

Aerosol Impacts on Climate and Biogeochemistry

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Abstract

Aerosols are suspensions of solid and/or liquid particles in the atmosphere and modify atmospheric radiative fluxes and chemistry. Aerosols move mass from one part of the earth system to other parts of the earth system, thereby modifying biogeochemistry and the snow surface albedo. This paper reviews our understanding of the impacts of aerosols on climate through direct radiative changes, aerosol-cloud interactions (indirect effects), atmospheric chemistry, snow albedo, and land and ocean biogeochemistry. Aerosols play an important role in the preindustrial (natural) climate system and have been perturbed substantially over the anthropocene, often directly by human activity. The most important impacts of aerosols, in terms of climate forcing, are from the direct and indirect effects, with large uncertainties. Similarly large impacts of aerosols on land and ocean biogeochemistry have been estimated, but these have larger uncertainties.

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INTRODUCTION

Aerosols are solid or liquid particles suspended in the atmosphere. They represent a small fraction of atmospheric mass but have a disproportionately large impact on climate and biogeochemistry. These particles modify atmospheric radiation in both the short- and longwave as well as alter cloud properties (**Figure 1**) (1). Variations in radiation and clouds, in turn, impact climate (2). While in the atmosphere, particles take part with photochemical reactions (3) and impact public health (4). When aerosols fall onto the surface, they darken snow albedo (5) and modify both land (6, 7) and ocean biogeochemistry (**Figure 1**) (8, 9). Because of aerosols' importance to climate and biogeochemistry, as well as impacts on human health, they have been studied for many years; yet significant

uncertainties remain. Human activities, such as energy production, industrial production, and land-use management, have radically altered the emission of many aerosols over the past 160 years (**Figure 1**) (1, 2). Most anthropogenic aerosols tend to cool the climate (10), thus they potentially hid the twentieth-century warming that occurred as a result of increasing greenhouse gases (2). Because of this, the magnitude of anthropocene changes in aerosol climate forcing in the current climate is very important for understanding climate sensitivity and for projecting future climate (11).

In the first section of this paper, we introduce aerosols, their properties, and their transport and removal processes. In the second section, we present the main mechanisms through which both natural and anthropogenic aerosols interact with climate and biogeochemistry. This is in contrast to previous reviews, which focused either on natural or anthropogenic aerosols and rarely considered aerosol impacts on biogeochemistry (12–16). We then look at different sources of aerosols from land and oceans and assess their natural contribution to climate and biogeochemistry as well as how humans have changed, and are likely to change, their impacts on climate and biogeochemistry. Many of these mechanisms operate on long timescales and are poorly understood. Here, we highlight the uncertainties. Important impacts on land and ocean ecosystems from aerosols are reviewed and highlighted in the sections on biogeochemistry.

To compare the effects of aerosols on climate and biogeochemistry, we estimate their impacts on radiative forcing, based primarily on literature results, but when not available, we make simple estimates and include these in tables and figures. Because the literature does not usually provide information on the relative importance of various aerosols on different impacts, we use a recent model simulation to calculate relative importance and changes since preindustrial times (17) (more details in the Supplement; follow the **Supplemental Material link** from the Annual Reviews home

Aerosols: liquids or solids suspended in the atmosphere

Climate sensitivity: the change in equilibrium surface temperature in response to a doubling of carbon dioxide

Radiative forcing: the change in the radiation budget (both short and longwave) owing to the addition of a constituent, either natural or anthropogenic

page at <http://www.annualreviews.org>). We do not discuss the potentially important impact of aerosols on health.

Aerosol Characteristics

Aerosols vary widely in their impact on climate and biogeochemistry because of substantial variability in their size, composition, and location in time and space. Here, we describe in more detail some of the attributes of aerosols. The impact of aerosols on various atmospheric and biochemical processes are determined by their number, size, volume, and composition (**Figure 2**). One of the most important attributes of aerosols for both impacts and their atmospheric lifetime is the aerosol size (**Figure 2**). Aerosols vary in size from a diameter of 1 nm up to 100 μm (**Figure 2**). Particles with diameters less than 1 μm are fine aerosols and are divided into the Aitken mode (diameter $<0.1 \mu\text{m}$) and the accumulation mode (diameter $>0.1 \mu\text{m}$ and $<1.0 \mu\text{m}$), and particles greater than 1 μm comprise the coarse aerosols. Coarse aerosols are usually primary aerosols, meaning they were directly emitted and are often entrained into the atmosphere by the wind [e.g., desert dust or sea-salt particles (18)]. In contrast, many anthropogenic aerosols occur in the fine mode. A substantial portion of fine aerosols are not emitted directly but are formed in the atmosphere (sometimes directly after high-temperature combustion). These secondary aerosols are discussed in more detail below. Although there tends to be a higher number of aerosols in the small size fractions, they contribute less mass, integrated over all the particles, because of their small size (**Figure 2**). In terms of mass, coarse particles dominate (**Figure 2**).

The aerosol composition is also variable (**Table 1**). Some aerosols are solid crustal material, whereas others are in aqueous solution (e.g., sulfuric acid), and still others are a mixture of solids and liquids. Characteristics of color, hygroscopicity, and chemical composition strongly determine the impacts of aerosols on climate and biogeochemistry, as described

in the next section (**Figures 2 and 3**). In addition, aerosols can interact with gas phase chemistry through the following processes: (a) equilibrium partitioning between the gas and particle phase (e.g., nitrate uptake onto aerosols), (b) irreversible partitioning from the gas to the particle phase (e.g., formation of ammonium sulfate), (c) aerosol liquid phase reactions of partitioned gases [e.g., organic polymerization and secondary organic aerosol (SOA) formation], and (d) aerosol surface phase reactions for particles in the gas phase (18, 19). Chemical reactions within aerosols can change their properties substantially. For example, the ability of an aerosol to take up water (hygroscopicity) can be modified by adding organic compounds (20). More discussion of these reactions are described in sections below.

Finally, the aerosol spatial and temporal variability is important for climate impacts. Two identical aerosols, one close to the ground and the other at high altitude, have differing climate impacts and lifetimes. Thus, an important property of aerosols is their location in space and time (e.g., **Figure 3**). The impact of an aerosol on the atmosphere is restricted to its generally brief time of actual residence in the atmosphere, but once deposited on the surface, an aerosol's effects can endure much longer than its coherence as a particle (**Figure 2**).

Aerosol Transport and Lifetime

Aerosol sources (described in more detail below) are ubiquitous on and over the earth's surface, spanning deserts, oceans, forests, grasslands, and areas of human habitation. Aerosols are removed from the atmosphere by two processes: wet and dry deposition. Wet deposition processes are associated with precipitation and are very efficient at removing most aerosols (18). Aerosols may be incorporated within a forming cloud droplet, collide with a cloud droplet already formed within the cloud, or may be hit by falling rain or ice particles below the cloud itself. Many cloud droplets evaporate, allowing the aerosols inside them to remain in the atmosphere, though possibly with their

Lifetime: time that an aerosol particle resides in the atmosphere (e.g., if the lifetime is 10 days, 1/e of the aerosol is left after 10 days)

Primary aerosols: aerosols entrained into the atmosphere as solids or liquids

Secondary aerosols: aerosols formed in the atmosphere by chemical reaction or condensation of gases

SOA: secondary organic aerosol

Table 1

Aerosol type	Emissions (Tg/year) (15) ^a	Emissions (Tg/year) (17)	SW AOD
Organic aerosols	95	35	0.0025
Biomass burning	54		
Fossil fuel	4		
Biogenic	35	0.2	
Black carbon	10	7.7	0.05
Biomass burning	6		
Fossil fuel	4.5		
Secondary organic aerosol	28	36 ^b	0.006 ^b
Biogenic	25		
Anthropogenic	3.5		
Sulfates	200	170	0.048
Biogenic	57		
Volcanic	21		
Anthropogenic	122		
Nitrates	18	16	0.003
Industrial dust	100		
Sea salts	10,100	5,100	0.030
Sea salt size <1 μm	180	98	0.019
Sea salt size 1–10 μm	10,100	5,000	0.012
Desert dust	1,600	2,600	0.030
Desert dust size <1 μm	170	98	0.012
Desert dust size 1–2.5 μm	500	280	0.011
Desert dust size 2.5–10 μm	990	2,200	0.006
Primary biogenic particles		110	0.0002

^aAbbreviations: SW AOD, shortwave aerosol optical depth; Tg, teragram; a blank entry; no information is available.

^bModel output multiplied by three for use in this study to better match estimates.

properties modified (21). Outside of precipitating clouds, aerosols are removed by dry deposition, which includes both the turbulent collisions of the aerosols with the surface and the gravitational settling of the aerosols (18). Because of the role of gravitational settling, larger particles are removed by dry deposition much more quickly than smaller particles.

The average atmospheric lifetime of aerosol particles varies from <1 day for very large particles (>10 μm) to 2–4 weeks for smaller particles far from precipitating regions (18, 22, 23). There are many uncertainties in aerosol removal processes, resulting in large uncertainties in model assessments, not just in the lifetime of the aerosols but also in the

resulting aerosol distributions (22, 23). These uncertainties derive from our poor understanding of aerosol-cloud interactions as well as from the paucity of high-quality deposition and vertical distribution observations (18, 22, 23). In clouds, many processes usually occur simultaneously. For example, aerosols are involved in cloud droplet formation, can be chemically modified in the cloud droplet, and are released back into the atmosphere when the droplet evaporates. Deducing exactly which process happened where is difficult (24).

Although some particles are removed close to the source area, many aerosols are transported long distances before being removed, and here, we discuss the primary pathways

for long-range transport. Once emitted into the atmosphere, aerosols mix quickly into the atmospheric boundary layer (the lowest 1–3 km of the atmosphere, where mixing is strongest) and can be transported long distances. Air parcels stay on a constant buoyancy surface (called isentropes), unless they are heated or cooled (25). These surfaces of constant buoyancy tilt upward between the tropics and the high latitudes because of temperature gradients (**Figure 4a**). Thus, a parcel emitted in the tropical boundary layer can reach the stratosphere (>10 km high) in the high latitudes by moving along an isentropic surface. Most of the atmosphere is radiatively cooling slightly on average during nonstormy conditions; so, over time, parcels of air become slightly heavier and move downward. In addition, aerosols tend to move slightly downward by gravitational settling. But if the parcel encounters a storm system with clouds and precipitation, the parcel can experience large heating or cooling and move vertically very quickly, perhaps traveling from the boundary layer to 10 km high in less than 1 h (24). At the same time, the precipitation within the storm system can cause wet deposition. On the basis of atmospheric heating rates, the residence time for a parcel on a given isentropic surface varies between <1 day for the tropics to 3–6 days in midlatitudes (based on 75 percentile heating rates) (**Figure 4a**). If a parcel moves away from the pole, the tilting of the neutral buoyant surfaces directs the parcel downward vertically and vice versa. Horizontally, there tend to be particular regions in midlatitudes that have more storms (**Figure 4b**), called the storm tracks. Winds in the midlatitudes (30°–60°) tend to be from the west, and winds in the tropics and high latitudes (>60°) are less uniform. Tropical aerosols are more likely to encounter storms, resulting in vertical mixing or quick removal (**Figure 4b**). Transport across latitudes tends to occur less often than transport along a latitude (26). Thus, in the mid- and high latitudes, one can think of a parcel containing aerosols moving along isentropes, following the local winds, slowly cooling, and moving to a lower isentropic

surface until it encounters a storm system, at which time it can be quickly removed or vertically transported.

IMPACTS OF AEROSOLS

Direct Radiative Forcing

Aerosols have substantially different optical properties than atmospheric gases and thus alter the reflection, absorption, and transmission of radiation in air layers. This results in a direct modification to the radiative budget of the atmosphere and the surface below, known as the direct radiative effect, which is important for both weather and climate as long as the aerosols are in the atmosphere (**Figure 5**). The magnitude and sign of direct radiative forcing owing to an aerosol is a strong function of its optical properties, which dictate what fraction of the light aerosols absorb or reflect. The optical properties of an aerosol can be summarized in terms of specific extinction (the fraction of the light at a particular wavelength the aerosol absorbs or reflects per unit mass) and of the single-scattering albedo (fraction of intercepted light that is reflected) (27). The integral of an aerosol concentration profile weighted by the specific extinction is equivalent to the remote sensed quantity of aerosol optical depth (AOD) (28).

To a first approximation, the extinction per unit mass (specific extinction) caused by an aerosol is maximized when the wavelength of light is similar in size to the particle diameter. Because of the difference in wavelengths between radiation from the sun in the shortwave (SW) wavelengths (0.1–5 μm) and the longwave (LW) emissions from the earth (5–100 μm), we normally consider these wavelengths separately when considering the earth's radiation budget (28). Therefore, SW AOD is most heavily influenced by particles with diameters of ~ 0.1 –1 μm (mostly fine particles, which are heavily influenced by anthropogenic activity) (**Tables 2 and 3, Figure 6**) (10, 27, 29), whereas LW AOD is dominated by particles roughly an order of magnitude larger (which are more likely to be natural) and likely

Stratosphere: the atmospheric layer above the troposphere, characterized by its stratification

AOD: aerosol optical depth

SW: shortwave radiation (visible light)

LW: longwave radiation (infrared)

Table 2 Net impact of different aerosol types in preindustrial conditions with climate forcing (W/m^2) and uncertainty bounds^a

Aerosol	Shortwave	Longwave	Indirect effect	Atmospheric chemistry	Snow albedo	Land biogeochemistry	Ocean biogeochemistry	Total forcing
Soils:								
Desert dust	-0.55+/-0.1 ^b	+0.19+/-0.1 (10)			0.02 (74)	0.3+/-0.3	-0.17 to 0.50	
NO _x and NH ₄	-0.1 to -2 (142)							
Vegetation:								
Fires					0.03 to 0.11 ^c			
Primary biogenic	-0.005+/-0.01	+0.005+/-0.005						
Biogenic secondary organic			-0.05 to -0.20 Wm^{-2} (226)					
Oceans:								
Sea salts	-0.5 to -6.0 (13)	+0.2+/-0.2						
Dimethyl sulfide			-2.0 (227) ^d					
Volcanic:								
Dispersive								
Explosive	0 to 3					0 to 0.08		
Total natural aerosols		+0.2 to 0.45 (228, 229)	0 to 3.4	0.3+/-0.15	0.03 to 0.11 ^c	0 to 0.38	-0.17 to -0.50	

^aNumbers without citations are derived in the supplemental methodology section; a blank entry, no information is available.

^bFrom Reference 10 but only includes models with optical properties consistent with recent observations.

^cFrom References 2, 65, 74, 121, 230, and 231 but include all forms of black carbon.

^dIndirect plus direct impacts.



Supplemental Material

Table 3 Net impact of different aerosol types with anthropocene radiative forcing (W/m^2) and uncertainty bounds

Aerosol	Shortwave	Longwave	Indirect effect	Atmospheric chemistry	Snow albedo	Land biogeochemistry	Ocean biogeochemistry	Total forcing
Sulfate	-0.4 ± -0.2 (10)							
Fossil fuel black carbon	$+0.2 \pm -0.15$ (10)				0.03 to 0.11 ^a			
Organic carbon	-0.05 ± -0.05 (10)							
Secondary organic carbon	-0.25 to -0.06 (232, 233)		-0.46 to 0.84					
Desert dust	-0.07 ± -0.02 (10, 136) ^b		-0.06 ± -0.03 (136)			$+0.05 \pm -0.01$ (136)	-0.04 ± -0.01 (136)	-0.08 ± -0.06 (136)
NO_x and NH_4	-0.10 ± -0.10 (10)					-0.12 to -0.37 (95) ^c		
Vegetation:								
Fires	0.03 ± -0.12 (10)							
Dimethyl sulfide	-		-0.05 to -0.3 (234, 235) ^d					
Total anthropocene aerosol changes	-0.50 ± -0.40 (10, 236)	0.02 (228)	-0.7 (-0.3 to -1.8) (10, 236)	0.3 ± -0.15 ^e	0.03 to 0.11 ^a	0 to -0.24 (105–107) ^f	-0.04 ± -0.01	
Sum of effects						-0.07 to -0.62 W/m^2		

^aFrom References 2, 65, 74, 121, 230, and 231 but includes all black carbon, not just anthropogenic.

^bShortwave plus longwave impacts.

^cAssumes one-half of the nitrogen deposition is from aerosols.

^dIndirect plus direct impacts.

^eFrom physical climate forcings.

^fIncludes all aerosols (natural and anthropogenic).

^gFrom physical climate forcings.

Troposphere: the lower 10–16 km of the atmosphere, where humans live and most weather takes place

Anthropocene radiative forcing: the change in radiative forcings over the anthropocene (late 1800s to current climate), including human-caused and natural changes

CCN: cloud condensation nuclei

IN: ice nuclei

is determined by the volume of the aerosol (Figures 2 and 6) (27). In an average sense, SW AOD is typically greater than LW AOD. Direct radiative forcing, however, is not a simple function of AOD, but it is complicated by the location of clouds above and below the aerosols as well as by changes in the land surface characteristics. The size of a particle is extremely important in calculating aerosol forcing, but the composition of the aerosol is also important for two reasons. First, aerosol composition determines how much aerosols will grow in the presence of water, which can dominate the aerosol mass (20). Aerosol composition also controls the “color” or fraction of light absorbed compared to light scattered (Figure 2). Direct radiative forcing from aerosols occurs as long as the aerosols reside in the atmosphere: one day to four weeks in the troposphere, or years in the case of the stratosphere (Figure 5).

There are significant uncertainties in calculating the direct radiative forcing of aerosols (12); many of these have to do with uncertainties in the chemical composition of the aerosols (30) as well as in the nonsphericity of aerosols (31, 32). Moreover, how the different aerosol chemical species are mixed in the atmosphere matters: Whether they are mixed within each particle (internal mixture) or only externally mixed (each particle has a distinct composition) makes a large difference in the impact on direct radiative forcing (33). Thus, although direct radiative forcing by aerosols is probably one of the best known impacts of aerosols, there are substantial uncertainties (Tables 2 and 3). Note that SW AOD is not simply related to aerosol mass, because smaller particles, with less mass, have more impact on the SW AOD (Figures 3 and 6).

As stated above, calculating the radiative forcing from the AOD is not straightforward because of the presence of clouds and changing surface properties. However, for purposes of apportioning the radiative forcing, we can assume that it is related to AOD (Figure 6). For natural aerosols, radiative forcing is dominated in the SW and LW by desert dust and

sea-salt aerosols (Table 2, Figure 6), whereas for anthropocene radiative forcing, sulfate aerosols are the most important (Table 3, Figure 6).

Cloud-Aerosol Interactions: Indirect Effects

Aerosols also affect the global radiation balance indirectly through their interactions with clouds by acting as nucleation sites for cloud droplets and ice crystals, known in this capacity as cloud condensation nuclei (CCN) or ice nuclei (IN), respectively. Clouds and water vapor are the atmospheric constituents that most strongly interact with radiation, and thus, any change in clouds and cloud-water vapor cycling will significantly change the radiative budget of the globe and therefore the climate (1). Cloud droplets grow by the diffusion and condensation of water vapor onto CCN in a vapor pressure equilibrium relationship described by extensions of the Köhler theory (18). If the amount of water vapor in the environment exceeds a threshold value of supersaturation (SS), the droplet solution enters into an unstable growth regime and will rapidly grow to cloud droplet size (diameter ~ 4 to 30 μm). The critical SS of a would-be CCN depends on the initial particle size and also its hygroscopicity, a function of the particle composition. Hygroscopicity is a measure of a particle’s ability to reduce the equilibrium vapor pressure above a droplet when in solution, thus enhancing the diffusion of vapor toward the droplet. Therefore, aerosol number, size, and composition are all factors for determining the droplet number concentration of a cloud for a given environmental temperature and SS (18). However, SS fluctuates over small spatial scales and is itself dependent on the number of cloud droplets that form because cloud droplet formation depletes the available water vapor, introducing a negative feedback into the system.

In modeling studies in which the environmental conditions are strongly constrained, the fraction of aerosols that can act as CCN (the

activated fraction) increases if the aerosols are larger or have a larger hygroscopicity (**Supplemental Figure 1**), although nonequilibrium effects can be important (34). Recent observational and modeling studies have shown that size tends to be a more important factor than composition for determining CCN activity (35, 36), even though the relative importance of these parameters depends on aspects of the aerosol population and environment (37). Some studies have argued that the number of CCN (at a SS of 0.4%) is roughly proportional to the SW AOD (38) because the largest number of aerosols that are large enough to be CCN are in the fine aerosol mode.

Unlike cloud droplets, ice crystals will form in the atmosphere homogeneously (without a solid to freeze onto), but crystal formation can be facilitated at warmer temperatures by a particle nucleus. Laboratory experiments have shown that ice grows effectively by vapor deposition onto dust (39) and potentially onto some black carbon (BC) particles (40). There has also been interest in the IN activity of primary biogenic particles (41) and speculation that heavy metals may play a vital role in ice cloud formation (42). A recent study (43) proposed a parameterization for predicting IN activity based solely on the particle size, noting that large particles tend to act as IN regardless of composition (**Figure 3c**).

Cloud particle formation, both liquid and ice, occurs on a micrometer scale, and yet, as a major link between aerosols and cloud microphysics, it influences cloud-scale and larger-scale processes. These influences, known collectively as the indirect effects, are reviewed in great detail elsewhere (16, 38, 44). The first indirect effect results from changes in cloud droplet size (45). Increases in aerosols acting as CCN (such as by anthropogenic emissions) can lead to increases in cloud droplet number and, for a fixed liquid water content, a decrease in droplet size (46). Observations have shown that this relationship is nonlinear and that the cloud droplet number tends to be less sensitive to changes in aerosol number when aerosol concentrations are high (38). As the

median droplet size decreases, the cloud albedo increases, linking increased aerosols to a negative indirect radiative forcing. Anthropogenic aerosol forcings from the indirect effect are likely to be similar in magnitude to those from the direct effect, although current estimates are more uncertain (**Table 3**). The indirect effect caused by anthropogenic activity is dominated by sulfate aerosols (**Table 3, Figure 6**). Note that because the number rather than the mass of large particles is important for predicting ice crystal number, IN numbers tend to look like a combination of SW AOD and mass (**Figure 3**).

Changes in the droplet size distribution also impact cloud dynamics and precipitation formation, potentially suppressing precipitation (47), which can increase cloud lifetime, an impact known as the second indirect effect of aerosols (48). The overall impact of aerosol/cloud effects on the earth's radiation budget is still uncertain, particularly with regard to the second indirect effect (10). The timescale of this effect is similar to the residence time of aerosols in the atmosphere (**Figure 5**). The extent of the second indirect effect may vary substantially between cloud types and may be partially or wholly offset by compensatory cloud processes (30). Finally, the magnitude of the aerosol effects must be evaluated against the role of turbulence in determining cloud structure across multiple scales (49, 50).

Even more difficult than understanding the impact of anthropogenic aerosols on clouds would be envisioning a planet without aerosols or with a very different aerosol distribution to assist the condensation of supersaturated water vapor or freezing of supercooled liquid water (**Table 2**). Paleoclimate studies have argued that there were reduced aerosols to modify cloud properties during warm climates in order to explain the "warm" poles (51). Calculations here (described in the Supplemental text) suggest that the sea-salt and desert dust aerosols are likely to be globally the dominant aerosol interacting with clouds over much of the planet, with anthropogenic sulfate aerosols and carbonaceous aerosols [black and organic carbon (OC) also being important (**Figure 6**)],

BC: black carbon

OC: organic carbon

 Supplemental Material

Photolysis rate: the rate of disassociation of a molecule caused by absorption of radiation

but these results are sensitive to the CCN activation algorithm assumed (52).

Aerosol Impacts on Atmospheric Gases

Aerosols impact atmospheric gas concentrations through three main mechanisms. First, the chemistry of the atmosphere is largely photochemistry (18), so aerosol-induced changes in incoming solar radiation can have a large impact. Absorbing aerosols have the largest impact on photolysis rates; dust also plays the major role, but BC and carbonaceous aerosols are locally important (53, 54). Under biomass-burning plumes or downwind of intensely polluted regions, up to 30% to 40% reductions in photolysis rates are estimated (53, 55). The impacts of altered photolysis rates on ozone are reported to be relatively modest; global changes range from -4% to 5% (54) to an average of $+1$ ppbv (parts per billion by volume) (56). The impacts on oxidant (HO_x) concentrations are generally more significant; global reductions are generally less than 10%, but changes of 40% occur locally (54, 56). Changes in HO_x levels change the methane lifetime and thus its radiative forcing. The net effect of these changes is smaller than estimates from direct or indirect effects (Tables 2 and 3) (56).

Second, heterogeneous reactions that occur on the surface of aerosols have important consequences for both stratospheric and tropospheric chemistry (19, 57). Important reactions are associated with the uptake of nitrogen and hydrogen oxides onto aerosols (19, 53, 58). The net impact of heterogeneous reactions on aerosols are reductions in tropospheric ozone and related oxidants of 7% to 13.5%, resulting in a 20% to 45% reduction in ozone radiative forcing (58, 59) as well as a factor of two decrease in aerosol forcing because of reductions in aerosol mass (60). By contrast, reductions in atmospheric oxidants owing to heterogeneous chemistry are likely to increase the methane lifetime and its radiative forcing by 10% (58). The heterogeneous chemistry impact

on atmospheric oxidants is similar in size to the impacts from changes in photolysis rates (53).

In the lower stratosphere, heterogeneous reactions on liquid sulfuric acid aerosols are of particular importance. Nitrogen species tend to react on stratospheric aerosols, enhancing ozone destruction by a variety of complex pathways (57). Lower stratospheric ozone reductions have been found to be very large following the Mount Pinatubo eruption (57).

In a third mechanism, aerosols impact the concentration of atmospheric species by acting as a sink for them. This dramatically alters their cycling through the atmosphere, their wet and dry composition, and their chemical reactivity. Over half of atmospheric SO_2 is converted into sulfate (61), and half of emitted ammonia (NH_3) is converted into ammonium aerosol (62). Estimates suggest that between 13% and 29% of emitted volatile OC is deposited as aerosol (see the Section on Secondary Organic Aerosols, below) (63, 64).

For the atmospheric chemistry interactions described here, the surface area and composition of the aerosol are very important (Figure 2), and the reactions occur very quickly, although the resulting changes in chemistry could have lasting impacts on greenhouse gas concentrations (Figure 5).

Snow-Albedo Interactions

Very small concentrations of absorbing aerosols deposited onto snow and sea ice can reduce albedo because (a) the visible absorptivity of ice is extremely small and (b) multiple scattering within surface snow greatly increases the probability that a photon will encounter a non-ice particle (65, 66). Moreover, small changes in snow albedo can exert a large influence on climate by altering the timing of snow melt and triggering snow/ice-albedo feedback (5, 67, 68). When compared with other radiative forcing mechanisms, snow darkening drives an equilibrium temperature response several times greater than equal forcing by carbon

dioxide (CO₂) (5, 68). Most of this can be attributed to the fact that, by definition, all of the energy associated with the forcing is deposited directly in the cryosphere where changes exert a powerful positive feedback on the earth's radiation budget (69, 70). The efficacy of aerosol-induced changes of snow albedo also may arise from snow metamorphism feedback in which accelerated snow aging leads to both darker snow and greater albedo perturbation from absorbing impurities (71). Snow-albedo impacts by aerosols are dominated by particles with a large absorption in the visible short wavelengths; thus, composition and size are important (**Figure 2**). Most global aerosol/snow modeling studies to date have focused on BC which is highly absorbing in the SWs. Snow-albedo feedbacks through deposition are likely important on the seasonal timescale, but in glaciated regions, accumulated deposition over multiple years may also be important because of cycling with ice dynamics (**Figure 5**).

Recent observational and modeling studies suggest that one of the most vulnerable regions to snow darkening is the Himalaya/Tibetan Plateau/Hindu Kush region (72–77), owing to its proximity to large emission sources, high insolation, and importance as a source of freshwater to large populations. The global forcing and response both exhibit strong seasonal cycles and are largest in spring, when Northern Hemisphere snow cover and solar radiation are both high (74).

There is strong evidence that both BC and dust contribute to regional forcing through snow-albedo interactions (68, 74, 78–81). Another agent that can darken snow is volcanic ash (82, 83), although this effect is highly transitory, and no global modeling studies have been conducted to assess its importance.

During the dusty last glacial maximum (~20,000 years ago), the impact of dust interactions with snow and ice albedo was likely larger. Increased dust deposition onto snow has been invoked to explain snow-free conditions over much of Asia during summertime in the last glacial maximum (84).

Land Biogeochemistry Impacts

Land biogeochemistry refers to the cycling of nutrients, especially carbon (C), nitrogen, and phosphorus, on land in vegetation and soils. This cycling is key to sustaining the biological diversity of many types of land ecosystems and the many services these ecosystems provide to humans (85). Aerosols impact land biogeochemistry in two main ways: directly, by changing the gain and loss of nutrients through the atmosphere, and indirectly, by modifying climate. The direct biogeochemical effect is primarily dependent on the mass of the aerosol exchanged (which is usually much smaller than the nutrient fluxes through soils and water). Therefore, land biogeochemistry is affected preferentially by large particles, and aerosol composition is of vital importance (**Figure 2**). These effects tend to operate on timescales of tens to millions of years (**Figure 5**) (86, 87). Of the main nutrient cycles of C, nitrogen, and phosphorus (88), all three have some component in aerosol form. For C, CO₂ gas represents an easily available form of C, and the fraction of C deposited or removed from ecosystems by aerosols is not substantial [120 PgC (petagrams carbon) flux of CO₂ (89) compared with tens of TgC (teragrams carbon) organic aerosols (**Table 1**)]. Direct deposition of nitrogen onto the surface is being substantially modified by humans primarily because of nitrogen generated by combustion and the application of artificial nitrogen fertilizer to soils (**Figure 6**) (90). Because many ecosystems are nitrogen limited (91), this results in an increase in productivity and thus a sequestering of C of between 0.24 and 0.7 PgC/year (**Table 3**) (87, 92–95).

Phosphorus limitation is common in tropical forest (96) and savannah ecosystems (97). Inputs from long-range transported desert dust aerosols have been argued to be important for phosphorus cycling on long timescales in Hawaii (86) and the Amazon (6). Globally, the deposition of phosphorus may be important in many ecosystems on a 1,000- to 100,000-year time frame in maintaining soil fertility (98) and may be responsible for long-term

nitrogen availability because of linkages between nitrogen fixation and phosphorus (99). Desert dust aerosols dominate the deposition of phosphorus, but primary biogenic particles and biomass-burning aerosols also contain phosphorus, whereas sea spray and volcanoes supply trace amounts (100). The impact of natural phosphorus deposition onto land ecosystems and the resulting sequestration of C have not been previously estimated, but they could be important because of large C storage and biodiversity in tropical forests (**Table 3**) (101), which may be phosphorus limited without deposition from aerosols.

Aerosol deposition can add nutrients, but it also can add toxins or acidity that the ecosystem is not well buffered against. Acid rain is a well-documented phenomenon that is primarily caused by the deposition of anthropogenic sulfate and nitrate aerosols and results in the leaching of micronutrients such as calcium and magnesium from the soils (102–104). There are no published estimates of the impact of acid rain on C uptake, but it could be large (**Table 3**).

The direct effects of aerosol on the sunlight, temperature, and precipitation conditions

of terrestrial ecosystems likely impact land biogeochemistry. These impacts have not been well documented. Changes in anthropogenic aerosols have been shown to decrease direct radiation and increase diffuse radiation (radiation that has been scattered at least once and thus comes in from a different direction), which tends to increase productivity (105). The net effect of anthropogenic aerosols in coupled-C-climate simulations has been assessed in two studies (106, 107). Most of the impacts of aerosols on the C cycle in these simulations are due to the cooling of the climate, although there can be significant regional-scale changes in climate.

Overall, both natural and anthropogenic aerosol interactions with land biogeochemistry are relatively poorly understood, but these interactions have the potential to be as important as the better-understood direct and indirect radiative forcing of aerosols (**Tables 3 and 4**).

Ocean Biogeochemistry Impacts

Ocean biogeochemistry is the interaction of the ocean circulation, biota, and chemistry and

Table 4 Sources of different types of aerosols in 1990 (17)

Sources of aerosols	Black carbon (Gg/year)	Amonia (Gg/year)	Nitrogen oxides (Gg NO ₂ /year)	Organic carbon (Gg/year)	Sulfur dioxide (Gg/year)
Energy sector and distribution	43	28	20,000	300	6,000
Industry	1,200	130	17,000	1,700	36,000
Land transport	1,100	460	37,000	1,100	5,500
Air transport	4	0	2,000	0	0
Shipping	95	0	13,000	100	8,000
Residential	2,300	480	9,000	8,000	14,000
Solvents	0	0	0	0	0
Agricultural	0	41,000	7,800	0	0
Agricultural waste burning	150	580	640	720	200
Waste	29	0	270	41	49
Biomass burning	2,800	11,000	5,600	23,000	3,800
Soils	0	0	15,000	0	0
Volcanoes	0	0	0	0	18
Oceans	0	8,000	0	0	30,000
Other natural	0	2,300	0	0	0
Lightning	0	0	13,000	0	0

is important both for the diverse species supported (e.g., corals and whales) as well as for the ecosystem services provided (85). Substantially more C is held within the oceans than either on land or in the atmosphere (89), and the C is entrained into the deep ocean either through physical processes, such as the ocean overturning circulation, or through the biological pump, whereby ocean productivity moves C from the upper ocean to the deep ocean (89). As on land, aerosol impacts on ocean biogeochemistry can be due to either direct deposition of nutrients or the effects of aerosol on the physical forcing of the ocean by the atmosphere. The most important nutrients delivered by the atmosphere to the ocean are thought to be nitrogen, phosphorus, and iron (**Table 3**) (100, 108–110), although deposition of other species may also be important (111). For biogeochemical impacts, the mass and type of aerosol are important (**Figure 2**).

The most important nutrient for ocean biogeochemistry appears to be iron because it is an essential micronutrient for many biota (8, 112). The potentially large iron requirements in nitrogen-fixing organisms make iron availability a strong control on nitrogen fixation (conversion of molecular nitrogen gas into the bioavailable fixed-nitrogen species) (113, 114). Thus, additions of iron to the ocean are more important than the large increases in nitrogen deposition for changing the nitrogen cycle of the ocean (108, 114). Some iron may be chemically bound too tightly for ocean biota to access (115), and although desert dust dominates the iron deposition to the oceans, human activities, especially the added acidity in the atmosphere from pollution, may be doubling the bioavailable iron (116).

Aerosol deposition to the ocean not only adds important nutrients, but also contributes to an increase in toxic metals (117) or acidity (118). The negative impact of the deposition has not been assessed in great detail, and it is unclear whether it is globally important, although it is likely that these effects can be important locally. The physical climate forcing of aerosols and anthropogenic aerosols and

their impact on ocean biogeochemistry have not been assessed, although significant changes were reported in one model study (107).

Climate Effects

The globally averaged surface temperature effects of different aerosols tend to be proportional to the radiative forcing (119), with the exception of snow-albedo feedbacks, which tend to be much larger than their radiative forcing would indicate, as discussed above. The details of how aerosols affect the regional climate, precipitation, or cloud properties depend on whether they are absorbing or scattering, the net top of atmosphere balance of radiation, and what is happening at the surface (2, 13, 120, 121).

NET EFFECT OF AEROSOLS ON CLIMATE AND BIOGEOCHEMISTRY

Different sources emit aerosols of different chemical composition, size, and spatial and temporal distribution. Here, we summarize the net effect of aerosols from different sources and what is known and unknown about them on the basis of available literature and simple calculations (**Table 2** and **3**; described in more detail in the Supplemental text). The climate impacts of different aerosols are quite nonlinear (120). For the sake of simplicity, we neglect discussion of these nonlinearities in order to attribute various impacts to different sources (**Figure 6**). In addition, we look at the relative increase in aerosol impacts caused by humans (**Figure 6**) and apportion the type of aerosol responsible for these changes. Note that some sources of aerosols produce aerosols of different compositions (**Table 4**).

Aerosols from Land Sources

Desert dust. Desert dust aerosols are mineral particles suspended in the atmosphere, and these particles are entrained in regions with little vegetative cover, dry soils, and strong

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winds. North Africa dominates the emission of desert dust, providing about 50% of global atmospheric dust (122). Owing to the mechanical nature of its emissions, there are strong seasonal and diurnal cycles in desert dust sources and transport pathways (123). Because of the large variability in sources and transport pathways, as well as uncertainty in the size distribution, estimates for the amount of mass in the atmosphere vary substantially (124). Desert dust particles consist of various minerals of different shapes, and thus there is considerable variability in their potential radiative properties (31, 125). While highly variable, desert dust aerosols are one of the most important aerosols for mass and radiative forcing (**Figure 6**, **Tables 1**, **2**, and **3**). Moreover, because crustal materials contain 3.5% iron and about 700 ppm phosphorus, desert dust is one of the most important aerosols for the transfer of iron and phosphorus into the atmosphere (**Figure 6**) (100, 116).

Desert dust aerosols are well archived in paleorecords in ice, marine sediments, terrestrial sediments (126), and even corals (127) and lake records (128), providing more information about the climate variability in desert dust than any other aerosol species. These records suggest that atmospheric desert dust concentrations are very sensitive to climate shifts, becoming two to four times larger during glacial versus interglacial time periods (129) or within glacial periods recorded at many sites (130, 131). In situ concentration observations suggest that dust concentrations can change regionally by a factor of four (132). In addition, humans can modify dust sources by land use (133, 134) or by fertilizing terrestrial plants with higher CO₂ (135). Recent estimates based on paleoclimate observations and modeling suggest a doubling of dust over the twentieth century (136). Future projections show wide variability in these estimates (116).

Nitrogen emissions from soils and agriculture. There are two main forms of nitrogen aerosols in the atmosphere: nitrates and ammonium. Both of these are secondary aerosols, formed from nitrogen oxides (NO) or

NH₃ emissions, respectively (**Tables 1** and **4**, **Figure 6**). There remain large uncertainties in emissions of these species from soils because of their dispersed nature (137–139). NO production in soil is predominately due to the bacterial processes of nitrification and denitrification (137). The emissions are controlled by soil conditions, including soil water, oxygen, organic matter, pH, and temperature. Emissions of NO are also regulated by the state and amount of nitrogen in the soil, which depend on the application mode and rate of artificial fertilizer and manure use, biological nitrogen fixation, and deposition of atmospheric nitrogen. Increased fertilizer use appears to result in significant increases in NO emissions (**Figure 6**, **Tables 2** and **3**) (140).

NH₃ is largely a product of soils and agriculture (**Table 4**). Between 10% and 30% of the fertilizer nitrogen may be volatilized as NH₃; the nitrogen loss is dependent on the type of fertilizer, properties of the soil, meteorological conditions, and agricultural management practices (138, 139).

Once in the atmosphere, sulfates and NH₃ partition between the aerosol and gas phase. Unneutralized nitrate aerosol can be found in regions with cold temperatures (e.g., the upper troposphere), but nitrate aerosol is generally found in conjunction with ammonium (141). Nitrate also partitions onto dust but with little direct radiative effect (59). Ammonium nitrate concentrations are sensitive to the amount of sulfate in the atmosphere because ammonium preferentially partitions into ammonium sulfate aerosol. Although the global nitrate to sulfate ratio is generally low [on the order of 10% (141)], nitrate concentrations can be particularly high at the surface, exceeding sulfate regionally in the U.S. Midwest, Eastern Asia, and Eurasia. This suggests nitrate can have important consequences for air quality (142). Nitrate forcings may increase considerably in the future (-1.28 W m^{-2}) as the ratio of sulfur to nitrogen emissions changes and as ammonium nitrate replaces ammonium sulfate (143). NH₃ changes the hygroscopicity of sulfate as well as its refractive index and can change its

climate forcing by 25% (141). Assumptions about internal and external aerosol mixtures have been found to change the sign and magnitude of anthropogenic aerosol forcing by almost up to 1 W m^{-2} (141). Ammonium and nitrate aerosols are also important for land and ocean biogeochemistry because they carry nitrogen (**Figure 5, Tables 2 and 3**) (95, 108).

Other aerosol precursors are also emitted from soils (and plants) in small quantities, for example, organic carbonyl sulfide and dimethyl sulfide (DMS), described in more detail under Aerosols from Ocean Sources, below.

Biomass burning. The burning of vegetation by natural wildfires or humans has enormous impacts on climate and biogeochemistry (144). The best estimates of biomass burning in the current climate rely on a combination of satellite and models (17, 145). At present, different biomass-burning emission inventories agree within 50% to 80% on a global scale, depending on the year and season. The large differences are mainly because of differences in the estimates of burned areas and amount of biomass burned (146). Biomass-burning emissions follow a strong seasonal cycle, unlike anthropogenic aerosol emissions. Biomass-burning inventories that cover multi-year time spans all show very high interannual variability of the emissions. For example, for the time period 1997 to 2009, biomass burning is estimated to emit 2.1 Tg BC/year on average, with maximum emissions of 2.9 Tg BC/year in 1998 and lowest emissions in 2009 with 1.6 Tg BC/year (147).

The chemical composition of biomass-burning aerosols depends on the fuel type and combustion conditions (148, 149) and consists mainly of BC and OC. Major biomass-burning events can emit aerosols into the free troposphere (150, 151), which has consequences for the lifetime and long-range transport of biomass-burning aerosols, and, as a result, the net radiative forcing of biomass-burning aerosol. Biomass-burning aerosols are thought to contain both phosphorus and soluble iron, making them important for land and ocean

biogeochemistry (**Figure 6, Tables 2 and 3**) (100, 116, 152, 153).

Preindustrial biomass-burning emissions are highly uncertain. It is often assumed that present-day biomass-burning emissions are highly impacted by anthropogenic activity and that preindustrial levels were substantially lower. Although several previous studies assumed that preindustrial levels of biomass-burning emissions relative to present-day sources ranged between 10% and 50% (17, 109), more recent estimates based on historical records (154, 155) and modeling studies (156, 157) suggest that preindustrial levels of biomass-burning emissions were comparable to present-day emissions or even slightly higher. Consequently, the anthropogenic radiative forcing caused by biomass-burning emissions is very uncertain (**Table 3**).

Primary biogenic particles. Primary biogenic particles are biotic-derived particles $<10 \mu\text{m}$ in size that are emitted directly into the atmosphere and can be parts of plants, spores, pollens, bacteria, or other biotic material. The global emission and distribution of primary biogenic particles are very uncertain with estimates between 100 and 1,000 Tg/year (100, 158) with some attempts to separate out the different components of primary biogenic aerosols (159, 160). These aerosols likely interact with radiation in the SWs and LWs (because of their size) and with clouds (161–163), but our estimates suggest these impacts are small (**Table 2, Figure 6**). Primary biogenic aerosols are not thought to strongly impact CCN or IN populations because they exist in small numbers (**Figure 6**) (164), but they may play a large role in biogeochemical cycles because they can contain significant phosphorus content (**Figure 6**) (100).

The response of primary biogenic particles to changes in climate or human land use is not known, but it could be strong. Emission of primary biogenic particles appears to be sensitive to atmospheric moisture (161) and aboveground biomass (160). Therefore anthropogenic changes in climate and shifts in plant

species are likely to modify primary biogenic particles, although this has yet to be estimated (14).

Aerosols from Ocean Sources

Sea spray. Earth's oceans emit both organic and inorganic aerosols, collectively known as sea-spray aerosol, directly into the atmosphere. Sea-spray aerosol is produced mechanically as a result of wind stress on the ocean surface (165). As such, the number and mass emission flux of marine aerosols have been observed to increase with increasing surface wind speed (166) and temperature (167). Source functions for sea-spray aerosol typically include multiple size modes that span a large diameter range from 20 nm up to 100 μm , with a greater number of supermicrometer particles relative to other species (165). Therefore, although aerosols in marine air masses are typically lower in number compared to that of continental air masses, they tend to have greater total mass (volume) (Figure 2).

The wind-driven mechanism of sea-spray aerosol emission leads to the production of internal mixtures of the inorganic and organic compounds. The largest inorganic species emitted from the ocean in terms of mass is sodium chloride, or sea salt. Organic compounds at the ocean surface that are accessible for emission as aerosol are produced by marine biological activity (168–170). Trace amounts of phosphorus are also emitted in sea spray (Figure 6) (100). Both organic aerosol and phosphorus emissions are likely related to the concentration of phytoplankton in the ocean surface layer (100, 171). Studies of the quantity of organic mass in sea-spray aerosol relative to sea salt are few and vary in their conclusions (reviewed in Reference 172), but some measurements, notably those in References 169 and 173, show a substantial organic mass fraction. These studies observed a sea-spray aerosol distribution in which the smaller particles (generally with diameter $<0.5 \mu\text{m}$) consisted mainly of organic matter, and the large (especially supermicrometer) particles were almost

entirely sea salt. Sea salts are likely to be one of the most important natural aerosols for direct and indirect radiative forcing, although uncertainties remain surprisingly large for such an important species (Figure 6; Table 2).

Evidence from ice cores suggests that sea-salt aerosols vary dramatically between glacial and interglacial climates (174); however, the interpretation of these records is confounded by the existence of aerosols that are high in sea-salt content but are derived from the formation of new sea ice (175). Some model simulations suggest that there can be high sensitivity of sea salts to variations in wind speeds, whereas others see little response (29, 176, 177). Since sea salts represent a significant source of natural aerosols, uncertainties in the radiative forcing and indirect effect of sea salts, as well as potential human impacts on sea salts, are important to resolve (Tables 3 and 4)

Ocean sources of sulfides. Dimethyl sulfide (DMS or CH_3SCH_3) is produced by phytoplankton and is the most abundant chemical form in which the ocean releases gaseous sulfur. In the atmosphere, DMS is oxidized to SO_2 . SO_2 itself reacts with OH to produce SO_4^{2-} , which can nucleate to form sulfate aerosol particles. Because phytoplankton respond to climate change, DMS emissions are sensitive to climate. Changes in DMS emissions, in turn, affect the abundance of sulfate aerosols, and hence cloud properties and climate. This DMS-climate feedback cycle is often cited as the CLAW hypothesis, named after authors who first proposed it (178). It was initially proposed to be a negative feedback in the earth system, which stabilizes the earth's climate against external perturbations. However, several modeling studies revealed that the interplay between climate and the marine production of DMS is very complex and strongly depends on the region and season (179–182). The direction and the magnitude of the feedback on a global scale still remains uncertain, despite more than 20 years of research (14). Note that there are minor emissions of DMS from soils and vegetation (183). DMS is most important for

its influence on aerosols (**Figure 6**, **Tables 2** and **3**).

The ocean (as well as some soils and marshes) also emits organic carbonyl sulfide, which is a relatively stable compound, until it reaches the stratosphere, where it reacts to form sulfate aerosol (184). Approximately 50% of the stratospheric aerosol derives from OCS (185). This stratospheric aerosol plays a role in cooling the atmosphere and in reducing ozone, as described in the Aerosol Impacts on Atmospheric Gases section, although a magnitude of this has not been estimated.

Lightning

Lightning is another natural process that acts as a source of atmospheric NO (precursors to nitrate aerosols) (**Table 4**) (186). Even though it is a relatively small source of NO, lightning directly provides NO to the upper troposphere, where chemical lifetimes are much longer than at the surface, which enhances the importance of lightning to nitrate aerosol formation. The change in lightning NO with climate is also highly uncertain but could be large (187).

Volcanoes

Explosive volcanic eruptions fill the atmosphere with ash and gases, most of which fall close to the eruption. However, sulfur dioxide gas emitted by volcanoes, sometimes directly into the stratosphere (>10 km high), is converted to sulfate aerosols; these have a residence time in the stratosphere of 1–2 years and produce strong radiative forcing (2) and a corresponding response in global surface temperatures of a few tenths of a degree for some strong eruptions (**Table 2**) (188). Changes in the number of volcanic eruptions have contributed to a change in radiative forcing over the anthropocene (10).

Stratospheric aerosols impact ozone chemistry in the lower stratosphere through aerosol-chemistry interactions and by cooling the atmosphere (see atmospheric chemistry section, above, and Reference 57). Volcanic explosions dominate the stratospheric aerosol signal in

available observations because it takes approximately eight years for volcanogenic sulfate aerosols to be fully removed from the stratosphere (57). After the Mount Pinatubo eruption, stratospheric ozone was reduced by ~6% in the Arctic, thus impacting radiative forcing (57).

In addition, there is an appreciable reduction in atmospheric CO₂ accumulation after an explosion owing to changes in land and/or ocean biogeochemistry (183). The changes could be the result of cooler and wetter tropics (189), increases in diffuse radiation (105), and/or fertilization of the ocean by iron or phosphorus (**Table 2**) (190–192).

Even when not erupting, small amounts of sulfur dioxide gas leak from volcanoes into the troposphere and form sulfate aerosol (**Table 4**) (193). Volcanoes are also likely to emit small quantities of nutrients such as phosphorus (100).

Secondary Organic Aerosols

SOAs derive from both natural and anthropogenic sources. Because of our poor understanding of these aerosols, we discuss both natural and anthropogenic secondary organic aerosols together in one section. Biogenic sources of volatile organic compounds (BVOCs), such as isoprene, monoterpenes, and sesquiterpenes, dominate the SOA budget simulated in models, based on laboratory yields (194), and are compared to anthropogenic SOA precursors (aromatics, alkanes, and alkenes). However, the oxidation of intermediate volatility compounds, which are traditionally not included in global models, may be an important additional source of SOA mass (195). The climate impact of SOAs is difficult to assess given our poor understanding of the formation and removal processes that control the present-day atmospheric loading of these particles (**Table 2** and **3**, **Figure 6**, Supplemental discussion) (196).

Estimating the response of the biosphere to changes in climate and atmospheric composition and the resulting feedback on SOA

BVOC: biogenic volatile organic compound

is a challenge. Global vegetation density and plant phenology are the first-order predictors of BVOC emissions. Both natural and anthropogenic land-use modifications, such as CO₂ fertilization (197, 198), ozone plant damage (199), deforestation, and plantation development, modulate the BVOC source intensity and distribution around the globe. Furthermore, plant productivity and trace gas emissions respond to changes in climate variables, such as temperature, light, soil moisture, and other environmental stresses (200). Finally, BVOC emissions have recently been shown to respond to changes in the chemical composition of the atmosphere, including CO₂ (201–204) and ozone concentrations (205–209).

Attempts to quantify the anthropogenic radiative forcing of SOA are complicated by the blurred lines between natural and anthropogenic origin that result from the interaction of emitted organics with the ambient atmosphere, where oxidant levels, particle acidity, and nitrogen oxide levels can significantly influence aerosol yields (210–213). Furthermore, the nonlinearities of organic species gas/particle partitioning suggest that, for example, SOA formation from biogenic sources can be significantly enhanced in a region of high anthropogenic organic aerosol loading. Although radiocarbon measurements indicate that much of the C in organic aerosol is modern (i.e., from nonfossil sources) (214, 215), the anthropogenic influence on the formation of these particles may be considerable. Recent evidence that SOAs can exist as an amorphous solid may also have implications for the size, scattering properties, and the direct radiative effect of these particles (216).

The indirect effect of SOAs depends largely on the relative importance of condensation of organics on existing particles versus nucleation of new particles. Laboratory SOA is not very hygroscopic (**Supplemental Figure 1**) (217, 218). Thus, small, freshly nucleated SOAs are not likely to be activated as CCN unless exposed to high in-cloud SSs (219). However, in clean remote regions, SOAs may be an important source of new particles, which may

grow into the more CCN-active accumulation mode (220, 221). Similarly, the condensation of organics can help grow inorganic particles to more CCN-active sizes. Finally, laboratory-generated SOAs do not appear to serve as a source of heterogeneous IN in the atmosphere (222), except perhaps owing to secondary phase transitions at low temperatures (223); however, the role of organic coatings on particle ice nucleation ability have not yet been fully explored.

Other Anthropogenic Aerosols

In addition to the varied natural sources of aerosols, humans directly emit aerosols, generate their precursors by combustion to create energy or materials, and influence natural sources of aerosols by manipulating the land surface. BC and OC aerosols are emitted predominately from biomass burning, industry, and land transportation (**Table 4**). NH₃ comes predominately from agriculture (as described above); NO (precursors for nitrates) are emitted largely from land transport; and sulfur dioxide emissions come primarily from energy production (**Table 4**). There have been significant changes in emissions of these constituents since preindustrial times (**Figure 6**, contrast **Table 4** with **Supplemental Table 2**), with the exception of biomass burning (as discussed above). In addition, humans can indirectly perturb both desert dust and biogenic primary and secondary emissions (discussed above).

The sources of directly emitted anthropogenic aerosols are often easier to estimate than emissions of natural aerosols (e.g., sea salts or desert dust), partly because they are proportional to activities that humans keep track of (coal burned, for example), in contrast to the lack of observation that occurs over the open ocean or desert. There are multiple estimates of past emissions of these compounds, showing the uncertainty in these estimates (17).

The impact of anthropogenic aerosols on direct and indirect radiative forcing has been intensely studied and reviewed over the past few years (10, 12, 224), because studies suggest that these forcings are of the same order as the

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warming of CO₂ and opposite in sign (**Table 3**) (10). In addition to these important effects, it is likely that anthropogenic aerosols impact atmospheric chemistry and snow albedo (**Table 3**), although the magnitude of these appears to be an order of magnitude smaller than the direct or indirect effects of aerosols (**Table 3**). However, the impact of aerosols onto biogeochemistry, especially on land, is estimated here to be potentially as large as the direct and the indirect effect of aerosols, especially for sulfate and nitrogen-containing aerosols, with very large uncertainty (**Table 3**).

SUMMARY/CONCLUSIONS

Aerosols are liquid or solid particles suspended in the atmosphere. Aerosols can be entrained into the atmosphere (through wind, for example) or formed in the atmosphere from chemical reactions. They vary in size from 1 nm to 100 μm and, especially small particles (**Figure 2**), can be transported long distances from their sources. Aerosols can be composed of inorganic salts, minerals, biotic materials, soot, or organic materials, as well as a combination of these. They follow prevailing winds along neutrally buoyant atmospheric layers until they encounter storm systems, where they are removed or transported vertically (**Figure 3**).

Aerosol impacts on climate and biogeochemistry are as diverse as aerosols themselves (**Figure 1**). Some processes, such as the direct radiative forcing of aerosols (interacting with incoming and outgoing radiation), aerosol-cloud interactions, and atmospheric chemistry, occur on short timescales and last as long as the aerosols are in the atmosphere (1 day to 4 weeks) (**Figure 5**). Once aerosols are deposited onto the earth's surface, these aerosols can interact with snow albedo to darken surfaces or can

provide a source of nutrients to land or ocean biogeochemistry for thousands of years (**Figure 5**). In addition, the deposition of aerosols on the land or ocean can carry acids or heavy metals that harm the biota. Thus, aerosols have strong interactions with climate and biogeochemistry (**Tables 2 and 3**).

During preindustrial times, the largest sources of aerosols were desert dust and sea salts, and these dominated the impacts on climate through direct and indirect forcing (**Tables 1 and 2, Figure 6**). However, ocean biota sources of aerosols, especially DMS, although small, may have provided important CCN as well (**Table 2, Figure 6**). Desert dust aerosols also may have been important for redistributing nutrients, such as phosphorus and iron, whereas fire aerosols altered snow albedo and carried nutrients (**Figure 6**). Nitrogen aerosol species were important for redistribution of nitrogen in the earth system. It is likely that explosive volcanic aerosols dominated the direct radiative forcing for up to one to two years following explosions and modified climate and biogeochemistry.

Anthropocene changes in aerosols have been large (**Figure 6**) and have substantially altered (>40%) SW radiative forcing as well as the number of CCN (**Figure 6, Table 3**). In addition, land and ocean biogeochemistries are likely to have been modified by changes in nutrients (**Figure 6**). The changes in snow albedo caused by anthropogenic aerosols are poorly constrained and important because of the sensitivity of the cryosphere to albedo change (**Tables 2 and 3, Figure 6**). Of these processes, land and ocean biogeochemistry responses to aerosol deposition of nutrients as well as the more studied aerosol-cloud interactions are likely to be the most important and poorly understood processes (**Table 2**).

SUMMARY POINTS

1. Aerosols impact climate and biogeochemistry while in the atmosphere and after they are deposited onto the earth's surface.

2. In the preindustrial climate, aerosol direct radiative forcings, interactions with clouds, and the resulting impact on climate are likely to be important climate impacts, but natural aerosols have not been well studied.
3. Anthropogenic changes in aerosols cause up to an ~40% change in SW radiative forcing from aerosols and ~60% increase in the number of CCN.
4. Aerosol interactions with land and ocean biogeochemistry, both in the natural climate system and under anthropocene changes, could potentially be as important as the cloud-aerosol interactions, although few studies focus on this issue.
5. Snow-albedo changes from atmospheric deposition of absorbing aerosols represent an important mechanism whereby polar climates in particular can be preferentially warmed by aerosols and thus represent an important new area of research.
6. Aerosols and atmospheric chemistry are intrinsically linked, making changes in aerosols important for greenhouse gas concentrations (e.g., ozone and methane) and making changes in atmospheric chemistry important for aerosols, such as SOA, nitrates, and ammonium.

FUTURE ISSUES

1. Aerosol direct radiative forcing, especially of SOAs, biomass-burning aerosols, and desert dust, is uncertain and has important impacts on climate.
2. Aerosol indirect radiative forcings, from either natural aerosols or anthropogenic aerosols, are likely to be some of the most important interactions with climate, and remain, despite considerable research, uncertain.
3. The impact of aerosols on land and ocean biogeochemistry comes from both changes in nutrient cycles as well as physical climate forcing. These impacts may be important for climate and the indirect effect but are so poorly understood that estimates are difficult to make.

DISCLOSURE STATEMENT

The authors are not aware of any biases that might be perceived as affecting the objectivity of this review.

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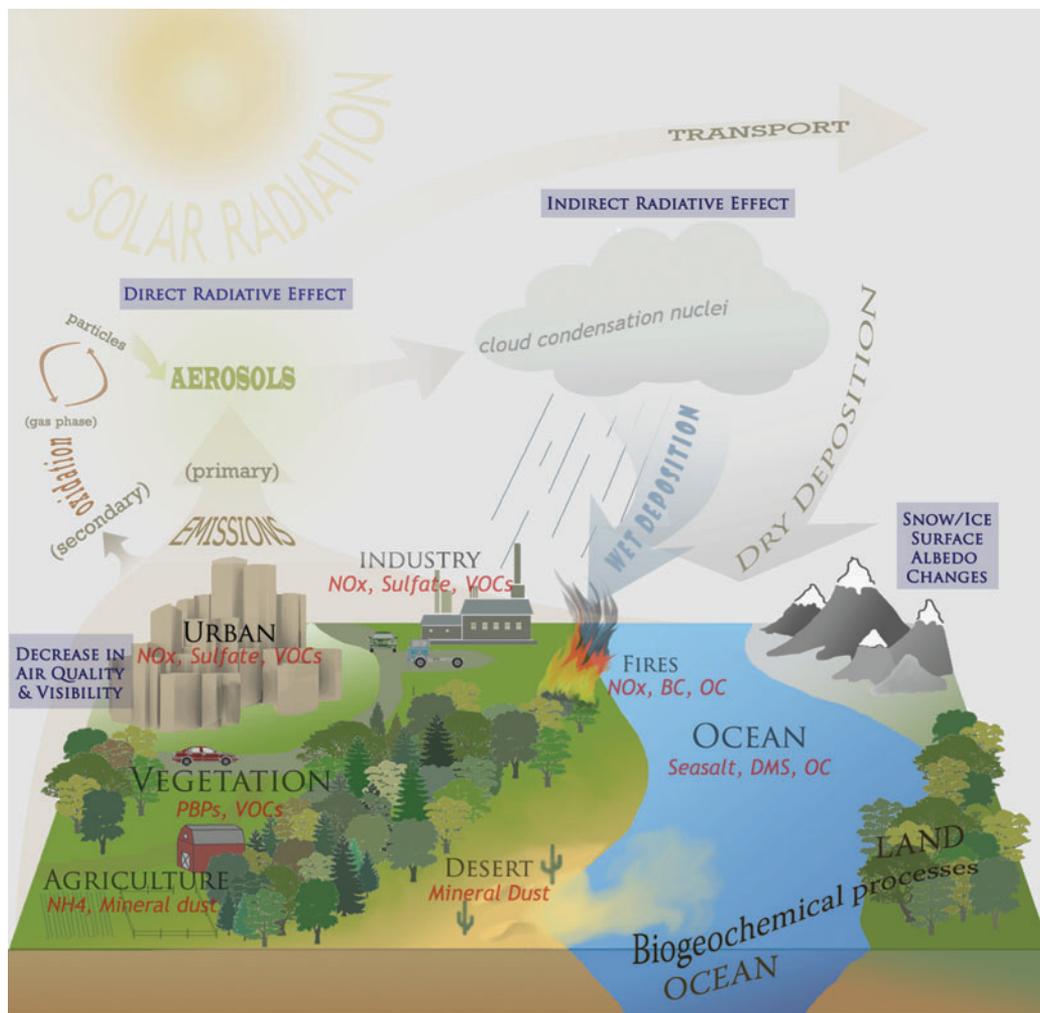


Figure 1

Schematic of aerosol sources, composition, transport, deposition, and impacts on climate and biogeochemistry.

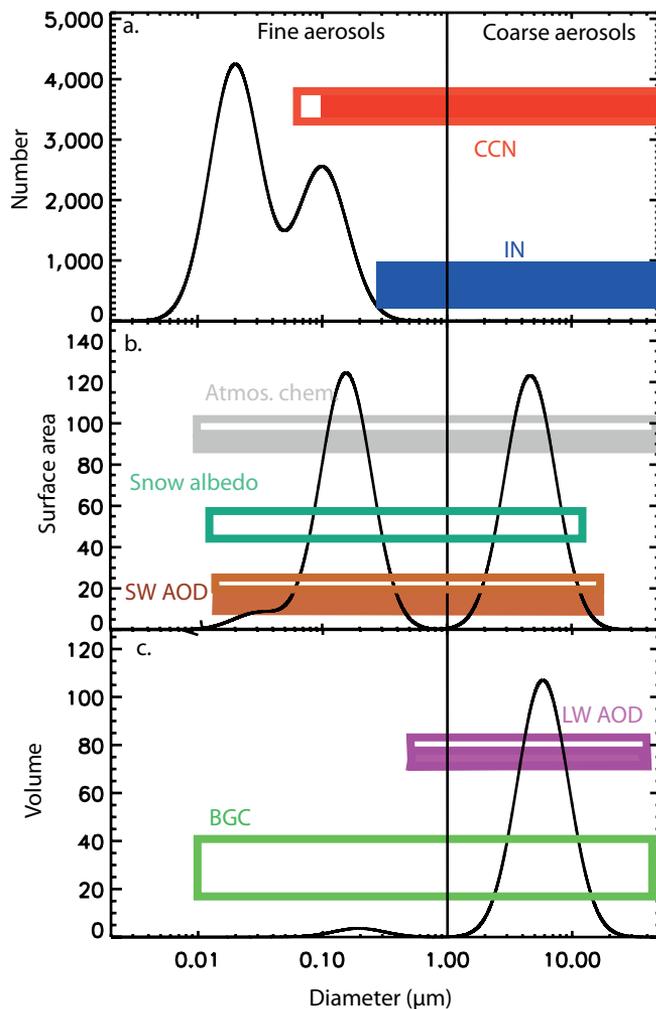


Figure 2

(a) Aerosol number, (b) surface area, and (c) volume for a typical trimodal aerosol distribution (based on information in figure 7.6 in Reference 18 and on information in Reference 225). Also shown in the boxes is a schematic representation of the typical aerosol diameter range impacting various processes as described in the text. Each process is assigned a panel depending on whether the impacts are primarily dependent on number (CCN and IN), surface area (SW AOD and LW AOD) or mass (biogeochemistry). Abbreviations: CCN, cloud condensation nuclei (*red*); IN, ice nuclei (*blue*); SW AOD, shortwave aerosol optical depth (*brown*); LW AOD, longwave aerosol optical depth (*purple*); and BGC, biogeochemically relevant species (*green*). Solid boxes represent only size-dependent processes, and the outlined boxes represent the part of impact that is composition dependent.

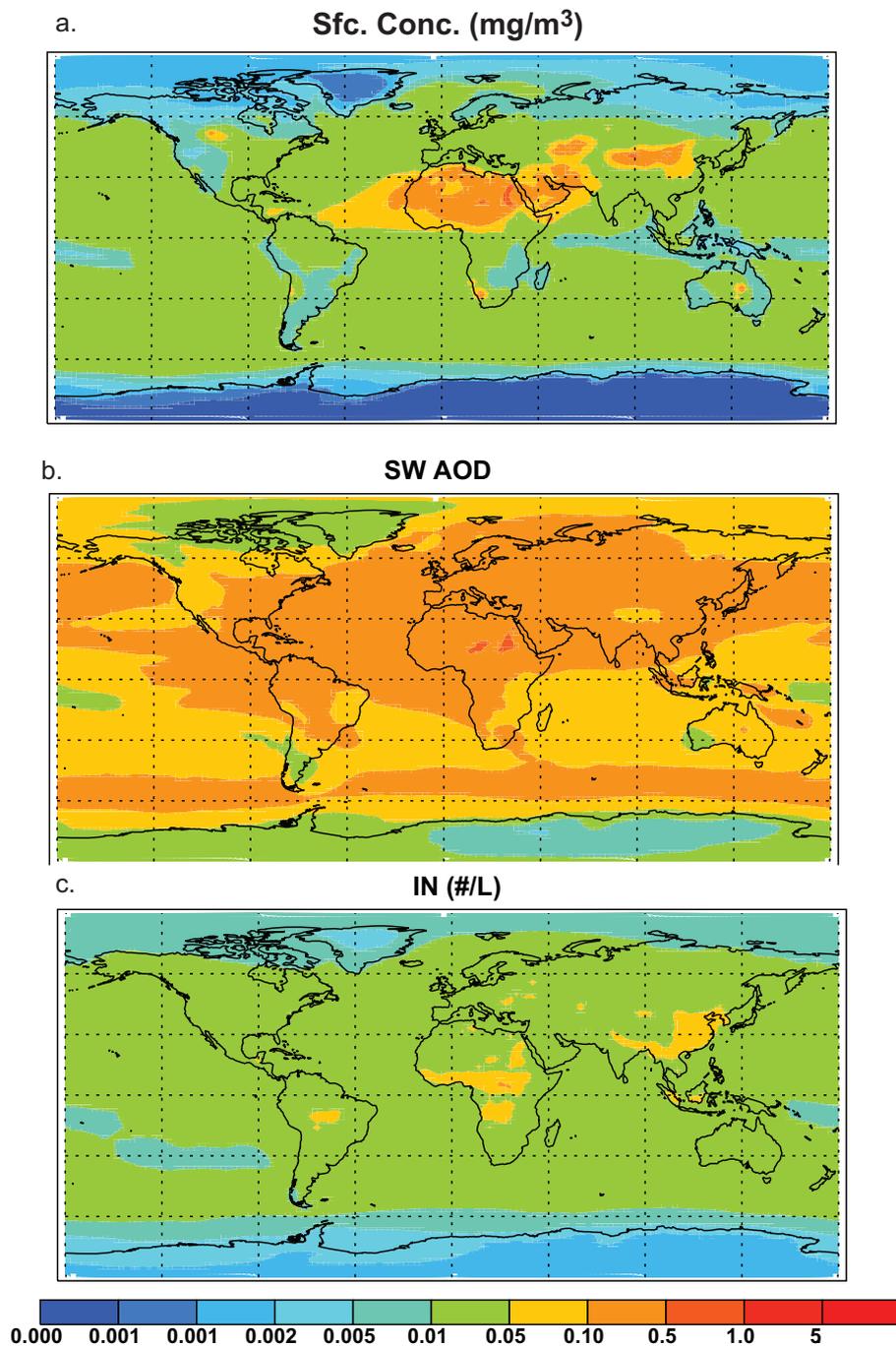


Figure 3

Spatial distribution of aerosols relevant for different effects, including (*a*) the surface concentration (mg/m³), (*b*) SW AOD (unitless), (*c*) and ice nuclei (IN) in the surface layer (#/L). Details on how these are calculated are discussed in the Supplemental text. The color scale at the bottom applies to all three aerosol measures. Abbreviations: Sfc. Conc., surface concentration; SW AOD, shortwave aerosol optical depth.

 Supplemental Material

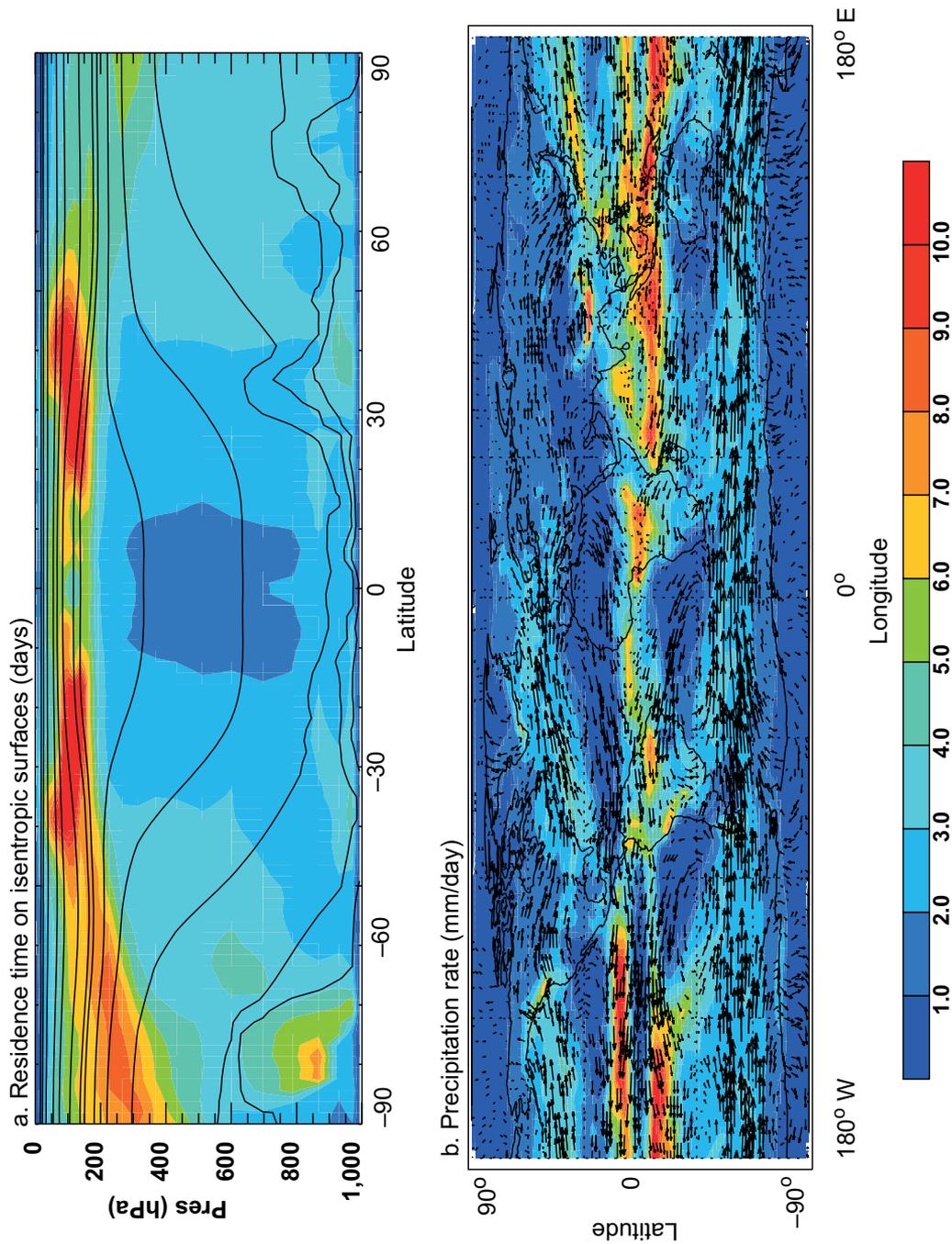


Figure 4

Transport pathways of aerosols. (a) Vertical distribution with latitude of the residence time of aerosols along a neutrally buoyant layer (isentropes), considering only vertical motion by heating in colors, with black lines marking surfaces of constant buoyancy (isentropes). (b) Annually averaged precipitation rate (mm/day) in color, with annual mean wind vectors for 850 hPa (hectopascals) (just above the boundary layer). The color scale at the bottom of the figure applies to both panels *a* and *b*, with applicable units.

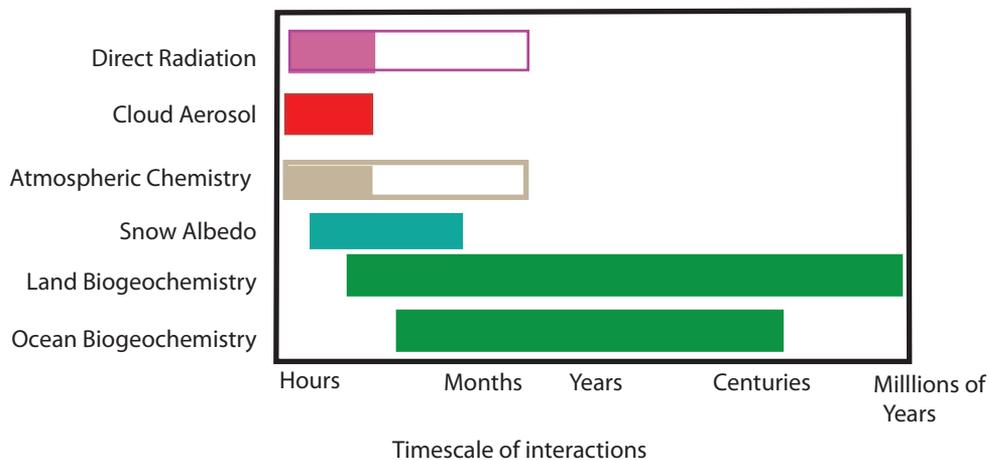


Figure 5

Timescale of different impacts on biogeochemistry from minutes to thousands of years. For impacts that occur while the aerosols are in the atmosphere, the timescale is relatively quick (one day to a few weeks), whereas for impacts that occur when the aerosols are deposited, the timescale can be much longer. Open boxes represent processes operating for stratospheric aerosols, and colored boxes represent processes operating for tropospheric aerosols.

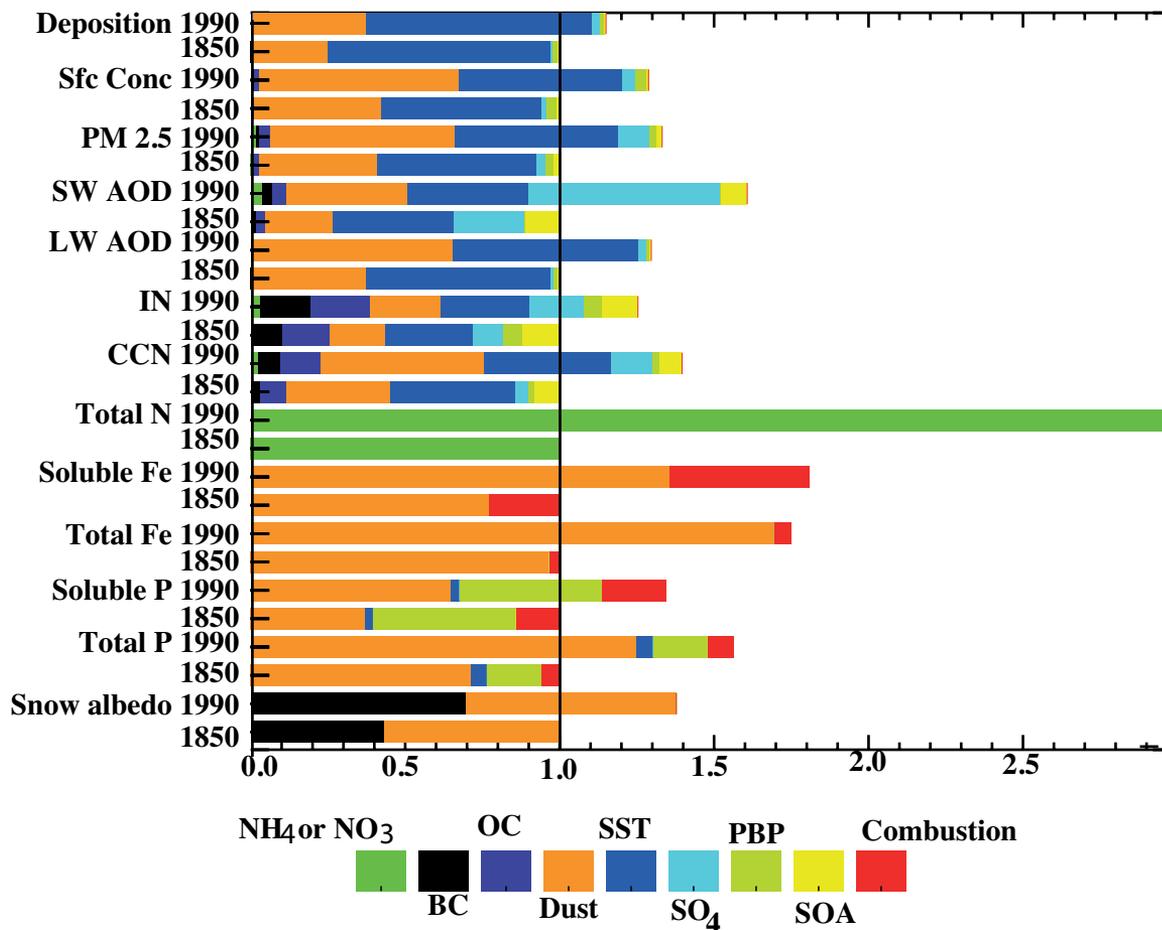


Figure 6

Fractional contribution to each climate or biogeochemical effect for each type of aerosol deposition. Abbreviations: Sfc Conc, surface concentration; PM 2.5, particulate matter less than 2.5 μm ; SW AOD, shortwave aerosol optical depth; LW AOD, longwave aerosol optical depth; IN, ice nuclei; CCN, cloud condensation nuclei; Total N, total nitrogen deposition; Soluble Fe, soluble iron deposition; Total Fe, total iron deposition; Soluble P, soluble phosphorus deposition; Total P, total phosphorus deposition; Snow albedo, snow-albedo impacts from deposition; BC, black carbon or soot; NH₄ or NO₃, ammonium and nitrate; OC, organic carbon; PBP, primary biogenic particles; SOA, secondary organic aerosols; SO₄, sulfate; and SST, sea salt. The numbers are normalized by the estimates for the preindustrial climate, so that increases since the preindustrial period are shown. Many of the proportions are calculated here, as described in the **Supplemental text** (Deposition, surface concentration, PM 2.5, SW AOD, LW AOD, IN, CCN, Total N), and others result from the literature review, as described in the **Supplemental text** (Soluble Fe, Total Fe, Soluble P, Total P, Snow albedo). Some previous studies did not use the same division of composition as used in this study, and therefore, these are put in a separate category (combustion).

 [Supplemental Material](#)