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Atmospheric Transport of Silicon

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Silicon (Si) can be transported through the atmosphere associated with airborne particles. Fly ash produced by industrial burning can contain Si, and those particles are abundant in heavily industrialized areas. However, global estimates of industrial Si emissions are not available. On the other hand, abundant amounts of Si are transported with airborne soil particles. Soil-derived mineral dust contributes significantly to the global aerosol load; estimates of global dust emissions range from 1,000 to 3,000 Mt/yr (Houghton et al. 2001). Dust aerosol consists of micrometer-size windblown soil particles that can be carried over thousands of kilometers through the atmosphere, and dust plumes are predominant features in satellite retrievals of global aerosol patterns (Husar et al. 1997; Herman et al. 1997). Silicon is one of the largest constituents of soil-derived mineral aerosol. Estimates of crustal Si content range between 26 and 44 wt% (Orlov 1992), where most Si is present in the form of quartz (SiO_2). The mean value of crustal Si has been estimated at 26.9 percent (Wedepohl 1978). Tréguer et al. (1995) estimate that about 10 Tmol (280 Tg) particulate Si reaches the ocean surface by eolian dust transport, of which they assume 5–10 percent to be dissolved in seawater. Thus, the transport of soil-derived dust in the atmosphere is an effective means of redistributing Si in the environment.

Global Cycle of Soil Dust Aerosol

Dust aerosol has a high spatial and temporal variability, and large uncertainties exist in quantitative estimates of large-scale dust loads. Modern global dust distribution and properties can be characterized by satellite retrievals and by concentration measurements at surface stations. So far, quantitative estimates of dust optical thickness by satellites are possible only over ocean surfaces, although qualitative patterns of dust distribution can be observed over land, for example, by the Total Ozone Mapping Spectrometer satellite instrument (Herman et al. 1997). In contrast, surface station measurements of dust concentrations (e.g., Prospero and Lamb 2003) and deposition flux measurements are too sparse to obtain regional averages through extrapolation of the local observations.

Therefore, assessments of dust deposition and of the effect of dust on climate rely on distributions computed with global dust cycle models, which predict emissions, transport within the atmosphere, and deposition. These models must be constrained and validated with available in situ observations or remotely sensed aerosol products.

Mineral aerosol is generated in arid and semiarid continental regions. Major dust sources include the Saharan and Sahelian region, the Arabian Peninsula, the Gobi and Taklamakan deserts in Asia, and the Australian desert (Duce 1995). The majority of atmospheric dust is emitted from North African (50–70 percent) and Asian (10–25 percent) deserts. Dust emission occurs when the surface wind shear velocity in the source area exceeds a threshold value, which is a function of surface properties such as the presence of roughness elements, the size of soil grains, and soil moisture. With the parameterization given by Marticorena and Bergametti (1995), the threshold wind shear velocities to lift grains from the ground are approximately 60 cm/s, 30 cm/s, 40 cm/s, and 150 cm/s for particles with diameters of 1,000, 100, 20, and 2 μm , respectively. Theoretical considerations and wind tunnel experiments show a dependence of dust emissions to the third or fourth power of the surface wind shear velocity (Gillette and Passi 1988; Shao et al. 1993). Fine soil particles (less than 5 μm) that can be transported over large distances are released when larger windblown sand grains impact on the soil surface, mobilizing the smaller particles through the process of saltation. Dust emissions occur preferentially in areas that contain fine and loose sediment. Prospero et al. (2002) find a good agreement of satellite-observed dust maxima on land and the location of topographic depressions, where fine sediment could accumulate either because such depressions were lakes during wetter climate periods or through riverine sediment input into those areas. Rough surfaces containing structural elements such as rocks and vegetation increase the threshold wind velocity needed for dust emission (Marticorena and Bergametti 1995) because the obstacles absorb wind energy. Most of these relevant processes have been incorporated into source parameterizations of models of the global dust cycle. Recently developed models include the use of an offline vegetation model to prescribe vegetation cover and therefore the extent of dust sources (Mahowald et al. 1999) and the occurrence of topographic depressions in dry, unvegetated areas as indicators of preferential sources for fine, loose sediment that can be easily deflated under strong wind conditions (Ginoux et al. 2001; Zender et al. 2003) and explicit simulation of a seasonal vegetation distribution to determine potential dust source regions (Tegen et al. 2002; Werner et al. 2002). Crusting of soil surfaces can reduce dust release from a source region. In addition, when fine soil material is lost by deflation, dust emission from a soil can decrease over time. Because of the lack of appropriate input data, those processes have not yet been implemented into global dust emission models. Surface disturbance as a consequence of cultivation in dry regions can increase dust emissions (e.g., during the Dust Bowl events in the United States in the 1930s and 1950s). Currently available estimates suggest that up to 50 percent of the global dust emissions could originate from such anthropogenically disturbed surfaces (Tegen and Fung 1995; Sokolik and Toon 1996). However,

newer research suggests that such sources probably contribute much less to the global dust burden than the percentage given in those estimates.

Dust injected at altitudes of several kilometers can be transported over thousands of kilometers by strong wind systems. Most prominently, dust emitted in the Sahara desert (where at least 50 percent of the global dust aerosol is produced) is transported across the North Atlantic to North and Central America and is found as far downwind as the Amazon Basin. Also, large amounts of Asian dust (Asian deserts produce about 10–25 percent of the global dust aerosol) are transported over the North Pacific toward the mid-Pacific islands and North America. State-of-the-art global dust models reproduce the large-scale regional and seasonal patterns (e.g., the Saharan dust plume over the North Atlantic, the transport of Asian dust to the Pacific, and the Arabian dust plume in the Northern Hemisphere summer) of the atmospheric dust loads reasonably well. However, none of them are able to realistically reproduce the magnitude of both Saharan and Asian dust export at the same time. These shortcomings reflect oversimplifications in the treatment of dust emissions in the current generation of models.

Dust is removed from the atmosphere by wet deposition (i.e., scavenging through precipitation in the water or ice phase) or through dry deposition, which is the gravitational settling and turbulent mixing toward the surface. Close to source regions, gravitational sedimentation is responsible for the major part of dust deposition, whereas wet deposition dominates the removal of far-traveled dust particles over remote ocean areas (Figure 7.1).

Although global models simulate the process of scavenging by dry sedimentation and subcloud removal, in-cloud removal of dust usually is neglected. Mostly, the process of subcloud removal is not simulated explicitly. Instead, a so-called scavenging ratio (i.e., the ratio of the dust concentration in rainwater and the dust concentration in air) is used to determine the efficiency of aerosol removal by rain.

Duce et al. (1991) report observed scavenging ratios between 200 and 1,000 for different regions of the world. Global models generally use a single scavenging ratio from within this range. Uncertainties in the choice of an appropriate scavenging ratio and failure to take into account changes in dust hygroscopicity increase the uncertainty of dust distributions in remote regions. Whether dust is removed from the atmosphere by dry or wet processes probably will affect the solubility of any trace elements transported with the dust. Unless they are removed by precipitation, the length of time that dust aerosol particles remain in the atmosphere depends on the deposition velocities of the different particle sizes (Figure 7.2). Atmospheric lifetimes of dust range from few hours for particles larger than 10 μm to 10–15 days for submicron particles (Ginoux et al. 2001).

Table 7.1 summarizes estimates of dust deposition into different ocean basins, derived from an extrapolation of dust concentration measurements combined with a simple model of dust deposition (Duce et al. 1991; Prospero 1996) and from results of recent global models of the dust cycle (Ginoux et al. 2001; Werner et al. 2002; Zender et al. 2003). For the global dust models, the total annual dust emission is given as well.

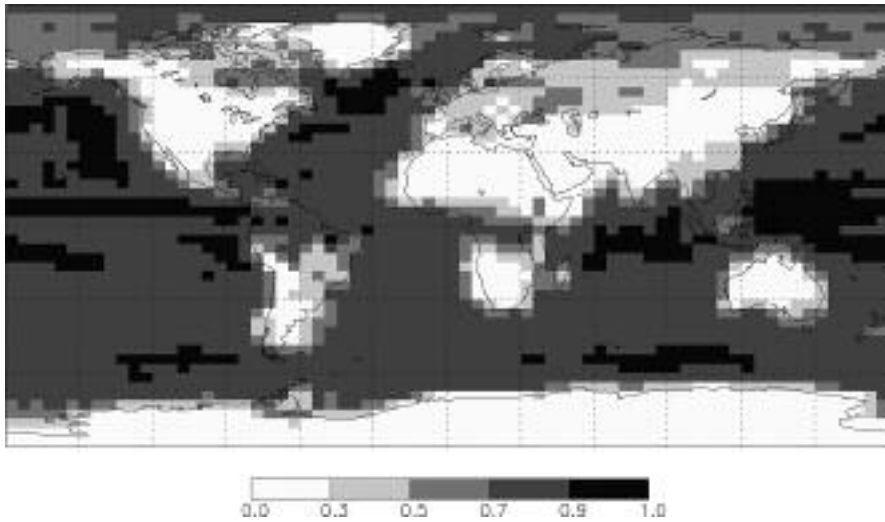


Figure 7.1. Ratio of wet to total deposition of dust aerosol, from the results by Tegen et al. (2002). Whereas over source regions more than 90% of dust is removed from the atmosphere by dry deposition, the main removal mechanism over remote ocean regions is wet deposition.

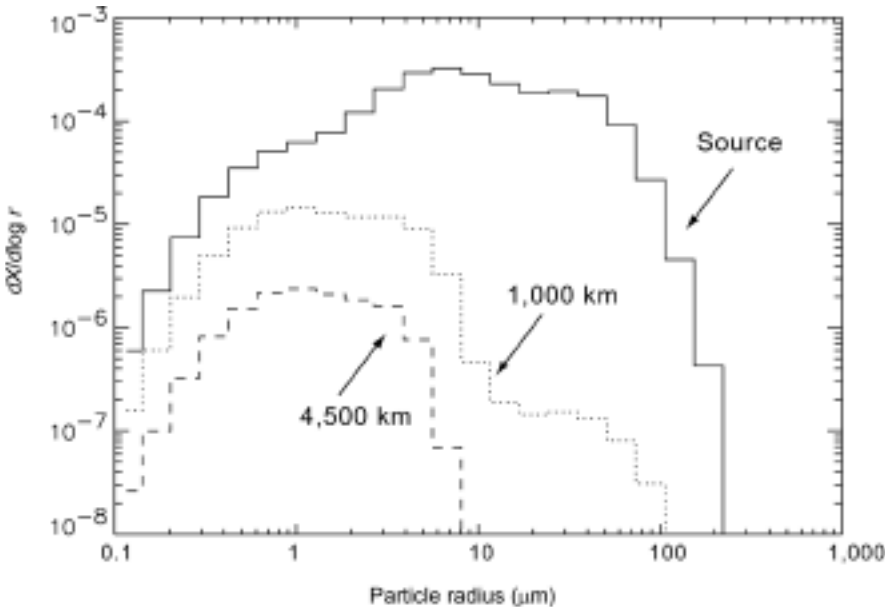


Figure 7.2. Modeled mass-size distribution of global dust particles (X = mass mixing ratio, in g dust/kg air) near the source (*solid line*), about 1,000 km downwind (*dotted line*), and about 4,500 km downwind of a source area (*dashed line*). Model results are from Tegen et al. (2002). Particles larger than 10 μ m are quickly removed from the atmosphere.

Table 7.1. Deposition of dust and Si into ocean basins extrapolated from observations or derived from global dust cycle models. Estimates of Si deposition and emission fluxes were computed assuming that dust contains 26.9 wt% Si.

	Extrapolated from Observations		Global Dust Cycle Models		
	Duce et al. 1991 (dust/Si, Mt/yr)	Prospero 1996 (dust/Si, Mt/yr)	Ginoux et al. 2001 (dust/Si, Mt/yr)	Werner et al. 2002 (dust/Si, Mt/yr)	Zender et al. 2003 (dust/Si, Mt/yr)
North Pacific	480/129	96/26	92/25	24/6	31/8
South Pacific	39/10	8/2	28/8	17/5	8/2
North Atlantic	220/59	220/59	184/49	201/54	178/48
South Atlantic	24/4	5/1	20/5	18/5	29/8
Indian Ocean	144/39	29/8	154/41	70/19	48/13
Global emissions	—	—	1,814/489	1,035/278	1,490/401

The estimates from Duce et al. (1991) and Prospero (1996) are based on extrapolation of the same measurements; however, Prospero computed his results using lower, more realistic scavenging ratios than Duce et al., except in the North Atlantic. The high scavenging ratio of 1,000 used by Duce et al. is the reason for their unusually high estimate of dust deposition in the North Pacific. The estimates of dust deposition in the North Atlantic agree well because a reasonable amount of data exists to constrain estimates of dust export from the Sahara.

Changes in Soil Aerosol in Past Climates

The atmospheric cycle of soil-derived dust aerosol is expected to respond to changes in global climate conditions, such as the overall intensity of winds that entrain dust into the air, and changes in the hydrological cycle that can affect the extent and dryness of source areas and the length of time that dust remains in the atmosphere. At the same time, dust can affect global and regional climate conditions in several ways, by affecting the radiative balance of the atmosphere or by providing micronutrient fertilizers to marine and terrestrial plants, thereby influencing the uptake of CO₂ in the biosphere and atmospheric CO₂ concentrations.

Dust deposition data from ice cores, marine sediments, and terrestrial sites provide information on dust in past climate periods (Kohfeld and Harrison 2001). The longest land records suggest that continuous atmospheric dust deposition has occurred on the Chinese loess plateau since about 7 million years ago (Ding et al. 1999). Significant efforts have been made to quantify changes in dust deposition through time. From these

records we know that during glacial periods, dust deposition was about 2–10 times higher than in interglacial periods (Figure 7.3).

The reason for the greater dust deposition probably is a combination of different factors, such as higher aridity, lower vegetation, greater availability of fine sediment, and higher surface wind speeds in glacial climates. Higher surface wind speeds could lift greater amounts of dust aerosols from surfaces than modern conditions, greater aridity would increase the areas with drier soils and lower vegetation, where soil deflation by wind is possible, and increase availability of fine sediments that could easily be picked up by surface winds. Atmospheric loadings of dust particles could also be higher under cold climate conditions as a consequence of the weakened hydrological cycle. Lower atmospheric water content leads to less precipitation, reducing the washout of atmospheric dust particles and thus leading to longer atmospheric lifetimes.

The best-documented period of high dust deposition in the Quaternary is the last glacial period (approximately 17,000–23,000 years ago). The last glacial maximum (LGM) has been a major focus of dust cycle modeling (e.g., Andersen et al. 1998; Mahowald et al. 1999; Lunt and Valdes 2002; Werner et al. 2002), partly because dust deposition rates were high and well documented for comparison (e.g., Kohfeld and Harrison 2001) and partly because the changes in boundary conditions (e.g., ice sheet extent, atmospheric composition) are well known. An LGM simulation using a full seasonal cycle of vegetation cover as a mask for dust emissions, as well as dried lake beds

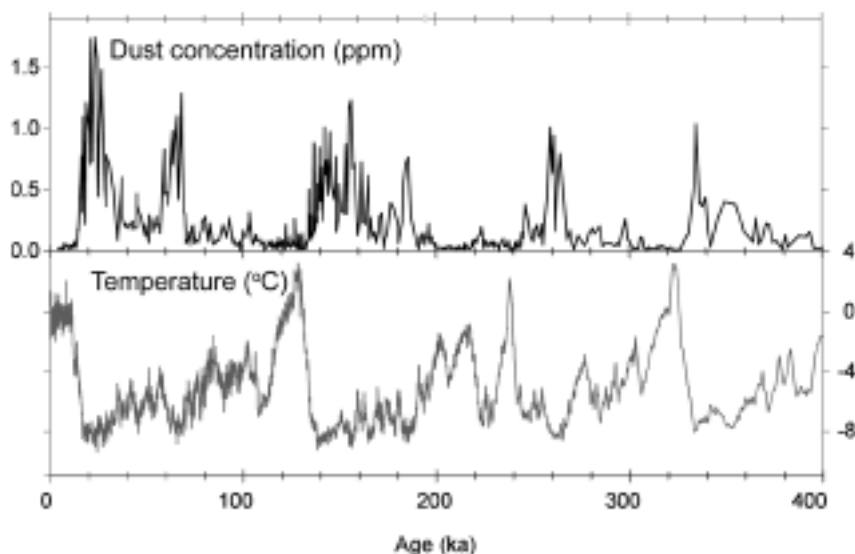


Figure 7.3. Records of dust concentrations and temperature, taken from the Vostok ice core, Vostok, Antarctica (Petit et al. 1999). Dust concentrations are much higher during glacial periods.

as preferential dust emission regions, shows a more moderate increase in dust emissions of a factor of two to three during the LGM, in agreement with the available marine sediment and ice core data (Werner et al. 2002). The potentially large effect of dust supplied by glacial outwash and the increase in dry lake beds at the LGM has not yet been evaluated by such global dust cycle models.

Properties of Soil Dust

The sizes of soil dust aerosol particles determine their atmospheric lifetimes and thus the distance that the particles can travel. Close to source regions the median dust particle radii are around 30–50 μm because particles in this size range can be lifted most effectively by surface winds. Because large dust particles fall out quickly, dust that has been transported away from the source has a median radius of about 1 μm (Duce 1995; see also Figure 7.2).

The mineral composition of dust particles reflects the mineralogy of the rocks at the earth's surface; some main constituents are quartz, feldspars, carbonates, and clays. Dust particles from different source regions have specific mineralogical composition and, as a consequence, different chemical composition. Claquin et al. (1999) summarized available information to compile global maps of mineral composition of soils in dry areas by relating soil mineralogy to soil types, for which global maps were available (Zobler 1986). They find that quartz makes up about 70 percent of the mass in the silt fraction of the soil (particles 2–50 μm diameter), and illite is the most abundant clay mineral (which contributes up to 50 percent regionally to the clay fraction). However, many uncertainties remain in that investigation. Additionally, it can be expected that during atmospheric transport the mineral composition of dust would change when the size distribution shifts toward smaller particles.

The Si content of different minerals can vary by a factor of approximately two. The average crustal Si content has been estimated to be 26.9 wt%. Whereas magmatites can contain 21–35 wt% Si and sediments 28–33 wt% (Wedepohl 1978), the Si content of clays is lower, on the order of 19–28 wt% (Newmann 1987).

Similar to the method used to estimate iron deposition by dust for investigations of the role of iron fertilization of the oceanic biosphere (e.g., Archer et al. 2000), we can obtain an estimate of the amount of Si deposited by dust aerosol by multiplying the deposited dust mass by the crustal abundance of 26.9 wt% Si. With this approximation we disregard the regional variability of the Si content in dust. However, as long as no more detailed information on the global mineral composition of atmospheric dust is available, this is a useful first-order estimate. With this assumption, global Si emissions of 250–500 Mt/yr associated with soil dust particles less than 10 μm (which can be transported over large distances) are computed with global dust cycle models (Table 7.1). Estimates of Si deposition into the ocean show that the highest amounts of dust Si are deposited into the tropical North Atlantic and the western North Pacific (Figure 7.4, Table 7.1).

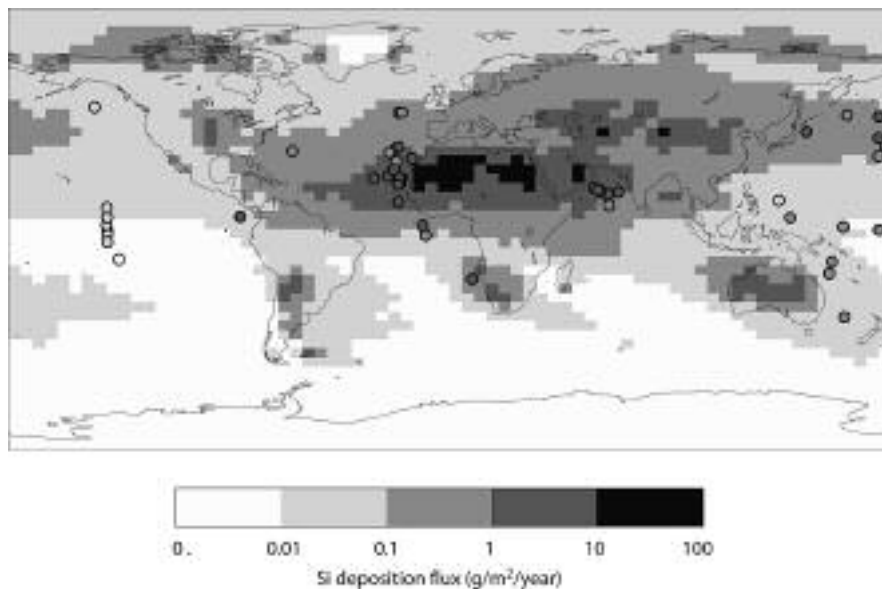


Figure 7.4. Deposition pattern of atmospheric Si, from global model results of dust deposition (*shaded areas*, Werner et al. 2002) and compiled sediment trap deposition fluxes (*symbols*, Kohfeld and Harrison 2001). Si content of 26.9% in the dust particles was applied globally.

Si deposition into the Southern Hemisphere ocean basins is low, with a total of probably less than 20 Mt/yr. If we assume that far-traveled dust consists mainly of clay particles, which have lower Si contents than the average crustal abundance, the deposition fluxes of Si may actually be lower than the estimates given in Table 7.1. However, this assumption would be based only on the median particle size of far-traveled dust, which may not reflect the mineral composition. The results in Table 7.1 show that the uncertainties associated with the particulate Si deposition into the different ocean regions are large; the estimates differ up to an order of magnitude for the North Pacific. This is a consequence of the large uncertainties in the methods that are used to estimate the global patterns of dust deposition.

Only a small fraction of the deposited Si dissolves in seawater. The percentage of particulate Si that dissolves in the water column depends on the mineral composition and size of the dust particles and is also uncertain. Whereas Tréguer et al. (1995) assume that 5–10 percent of the Si transported to the ocean surface by aeolian dust dissolves in the surface water, Guerzoni et al. (1999) report 1 percent solubility for Si in Saharan dust samples collected at Sardinia. Gehlen et al. (2003) assume a 3 percent solubility of aluminosilicates in seawater in their model investigation. As for the total Si content in dust

particles, the solubility is likely to change during transport, with the removal of large, insoluble quartz particles and possible chemical transformation during the cycling of dust particles through clouds.

Conclusion

The deposition of atmospheric Si into the global oceans can be estimated from oceanic dust deposition fluxes. According to the estimates given in Table 7.1, the annual eolian deposition of dust into the oceans ranges between approximately 300 and 900 Tg per year (i.e., between 80 and 240 Mt particulate Si). This flux is smaller than the estimate of the particulate riverine transport into the oceans. However, whereas the riverine flux of Si influences mostly the coastal regions, eolian dust can be deposited on remote ocean surfaces. The eolian oceanic Si input is equivalent to about 3–9 Tmol Si per year. Assuming solubilities of particulate Si between 1 and 10 percent, 0.03–1 Tmol of dissolved Si is added by deposited soil dust to the oceans each year. During glacial times, this value would have been two to three times higher. This is a small fraction of the total content of dissolved Si of approximately 10^5 Tmol (Tréguer et al. 1995), but regional variations may be of importance. For example, in the North Atlantic, up to 50 g/m²/year is deposited at some locations (Tegen et al. 2002), which would result in a regional flux of up to 0.05 mol Si at the locations of maximum dust deposition.

Given those uncertainties, it is unlikely that these estimates could be much improved with the implementation of a more detailed mineralogy to better represent the regional variability in Si content in the dust before the uncertainties in the estimates of dust deposition themselves can be reduced.

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