Atmospheric global dust cycle and iron inputs to the ocean

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[1] Since iron is an important micronutrient, deposition of iron in mineral aerosols can impact the carbon cycle and atmospheric CO_2. This paper reviews our current understanding of the global dust cycle and identifies future research needs. The global distribution of desert dust is estimated from a combination of observations of dust from in situ concentration, optical depth, and deposition data; observations from satellite; and global atmospheric models. The anthropogenically influenced portion of atmospheric desert dust flux is thought to be smaller than the natural portion, but is difficult to quantify due to the poorly understood response of desert dust to changes in climate, land use, and water use. The iron content of aerosols is thought to vary by a factor of 2, while the uncertainty in dust deposition is at least a factor of 10 in some regions due to the high spatial and temporal variability and limited observations. Importantly, we have a limited understanding of the processes by which relatively insoluble soil iron (typically $\sim$0.5% is soluble) becomes more soluble (1–80%) during atmospheric transport, but these processes could be impacted by anthropogenic emissions of sulfur or organic acids. In order to understand how humans will impact future iron deposition to the oceans, we need to improve our understanding of: iron deposition to remote oceans, iron chemistry in aerosols, how desert dust sources will respond to climate change, and how humans will impact the transport of bioavailable fraction of iron to the oceans.

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1. Introduction

[2] In most regions of the world ocean photosynthetic production (i.e., ''primary'' productivity) is limited by the availability of the nutrients nitrate and phosphates. Regions where nutrients concentrations are high are usually characterized by high concentrations of chlorophyll in surface waters. There are, however, large areas of the world ocean where the concentrations of nutrients are high yet chlorophyll is low (that is, high nutrient low chlorophyll (HNLC) waters), for example the equatorial Pacific and much of the southern oceans. *Martin* [1990] hypothesized that primary productivity in HNLC regions was limited by the availability of iron. Deposition of iron to these regions also has important implications for the CO_2 budget, as increases in iron to the oceans may result in increased productivity and hence a decrease of CO_2 in the atmosphere. A pattern consistent with these interactions is observed on glacial timescales [e.g., *Martin*, 1990; *Watson and Lefevre*, 1999; *Archer et al.*, 2000]. Additionally, certain nitrogen fixing organisms such as *Trichodesmium* [*Falkowski et al.*, 1998; *Mills et al.*, 2004] have higher iron requirements; thus increased supplies of iron may impact the production of the macronutrient fixed nitrogen and influence productivity in oligotrophic tropical waters.

[3] In many open-ocean regions the input of new iron to the surface waters is dominated by the atmospheric deposition of soluble iron in mineral aerosols [e.g., *Fung et al.*, 2000; *Sarthou et al.*, 2003]. In terms of the biogeochemical response to atmospheric deposition, the aerosol iron fraction of importance is that which is bioavailable and this may be

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different than the soluble fraction (see section 4.3). Mineral aerosols consist of soil particles that are lifted into the atmosphere when high winds occur over erodible surfaces. These particles can be transported long distances from their source regions before being deposited via settling, turbulent deposition or precipitation processes. Much of this paper describes our understanding of dust sources, transport and deposition. Changes in these processes in the future will play a large role in controlling changes in atmospheric deposition of bioavailable iron. We also review the less understood issue of how insoluble soil iron is processed in the atmosphere to become more soluble, and presumably more readily utilized by ocean biota.

[4] This paper is a product of the IGBP Fasttrack iron meeting, held during April 2004 in Norwich, England. Our goal is to highlight the current understanding of atmospheric iron deposition to the oceans and identify gaps in our current understanding in order to promote studies that will complete our picture of the transport of iron from the terrestrial sources to the oceans. Additionally, we want to understand how humans and climate change will alter dust-iron fluxes to the ocean in the future. This is a prerequisite for the appropriate inclusion of iron in earth system models. This paper represents the first synthesis of our knowledge of desert dust sources, transport and deposition processes, as well as estimates of the amount of soluble and bioavailable iron in desert dust deposited in the source regions, although there are large uncertainties in what is bioavailable. Related papers overviewing the whole iron cycle [*Jickells et al.*, 2005], as well as the ocean cycle and paleoclimate data showing relationships between iron and carbon dioxide, are planned.

[5] This paper will review dust source areas, emission processes (section 2.1), transport and deposition mechanisms (section 2.2) and dust distributions as observed and modeled (section 2.3). Section 3 reviews variability of dust fluxes, including the role of humans in modulating dust (sections 3.1 and 3.2). Section 4.1 reviews our understanding of the amount of iron in mineral aerosols, while section 4.2 focuses on dust deposition estimates from modeling and observations. Section 4.3 addresses the atmospheric processing of iron to become soluble. Section 5 contains recommendations for future research areas. We do not address the important processes occurring in the oceans which control the solubilization and utilization of iron in the upper ocean, which are addressed in papers related to this one [e.g., *Jickells et al.*, 2005]. A brief review of dust interactions with climate is including in the auxiliary material (text, section S1.0).[1]

2. Controls on Distribution of Atmospheric Mineral Aerosols

[6] Atmospheric deposition of iron to the open ocean is dominated by the iron contained in mineral aerosols (sometimes referred to as soil dust or desert dust) [*Fung et al.*, 2000]. Here we discuss the sources, transport and deposition processes controlling the entrainment of soil particles and their deposition downwind in ocean regions.

2.1. Sources of Atmospheric Mineral Aerosols

2.1.1. Geographical Source Areas

[7] The dominant sources of mineral aerosols are the arid regions of North Africa, the Arabian Peninsula, Central Asia, China, Australia, North America, and South Africa. While general sources were well known, satellites are providing new information on sources and their relative importance [e.g., *Prospero et al.*, 2002; *Washington et al.*, 2003]. Auxiliary Figure S1 shows an estimate of the largest source areas from *Prospero et al.* [2002] and their downwind trajectories. While the relative magnitude of the sources are difficult to constrain using only satellites, the strength of the retrieved aerosol optical thickness in nearby ocean regions suggests that mineral aerosols are one of the dominant aerosol types worldwide (see auxiliary Figure S2).

[8] There are a few satellites that can detect aerosols over land, such as the Total Ozone Mapping Spectrometry absorbing aerosol index (TOMS AAI) [*Torres et al.*, 1998] or Infra-red Difference Dust Index (IDDI) [*Legrand et al.*, 1994]. The majority of the sources seen from satellite are centered over basin regions which drain from highlands [e.g., *Prospero et al.*, 2002; *Goudie and Middleton*, 2001; *Brooks and Legrand*, 2000; *Washington et al.*, 2003]. These highland regions serve as a source of a steady supply of small particles, which are moved down into the basin after rain events. When the soils dry, these relatively small alluvial particles (typically under tens of micrometers diameter) can be easily entrained into the atmosphere. Thus, while in the dust source itself precipitation must be low so that the soils remain free to erode, the greater precipitation in adjacent highlands increases the production of small particles through weathering and the subsequent transport to deflation basins. For example, the runoff from the Tibesti and Ahaggar mountains in central Africa contributes to the dominant dust sources observed in adjacent low-lands [e.g., *Prospero et al.*, 2002]. Additionally, many of the important dust sources were flooded during the Holocene and Pleistocene (e.g., Lake Chad Basin in North Africa [*Prospero et al.*, 2002; *Washington et al.*, 2003]). Emissions are strongest at the edges of these formerly wet basins [e.g., *Prospero et al.*, 2002; *Reheis et al.*, 1995; *Yaalon*, 1987].

[9] Further studies based on the inspection of satellite images (especially MODIS) suggest that many large dust events are comprised of dust derived from "point" sources. The classic examples are the sources in the Bodele Depression in Africa. *Prospero et al.* [2002] identify the Bodele as one of the most persistently active dust sources in the world. Close examination of the dust events in the Bodele show that the dust cloud is comprised of a multitude of discrete plumes. MODIS shows that the plumes are long and narrow with very little dispersion over a distance of 100 km or more (e.g., Figure 1). The main source area lies within a region identified as playa (dry lakes). Furthermore, the plumes tend to start at the eastern (windward) edge of the playa. Although we lack explicit data on the site we can imagine a scenario for these dust events. There is often a strong easterly wind flow in this region augmented by the

[1] Auxiliary material is available at ftp://ftp.agu.org/apend/gb/2004GB002402.

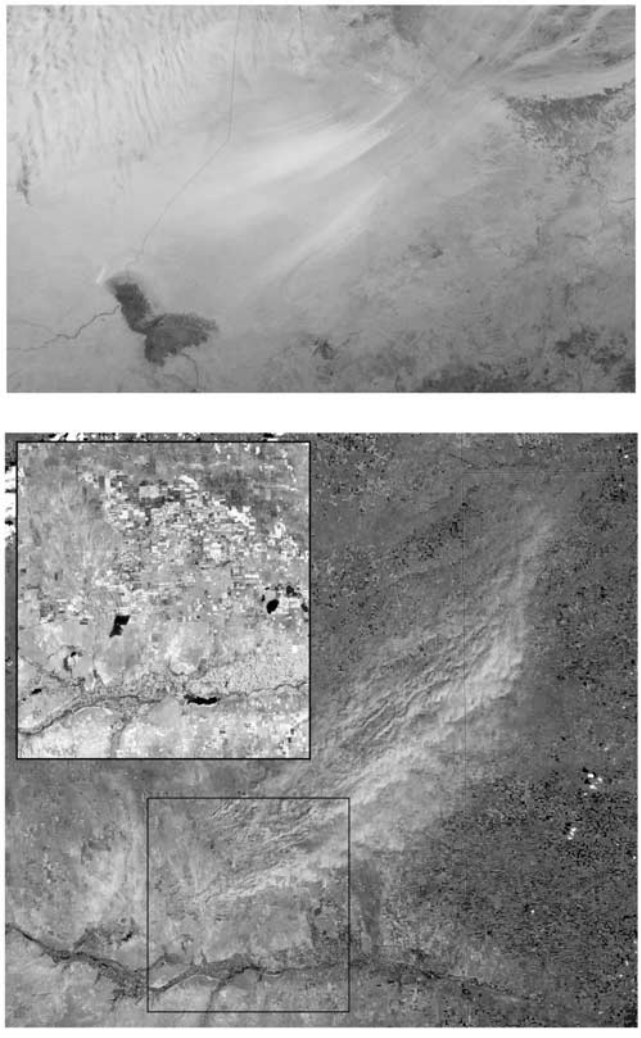

Figure 1. (top) TERRA MODIS (moderate resolution imaging spectroradiometer) image of Bodele basin in North Africa (approximately 17°N, 18°E) on 11 February 2004 [*Koren and Kaufman*, 2004] taken from http://earthobservatory.nasa.gov/Newsroom/NewImages/images.php3?img_id=16534. (bottom) Colorado dust storms, 18 April 2004. This MODIS image shows a cluster of dense dust plumes that erupted from fields in southeastern Colorado, to the north of the Arkansas River visible at the bottom of the image. The dust cloud extends into Kansas (border in the right of the image); later in the afternoon, dust was carried deep into Nebraska (border in the upper right corner). The inset shows a Landsat image of the area demarcated by the box. The grid pattern in the image indicates the presence of agricultural fields that cover much of this area. These fields seem to be the source of the major dust plumes seen in the image. (Landsat image is courtesy of Max Bleiweiss, New Mexico State University. The MODIS image is from the NASA MODIS Rapid Response System Gallery web site: http://rapidfire.sci.gsfc.nasa.gov/gallery/). See color version of this figure at back of this issue.

funneling of winds between the Tibesti Mountains and the Ennedi Plateau. In MODIS and Landsat, one can see long linear dune formations extending from Egypt through this "gap" and into Chad. Winds or infrequent rainstorms constantly move sand into the Bodele playa system. This sand blasts the surface of the playas (as described in the following section) releasing a large and steady flow of dust to the atmosphere. The importance of small-scale dust sources dispersed in large-scale basins has been well documented in extensive field studies in various regions including the Southwestern United States [e.g., *Gillette*, 1999]. Additionally, it is likely that sources evolve during the year as changing seasons bring strong winds and low precipitation to different regions [e.g., *Marticorena and Bergametti*, 1995]. Another example of "point sources" of dust is shown in Figure 1 (bottom), where winds from the west drive a desert dust event in an agricultural region of Colorado in the United States. Note that not all the agricultural fields are equally good at producing dust.

[10] Satellite data [e.g., *Prospero et al.*, 2002] suggest that the largest and most intense dust sources lie in the Northern Hemisphere (e.g., auxiliary Figure S1 and S2). Nonetheless it is difficult to assess the importance of specific dust sources on a global scale. This is especially true for smaller sources that could be important for transport to low dust deposition regions (e.g., off the coast of South America or South Africa). In contrast to the Northern Hemisphere where dust often dominates total column optical thickness, over the southern oceans dust contributes a small fraction of the aerosol optical thickness relative to other aerosols (e.g., sea salt aerosols [*Tegen et al.*, 1997; *Reddy et al.*, 2005]). Also the optical depths are close to the detection limit of the satellites, although new satellites have improved capabilities [*Barnaba and Gobbi*, 2004; *Kaufman et al.*, 2005].

2.1.2. Dust Emission Processes

[11] Dust generation is a highly complex process that responds in a non-linear way to a variety of environmental factors. For this reason, dust emissions are highly variable on spatial and temporal scales. The first requirement is that the soil surface be dry and sparsely vegetated. Secondly there must be present on the surface substantial concentrations of relatively large particles (tens to hundreds of μm diameter) that the wind can "grip" and force into movement. Once these particles start to move, they bounce (saltate) across the surface, dislodging smaller particles (under 10–20 μm diameter) which can be lifted into the atmosphere and subsequently carried great distances as "dust." Auxiliary Figure S3 shows a simple diagram of these processes.

[12] The modeling of the soil deflation process begins with the saltation flux, the horizontal mass flux over the surface. This flux is proportional to the third power of the wind friction velocity [e.g., *Bagnold*, 1941; *White*, 1979; *Gillette*, 1974, 1979; *Shao et al.*,1993], a measure of the wind shear stress on the soil surface; it is a function of planetary boundary layer winds, surface roughness and atmospheric stability [e.g., *Garratt*, 1992].

[13] The threshold friction velocity is defined as the friction velocity above which soil particles begin to move in saltation flux. Work by *Bagnold* [1941], *Chepil* [1945],

Iversen et al. [1976] and *Iversen and White* [1982] suggests that the threshold friction velocity increases as grain size increases owing to gravity, and it also increases for the smallest particles owing to particle cohesion. These two effects lead to an optimum particle size ($\sim$60–80 μm) for which the threshold friction velocity is minimum.

[14] A second factor that strongly affects the erosion threshold in natural situations is the presence of non-erodible elements (such as vegetation or rocks). They affect the erosion threshold in two ways. First, the non-erodible elements cover part of the surface and thus protect it from the aeolian erosion; second, they consume part of the wind momentum that would otherwise be available to initiate particle motion. Thus an increase in non-erodible elements decreases the emissions of dust [*Schlichting*, 1936; *Marshall*, 1971; *Marticorena and Bergametti*, 1995]. Agriculture can change non-erodible elements [e.g., *Kardous et al.*, 2005].

[15] Another factor affecting the erosion thresholds is soil moisture. Briefly, the soil water reinforces the cohesion forces between the soil grains and thus increases the erosion thresholds. Soil water retention consists of molecular adsorption on the soil grains' surface and capillary forces between the grains. Inter-particle capillary forces are the main factor responsible for the increase of the wind erosion threshold observed when the soil moisture increases. Below a soil moisture content close to the maximum amount of adsorbed water, these capillary forces are considered as being not strong enough to significantly increase the erosion threshold. Since the total potential soil moisture content depends on the soil texture, the increase in erosion threshold is different for the same soil water content, depending on the soil type. A parameterization of the influence of the soil moisture on the erosion threshold has been proposed by *Fécan et al.* [1999]. It enables the computation of the increase of the erosion threshold in wet conditions by reference to dry conditions as a function of the soil moisture and the residual soil moisture defined as a function of the soil clay content.

[16] These various factors affecting the aeolian erosion thresholds are responsible for a large part of the observed heterogeneity of dust emissions in time and space from a dust source region. Thus a correct assessment of dust fluxes requires a precise knowledge of the surface properties of erodible terrains. Such detailed knowledge is unavailable for most of the Earth's surface.

[17] Once the threshold friction velocity is exceeded, the horizontal flux (saltation flux) begins. Only a small fraction (order 10^{-6}) of the saltation flux is converted into a vertical flux, i.e., the fraction that becomes suspended dust in the atmosphere [*Marticorena et al.*, 1997]. The boundary between suspension and saltation is controlled by the ratio of the threshold friction velocity and the terminal velocity of the particles. The terminal velocity is defined by the equilibrium of the particle weight and the wind drag and so depends on the particle diameter and density and on atmospheric conditions. Above 50 μm, particles are infrequently suspended into the vertical dust flux, and remain in the horizontal saltation flux [*Greeley and Iversen*, 1985]. However, mobilization of particles with a diameter smaller than 50 μm requires very high threshold friction velocities owing to the strong cohesion forces between them. Thus the movement of these fine particles is not initiated directly by the wind friction on the erodible surface, but requires sandblasting or bombardment processes to disrupt the aggregates and produce dust particles in the saltation flux [e.g., *Gillette*, 1979; *Gomes et al.*, 1990; *Shao et al.*, 1993] (see auxiliary Figure S3). Once the smaller particles are created in the saltation flux by the larger particles, the smaller particles are more easily suspended into the atmosphere and thus they dominate the vertical flux. There is evidence for generation of fine particles during abrasion processes that can occur in dust storms [*Bullard et al.*, 2004]. Additionally, the shorter lifetime of larger particles due to gravitational settling produces downwind a transported size bin closer to 2–3 μm [*Schulz et al.*, 1998]. Since dust production requires saltation as an intermediate process, the vertical flux (i.e., the flux of dust that becomes available for long-range transport) is generally derived from the horizontal flux through a coefficient of proportionality. Most of the recent experimental and theoretical studies attempt to express this coefficient as a function of soil characteristics [*Alfaro et al.*, 1997; *Shao*, 2001; *Alfaro and Gomes*, 2001; *Shao*, 2004]. These processes are visualized in auxiliary Figure S3.

2.1.3. Atmospheric Modeling of Dust Sources

[18] A variety of regional and global models of the dust aerosol cycle have been developed since the early 1990s. Within these models dust emissions are either prescribed, or (more often) computed using model-predicted meteorological fields. Dust is generally transported as a passive tracer and removed from the atmosphere by dry and wet deposition processes. While in earlier global models [e.g., *Joussaume*, 1990; *Tegen and Fung*, 1994; *Andersen et al.*, 1998; *Mahowald et al.*, 1999] all desert surfaces were assumed to be potential dust sources, recent models [e.g., *Ginoux et al.*, 2001; *Zender et al.*, 2003b; *Tegen et al.*, 2002] make an effort to take into account the influence of the different surface conditions on dust emissions. Most of the current generation of dust models include the effects of erodible soil elements, soil moisture, and other wind tunnel derived impacts described in section 2.1.2 [e.g., *Zender et al.*, 2003a]. However, simplifications are required at the global level because of the lack of good data sets of soil texture, non-erodible soil elements and other properties required at the fine scales. Moreover, it is not clear how relationships derived in wind tunnel studies at a small scale level (section 2.1.2) should be extrapolated to wind, surface and soil conditions specified in a global or regional model-sized grid-box. Such extrapolations have not been explicitly studied and thus are only crudely parameterized in models. There are likely to be regional effects (such as funneling through topography) as well as microscale turbulent effects which are important when scaling from wind tunnels to regional or global scale models that are well quantified. Many modelers parameterize this by assuming a globally constant dust emission factor, which is modified to result in global dust concentrations, deposition fluxes and optical depths (a measure of the global attenuation of direct solar radiation) that agree with observations.

Table 1. Deposition of Dust Into Various Ocean Basins[a]

	Duce et al. [1991]	*Prospero* [1996]	*Ginoux et al.* [2001]	*Zender et al.* [2003a]	*Tegen et al.* [2004]	*Luo et al.* [2003]	Composite Dust Deposition [*Jickells et al.*, 2005]
North Pacific	480	96	92	31	56 (0.29)	35 (0.13)	72
South Pacific	39	8	28	8	11 (0.47)	20 (0.22)	29
North Atlantic	220	220	184	178	259 (0.21)	230 (0.14)	202
South Atlantic	24	5	20	29	35 (0.20)	30 (0.17)	17
Indian Ocean	144	29	154	48	61 (0.12)	113 (0.10)	118
Global emissions	-	-	1814	1490	1800	1650	1790

[a]Units are Mt/yr. The numbers in brackets are variability estimates (stdv/mean).

[19] Apart from the global scale dust models, a number of models have been developed to describe the dust cycle for key regions like the Sahara [*Westphal et al.*, 1988; *Marticorena et al.*, 1997; *Nickovic and Dobricic*, 1996] and East Asia [e.g., *Zhang and Carmichael*, 1999; *Wang et al.*, 2000]. Such regional-scale models are well suited for simulation of individual dust storm events or for comparisons with in-situ observations, made during field experiments. While the topography, soil conditions and small-scale extreme wind events are described better in regional than in global models, there are still uncertainties in describing the dust source area, the surface conditions in the source area, and changes in size distribution and chemical composition during transport. Currently, global models predict dust emissions of between 1000 and 2000 Mt/yr for particles with less than 10 μm radius (Table 1).

2.2. Dust Transport and Deposition

[20] Dust atmospheric lifetime depends on the particle size, ranging from a few hours for particles larger than 10 μm, to up to several weeks for submicrometer-sized dust particles. The lifetime of submicrometer sized particles depends partly on the precipitation efficiency, which in turn depends on the type of precipitation, on the vertical distribution of dust, and on particle properties (which may change during transport). In the atmosphere, dust is moved by the prevailing winds and transported vertically by moist and dry convective processes, as well as adiabatic vertical motion associated with frontal systems. Once lifted into the free troposphere, dust can be transported over thousands of kilometers from the source areas. Thus dust can be transported out of the Sahara with the Easterly jet over the Atlantic, often reaching the Caribbean [e.g., *Prospero and Nees*, 1986], southern Florida [*Prospero*, 1999], and the eastern United States as far north as New England [*Perry et al.*, 1997]. Asian dust often crosses the Pacific to North America, resulting in significant concentrations over large areas [*VanCuren*, 2003]. One dust event from China is documented to have continued across the Atlantic and deposited measurable dust amounts in the Alps [*Grousset et al.*, 2003].

[21] A major mechanism leading to dust deposition is the sedimentation due to gravity. Because of this process, large particles sediment out more quickly than smaller particles, which leads to a shift toward smaller particle sizes during transport. Close to the source areas dry deposition is the dominant deposition mechanism for dust particles. While dust particles with sizes of about 70 μm are picked up most easily by the winds, dust transported up to thousands of km usually has a mode diameter of approximately 2 μm [e.g., *Schulz et al.*, 1998]. Recent measurements during the PRIDE experiment show larger mode sizes, typically 3.5 μm, for dust that has traveled from North Africa to Puerto Rico [*Reid et al.*, 2003; *Grini and Zender*, 2004]. Comparisons of the size distribution of African dust measured in the Canary Islands off the coast of West Africa with those in Puerto Rico show only a relatively minor shift to smaller sizes after a transport of about 4000 km [*Maring et al.*, 2003]. While typically the dust mass median diameter is relatively small, large dust particles (>100 μ m) have been carried great distances to remote ocean regions [e.g., *Betzer et al.*, 1988]. The long-range transport of such large particles cannot be simulated using conventional knowledge of dry-deposition processes [e.g., *Slinn and Slinn*, 1980; *Colarco et al.*, 2002; *Reid et al.*, 2003; *Ginoux*, 2003; *Grini and Zender*, 2004]. Additional processes need to be included in models to account for the transport of large particles.

[22] Dust is also washed out of the atmosphere by precipitation. The interaction of dust with clouds is not well understood, partly because, in general, aerosol-cloud interactions are not well understood, and particularly because of a dearth of cloud microphysical measurements in dust dominated regions. This lack of a well-known relationship makes different researchers make very different assumptions about cloud-dust interactions. Mineral aerosols are not readily soluble in water, leading many researchers to assume that mineral aerosols do not interact with clouds directly, but rather that they are scavenged via sub-cloud removal mechanisms. Mineralogical evidence suggests, however, that dust particles readily attract water [*Koretsky et al.*, 1997], and thus some investigators assume that mineral aerosols are readily incorporated into clouds and can act as cloud condensation nuclei (CCN) [*Rosenfeld et al.*, 2001]. To simulate the poorly understood wet removal process, many researchers use simple scavenging ratios [e.g., *Tegen and Fung*, 1994; *Mahowald et al.*, 2003] of between 200 and 1000 based on observational estimates of between 200 and 1000 [*Duce et al.*, 1991]. However, other models use more explicit microphysics to incorporate aerosols into clouds and wash them out [*Rasch et al.*, 2001]. Recent studies have also included the impact of mixing dust with soluble sulfate thereby increasing the hygroscopic properties of the dust; this work suggests that this improves the comparison of their model results with in situ measurements [*Fan et al.*, 2004]. Wet deposition is a difficult process to simulate in models, and comparisons of wet deposition

fluxes in models show that very different vertical profiles result from different parameterizations that have similar surface deposition [*Rasch et al.*, 2000]. Simulations of dust in the current generation of dust models show large differences in their wet deposition lifetimes that range from 56 days [*Ginoux et al.*, 2001] to 10 days [*Mahowald et al.*, 2002]; these discrepancies suggest that further work in this area is required. Unfortunately, explicit measurements of wet and dry deposition are rare [e.g., *Sarthou et al.*, 2003], and more observations of both wet and dry deposition are needed. Despite the differences in simulating wet and dry deposition, current models yield a reasonable match with the few observations of total deposition [e.g., *Ginoux et al.*, 2001; *Tegen et al.*, 2002; *Luo et al.*, 2003]. The few observationally based estimates we have suggest that wet deposition is more important than dry deposition over ocean regions [*Hand et al.*, 2004] (auxiliary Table S1), although these are limited in both space and time.

2.3. Dust Distributions: Observations and Models

[23] Owing to the short atmospheric lifetime of dust (days to weeks) many observations are required to adequately characterize dust concentration and flux distributions. At present, there are approximately two dozen long-term data sets obtained at in situ aerosol measurement stations [e.g., *Prospero and Nees*, 1986; *Arimoto et al.*, 1990, 1997] (University of Miami network). Some additional data sets of in situ deposition flux are compiled by *Ginoux et al.* [2001], and ice core and ocean sediment flux (e.g., compiled by *Kohfeld and Harrison* [2001]). The difficulty of interpreting ocean sediment flux data in terms of dust deposition is detailed in section 4.2).

[24] Because of their large spatial coverage, satellite data have proven useful in evaluating dust sources, transport and deposition in global models. Both AVHRR [*Husar et al.*, 1997] and TOMS AI [*Herman et al.*, 1997] have yielded long term records of aerosol optical depth, but it is difficult to retrieve dust properties quantitatively from these records. Dust retrievals are often difficult because of the presence of aerosols other than dust and of clouds and because of the assumptions that must be made about the physical properties of the aerosols. New satellite observations from POLDER, MODIS and MISR will likely provide greater insights into dust distributions because of increased spectral, spatial and temporal resolution [*Barnaba and Gobbi*, 2004; *Ginoux and Torres*, 2003; *Kaufman et al.*, 2005]. Large dust plumes can often be observed in satellite images; in some cases, plumes can be followed for a week or more during which time the dust cloud can span an entire ocean. A number of web sites present collections of satellite images that can be searched for specific types of events including dust storms (e.g., http://visibleearth.nasa.gov/, http://rapidfire.sci.gsfc.nasa.gov/products/, http://jwocky.gsfc.nasa.gov/aerosols/aerosols.html, http://www.osei.noaa.gov/). While quantitative retrieval of dust properties is still difficult, the areal coverage and progress of dust clouds provides useful information for the validation and improvement of dust models [e.g., *Tegen et al.*, 2002].

[25] Many global dust models are based on forecast center meteorological analysis, which are an optimal combination of model and meteorological observations. These models yield dust distributions that match satellite dust observations reasonably well [e.g., *Ginoux et al.*, 2001; *Tegen et al.*, 2002; *Zender et al.*, 2003a; *Luo et al.*, 2003]. In contrast model studies based on general circulation winds may have biases due to problems in model surface winds [e.g., *Tegen and Miller*, 1998]. Model studies allow us to extrapolate existing measurements (usually dust concentration or optical depth) to larger geographical areas; these extrapolated concentration data can then be used to calculate dust deposition. Correlations between model predicted values of concentration at observational sites against nearby grid boxes suggest that observations of concentration represent regional pictures of concentration and, to a lesser extent, deposition [*Mahowald et al.*, 2003]. In contrast, satellite-based estimates of optical depth provide a global picture of aerosol distribution. But in studies of the correlation between modeled optical depth, mobilization and deposition, they only capture about 50% of the variability in deposition or mobilization. Thus estimates of dust mobilization or deposition based on satellite optical depths are subject to uncertainty [*Mahowald et al.*, 2003]. These differences are partly due to the fact that dust plumes are often traveling at altitude and partly because most dust deposition occurs during precipitation events that are unrelated to satellite optical depth.

3. Dust Variability and Anthropogenic Influences

3.1. Temporal Variability

[26] Dust source activity, and dust transport and deposition are highly variable on timescales ranging from minutes to centuries to millennia. The dust events shown in Figure 1 highlights the temporal-spatial heterogeneity of one desert dust event. Besides variability in winds, it is not clear what other factors are the major drivers of this variability on longer timescales. Given the highly non-linear character of dust generation processes, relatively small changes in the source region environment can have great consequences.

[27] Studies suggest that there were up to 100-fold changes in the mineral aerosol deposition to ice cores between cold and warm periods. For example, dust deposition was much greater during the Last Glacial Maximum, $\sim$21,000 years before present compared to the present-day warm climate [e.g., *Petit et al.*, 1990; *Legrand*, 1995; *Steffensen*, 1997; *Rea*, 1994]. Globally averaged deposition changes are likely to be on the order of 2–3 fold greater during cold periods [e.g., *Rea*, 1994; *Mahowald et al.*, 1999]. Within glacial periods dust concentrations in ice showed very strong variability [e.g., *DeAngelis et al.*, 1997]. The increase in dust at the time of the last glacial maximum could have been caused by several mechanisms. Stronger surface wind speeds could lift greater amounts of dust aerosols from surfaces compared to modern conditions. Increased aridity could have expanded source areas. Because of lower sea levels, large areas of sediments on the continental shelved are exposed and could be eroded by winds. Atmospheric loadings of dust particles could also be increased under cold climate conditions as a consequence of the weakened hydrological cycle. The decrease in atmo-

spheric water content leads to decreased precipitation, reducing the washout of atmospheric dust particles and thus leading to longer atmospheric lifetimes. Model investigations used to assess the impact of those processes on the increase in glacial dust deposition, conclude that all of these above mentioned processes may play important roles for glacial-interglacial changes in the dust cycle [e.g., *Joussaume*, 1990, 1993; *Andersen et al.*, 1998; *Reader et al.*, 1999; *Mahowald et al.*, 1999; *Lunt and Valdes*, 2002; *Werner et al.*, 2002]. The potentially large effect of dust supplied by glacial outwash has not yet been evaluated. It is perhaps significant that the huge areas of loess were laid down during glacial times, for example the massive deposits in what is now the central United States [*Bettis et al.*, 2003]. Dust coming from North Africa is likely to have decreased substantially during the climate optimum period of the Holocene (~6000 kybp), which is thought to be related to the greening of North Africa during that time period [e.g., *deMenocal et al.*, 2000]. Testing the performance of the dust models for past conditions is a valuable tool for evaluating the model performance in drastically different climates, and such tests will help to assess model performance for future predictions of the dust cycle.

[28] In recent history, dust coming from North Africa observed at Barbados has changed by a factor of 4 between the wet 1960s and the dry 1980s [*Prospero and Nees*, 1986; *Prospero and Lamb*, 2003]. Analyses of land based data [*Goudie and Middleton*, 1992; *N'Tchayi et al.*, 1994, 1997] demonstrate that the frequency of dust storms in the Sahel have increased since the 1950s. Many authors have attributed this increase in dust activity to widespread land degradation and desertification driven by a combination of climatic desiccation and human impacts: overgrazing, deforestation, population pressure and inappropriate land-use practices are often cited as causes of degradation [*Tegen and Fung*, 1995; *Tegen et al.*, 1996]. *N'Tchayi et al.* [1997] suggested that the Sahel had become a more significant source of dust than the Sahara.

[29] There is, however, no strong evidence to support claims of systematic, regional-scale desertification or land degradation. Many studies have concluded that there has been no long-term shift in the desert boundary between the Sahel and the Sahara, nor any net decrease in potential productivity in the Sahel [*Tucker et al.*, 1991, 1994; *Nicholson and Tucker*, 1998; *Prince et al.*, 1998; *Schlesinger and Gramenopoulos*, 1996]. While anecdotal evidence suggests that localized land degradation may present a problem in some areas, other studies suggest that the problem has been exaggerated [*Tiffen and Mortimore*, 2002]. Furthermore, it is sometimes difficult to decouple human impacts from the cycles in wind erosion associated with episodes of drought or climatic desiccation, which are natural features of semi-arid regions where rainfall is highly variable on multiple timescales [*Thomas*, 1997].

[30] It has also been suggested that changes in dust event frequency may be the result of changes in meteorological mechanisms associated with dust mobilization and transport rather than with changes in the land surface. During the late twentieth century in the Sahel, there was a shift from large, well-organized convective disturbances associated with both rainfall and dust mobilization, to weak, poorly organized disturbances associated with dry years. It has been hypothesized that increases in dust event frequency in the summer may be the result of a greater proportion of dry convection events associated with dust mobilization but not with rainfall and subsequent wet deposition [*Brooks*, 2000; *Brooks and Legrand*, 2000].

[31] Recently, *Prospero and Lamb* [2003] showed that dust concentrations in Barbados were highly correlated with prior-year rainfall deficits in the Sahel-Soudano region of North Africa. The correlation with rainfall does not necessarily mean that rainfall itself was the major factor. Other meteorological variables associated with changes in rainfall could be responsible, including changes in wind speed, gustiness, transport paths, etc. It was pointed out that the major dust peaks in the Barbados record were closely associated with major ENSO events, the ENSO generally leading by one year the peak dust concentrations. These fluctuations in dust observed far from the dust source regions can be due to a combination of source, transport and depositional changes. It is difficult to determine the relative role of these factors, including the role of humans, since the only available studies of human land use change in the Sahel also showed a large expansion in land use between the 1960s and the 1980s. Land use has remained relatively constant since then [e.g., *Stephenne and Lambin*, 2001], similar to the temporal behavior in dust at Barbados. Modeling studies have attempted to simulate this change, but were hampered by the accuracy of the forecast center surface winds and precipitation for this problem [*Mahowald et al.*, 2002].

[32] In contrast, 20 years of dust measurements at Midway in the North Pacific do not show any long term trends [*Prospero et al.*, 2003] although there have been some periods when dust activity did increase somewhat, especially in the late 1990s for several years. While dust measurements at Midway show no trends, *Prospero et al.* [2003] show that the concentration of anthropogenic sulfate at Midway doubled from 1981 to the early mid 1990s, closely matching the emissions of the pollutant SO_2 in China. The contrast between the trends in the concentration of a known pollutant ($SO_2 - SO_4^=$) and the absence of a trend in dust suggests that the lack of a trend in dust is not linked to changes in transport over time. Contrary to popular opinion, the frequency of severe dust storms in China over the period 1954–2002 was at a peak in the 1950s; it has steadily decreased, reaching a minimum in the 1990s although showing a relative increase in 2000–2002 [*Zhou and Zhang*, 2003]. *Sun et al.* [2001] show that dust storm variability in China is most strongly driven by changes in strong winds. Thus the recent increase in strong fronts along with dry conditions is believed to be responsible for the increase in dust storms in the late 1990s.

3.2. Quantifying Anthropogenic Impacts on Dust

[33] Section 2.1 shows that the dominant sources of dust in the current climate appear to come from natural topographic lows, where easily erodible alluvial soils were deposited during earlier pluvial periods and which are now replenished during rain or wind events [e.g., *Prospero*

et al., 2002; *Goudie and Middleton*, 2001; *Brooks and Legrand*, 2000]. However, the studies described in section 2.1 were not quantitative enough to predict anthropogenic impacts on radiative forcing, for example, and possible changes that may occur in the future to desert dust sources. In addition, studies such as these based on satellite observations may be sensitive to other factors such as boundary layer depth [e.g., *Mahowald and Dufresne*, 2004] as well as the low correlation between satellite optical depth and dust entrainment into the atmosphere [*Mahowald et al.*, 2003]. Anthropogenic sources include agricultural regions that become active during nonvegetated, dry seasons [e.g., *Gillette and Passi*, 1988], pastured regions in semi arid regions, and regions where the water table has been substantially modified [e.g., *Reheis*, 1997]. These regions may well overlap with sources identified as ''natural'' since they will all occur in arid basin regions with fine soils and highland drainage. Additionally, there is some evidence that some sources are very small (e.g., Figure 1), which highlights the strong hetereogeneities in the sources which suggests that it will be difficult for satellites to determine whether human activities impact sources on a global scale.

[34] Modeling studies have attempted to constrain the anthropogenic portion of present-day dust more quantitatively. *Tegen and Fung* [1995] suggested that a 30–50% land use source best matched the AVHRR data, but later studies showed that this was a model dependent result [e.g., *Schulz et al.*, 1998]. *Mahowald et al.* [2002] suggested that the 4x-fold increase at Barbados was best matched by a new source, but that the forecast center meteorological data set was not sufficiently robust to be definitive. *Luo et al.* [2003] showed that a 50% land use and a 0% land use source were equally good at matching available observations, and were less different than differences between source parameterizations and meteorological winds. *Tegen et al.* [2004] used dust storm frequency data [*Engelstaedter et al.*, 2003] to estimate that land use sources contributed less than 10% to the total source. In contrast, using another model and methodology but the same data as *Tegen et al.* [2004], a 0–50% land use source were found be statistically equivalent [*Mahowald et al.*, 2004]. These studies highlight the great uncertainties in using model simulations to quantify anthropogenic disturbances to atmospheric dust.

[35] Regional studies may allow us to constrain the land use sources of dust more conclusively. *Xuan and Sokolik* [2002] attempted to quantify the amounts of dust derived from various sources in China. They identify and characterize three broad types of dust sources in Northern China: Type 1, deserts in dry-agricultural areas; Type 2, deserts located on the plateaus; and Type 3, deserts located in topographical lows. They find type 1 sources (dry agricultural areas) only contribute 1% of the total annual particulate matter dust emissions while Type 2 and 3 contribute 35% and 64%, respectively. *Chin et al.* [2003] also identify limited regions of land use as sources in Asia. *Yoshioka et al.* [2005] suggest that land use represents between 0 and 25% of the North African source, using TOMS AI and simulated model absorbing aerosol index.

[36] In addition to direct land use changes to source areas, there are possible impacts through the hydrological cycle (e.g., the Aral Sea or Owens Lake [*Reheis*, 1997]) and through climate changes. Studies suggest that fluctuations in the water table are correlated with dust emissions [e.g., *Mahowald et al.*, 2004]. More studies on the role of changes in the hydrological cycle due to natural or human perturbations and their impact on desert dust generation are required.

[37] Determining the global anthropogenic portion of dust may not be possible using satellite data or model results because of the difficulty of discerning different types of sources using either model [e.g., *Mahowald et al.*, 2002; *Luo et al.*, 2003; *Mahowald and Luo*, 2003] or satellite data [*Mahowald et al.*, 2003; *Mahowald and Dufresne*, 2004]. Since most dust sources occur at very small scales (see Figure 1) it is possible that these sources represent dirt roads or other anthropogenic disturbances, even in very remote regions. Thus field studies in specific regions are required to address the global importance of anthropogenic disturbance.

[38] Only one study has attempted to quantify the impact of human induced climate change on historical changes in dust sources between the pre-industrial and current climate, and this study concluded that depending on the relative importance of carbon dioxide fertilization and land use, humans could have increased the desert dust source by 60% or reduced it by 20% [*Mahowald and Luo*, 2003]. Unfortunately, ice core data were unable to discriminate between these cases because of regional inconsistencies in the ice core records, and because the ice cores are located so far from the dust source areas.

[39] Two studies [*Mahowald and Luo*, 2003; *Tegen et al.*, 2004] of projections of dust emissions in the next 100 years have been published, one yielding 20–60% reductions in dust, and the other resulting in either a 10% decrease or 20% increase in dust. These models included the impacts of changes in vegetation cover as consequence of changes in meteorological parameters, and changes in vegetation cover caused by a warmer climate and increased CO_2 levels. Reducing the discrepancies in these estimates will require a better understanding of the changes in vegetation under warmer climate conditions, together with improvements in dust emission modeling. The impact of climate change on natural sources of dust is an important topic that has not yet been addressed in a systematic and convincing manner. It is possible that changes in vegetation may open up new sources (e.g., ''disturbed sources'' from *Tegen and Fung* [1995]). However, this impact is not well quantified now, in either the main desert dust source regions or the smaller dust source regions that control ocean deposition to some regions.

4. Iron/Dust Connections

4.1. Iron Content of Dust

[40] Iron rather than dust is the key issue for ocean biogeochemistry. The iron content of minerals varies considerably from place to place depending on the mineralogy of the source material [e.g., *Claquin et al.*, 1999; *Fung et al.*, 2000]. The average iron content of the Earth's crust is 3.5% [*Taylor and McLennan*, 1985] and this value is widely used in assessing global-scale iron inputs to the ocean [*Duce and Tindale*, 1991]. There may be some differentiation in

amount of iron depending on the size fraction [e.g., *Claquin et al.*, 1999]. A compilation of observations (auxiliary Table S2) as well as model results [*Hand et al.*, 2004] show that the factor of 2 differences in iron amounts in soils tend to cause smaller variability in iron content in aerosols, suggesting that uncertainties in dust deposition and iron solubility (below) are more important variables to understand than the regional differences in iron amounts in source soils. This would be the case even for aerosol particles collected off the coast of Australia, where the iron content from Australian soils is predicted to be 50% higher than the global average [*Hand et al.*, 2004].

4.2. Dust Deposition to the Oceans

[41] There have been a variety of estimates of dust deposition to the oceans broadly falling into two groups, the first based on extrapolation of measurements [e.g., *Duce et al.*, 1991] and the second based on modeling [e.g., *Tegen and Fung*, 1995] with some approaches based on refinements and combinations of the two approaches [*Gao et al.*, 2001]. In all these methods, an aerosol field is estimated and the flux derived by some sort of parameterization of deposition. Hence the key uncertainties arise in estimations of the dust concentrations and in parameterization of deposition. The various approaches all yield broadly similar fluxes (Table 1) which might suggest that total dust deposition amounts to different basins are fairly well understood. It should be noted, however, that many models are calibrated and to some extent tuned against the field data. Thus the apparent agreement is not a good test of the accuracy of our assessments. Global maps of aerosol abundance from satellites [e.g., *Husar et al.*, 1997] provide an approximate check on the aerosol distribution pattern though this is largely qualitative. The uncertainties of any of these approaches is likely to be of order a factor of 10, due to uncertainties in sources, deposition, dust distribution as well as uncertainties due to the high temporal and spatial variability in dust.

[42] One independent method with which to check the validity of the flux estimates and/or the deposition velocities is to compare these with directly estimated ocean fluxes using deep ocean sediment traps. This has been done in a semi-quantitative manner on a global scale [e.g., *Mahowald et al.*, 1999] and quantitatively at particular stations [e.g., *Jickells et al.*, 1998; *Uematsu et al.*, 2003; *Bory and Newton*, 2000]. The use of sediment trap data requires that the traps are deployed so as to avoid advective or resuspended sediment fluxes and are free of hydrodynamic artifacts; hence traps must be located in deep water well above the sediments and well away from ocean margins. Sediment trap fluxes are highly variable in time in association with biological cycles in surface waters; indeed dust may contribute to the flux pattern itself by acting as ballast for downward ocean fluxes [*Francois et al.*, 2002]. Dust fluxes in the water column vary depending on the local biological productivity [*Bory and Newton*, 2000] and hence interpreting sediment trap data is difficult. Additionally, advection of dust in surface waters before being deposited in the sediment trap increase the uncertainty associated with sediment traps [*Siegel and Deuser*, 1997]. In areas far away from dust sources, the dust component of sediment trap samples will be very small, which can lead to large errors in estimated fluxes. Aluminosilicate fluxes to sediment traps are usually determined by difference between total fluxes and those from the biogenic components organic C, $CaCO_3$, and opal [e.g., *Jickells et al.*, 1998]. Consequently estimates are particularly uncertain at low dust fluxes. Differences in local dust composition can also contribute to uncertainties in estimates of total dust deposition. For example, Al is often used as an indicator of dust; consequently, large systematic errors will result if an inappropriate dust/Al ratio is used [e.g., *Tindale and Pease*, 1999].

[43] Figure 2 shows a comparison of a model composite of dust fluxes to the ocean [*Jickells et al.*, 2005] and compiled sediment trap data [*Kohfeld and Harrison*, 2001] (some of which suffer from some of the problems identified above). This model composite is an average of three model frameworks including more than 10 years of forecast center winds-based simulations with extensive comparison to available in situ and satellite observations [*Jickells et al.*, 2005; *Luo et al.*, 2003; *Ginoux et al.*, 2004; *Tegen et al.*, 2004]. The model and sediment trap consistency suggests this estimate of dust deposition is a good extrapolation of available data, within the factor or 10 uncertainty in dust fluxes.

4.3. Chemistry and Solubility of Iron in Dust

[44] Not all iron is bioavailable in the ocean. Most researchers focus on soluble iron, which is usually considered to be Fe(II), although other forms of iron may also be bioavailable [e.g., *Barbeau et al.*, 2001]. Indeed, Fe(II) may not always be available [e.g., *Visser et al.*, 2003]. Soluble iron in soils represents about 0.5% of total iron [*Fung et al.*, 2000; *Hand et al.*, 2004]. But measurements of iron in aerosols suggest a much higher solubility [e.g., *Zhuang et al.*, 1992], implying substantial atmospheric processing. Soluble iron fractions reported in different experimental data sets are summarized in auxiliary Table S3. Solubilities range from 0.01% to 80% and are highly heterogeneous in space and time. The measurements in auxiliary Table S3 were made by a variety of techniques and different measurement techniques will yield different values of iron solubility (see *Spokes and Jickells* [1996] example in table). There is still no consensus about which method yields the value of iron solubility most appropriate for the in situ seawater environment. Additionally, some of the early work may be biased because of the photoreduction of iron in the aerosol suspension exposed to the sun (e.g., see *Zhu et al.* [1997] for discussion). Fine mode aerosols (<2.5 μm) tend to yield larger iron solubilities than coarse mode aerosols ($>2.5\mu$m) [e.g., *Siefert et al.*, 1999]. Solubility may be enhanced by higher net acid concentrations in fine mode aerosol, but there is no observable relationship between iron solubility and acid species concentrations in any aerosol fraction [*Hand et al.*, 2004; *Baker et al.*, 2005]. *Hand et al.* [2004] found that the longer lifetime of fine mode particles could explain the differences in solubility. Mineral aerosols in rainwater tends to have iron solubilities (between 4 and 26%) in regions close to industrial regions (reviewed by *Jickells and Spokes* [2001], *Kieber et al.* [2001], and *Ozsoy*

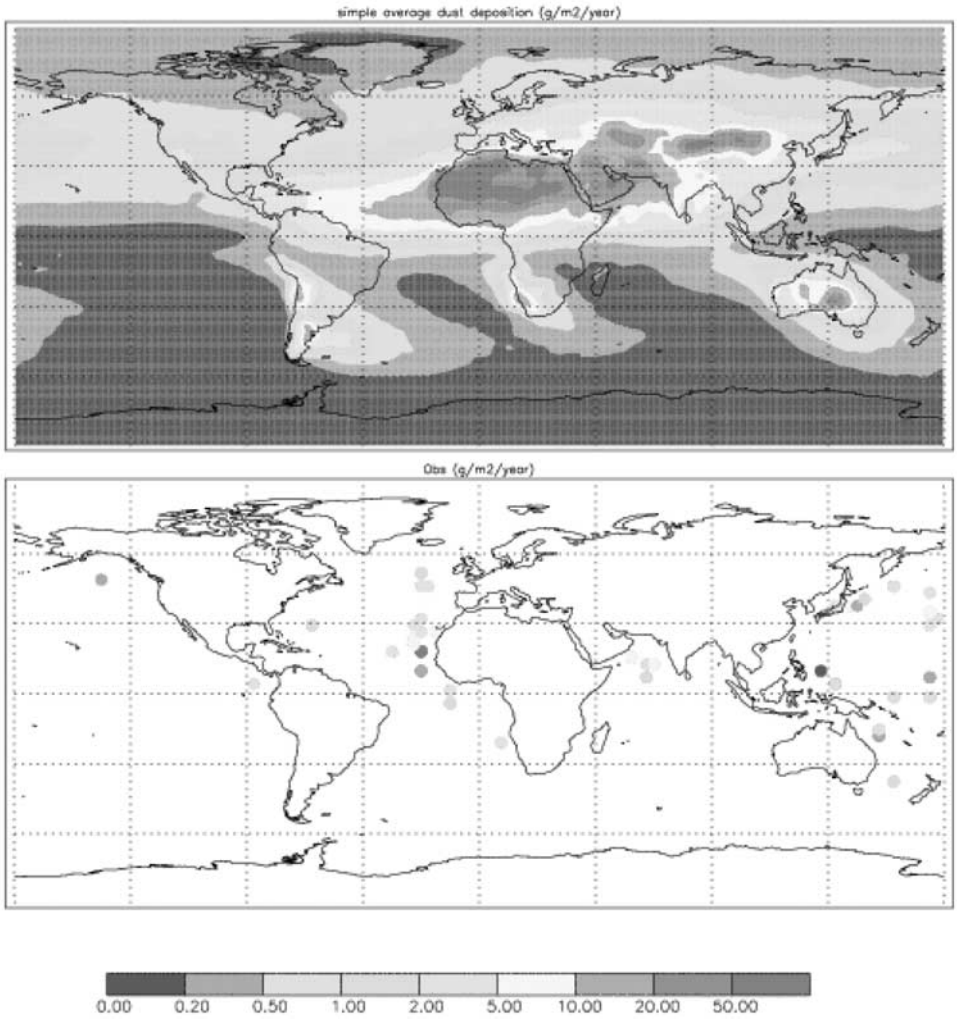

Figure 2. Desert dust deposition (g/m^2/yr) estimated from an average of three reanalysis based models simulated for 10+ years [*Luo et al.*, 2003; *Ginoux et al.*, 2004; *Tegen et al.*, 2004], and representing our best estimate of dust deposition. Models compare well to available in situ and satellite observations and are shown here compared against sediment trap data [*Kohfeld and Harrison*, 2001]. See color version of this figure at back of this issue.

and Saydam [2001]), and values as high as 50% near Bermuda [*Kieber et al.*, 2003].

[45] Although it is clear that atmospheric processing changes the solubility of iron as it moves from the sources regions to be deposited in oceans, it is not clear the relative importance of different mechanisms. Several studies have emphasized that photochemical and cloud processes in the atmosphere can modify iron solubility of desert dust [*Jickells and Spokes*, 2001; *Desboeufs et al.*, 2001; *Hand et al.*, 2004]. Laboratory studies suggest that reactions of iron with other aerosol species in solution could be very important in the enhancement of the bioavailable iron. Reactions of ferric iron with organic species such as oxalic acid could play a significant role in producing soluble iron [e.g., *Zuo and Hoigné*, 1992; *Zhu et al.*, 1993; *Pehkonen et al.*, 1993; *Siefert et al.*, 1994; *Zuo*, 1995]. Oxalic acid has anthropogenic sources such as incomplete combustion, ozonolysis and photooxidation of hydrocarbons and is common in cloud water [*Warneck*, 2000]. *Saydam and Senyuva* [2002] found increases in soluble iron during in-cloud photochemical reductions with oxalate and suggested that soil fungi were a natural source of oxalate. Inorganic aerosol solutions can also be important if mineral aerosols are coated with hygroscopic species such as sulfates and nitrates [e.g., *Zhuang et al.*, 1992; *Zhu et al.*, 1992]. Cycles of evaporation and condensation can result in very acidic solutions with high ionic strengths and can enhance the solubilization of Fe by photoreduction of Fe(III) [*Zhu et al.*, 1997]. Recent studies have suggested that pollution from Asia increases iron solubility [*Meskhidze et al.*, 2005; *Boyle et al.*, 2005], inconsistent with previous lower solubilities in the Pacific than the Atlantic [*Hand et al.*, 2004], but consistent with the recent high iron solubility measurements in the Pacific [*Ying*, 2004]. In situ aerosol data does not

support a clear relationship between either oxalic acids or sulfuric acids and iron solubility [*Chen and Siefert*, 2004; *Hand et al.*, 2004]. While photochemical reduction in dust aerosols over the Atlantic can yield significant concentrations of Fe(II) in situ, Fe(II) comprises only a very small fraction of the total soluble iron and the total soluble iron does not seem to be related to Fe(II) concentrations [*Zhu et al.*, 1997]. Modeling studies based on cloud processing and photolytic reactions have been unable to account for the great variability seen in observations; the models however did yield higher solubilities for fine particles, similar to observations, and attribute it to the longer residence time [*Hand et al.*, 2004]. *Baker et al.* [2005] suggest that Fe solubility may be systematically higher in areas remote from the desert dust plumes. Clearly we need much more study of the atmospheric processes that convert insoluble iron to soluble iron.

[46] Attempts have also been made to constrain the soluble iron amounts based on measurements in the ocean to complement the direct atmospheric measurements. *Jickells and Spokes* [2001] made some simple ocean model calculations to estimate the amount of soluble iron deposited to the oceans. Adapting the *Lefevre and Watson* [1999] model to estimate overall atmospheric iron, they obtained an iron solubility estimate of 2%. Aluminium solubility is generally found to be somewhat higher than that of iron [e.g., *Spokes and Jickells*, 1996; *Desboeufs et al.*, 2001; *Baker et al.*, 2005]. *Vink and Measures* [2001] estimated an overall aluminium solubility of 3.25%, which suggests an iron solubility similar to that calculated by *Jickells and Spokes* [2001]. In a rather detailed investigation of aluminium solubility *Gehlen et al.* [2003] considered a wide range of aluminium solubilties from dust and concluded that 1.5–3% best fitted the available data. The model of *Archer and Johnson* [2000] required the bioavailability of aerosol iron (which assumes that the bioavailability of aerosol iron is related to solubility) to be in the range of 2–12% depending on which atmospheric dust deposition simulation they used. *Fung et al.* [2000] assumed 1–10% solubility. *Wu and Boyle* [2002] report an iron solubility of 1% and *Bopp et al.* [2003] also assumed a 1% solubility.

[47] The uncertainty in the solubility of iron from dust is at least as important as the uncertainty and variability in dust fluxes themselves in determining the overall uncertainty of atmospheric bioavailable iron inputs to the oceans. Thus we require an improved understanding of the solubility of iron being deposited in mineral aerosols, as well as the atmospheric processing influencing iron solubility. The situation is further complicated once the mineral aerosol hits the surface ocean. Soluble iron delivered to the oceans from the atmosphere undergoes a dramatic change in physico-chemical environment. The effect of this on the bioavailablity of atmospheric delivered iron is poorly known [*Jickells et al.*, 2005].

5. Summary and Conclusions

[48] This assessment has focused entirely on desert dust sources that clearly dominate the global total iron/dust cycle. However, there are additional sources of iron emissions. In their review of crustal enrichment factors, *Wiersma and Davidson* [1986] reported an average enrichment factor for iron of 1.3 which implies that there are additional noncrustal sources (although this factor could be higher or lower in some regions). Iron from other sources, such as anthropogenic [*Spokes and Jickells*, 2002], meteoritic [*Johnson*, 2001] or volcanic sources [*Benitez-Nelson et al.*, 2003], is likely to be in the form of iron oxides rather than aluminosilicate phases and hence potentially more soluble [*Jickells and Spokes*, 2001; *Desboeufs et al.*, 2001]. Consequently these sources may contribute disproportionately to the soluble iron input. If industrial sources of iron aerosols are significant, it means that in addition to perturbing the iron cycle by changing dust source production, we may be influencing it by human activity that produces modest amounts of relatively soluble iron. More data is required to address the importance of alternative sources of iron.

[49] In order to understand how atmospheric iron will impact biogeochemistry in the future we need to improve our understanding of the following areas: (1) characterization of deposition of iron to remote regions of the oceans; (2) characterization of iron chemistry, including solubility, in aerosols and precipitation in remote regions; (3) characterization of desert dust source regions and their response to humans and climate change; and (4) how deposition bioavailability of iron will change with climate and human activity. The answers to all these questions are poorly constrained because of a dearth of observations. We have few observations of dust deposition in ocean environments, including the relative importance of wet and dry deposition. Additionally, there are few observations of iron solubility globally. Also we do not understand the processes that control how iron becomes more soluble in the atmosphere. More observations of dust concentration, deposition and iron solubility content are required in the remote ocean regions of the world.

[50] Furthermore, we need to improve our dust models that are essential to extrapolate our limited observations to global scales. In order to improve dust models we need a better understanding of dust source processes. One strategy would be to focus on major dust sources. Satellites can be used to identify sources in a variety of different terrains and environments, including those potentially impacted by human activities or climate change. Initial studies of these regions would be based on satellite products and supporting information on population, industrial and agricultural activities. These would be followed up by field expeditions to a selected subset of these sites. In addition to measurements related to the terrain and micrometeorological aspects of the study, aerosol chemical/physical properties and related soil characteristics should be studied. Measurements of optical properties of the soil and suspended dust particles should be combined with remote sensing retrievals of dust to develop improved algorithms for soil aerosols. Although dust sources in the Southern Hemisphere are relatively small compared to those in the Northern Hemisphere, they warrant study because they could have a disproportionate impact on ocean productivity due to their proximity to the large HNLC areas in the southern oceans. In contrast the large dust

sources in the Northern Hemisphere warrant study also because of their impact on radiative forcing and the impact on nitrogen fixing organisms [*Falkowski et al.*, 1998].

[51] Model predictions of dust deposition in remote regions are also uncertain owing to the long travel time and the consequent accumulation of errors in models. One of the great sources of uncertainty in estimating wet deposition of dust is our lack of knowledge about dust-cloud microphysics and precipitation formation processes. This problem could be partly addressed with more measurements of dust deposition as suggested above. Even so, there remains a major problem in model estimates because of the way in which they treat precipitation. Because in global models much precipitation occurs in sub-grid-scale events, the precipitation process and the consequent aerosol removal is highly parameterized. Thus we need improvement in parameterizations of cloud aerosols interactions, as well as the resulting aerosol wet deposition in models. Dust modeling studies as well as observational studies need to include uncertainty estimates so that we are better able to focus on critical processes in future studies in order to reduce our uncertainty.

[52] Dust models need to include iron chemistry and solubility processes. This requires additional observations of iron solubility and the processes controlling it. Only one modeling study has been published that includes iron solubility [*Hand et al.*, 2004]; clearly more work is required in this direction.

[53] Finally we need to understand how dust sources, transport, deposition and solubility might change in the future. This can best be done by understanding how these processes changed in the past with changing climate. This could be addressed by focusing on a few dust source regions in transitional environments. These would be monitored for changes over decade-long timescales to see how they respond to weather and climate variability and to climate change. It would be very interesting to see if we can retrospectively look at the dust variability over North Africa over the past decades to see if this can be linked to specific source regions or processes. Some attempts have been made in this direction, but more work is necessary to fully understand source change processes.

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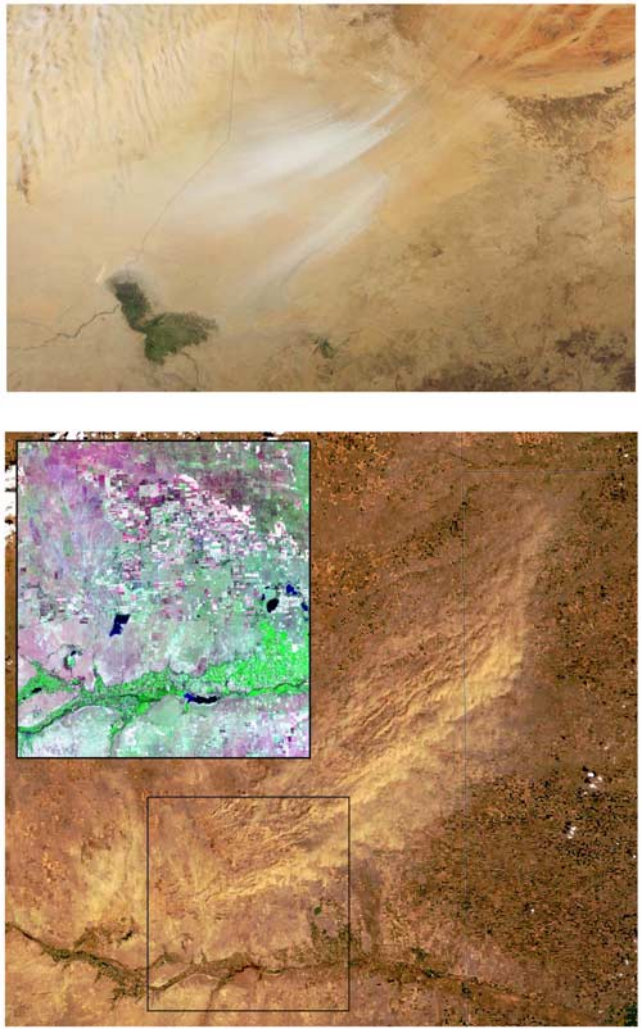

Figure 1. (top) TERRA MODIS (moderate resolution imaging spectroradiometer) image of Bodele basin in North Africa (approximately 17°N, 18°E) on 11 February 2004 [*Koren and Kaufman*, 2004] taken from http://earthobservatory.nasa.gov/Newsroom/NewImages/images.php3?img_id=16534. (bottom) Colorado dust storms, 18 April 2004. This MODIS image shows a cluster of dense dust plumes that erupted from fields in southeastern Colorado, to the north of the Arkansas River visible at the bottom of the image. The dust cloud extends into Kansas (border in the right of the image); later in the afternoon, dust was carried deep into Nebraska (border in the upper right corner). The inset shows a Landsat image of the area demarcated by the box. The grid pattern in the image indicates the presence of agricultural fields that cover much of this area. These fields seem to be the source of the major dust plumes seen in the image. (Landsat image is courtesy of Max Bleiweiss, New Mexico State University. The MODIS image is from the NASA MODIS Rapid Response System Gallery web site: http://rapidfire.sci.gsfc.nasa.gov/gallery/).

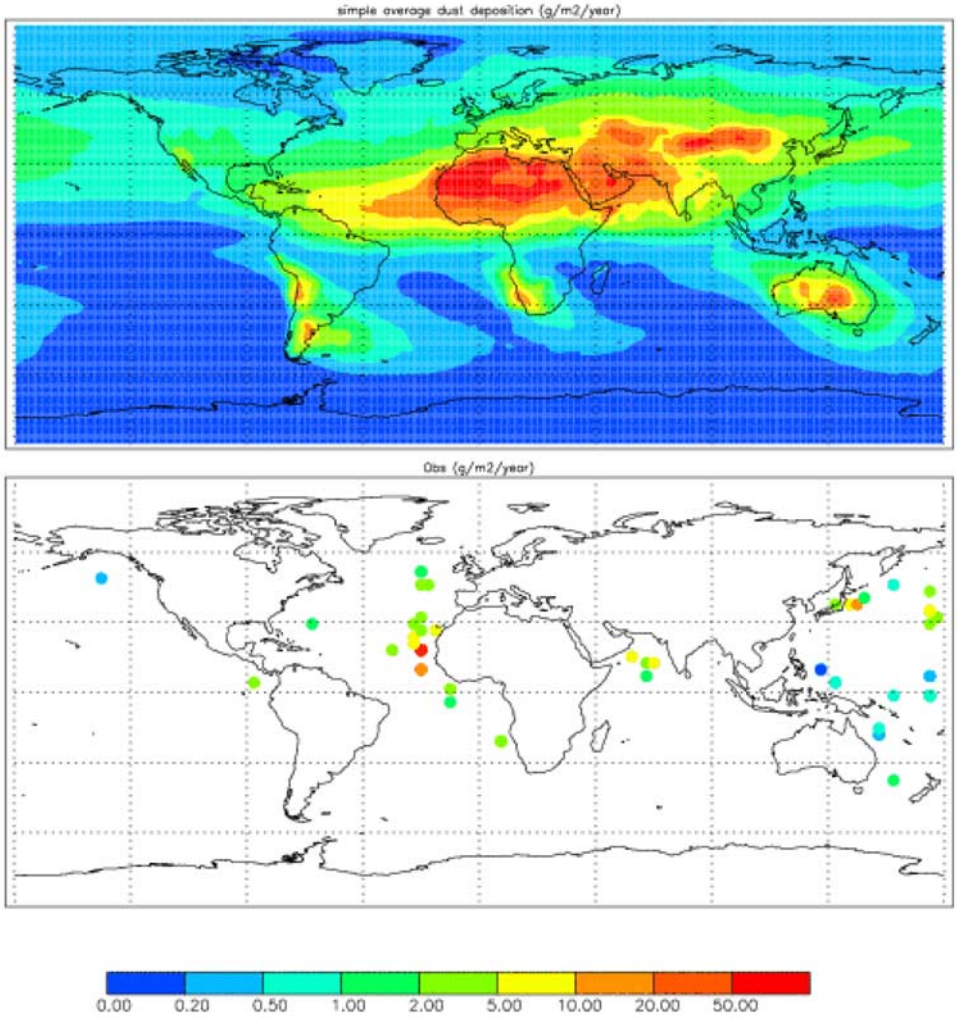

Figure 2. Desert dust deposition (g/m^2/yr) estimated from an average of three reanalysis based models simulated for 10+ years [*Luo et al.*, 2003; *Ginoux et al.*, 2004; *Tegen et al.*, 2004], and representing our best estimate of dust deposition. Models compare well to available in situ and satellite observations and are shown here compared against sediment trap data [*Kohfeld and Harrison*, 2001].