Atmospheric Aerosols: Clouds, Chemistry, and Climate

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Abstract

Although too small to be seen with the human eye, atmospheric particulate
matter has major impacts on the world around us, from our health to global
climate. Understanding the sources, properties, and transformations of these
particles in the atmosphere is among the major challenges in air quality and
climate research today. Significant progress has been made over the past two
decades in understanding atmospheric aerosol chemistry and its connections
to climate. Advances in technology for characterizing aerosol chemical com-
position and physical properties have enabled rapid discovery in this area.
This article reviews fundamental concepts and recent developments sur-
rounding ambient aerosols, their chemical composition and sources, light-
absorbing aerosols, aerosols and cloud formation, and aerosol-based solar
radiation management (also known as solar geoengineering).

INTRODUCTION

Atmospheric aerosols, also known as particulate matter (PM), are small (2 nm–10 μm) particles in the ambient atmosphere. These particles originate from human activities as well as from natural processes or sometimes a combination thereof (**Figure 1**). They may be emitted directly or formed in situ as a result of chemical and physical transformations of gas-phase emissions in the atmosphere. Although we generally associate the word *particle* with solid material, atmospheric aerosols may be liquid or solid or have exotic morphologies such as gels surrounding solid cores (1), two-phase liquid particles (2), or organic glasses (3). Owing to their small size, individual atmospheric aerosol particles have little inertia, and therefore they may stay airborne for several days after being emitted to or formed in the atmosphere. As a result, during their atmospheric lifetime they may be transported long distances, participating in atmospheric chemical reactions and impacting human health (see the sidebar titled Atmospheric Aerosols and Health), environmental quality, and climate.

In this article, I review fundamental concepts and recent developments surrounding ambient aerosols, their chemical composition and sources, and their multiple roles in climate. I focus in particular on our understanding of how the chemistry of atmospheric aerosols impacts their climate-relevant physical properties, such as their optical properties and their ability to nucleate cloud droplets or ice particles. I close with discussions of aerosol-based climate management (also known as solar geoengineering) and future opportunities and challenges in the field.

AEROSOL CHEMICAL COMPOSITION AND SOURCES

The chemical composition of atmospheric aerosol particles varies widely as a result of their diverse sources (**Table 1**). For example, marine aerosols are typically composed of sea salt and organics
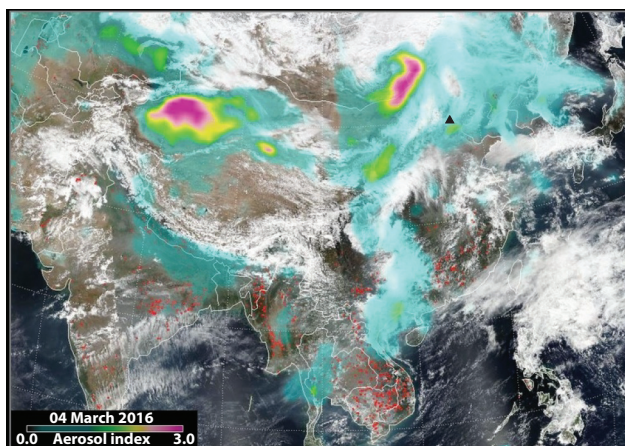

Figure 1

This composite image of data from the NASA Suomi NPP Satellite shows several different types of aerosols and clouds over Asia. The figure combines a Visible Infrared Imaging Radiometer Suite (VIIRS) RGB image with aerosol index (AI) data from the Ozone Mapping Profiler Suite (OMPS) for March 4, 2016. The red dots are fire location data from the moderate resolution imaging spectroradiometer (MODIS) satellite. The AI shows dust over the Taklamakan Desert in northwest China and eastern Mongolia and smoke from fires over Southeast Asia. Pollution-related aerosol can be seen over Beijing (*black triangle*), and there is a mixture of pollution and smoke from biomass burning over North India. Image courtesy of Colin Seftor for NASA's S-NPP OMPS Science Team.

ATMOSPHERIC AEROSOLS AND HEALTH

Air pollution contributes to 5.5 million premature deaths globally and 130,000 premature deaths in the United States each year (138, 139). Atmospheric aerosol particles less than 2.5 μm in diameter, also called $PM_{2.5}$, are singled out as being the most hazardous portion of the aerosol population because these small particles can penetrate deep into the lungs upon inhalation, whereas larger particles typically deposit in the upper respiratory tract. Atmospheric aerosols are associated with a wide array of acute and chronic diseases, from cardiovascular and respiratory issues (140) to mental health (141), dermatology (142), and reproductive health (143). The physiological mechanisms of aerosol impacts on health are just beginning to be understood. Aerosol particles can be a delivery system for cytotoxic or carcinogenic substances such as lead, polycyclic aromatic hydrocarbons and their derivatives, and transition metals (144). Some common aerosol organics have been connected to oxidative stress response in the lungs (145). Research also indicates that toxicity is connected to redox activity in combustion exhaust particles and that it increases as these particles age in the atmosphere (146).

from biological activity at the sea surface (4, 5). In contrast, continental aerosols may consist of dust or secondary inorganic salts mixed with organic material of biogenic or anthropogenic origin (6, 7). Particles containing both inorganic and organic material, along with significant liquid water (8), are typical. Aerosol particles are acidic (9, 10), especially over continental regions (11). They also have extremely high ionic concentrations; salt crystallization is suppressed in aerosols owing to their small size (12). In recent years, technological advances have allowed investigators to identify thousands of organic species that are present in aerosol particles (13–15), and statistical approaches have been used to derive deep insight from organic fragmentation patterns obtained via high-resolution mass spectrometry (16, 17). However, the majority of organic aerosol mass still often remains unspeciated.

Many aerosols are emitted directly into the atmosphere (known as primary emissions) through human activities or natural processes. However, most aerosol particles smaller than 2.5 μm in diameter (also known as $PM_{2.5}$) are formed via chemical and physical processing of gas-phase precursors in the atmosphere (known as secondary particles). Note that, even though natural emissions of sea salt and dust particles dominate aerosol fluxes on a mass basis, particles from these primary sources tend to be in the coarse size range (>2.5 μm in diameter). An important example of secondary inorganic aerosol is particulate sulfate formed by the oxidation of SO_2 gas to make aerosol sulfate, which may be neutralized by the uptake of NH_3 (18). Secondary organic aerosol (SOA) material forms via two major pathways: condensational and aqueous phase. Condensational SOA formation involves the thermodynamically driven partitioning of organic gases

$PM_{2.5}$: particles <2.5 μm in diameter

SOA: secondary organic aerosol

Table 1   Sources of atmospheric aerosols and estimates of their magnitudes (with ranges, where available)

Source	Magnitude (Tg year^{-1})	References
Secondary inorganic (anthropogenic sulfates, nitrates)	69.9	147, 148
Mineral dust (natural)	1,000	149
Sea salt	$1-3 \times 10^4$	150
Black carbon	7.5 (2–29)	36
Primary organic aerosol	33.9 (17–77)	151
Secondary organic aerosol	140 (50–380)	152

[e.g., lower-volatility species formed by the oxidation and functionalization of volatile organic compounds (VOCs)] onto existing aerosol particles (19). Aqueous SOA formation refers to the uptake of water-soluble VOCs by aqueous aerosols, cloud droplets, or fog water, followed by aqueous chemistry to form lower-volatility secondary organic material (20). It was recently proposed that photosensitized chemistry involving light-absorbing aerosol organics and VOCs can lead to autocatalytic formation of SOA (21–23).

AEROSOLS AND CLIMATE FORCING

Atmospheric aerosol particles impact Earth's radiation balance and, therefore, its climate. They alter Earth's albedo by scattering and absorbing solar radiation and by influencing cloud formation and properties. Aerosol effects provide a net negative radiative forcing that may be comparable in magnitude to the positive forcing by the greenhouse gas carbon dioxide (CO_2) (24). However, significant gaps in our scientific understanding of aerosol-related climate forcings have resulted in very large uncertainties in the estimates of their magnitudes. Because climate forcing is defined as the difference between forcing elements in the climate system today as compared to 1750 (24), the primary source of uncertainty in estimations of aerosol climate forcing is a lack of information regarding aerosol loadings in preindustrial times. Unlike many greenhouse gases, the short lifetime of aerosols means that they are not well mixed and therefore not uniformly distributed in the atmosphere. Therefore, it is difficult to infer aerosol loadings far from the locations of the few natural aerosol paleorecords that are available (25) and the baseline for climate forcing calculations must be estimated using models of aerosol sources in 1750. The sources and characteristics of aerosols in pristine regions are of particular interest because they may have similarities to preindustrial aerosols (26). Sources of aerosols in preindustrial times included marine aerosols, mineral dust, biomass burning, volcanos, and new particle formation from biogenic VOCs. When condensational sinks do not dominate, new particles can form from the condensation of gas-phase precursors. Investigators recently identified that highly oxidized products of biogenic VOCs can nucleate to form new particles (27–29). For many years, new particle formation was believed to require H_2SO_4, which is formed largely from anthropogenic SO_2 (30, 31). These recent findings have implications for models of aerosols in 1750 and, therefore, may help reduce uncertainty in estimates of aerosol climate forcing in modern times.

In this section, I review the main ways in which aerosols impact climate by directly scattering and absorbing solar radiation and nucleating clouds. I highlight black and brown carbon, two specific categories of aerosols that are important for climate. I close by discussing proposals to intentionally alter climate using aerosols.

Aerosol Direct Effects on Climate

Aerosols directly scatter and/or absorb solar radiation, depending on their optical properties. Aerosols are referred to as short-lived climate forcers because the atmospheric lifetime of aerosols (several days) is much shorter than that of greenhouse gases (tens to hundreds of years). Because of their short lifetime, atmospheric concentrations of aerosols respond relatively rapidly to changes in emissions.

Light scattering by aerosols increases Earth's albedo and therefore contributes negative radiative forcing. Because most aerosols are similar in size to the wavelengths of the solar radiation that arrives at the earth's surface, aerosol light scattering is described well by Mie theory. For simple spherical particles, if the complex refractive index ($N(\lambda) = n_\lambda + k_\lambda i$) is known, optical parameters for climate modeling, such as the single scatter albedo (SSA), may be calculated for a given particle

size. SSA describes the direct interaction of an aerosol particle with solar radiation and is given by

$$SSA = \frac{Q_{scat}}{Q_{scat} + Q_{abs}}, \qquad 1.$$

where Q_{scat} and Q_{abs}, are the scattering and absorption efficiencies of the aerosol, respectively. For SSA $= 1$, the particle is purely scattering. Below a critical value of SSA, the aerosols will have a net positive radiative forcing owing to their absorption of solar radiation. The critical value depends on surface reflectance, but it is estimated to be ~0.85–0.9 (32).

Since the introduction of the US Clean Air Act (CAA) in 1970, pollution controls designed to protect public health have successfully led to a decrease in scattering aerosols across the continental United States, saving lives and money (the US Environmental Protection Agency calculates that the economic benefits of the CAA far exceed the costs) (33). However, an unintended consequence of this reduction in aerosols has been a decrease in albedo, "unmasking" the radiative forcing caused by an increase in CO_2 over the same time period (34).

Black Carbon

Strongly absorbing carbonaceous aerosols, also known as soot or black carbon (BC), are generated via combustion processes (e.g., biomass or the combustion of fossil fuels). BC absorbs strongly across the visible spectrum and absorbs more light than it scatters (SSA of ~0.38) (35). As a result, it contributes a net positive radiative forcing (warming effect) second only to that of CO_2 (36, 37). Because BC is such a powerful short-lived climate forcer, it has been proposed that reductions in BC emissions could be a short-term pathway to climate change mitigation (36, 38); however, predicting the potential impacts of such a policy is complicated by the fact that BC is often coemitted with scattering aerosols (36, 37). The potential climate benefits of reducing BC emissions, in combination with the health benefits of reduced BC exposure, are why significant attention has been placed globally on the development and distribution of improved household energy solutions, such as clean cookstove technology, in developing countries (36).

Brown Carbon

Light-absorbing organic aerosol material, or aerosol brown carbon (BrC), is commonly observed in air masses that have been influenced by either pollution or the burning of biomass or biofuel (39). BrC has been observed in the form of amorphous spheres, or "tar balls" (40–43), or as water-soluble, light-absorbing "humic-like substances" (HULIS) in aqueous extracts of atmospheric aerosols (44–46).

BrC does not absorb as strongly as BC at long wavelengths (550 nm and higher), and therefore its contribution to total aerosol absorption in this range is relatively minor. However, unlike BC, the classic BrC absorption spectrum (**Figure 2**) features sharply increasing absorption with decreasing wavelength, without distinct peaks, below ~400 nm. An empirical parameter known as the Ångström exponent, α, is often used to describe the shape of the BrC absorption spectrum. This can be expressed as the wavelength dependence of the mass absorption efficiency (MAE) (m^2 g^{-1}):

$$MAE \propto \lambda^{-\alpha}. \qquad 2.$$

Typical values of α range between 2 and 7 (47, 48). The chemical composition of BrC determines its optical properties. Many organic species absorb at wavelengths below 400 nm, including carbonyl-containing species and some N-substituted organics. Most of the SOA formation processes that have been observed in laboratory studies to lead to light-absorbing (brown)
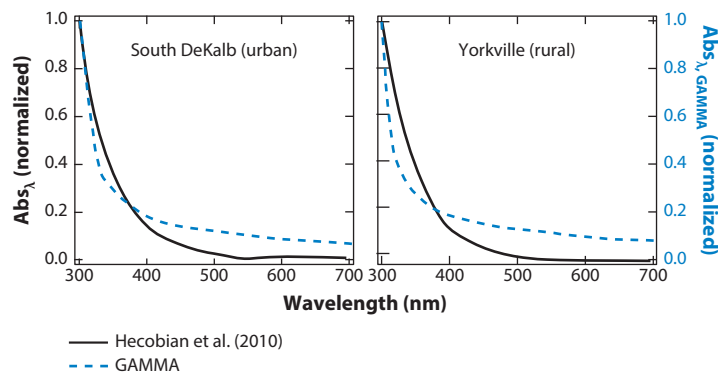
Figure 2

Wavelength dependence of light absorption by brown carbon in aqueous extracts of ambient aerosols sampled by Hecobian et al. (48) (*solid line*, *left axis*) at urban and rural locations in Georgia, United States. Composite absorption spectra predicted by the photochemical box model GAMMA (*dashed blue line*, *right axis*) are shown for comparison. Adapted from Woo et al. (2013) with permission from the Royal Society of Chemistry.

products involve the formation of N-substituted organic compounds from the reactions of organics with ammonia/ammonium or amines (49–54). Pi-conjugated or aromatic compounds, and/or N-substituted organic species, may have absorption peaks at wavelengths between 400 and 700 nm (55, 56). Phillips & Smith recently identified that ~50% of absorption between 300 and 600 nm in aqueous extracts of BrC collected from ambient aerosols in Athens, Georgia, could be attributed to charge transfer complexes formed by interactions between alcohol and carbonyl groups (57, 58).

Because the optical properties of aerosol BrC are connected to their chemical composition, it follows that they may evolve as a result of photochemical aging in the atmosphere. The optical properties of ambient aerosols have been observed to vary with photochemical age (7, 59). Oxidation may decrease (through, for example, the breaking of C=C or C−N bonds) or increase (through the creation of carbonyls) (60) BrC light absorption. Several classes of light absorbing aerosol organics have been observed to lose their brown color as a result of photolysis and oxidation, a process referred to as "photobleaching" (60–62). The potential impact of BrC on climate depends on the relative rates of BrC formation and photobleaching (63).

As a result of the uncertainty surrounding the sources and properties of BrC, and its tendency to be coemitted and mixed with BC, these aerosols are not well represented in atmospheric chemistry and climate models. It is becoming clear, however, that absorption by BrC has a significant impact on climate. Alexander et al. measured the complex refractive index of amorphous BrC particles from Asia to be $N = 1.67 − 0.27i$ at 550 nm, and they determined using Mie calculations that these particles have significant values of Q_{abs}, leading to SSA <0.9 at most wavelengths (42). Efforts to analyze BrC absorption using models and large observational data sets, including measurements of aerosol optical depth from the AERONET ground-based remote-sensing network (64), have yielded estimates of 0.77–0.85 for the SSA of BrC (35); these analyses indicate that BrC globally contributes 6–40% of the aerosol absorption at 440 nm (65) or 20% at 550 nm (66). Inclusion of BrC in the population of organic carbon (OC) aerosols changes the net effect of OC from scattering to close to zero, especially in areas with heavy open biomass burning (35, 66).

Aerosols and Cloud Droplet Formation

The tendency of aerosols to take up water vapor and form cloud droplets is central to climate and the hydrological cycle. Particularly because of the high surface tension of liquid water, and

therefore the high energetic cost of homogeneous nucleation of water vapor, cloud formation as we know it would not be possible without aerosols serving as cloud condensation nuclei (CCN). Increased aerosol concentrations due to human activities lead to an increase in cloud droplet numbers, and smaller cloud droplets (67), making clouds more reflective and increasing cloud lifetimes (68). Estimates of the climate impact of aerosol–cloud interactions are highly uncertain (24), owing in part to uncertainties in preindustrial CCN loadings and spatial distributions and in part to challenges in bridging scales between cloud droplets (∼10 μm) and the grid size of climate models (typically >50 km) (69). Reducing uncertainties in the aerosol impact on clouds is critical for improving climate predictions.

Cloud droplet formation depends on meteorological parameters, such as updraft velocity, as well as the intrinsic ability of an aerosol to take up water and serve as a cloud condensation nucleus (CCN ability) (70). CCN activation is described by the Köhler equation, which accounts for the competing influences of the Kelvin (curvature) and Raoult (solute) effects on the equilibrium vapor pressure of the aqueous aerosol (71):

$$ln\left(\frac{p_w(D_p)}{p^0}\right) = \frac{A}{D_p} - \frac{B}{D_p{}^3} \qquad \text{3.}$$

with

$$A = \frac{4M_w\sigma}{RT\rho_w} \text{ and } B = \frac{6n_sM_w}{\pi\rho_w}, \qquad \text{4.}$$

where D_p is the diameter of the droplet, p_w is the water vapor pressure over the droplet of diameter D_p, p^0 is the water vapor pressure over a flat surface, M_w is the molecular weight of water and ρ_w is its density, R is the gas constant, T is temperature, σ is the surface tension of the droplet, and n_s is the number of moles of solute. Some researchers have argued that particle size alone (which affects D_p and n_s) is the most important factor determining aerosol CCN ability (72), which, if true, would simplify the task of accurately representing cloud formation in models, since accounting for chemistry would not be necessary. However, a CCN's chemical composition clearly has a major impact on its hygroscopicity (73). Aerosol chemistry may alter the fraction of CCN material that dissolves during activation (n_s) (74, 75). Droplet surface tension (σ) can be depressed by organic material at the gas–aerosol interface (76, 77). Organic material can reach the gas–aerosol interface directly via adsorption of organic species from the gas phase, or by partitioning of amphiphilic, surface-active species from the aerosol bulk to the gas–aerosol interface. Bulk absorption of trace gases simultaneously with water uptake during activation (known as co-condensation) may enhance CCN activity (78, 79). Our work suggests that surface adsorption of organic gases can also enhance aerosol CCN activity (and therefore cloud droplet numbers) by depressing surface tension (**Figure 3**) (80).

Despite the potential for reduced droplet surface tension to mitigate the Kelvin effect, some models suggest that the partitioning of nonvolatile particle-phase organics to the gas–aerosol interface may not necessarily enhance aerosol CCN activity. This limitation is due to the partitioning of solute to the surface decreasing the amount of solute in the bulk (n_s), which offsets the decrease in critical supersaturation gained from the surface tension depression (81, 82). In the case of volatile solutes partitioning to the aerosol by adsorption or co-condensation from the gas phase, bulk depletion is not expected to be important.

Significant experimental challenges exist for measuring the surface tension of submicron-sized aerosol particles directly (83), and mounting experimental evidence indicates that it is not appropriate to extrapolate surface-activity parameters from macroscopic bulk solutions to aerosols, or micron-sized droplets, owing in part to the high surface area to volume ratio in those systems (80, 84, 85). Measurements of both the critical supersaturation and the particle's wet diameter at the moment of activation have the power to decouple the effects of surface tension reduction, bulk solute depletion, surface adsorption, and co-condensation on CCN activity (84–86).
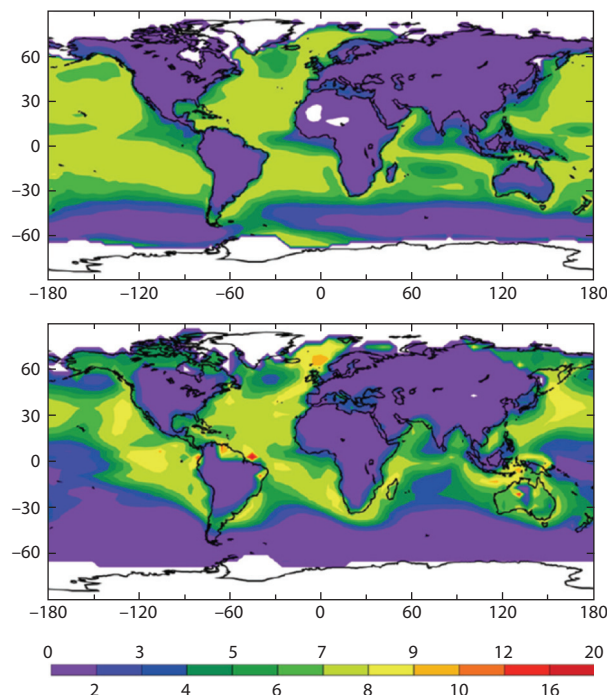
Figure 3

Stratus cloud droplet number concentration response (%) to an increase in aerosol hygroscopicity, κ, of 20%. A similar increase in aerosol hygroscopicity was observed when aerosols were exposed to gas-phase methylglyoxal in the chamber experiments of Sareen et al. (80). Results are shown for the GEOS-CHEM (*top panel*) and NASA-GMI (Global Modeling Initiative) (*bottom panel*) models coupled with the cloud droplet adjoint framework of Karydis et al. (70). Adapted from Sareen et al. (80) with permission from the National Academy of Sciences.

Aerosols as Ice Nuclei

Another way that atmospheric aerosols impact climate is by modulating ice formation in clouds. Owing to the Kelvin effect and freezing point depression by solutes, ice particles and supercooled liquid water may coexist in a cloud down to around $-20°C$. The formation of ice particles in clouds requires an ice nucleus (IN) except at very cold temperatures ($-40°C$). Aerosols may nucleate ice particles via three mechanisms: freezing of supercooled water droplets containing aerosol particles (immersion freezing), water vapor adsorption onto an aerosol particle at subfreezing temperatures to form ice (deposition mode), or collision of a supercooled water droplet with an aerosol particle (contact mode). Understanding the ice/liquid phase partitioning of water in clouds is important because, once formed, ice hydrometeors tend to grow very large at the expense of surrounding supercooled water droplets (87). This growth is due, in part, to the thermodynamic stability and lower equilibrium vapor pressure of the ice crystals. Therefore, ice nucleation leads to increased precipitation and can impact the radiative properties of clouds. It was previously believed that only a few types of aerosols with special properties could be effective ice nuclei. These were generally large, water-insoluble particles with a crystal structure and bond length similar to those of ice, so their surfaces could act as a template for ice crystallization (87). Examples include dust particles or metal oxide–containing combustion particles. Although this is largely true for cirrus ice formed via vapor deposition (88), in the past 15 years the definition of a "good" IN has
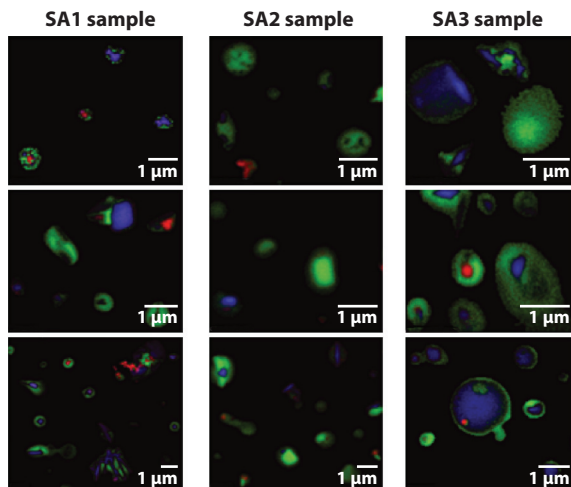
Figure 4

Scanning transmission X-ray microscopy with near-edge X-ray absorption fine structure spectroscopy (STXM/NEXAFS) component images for three populations of field-collected ice-nucleating aerosol particles. In each component image, the green, red, and blue colors map the dominating presence of the carboxyl functional group (organic material), carbon double bonding (soot-like), and inorganic material, respectively. Adapted from Knopf et al. (96) with permission from the American Geophysical Union.

been expanded considerably to include water-soluble species such as ammonium sulfate (89, 90), amorphous organics (91), soot (92), biological particles (93, 94), sea spray (94, 95), and mixed particles containing inorganic and organic material (**Figure 4**) (96).

Geoengineering

Some investigators have proposed that climate warming could be counteracted by increasing the Earth's albedo, an approach called "solar radiation management" or "solar geoengineering." The introduction of aerosols to the lower stratosphere is believed to be among the highest impact, most cost-effective, and most quickly deployable approaches to solar geoengineering (97). Although some researchers have suggested the use of engineered particles with special photophysical properties (98, 99), the most well-studied approach for solar geoengineering is the introduction of sulfate aerosols to the lower stratosphere. This approach mimics the formation of sulfate aerosols that follows the injection of sulfur dioxide into the stratosphere via volcanic eruptions. The cooling effects of volcanic eruptions on climate are well documented (100, 101) and have been carefully studied using climate models (102).

Significant downsides exist for this approach (103), leading many experts to recommend that solar geoengineering should be considered only as a short-term intervention in the case of a rapid climate change emergency (104, 105). Atmospheric chemists and climate scientists warn against side effects such as an enhancement of stratospheric ozone depletion due to the introduction of particle surfaces for heterogeneous chlorine activation (106, 107), stratospheric warming (108, 109), and changes in atmospheric dynamics, including an intensification of the stratospheric polar vortices (110). Increasing Earth's albedo with aerosols would do nothing to counteract ocean acidification associated with high CO_2 levels (111, 112). Significant ethical issues surround geoengineering as well. Geoengineering approaches could deflect efforts from more sustainable long-term solutions to climate change, i.e., reducing greenhouse gas emissions (105).

Furthermore, the large-scale environmental impact of solar geoengineering exceeds the boundaries of any one nation, which, coupled with the potential for negative consequences (anticipated and unanticipated), makes geoengineering an international political issue. In 2010, more than 190 nations adopted a prohibition on geoengineering under the 1992 United Nations Convention on Biological Diversity, citing the need for scientific consensus and risk assessment (113).

CHEMICAL AND PHYSICAL TRANSFORMATIONS OF ATMOSPHERIC AEROSOLS

The lifetime of a tropospheric aerosol is on the order of one week, and over this time period these particles are chemically transformed via sunlight, cloud processing, and reactions with gas-phase species. We are just beginning to understand how these chemical transformations impact aerosol physical properties. BrC formation in aerosols and uptake of surfactant gases, as discussed in the previous sections, alter the climate-relevant physical properties of the aerosol. The atmosphere is an oxidizing environment, and organic material in aerosols becomes more oxidized and less volatile with age (114). Volatility may also decrease owing to inorganic-organic interactions in the particle, such as organic salt formation (115, 116). Deposition of acid gases onto soot (117) and dust (118) particles leads to increased hygroscopicity and changes in aerosol optical properties. A typical aerosol may take up water to form a cloud droplet and dry out again several times over the course of one day, leaving in its composition the signature of chemistry in both cloud and aerosol modes (119).

FUTURE CHALLENGES AND OPPORTUNITIES

Significant progress has been made over the past two decades in understanding atmospheric aerosol chemistry and its connections to climate. Advances in technology used to characterize aerosol chemical composition (13, 14, 16, 120–126) and physical properties relevant for climate have enabled rapid discovery in this field over the past decade (93, 127–130). In the coming years, we will need strategies to manage and extract the full value from the overabundance of data being generated by ground-based measurements as well as by remote sensing. Although traditional statistical approaches have proven useful thus far (17), more sophisticated techniques from the field of data science, such as machine learning (131), may be in order. Advances in this direction will require close interdisciplinary collaboration between data scientists and atmospheric chemists. Similar collaborations between experts in machine learning and climate scientists have yielded the new field of climate informatics (132).

Bridging the scales between the molecular-level knowledge generated via laboratory and theoretical studies and the modest level of chemical detail possible in computationally complex large-scale models is an ongoing challenge. As our knowledge of atmospheric chemical mechanisms becomes more complex, and the pace of discovery more rapid, we will need to develop an approach to kinetic model reduction (133) that is more systematic than the empirical approach that is currently standard in the field.

Air quality has reached a state of crisis in many global megacities, including Beijing, New Delhi, and Tehran. These societies have an opportunity to design and implement new policies that simultaneously address air quality and climate goals (134). While some of the major barriers to developing and enforcing effective air quality and climate policies are political and economic in nature, continued improvement in the prognostic capabilities of three-dimensional atmospheric and Earth system models is also needed for the design and evaluation of these and other policies. For example, investigators recently showed that improvements to the sulfate production mechanism in

air quality models are necessary in order to accurately model persistent haze episodes in China, and the potential benefits from emission reduction policies (135, 136). Such improvements will require continued advances in our fundamental understanding of atmospheric chemistry and physics (137).

SUMMARY POINTS

1. Atmospheric aerosol particles impact Earth's climate by scattering and absorbing solar radiation and by influencing cloud formation and properties.

2. The chemical composition of ambient aerosols and their chemical transformations in the atmosphere impact their climate-relevant physical properties.

3. Rapid discovery in this area over the past two decades has been enabled by advances in technology for characterizing aerosol chemical composition and physical properties.

4. Future challenges and opportunities lie at the interface of atmospheric chemistry and computer science.

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