to capture episodic dust events, GEOS-Chem-predicted daily dust burden in Patagonia are compared to airport visibility reports (obtained from www.weatherunderground.com) and remotely sensed data. Although visibility reports give only a qualitative assessment of the days in which visibility in the area was restricted due to large amounts of suspended particles, with a virtual absence of surface flux measurements

¹Auxiliary materials are available in the HTML. doi:10.1029/2009JD013311.

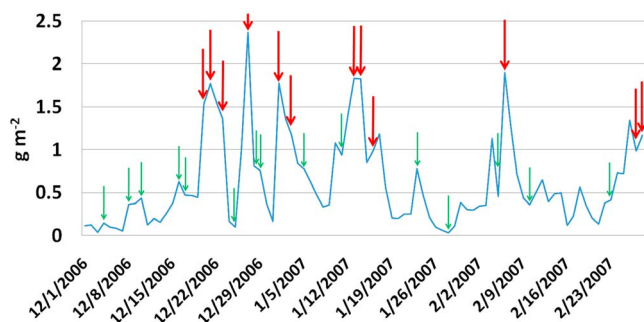


Figure 2. Model-predicted daily total dust burden (blue line) and Patagonian visibility reports during the austral summer of 2006/2007. Bold red arrows denote the days in which two or more stations reported low visibility and the green arrows denote days in which only one station reported the presence of dust.

and low anthropogenic influence, visibility reports near active dust regions are one of the best indicators of dust activity in the region [Mahowald *et al.*, 2007]. To estimate the extent of individual dust outbreaks, we further separate days in which only one station reports increased concentrations of suspended particles from days with two or more stations giving such reports. Figure 2 shows the comparison of GEOS-Chem-predicted daily dust burden in Patagonia with the local visibility reports from the four stations shown on Figure 1. The stations were selected based on their proximity to the major dust source regions simulated in Patagonia. Daily column dust burdens were calculated between 39°–47°S and 62°–68°W and the austral summer season was chosen due to the largest dust advection simulated from Patagonia during this season. According to Figure 2, simulated days with total dust burden significantly higher than the background level of dust ($\sim 0.5 \text{ g/m}^2$), correlate well with the days in which multiple stations reported elevated dust levels (red arrows). Analysis of the simulated dust event on Feb. 4, 2007, when no stations reported substantial dust, revealed that the model-predicted dust plume originated near Trelew (43.5°S, 68°W), but was quickly elevated above the surface boundary layer and transported over the stations at high altitude ($\sim 2 \text{ km}$) (see auxiliary material Figure S2). Little interaction with the surface downwind from the source region could be the reason for the discrepancy between model simulated dust outbreaks and surface visibility reports. Although the bulk of the model-predicted dust over the SAO domain during the high dust season (austral summer) is concentrated below 1 km (auxiliary material Figure S3), which is in agreement with Gassó and Stein [2007] and Li *et al.* [2008], a detailed analysis of the model-predicted vertical structure of Patagonian dust outflow suggests intermittent high-altitude (free tropospheric) pathways. Figure S2 illustrates satellite retrievals for the elevated aerosol plume on Feb. 5, 2007 off the coast of Patagonia the day after emission of the dust shown in Figure S2. Overall, Figure 2 suggests that the model can capture episodic dust outbreaks from Patagonia and is suited for the analysis of mineral dust and sol-Fe deposition to the SAO domain.

[17] Since model-predicted daily aerosol column burdens will be biased toward the outbreaks from the largest dust source region (i.e., source A on Figure 1), for further model comparison, GEOS-Chem-predicted AOD values were compared to the AERONET (500 nm) measured column-integrated daily aerosol optical properties near Trelew, Argentina (43.2°S, 65.3°W) [Holben *et al.*, 1998]. Analysis of satellite and model results showed, that before leaving the continent, dust plumes typically move in an easterly/northeasterly direction, therefore the Trelew station can be used to test the model's ability in capturing dust outbreaks from relatively smaller dust regions. Good correlation ($R = 0.58$) between GEOS-Chem-predicted AOD values and the ground-based measurements at Trelew (see Figure 3) suggests that the model is capable of capturing episodic events of dust outbreaks from smaller dust source regions in Patagonia. However, Figure 3 also shows that GEOS-Chem-predicted AOD values are somewhat higher compared to AERONET measurements, indicating potential overestimation of model-predicted dust concentration over the monitoring site.

[18] GEOS-Chem-predicted seasonal variability in AOD is further compared against surface (AERONET) and remotely sensed (MODIS and MISR) data. Figure 4 indicates that during the high dust activity season of the austral summer, the major fraction of total AOD is associated with mineral dust. The difference between MODIS and MISR retrievals could be attributed to the overestimation of AOD by MODIS over terrestrial regions with low aerosol loadings [Chu *et al.*, 2002; Martonchik *et al.*, 2004; Remer *et al.*, 2005]. Overall, Figures 1 to 4 indicate that GEOS-Chem is able to reproduce the locations and timing of Patagonian dust outbreaks and can be used for the assessment of dust and sol-Fe deposition to the SAO.

3.1.3. Dust Transport and Deposition to the SAO

[19] Between October 2006 and September 2007 GEOS-Chem predicted an annual mineral dust deposition of $\sim 22 \text{ Tg y}^{-1}$ to the surface waters of the entire SAO. About 7 Tg y^{-1} of this dust was to be deposited to the possible HNLC regions of the SAO. Figure 5 shows that the model-predicted yearly dust deposition to the SAO is generally consistent with previous estimates. Compared to dust deposition fluxes to the oceanic basins in the Northern Hemisphere, the SAO receives very little aeolian dust, accounting for less than 5%

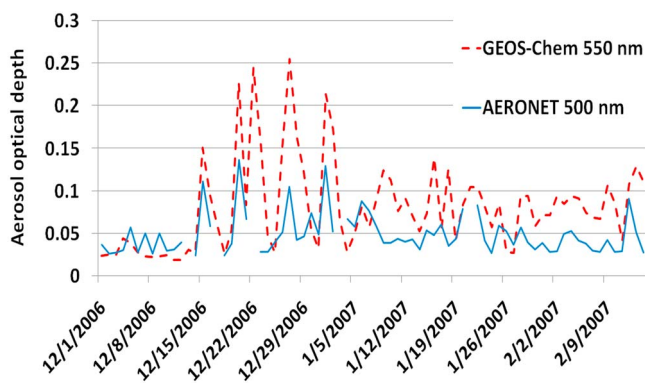


Figure 3. Daily averaged GEOS-Chem-predicted (550 nm) and AERONET-measured AOD (500 nm) values over Trelew, Argentina (43.2°S 65.3°W).

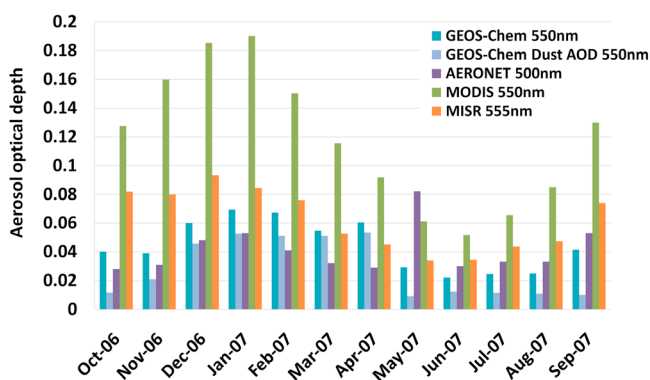


Figure 4. Comparison of model-predicted and satellite-derived monthly mean values of daily AOD averaged between 39°–47°S and 62°–68°W. AERONET ground-based measurements are shown for the Trelew station.

of global mineral dust inputs to the oceans [e.g., *Gao et al.*, 2001]. In addition to annual total dust deposition, dust seasonality and spatial distribution also play a significant role for ocean biogeochemical processes. Figure 6 shows that the largest amount of mineral dust is predicted to be deposited to the SAO during the austral summer season. Figure 6 shows that the summer fluxes can account for about one half of the total annual dust deposition to the SAO. Such strong seasonal variability of dust deposition is consistent with available meteorological data. Strong and persistent westerly winds dominate the Patagonian region throughout the year [*Paruelo et al.*, 1998], and with precipitation in the austral summer accounting for only about 10% of total annual rainfall [*Jobbagy et al.*, 1995], a maximum in dust advection from Patagonia during the DJF season is expected.

[20] Figure 7 shows the spatial distribution of total mineral dust fluxes (wet and dry deposition) to the surface waters of the SAO and possible HNLC regions below 42°S. According to Figure 7 the removal of Patagonian dust primarily occurs between 30°S and 60°S, while ~30% of the dust deposited to the SAO during this time period occurred in potential HNLC regions. Analyses of model results indicate that wet deposition is the primary removal mechanism accounting for 40% (near the coast) to 80% (open ocean) of total fluxes. Although these results are in good agreement with recent modeling studies of *Li et al.* [2008] and *Wagener et al.* [2008], due to the absence of in situ aerosol measurements in the SAO between October 2006 and September 2007, it is not possible to estimate the reliability of the GEOS-Chem-predicted dust fluxes over the SAO.

[21] The only available in situ measurements of surface level mineral dust concentrations are from the stations operated by the University of Miami [*Savoie et al.*, 1993] in Antarctica between 1990 and 1991. The two stations (King George Island (62.18°S, 58.3°W) and Palmer Station (64.77°S, 64.05°W)) recorded daily averaged dust concentrations from May 1990 to April 1991. When the annual-means of the daily averaged dust values were compared to model-predicted dust concentrations (not shown) GEOS-Chem was a factor of 5 and 8 lower at King George Island and Palmer Station, respectively. A possible source of

error in the GEOS-Chem prediction of dust deposition to Antarctica may be associated with the difficulty in simulating wet deposition in areas such as the higher latitudes on the coast of Antarctica [*Rasch et al.*, 2000; *Ginoux et al.*, 2001; *Mahowald et al.*, 2002].

3.2. Sol-Fe Deposition to the SAO

[22] The fluxes of sol-Fe advected from the continent and deposited to the surface waters of the open ocean depend on the amount of dust, which determines the total Fe concentration, and the DIF within the dust. As discussed above, part of the DIF (0.45%) is fixed in the model as the initial condition and the other part results from atmospheric transport and chemical aging of mineral aerosols. Comparison of Figures 6 to 8 shows that in the SAO domain the spatial distribution of sol-Fe fluxes closely relate with dust. The reason for this result lies in the amount of acidifying pollutants (e.g., SO_2) relative to the amount of mineral dust. Sulfuric acid produced from the oxidation of natural sources of SO_2 , such as DMS and volcanoes, is insufficient to overcome the buffering capacity of the CaCO_3 contained in the dust. Model predicted surface concentrations of nss-SO_4^{2-} and methanesulfonate (MSA), major atmospheric photo-oxidation products of gaseous DMS, at the two Antarctic sites compared well with the University of Miami data (auxiliary material Figure S4). Overall, the rate of Fe mobilization is small and the DIF of dust aerosols is primarily controlled by the initial value prescribed at the source region. Despite minor amounts of total sol-Fe resulting from the chemical processing of dust, Figure 9 shows that away from the source region, particularly near the Antarctic coastline, sulfuric acid produced from the oxidation of SO_2 is large enough to acidify deliquesced aerosols and initiate Fe dissolution. The model results clearly indicate elevated DIF (up to 1.0%) for the dust plumes over the Weddell Gyre, south of the southern ACC boundary [e.g., *Orsi et al.*, 1995]. However, it is important to notice that despite increased DIF values associated with mineral dust plumes

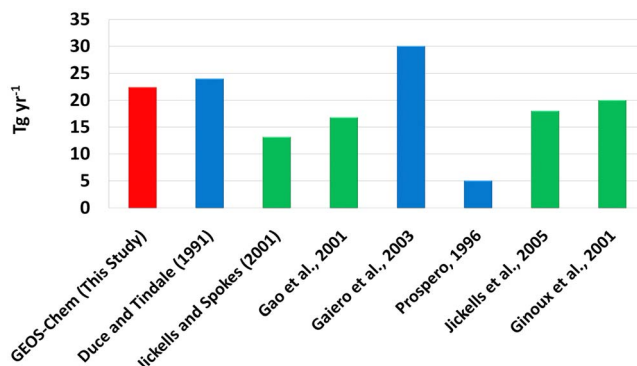


Figure 5. GEOS-Chem-predicted (red bar) annual dust deposition (Tg yr^{-1}) to the SAO compared to past estimates made from modeling (green) and measurement studies (blue). Estimates for *Duce and Tindale* [1991] and *Jickells and Spokes* [2001] are adopted from *Gao et al.* [2001]. Scavenging ratio = 200 was used by *Prospero* [1996], and 1000 by others.

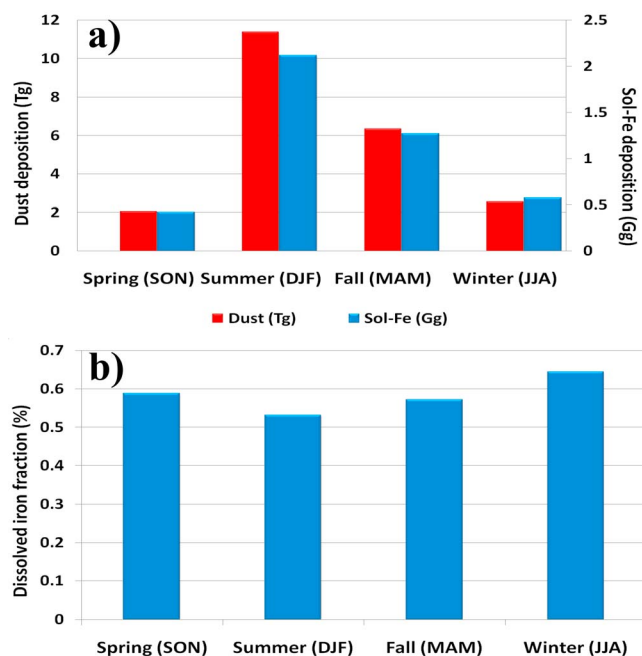


Figure 6. GEOS-Chem-predicted seasonally averaged (a) mineral dust (Tg) and sol-Fe (Gg), and (b) dissolved iron fraction of Patagonian dust deposited to the SAO.

far-removed from their source region, total fluxes of sol-Fe in Antarctic shelf waters remain very low (Figure 8). The analyses of our model results show that the annually averaged DIF of Patagonian dust is $\sim 0.57\%$, yielding $\sim 0.25 \mu\text{g m}^{-2} \text{ day}^{-1}$ atmospheric flux of sol-Fe to the SAO domain (surface area of the SAO is calculated to be $\sim 4.4 \times 10^{13} \text{ m}^2$). Our model simulations show that between October 2006 and September 2007 a total of $\sim 4 \text{ Gg}$ sol-Fe was deposited to the entire SAO domain; about 32% of this sol-Fe was predicted to be removed to the possible HNLC regions of the SAO below the ACC boundary.

[23] The inferred values of DIF over the SAO domain are significantly less than ones found downwind from east Asia [Solmon *et al.*, 2009] and are thought to be primarily attributed to low concentration of acidic trace gases available to be mixed with Patagonian dust. Considering that the initial DIF of Patagonian soil was prescribed to be the same as that of Solmon *et al.* [2009], such low DIF values suggest that acid mobilization may not be an important mechanism for producing sol-Fe in the SAO domain. Substantial acidification of aeolian dust prior to its deposition to the ocean may be required to solubilize the large fraction of mineral-Fe and make it bioavailable. This result is consistent with our earlier studies based on the analysis of remotely sensed data in the SAO suggesting that dust deposition may not be the controlling mechanism for biological activity in this region [Meskhidze *et al.*, 2007]. Our modeling results for the contribution of mineral dust to the total Fe budget is also consistent with previous oceanographic studies [e.g., Löscher *et al.*, 1997], indicating that aeolian input is likely to account for less than 10% of sol-Fe concentration in the surface waters of the SAO. Under such low dust-Fe inputs, other sources of bioavailable Fe (e.g., anthropogenic combustion [Luo *et al.*, 2008]) may become important. Overall,

our model simulations show that the distribution of DIF as well as total sol-Fe fluxes can be very complex and highly variable in both space and time, suggesting that detailed chemical evolution of dust plumes may be needed for the accurate representation of the role of mineral-Fe in different regions of the oceans. Furthermore, although dust plumes may not deposit sufficient quantities of sol-Fe to cause visible phytoplankton blooms, the episodic supply of dust from Patagonia may still be an important source of the micronutrient Fe for HNLC waters of the SAO. Detailed analyses of hydrographic data concurrently with in situ studies of mineral dust composition and sol-Fe fraction may be necessary to unambiguously identify the role of Patagonian dust for biological productivity in the SAO.

4. Model Sensitivity Studies

[24] In this section, sensitivity calculations are presented to illustrate how variations in key model parameters affect fluxes of sol-Fe. All sensitivity studies were conducted for December 2006 and sol-Fe deposition fluxes are compared to the baseline simulation. December 2006 was chosen because of active dust advection out of Patagonia and elevated dust fluxes to the SAO during this period. Total Fe and all other conditions remained unchanged between the runs; therefore, the differences in sol-Fe deposition between the baseline case and the sensitivity simulations can be interpreted as the relative changes due to the variations in the specific model parameter. Table 2 shows total sol-Fe fluxes (Gg) during December 2006 to the SAO predicted by GEOS-Chem/DFeS during the baseline and sensitivity simulations. According to Table 2, Fe-laden clay dissolution demonstrated a small effect ($\sim 6\%$) on total sol-Fe fluxes to the SAO. Such a small relative increase in sol-Fe is due to the minor dissolution rates of both Fe oxides and clay minerals for mineral aerosols characteristic to this region. This sensitivity study indicates that regardless of whether the sol-Fe originates from clays or Fe(III) oxides, the soluble fraction of mineral-Fe in the SAO domain is likely to remain low.

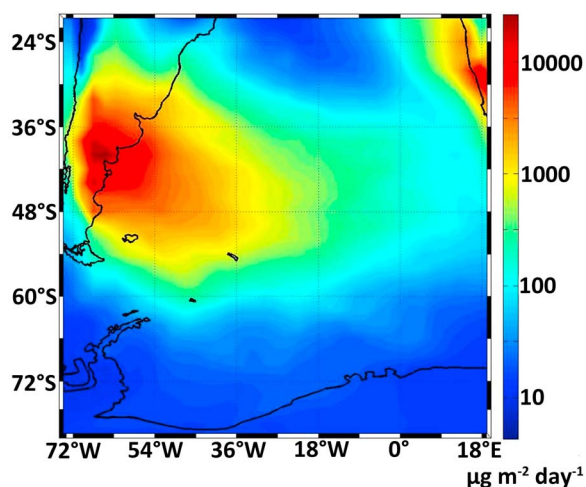


Figure 7. GEOS-Chem-predicted annually averaged spatial distribution of dust deposition to the SAO ($\mu\text{g m}^{-2} \text{ day}^{-1}$).

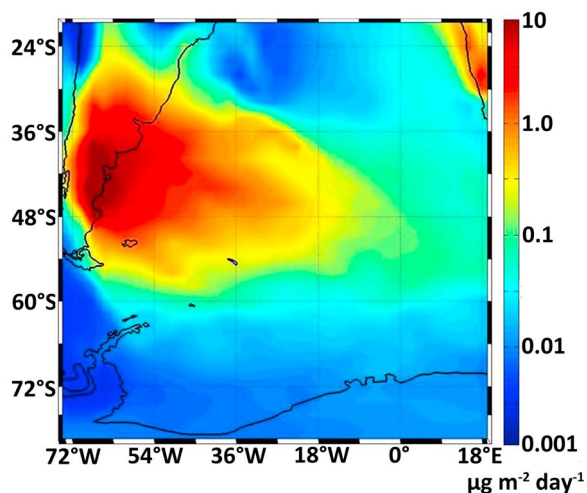


Figure 8. GEOS-Chem-predicted spatial distribution of annually averaged sol-Fe deposition to the SAO ($\mu\text{g m}^{-2} \text{day}^{-1}$) for the baseline simulation period.

[25] To understand the effects of natural sources of acidic trace gases on mineral-Fe dissolution, we carried out simulations with doubled SO_2 emissions from natural sources. The DMS emissions were uniformly increased in the model by a factor of two, and the emissions were doubled for the active volcanoes Lascar (23°S , 68°W), Llaima (39°S , 72°W) and Sabancaya (16°S , 72°W) volcanoes in the Patagonia/SAO domain that could have potential influence on Patagonian dust plumes [Chin *et al.*, 2000; Andres and Kasgnoc, 1998; Simkin and Siebert, 1994]. Table 2 shows a limited enhancement of $\sim 7\%$ in sol-Fe fluxes is predicted with doubling of natural sulfur emissions rates. Such a small increase in sol-Fe deposition rates can be viewed as an indication that during the high dust season, even with doubling natural emissions, the buffering capacity of mineral dust may remain large enough to counteract the acidification. Nevertheless, Figure 10 shows that despite little change in total monthly averaged fluxes (Table 2), considerable spatial variation in deposition rates can be observed, with the highest increase in regions of the SAO associated with DMS emissions. Figure 10 also indicates that for the years with no major volcanic perturbations, out of the two natural sources of SO_2 , DMS emissions are likely to have much larger impact on the mobilization of Fe within Patagonian dust. This result is likely attributed to the fact that South American volcanoes in the model have low SO_2 emission rates during this time period and higher vertical transport pathways in comparison to that of Patagonian dust plumes. This result highlights the importance of marine biological emissions of DMS for the sol-Fe budget in surface waters of the SAO and points the need for more field studies to better constrain the model-predicted fluxes of DMS.

[26] Under the conditions of high dust buffering capacity and low concentrations of acidic trace gases characteristic to the study domain, a major source of uncertainty in simulating sol-Fe fluxes could be attributed to the initial Fe solubility in mineral dust prescribed at the source region. Reported studies have shown a very large range (between $\sim 0.001\%$ to 2%) for the water soluble fraction of Fe in desert topsoils [Guieu and Thomas, 1996; Spokes and

Jickells, 1996; Zhuang *et al.*, 1990; Bonnet and Guieu, 2004; Ooki *et al.*, 2009]. To better understand how the DIF at the source region impacts the deposition of sol-Fe to the SAO, sensitivity study was conducted with initial DIF reduced to 0.1% . Table 2 shows that out of all the parameters examined, DIF at the source region has the dominant effect on the magnitude of bioavailable Fe deposited off the coast of Patagonia. Although studies show that a significant fraction of mineral-Fe can become soluble during atmospheric transport and transformation of Asian dust plume [Solmon *et al.*, 2009], our simulations suggest that prescribed DIF at the source region is likely to be the primary control for predicted Fe fertilization effect of Patagonian dust. Such high dependence on initial DIF can be explained by the relatively pristine nature of this region and highlights the need for improved quantification of the magnitude and chemical composition of water sol-Fe and reactive Fe nanoparticle formation at the dust source regions of South America.

[27] To illustrate how reasonable variations in the CaCO_3 composition of Patagonian-dust at the source region can affect sol-Fe production rates, sensitivity simulations were conducted in which the CaCO_3 concentration was prescribed to be 3% . According to Claquin *et al.* [1999] such low CaCO_3 concentrations are not typical for large regions of Patagonian surface soils; nevertheless, it is possible for the dust plumes emanated from the predominant source regions of Patagonia to have very low CaCO_3 concentration. The sensitivity test results, summarized in Table 2, reveal that the threefold reduction in CaCO_3 concentration yields up to a 60% increase in sol-Fe deposition to the SAO. Although this study demonstrates the sensitivity of sol-Fe production to the buffering capacity of mineral dust and the need for improved quantification of the mineralogical composition of Patagonian soil, our model results suggest that even with minimal CaCO_3 concentrations, aeolian input accounts for a small fraction of the sol-Fe budget of the SAO.

[28] The potential importance of RH dependent uptake of SO_2 onto Patagonian dust is examined in a sensitivity study using SO_2 uptake rates based on findings for CaCO_3 powder

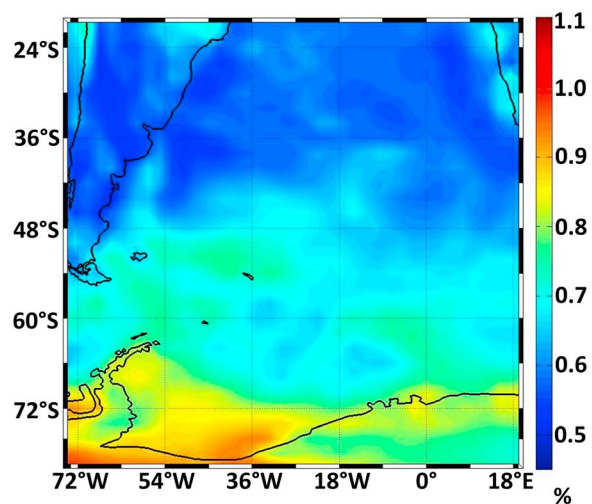


Figure 9. GEOS-Chem-predicted annually averaged dissolved iron fraction (%) of the dust deposited to the SAO.

Table 2. Total Sol-Fe Deposited to the SAO During the Baseline Simulations and Model Sensitivity Studies From December 2006

	Total Sol-Fe Deposition (Gg)	Percent Change (%)
Baseline	0.86	0.0
Fe-laden clay dissolution	0.91	5.8
Double natural SO ₂ emissions	0.92	7.0
Initial Fe solubility (0.1%)	0.17	-80.2
Reduce CaCO ₃ (3.0%)	1.37	59.3
RH dependent SO ₂ uptake	0.75	-11.8

by *Al-Hosney and Grassian* [2005] and *Preszler Prince et al.* [2007]. Table 2 illustrates that the implementation of RH dependent SO₂ uptake resulted in about 12% decrease in sol-Fe deposited to the SAO. One of the likely reasons for such a small change to sol-Fe production in Patagonian dust is the minor contribution from chemical aging due to pristine nature of this region. Nonetheless, future studies need to be conducted to better constrain SO₂ uptake rates on genuine mineral (and in particular Patagonian) dust in the presence of Fe-oxides and variable RH conditions [*Adams et al.*, 2005; *Baltrusaitis et al.*, 2007; *Fu et al.*, 2009].

5. Conclusion

[29] A year-long GEOS-Chem/DFeS simulation was conducted to quantify the magnitude and spatial distribution of Patagonian dust and associated sol-Fe fluxes to the SAO. The advantage of using GEOS-Chem/DFeS is the model's ability to predict DIF of mineral-dust and the fluxes of sol-Fe without making an a priori assumption of dust-Fe solubility. Currently there are few 3-D global models that can explicitly calculate dissolution-precipitation kinetics of Fe oxides based on gas- and aqueous-phase chemistry of reactive compounds within air masses containing mineral dust. Comparison of model simulations with available ground-level and remotely sensed data shows that GEOS-Chem/DFeS correctly identified dust emission regions, individual dust episodes, and dust outbreak seasonality from

South America. For a year-long simulation from October 2006 to September 2007 Patagonia is predicted to be the major contributor of dust to the SAO domain. In Patagonia, dry lake/river beds and low lying regions with little vegetative cover have been identified as the predominant sources of windblown dust [*Prospero et al.*, 2002]. Model-predicted yearly total dust emissions from South America ($\sim 30 \text{ Tg y}^{-1}$) should be viewed as the low estimate of mineral dust mass, as the model only considers dust particles with diameters up to $12 \mu\text{m}$. However, this limitation does not affect the prediction of dust and sol-Fe deposition to the SAO domain. The atmospheric lifetime of particles larger than $12 \mu\text{m}$ is not sufficient to have considerable impact downstream of the source [*Tegen and Fung*, 1994; *Ginoux et al.*, 2001]. At the seasonal timescale, the austral summer was identified as the strongest period of dust transport and deposition, contributing about one-half of the yearly total dust fluxes to the SAO. Such a strong seasonal pattern in dust mobilization and transport from Patagonia is consistent with the atmospheric circulation patterns and local meteorological data. The comparison of Patagonian visibility reports, as well as ground-level and remotely sensed data of AOD with the model-predicted daily dust abundances demonstrate that GEOS-Chem was capable of capturing the majority of reported large dust storms and the seasonality of mineral dust advection in the region.

[30] During the year-long simulation of GEOS-Chem/DFeS an annual mineral dust deposition of $\sim 22 \text{ Tg y}^{-1}$ to the surface waters of the SAO was predicted, and $\sim 30\%$ of this dust was simulated to be deposited to the possible HNLC regions of the SAO. Although the total mineral dust deposition to the SAO is consistent with the range of estimates made from past modeling and measurement studies [*Duce and Tindale*, 1991; *Jickells and Spokes*, 2001; *Gao et al.*, 2001; *Gaiero et al.*, 2003; *Ginoux et al.*, 2001], the extreme paucity of in situ dust measurements in Patagonia and the SAO and the difficulties associated with satellite retrievals of dust plumes in the region does not allow for the comprehensive evaluation of model-simulated spatial distributions of dust concentrations and fluxes.

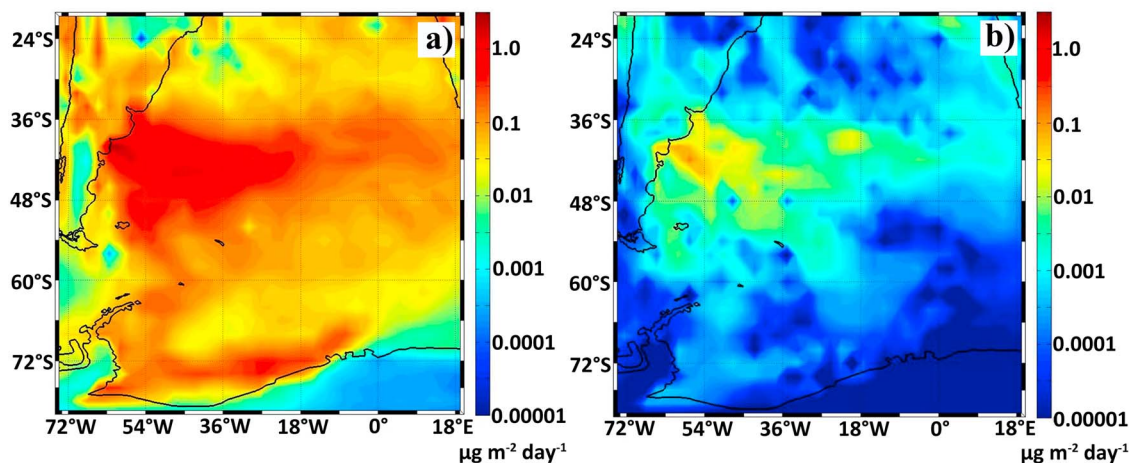


Figure 10. Difference between sensitivity simulations and default monthly averaged sol-Fe deposition rates ($\mu\text{g m}^{-2} \text{ day}^{-1}$) due to the doubling of (a) DMS and (b) volcanic SO₂.

[31] From October 2006 to September 2007 GEOS-Chem/DFeS predicted an average DIF of Patagonian dust to be $\sim 0.57\%$, leading to $\sim 4 \text{ Gg y}^{-1}$ of sol-Fe deposited to the surface waters of the SAO. It is important to note that $\sim 32\%$ of the sol-Fe produced within Patagonian dust plumes was deposited to the regions of the SAO considered to be possible HNLC areas. Compared to the measurement and modeling studies conducted in North Pacific [e.g., Luo *et al.*, 2005; Meskhidze *et al.*, 2005; Solmon *et al.*, 2009], the model-predicted DIF and sol-Fe fluxes are very low, leading to small fertilization of the surface ocean. The minor amount of sol-Fe formed during the baseline simulations is primarily explained by the low ambient concentrations of acidic trace gases that can acidify mineral aerosols and initiate Fe dissolution. Analyses of model results for sol-Fe deposition data suggest that Patagonian dust may not be the major controlling factor for ecosystem productivity in Fe-limited oceanic regions of the SAO, as dust is likely contributing less than 10% of the estimated yearly averaged sol-Fe flux. Although predicted sol-Fe results have not been tested extensively in the SAO and therefore should not be considered as conclusive, our simulation results for sol-Fe deposition data are consistent with recent in situ studies in the SO [Blain *et al.*, 2007; Wagener *et al.*, 2008] and corroborate hypothesis that large phytoplankton blooms in the SO are likely to be supported by the up-welled Fe-rich waters rather than dust deposition [Meskhidze *et al.*, 2007; Blain *et al.*, 2007; Wagener *et al.*, 2008]. Despite low average DIF of Patagonian dust, model results highlight the potential importance of episodic dust transport and deposition, as the total amount of sol-Fe deposited during such events can be orders of magnitude higher compared to the annually averaged dust flux. In addition, the model portrays a complex spatial pattern of DIF over the SAO, with DIF from 0.45% near the source regions to about 1.0% near the Antarctic shelf waters, highlighting the advantages of the prognostic approaches to Fe dissolution kinetics. Last, although the amount of sol-Fe formed during short-term atmospheric dust-Fe dissolution may not be the dominant source of bioavailable Fe leading to large-scale dust-mediated blooms in the SAO, a study by Boyd *et al.* [2009] shows that long-term oceanic dissolution of dust-Fe in the euphotic zone through grazer/particle interactions, photo-reduction in conjunction with siderophores, and reduction of Fe within particle micro-zones, may also contribute to the oceanic sol-Fe concentrations.

[32] If we presume that the acidity of particles is an essential factor to determine Fe dissolution, our sensitivity analyses suggest that considerable changes in biogenic emission sources (e.g., much higher emissions of DMS or large volcanic eruptions co-located with dust) may be needed for sizeable enhancement of sol-Fe in Patagonian dust. Model sensitivity studies also show that in pristine environments with little anthropogenic influence, initial DIF and the dust mineralogical composition (specifically CaCO_3) could have a dominant effect on predicted sol-Fe formation. In this study we primarily focused on acid mobilization of Fe in deliquesced mineral aerosol solution and did not explicitly consider enhancement of Fe dissolution rates by cloud cycling and photo-reductive dissolution in the pres-

ence of organic complexing agents [Cornell and Schindler, 1987; Suter *et al.*, 1988; Siffert and Sulzberger, 1991; Pehkonen *et al.*, 1993; Spokes *et al.*, 1994; Desboeufs *et al.*, 2003; Spokes and Jickells, 1996; Hand *et al.*, 2004; Mackie *et al.*, 2005]. Despite the great advancement in our capability of modeling enhancement of Fe dissolution, the mechanistic treatment of photo-chemical reductive dissolution of Fe in the presence of strong organic ligands remains a challenging issue for regional and global 3-D chemical transport models [Solmon *et al.*, 2009]. Additional in situ studies in remote waters of SAO are needed to increase the knowledge of mineral dust and sol-Fe deposition, thus furthering the understanding of the implications these processes have on phytoplankton primary productivity, atmospheric CO_2 concentrations, and global climate.

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C. Carouge and R. M. Yantosca, School of Engineering and Applied Sciences, Harvard University, 29 Oxford St., Cambridge, MA 02138, USA.
 P. Y. Chuang, Earth and Planetary Sciences Department, University of California, Santa Cruz, 1156 High St., Santa Cruz, CA 95064-1099, USA.
 D. M. Gaiero, CIgES, FCEfyN, Universidad Nacional de Córdoba, Enrique Barros, 5000 Córdoba, Argentina.

S. Gassó, Goddard Earth Sciences and Technology Center, University of Maryland Baltimore County, 5523 Research Park Dr., Ste. 320, Baltimore, MD 21228, USA.

M. S. Johnson and N. Meskhidze, Department of Marine, Earth, and Atmospheric Sciences, North Carolina State University, Jordan Hall, Rm. 1125, 2800 Faucette Dr., Raleigh, NC 27695-8208, USA.

F. Solmon, CNRS, 3, rue Michel-Ange, F-75794 Paris CEDEX 16, France.

Y. Wang, Department of Environmental Science and Engineering, Tsinghua University, Beijing 100084, China.

S. Wu, Atmospheric Science Program, Department of Geological and Mining Engineering and Sciences, Michigan Technological University, 6340 Dow Environmental Sciences and Engineering Bldg., 1400 Townsend Dr., Houghton, MI 49931-1295, USA.