Scattering and Absorbing Aerosols in the Climate System

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Abstract

Tropospheric anthropogenic aerosols contribute the second-largest forcing to climate change, but with high uncertainty owing to their spatio-temporal variability and complicated optical properties. In this Review, we synthesize understanding of aerosol observations, and their

radiative and climate effects. Aerosols offset ~1/3 of the warming effect by anthropogenic greenhouse gases. Yet, in regions and seasons where the absorbing aerosol fraction is high—such as South America, and East and South Asia—substantial atmospheric warming can occur. The internal mixing and the vertical distribution of aerosols, which alters both the direct effect and aerosol-cloud interactions, might further enhance this warming. Despite extensive research in aerosol-cloud interactions, there is still at least a 50% spread in total aerosol forcing estimates. This uncertainty is linked, in part, to the poor measurement of anthropogenic and natural aerosol absorption, as well as the little-understood effects of aerosols on clouds. Next generation space-borne multi-angle polarization and active remote sensing, combined with *in situ* observations, offer opportunities to better constrain aerosol scattering, absorption and size distribution, thus improving models to refine estimates of aerosol forcing and climate effects.

Key points

- Climate models indicate at least a 30% uncertainty in aerosol direct forcing and 100% uncertainty in indirect forcing due to aerosol-cloud interactions.
- The amount of aerosol light-scattering and absorption, expressed as the aerosol single scattering albedo (SSA) parameter, is critical in affecting both aerosol radiation interaction and aerosol–cloud interactions.
- Current satellite sensors cannot provide global-scale 3-D SSA measurements. Future observational efforts should combine satellite-based multi-angle polarization sensors and high spectral resolution lidars with international aircraft and surface *in situ* observation networks.
- Direct comparison of radiation properties observed by satellites with those derived from climate models that assimilate aerosol parameters will improve the understanding of aerosol microphysical properties.
- Future work should investigate the mechanisms underlying aerosol-cloud interactions, especially the adjustment of cloud fraction and water for warm clouds and the microphysical processes in ice and mixed-phase clouds.

Introduction

Aerosols are small liquid or solid particles suspended in the atmosphere[1]. They can be emitted directly (such as dust, sea salt, black carbon and volcanic aerosols) or formed indirectly through chemical reactions (including sulphate, nitrate, ammonium and secondary organic aerosols. Owing to their relatively short lifetime, aerosol concentrations typically peak near their sources. Desert regions (such as North Africa and the Middle East), industrial regions (such as East and South Asia), and biomass burning regions (such as South America and South Africa) are therefore characterized by high mass concentrations (**Fig. 1**). Aerosols exhibit complicated compositions and vary substantially in shape and size, typically ranging between 0.01-10 μm^2. Depending on these structural and compositional characteristics, aerosols can scatter and/or absorb shortwave radiation, as quantified through the single scattering albedo (SSA; **Table 1**). Purely scattering aerosols include sulphates, nitrates, ammonium and sea salt particles, whereas absorbing aerosols are primarily black carbon (BC), with dust and organic carbon partly absorbing in the ultra-violet spectrum[3].

Aerosols have a direct bearing on Earth's energy balance, and therefore, on climate. For instance, aerosol scattering and absorption alters the radiation balance and atmospheric stability through perturbations to the vertical temperature profile. Aerosols can further serve as cloud condensation nuclei (CCN) or ice nucleating particles (INPs), which modify the reflectivity and lifetime of clouds through microphysical processes. Collectively, these influences are quantified as aerosol forcing: the change of net radiative flux at a specified level of the atmosphere, often assessed relative to estimated preindustrial conditions[4].

Globally, anthropogenic aerosols are estimated to produce a net cooling ~ -1.3±0.7 W m^{-2} at the top of atmosphere; -0.3±0.3 W m^{-2} is attributed to the aerosol-radiation interaction (ARI), -1.0±0.7 W m^{-2} to aerosol-cloud interactions, ~-1.15 W m^{-2} to total forcing from scattering aerosols, and ~+0.12 W m^{-2} to BC[4]. This combined aerosol forcing offsets roughly one third of the warming from anthropogenic greenhouse gases (GHGs). However, the large spread in the estimated aerosol forcing leads to large discrepancies in climate sensitivity[5,6]. Thus, aerosols are

considered to be the largest contributor of uncertainty in quantifying present day climate change[4].

Much of this uncertainty in aerosol forcing arises from both the lack of separate global constraints on aerosol optical and microphysical properties (optical depth, size distribution, hygroscopicity, and mixing state, among others) and the inaccurate representation of them in climate models[7-10]. In particular, aerosol SSA is further thought to contribute substantially to the uncertainties in direct aerosol forcing[11,12] , and might even change in sign[13-15]. Despite its importance, aerosol SSA is largely unmeasured by current satellite sensors. However, emerging techniques using high accuracy, multi-angle polarimetry measurements combined with space-based lidars, and constrained by ground-based remote sensing and detailed *in situ* measurements of particle microphysical properties, represent a promising way to establish a consistent 3-D global SSA record[16-21]. Combined with models, this progress will thus improve the modelled aerosol parameters, better quantify aerosol forcing estimates and even climate projections. Moreover, consistent aerosol observational records will allow improved quantification of the broader environmental impacts of aerosols, including on air pollution and haze, and in turn, visibility and human health.

In this Review, we focus on the aerosol impacts on climate, and synthesize the latest progress in measuring aerosol properties, understanding their spatio-temporal variability, and efforts taken to quantify their radiative and climate effects. We describe the challenges remaining in understanding the physical properties and space-time variability of different aerosol types, and propose possible approaches to better constrain aerosol forcing.

Physical processes impacting climate

Determining the ultimate impact of aerosol forcing on climate is complicated, as aerosols impact the energy balance and the climate system through various pathways. These pathways include direct scattering and absorption of radiation (direct effects), interaction between these two effects, as well as with clouds, as will now be discussed (**Fig. 2**).

Aerosol scattering effect

The primary effect of aerosols in the climate system is the scattering of solar radiation[1], which means that only the direction of the radiation changes. Sulfate, nitrate, and sea salt aerosols can be considered purely scattering at visible wavelengths[22]. Most aerosols types are relatively small, generally comparable to or smaller than the wavelength of visible light. As a result, their scattering effect is strongest in the shortwave spectrum and negligible at wavelengths longer than about a micron, with the exception of some large dust particles[23]. This spectral characteristic is distinct from many GHGs, which primarily absorb in the thermal infrared. The scattering increases the fraction of solar radiation reflected back to space, cooling the climate system. However, high surface albedo and the presence of clouds tend to reduce the net effect[12,14,24]. The vertical distribution of aerosols does not change their scattering effect; however, it can impact the radiation balance, for example, by changing the optical path over which water vapor absorption occurs[25] or the aerosol layer vertical location relative to clouds.

The scattering of aerosols also exhibits directional and polarization characteristics. In climate modeling, aerosols are usually assumed to be homogeneous spheres. Light interactions with spherical aerosols are governed by the Mie scattering solution; the degree of forward scattering is often represented by the asymmetry parameter (**Table 1**). Typical *g* values for aerosols are between 0.6 and 0.8, indicating primarily forward scattering[26]. Non-spherical particles are generally less efficient at backscattering, resulting in an increased *g* value, although the exact phase function can be much more complicated depending on the particle size, morphology, orientation, surface roughness, and so on[27,28]. In addition, radiation scattered by aerosols also exhibits polarization features, and the degree of polarization, as well as the phase function of the polarized component, are sensitive to aerosol type, especially aerosol shape, and absorption[29]. The linear depolarization ratio (**Table 1**) is zero for homogeneous spheres but can be much larger for non-spherical particles[28], which has been used by polarized lidars to distinguish non-spherical dust from spherical particle types[30]. The sky polarization pattern of scattered radiation by absorbing aerosols is mostly different from that of purely scattering aerosols and can be used to retrieve SSA in remote sensing applications[29,31].

Aerosol absorption effect

Some aerosols also absorb radiation. BC in aerosols makes the largest contribution to aerosol absorption[32], with nearly flat spectral behavior at visible wavelengths[33]. Dust and organic carbon aerosols strongly absorb in the Ultraviolet (UV) range, but this absorption quickly becomes negligible beyond ~600 nm[33,34]. Aerosol absorption leads to a positive radiation balance anomaly in the climate system and contributes to atmospheric warming. At the surface, however, the effect is generally still cooling, as the aerosol heating occurs higher in the atmosphere[35]. The atmospheric heating tends to increase atmospheric stability, degrade air quality, and slow the hydrological cycle[15,36-38] , which might induce a positive feedback of aerosol climate effects by decreasing aerosol wet deposition.

The effect of aerosol absorption also depends on the surface albedo and the presence of clouds, and can, therefore, be more complicated. In general, absorbing aerosols appear relatively darker over brighter surfaces, which tends to enhance the net effect of atmospheric absorption[12]. Under a cloudy sky, when the aerosols are located above clouds, the atmospheric absorption is greatly enhanced similar to the case of aerosols above a bright underlying surface. In contrast, aerosol-induced absorption will be weakened if the cloud is above the absorbing aerosol layer and even approaches zero as the cloud becomes thicker[39], as much of the incoming radiation will be reflected or attenuated by the cloud before reaching the aerosols[40,41].

The vertical distribution of absorbing aerosols is also critical in determining their radiative effect[42], and this factor is coupled with the surface albedo effect. Under clear sky, with a lightly-reflecting surface, if absorbing aerosols are located at a higher altitude, more incoming radiation is available there than at low altitudes for absorption[43], which induces a stronger warming effect. However, if the surface is sufficiently bright, such as over snow, the interaction of absorbing aerosols with surface-reflected radiation can become the dominant term, and thus a lower absorbing aerosol layer height might induce a stronger warming effect[25].

Absorbing aerosols themselves can change the underlying surface albedo by depositing on bright surfaces such as snow and ice[41,44,45]. This albedo reduction results in a warming of the surface, and contributes to processes such as Arctic ice melting[46] and Himalayan glacier retreat[47].

Combined effect of scattering and absorbing aerosols and their interaction

Purely scattering and light-absorbing aerosols frequently coexist, and because many of their radiative effects are opposite, their combined effect is complicated and uncertain. The ratio of aerosol scattering to absorption, characterized by the SSA, thus critically determines the magnitude and the sign of the aerosol forcing[48]. Typical SSA values found at worldwide locations are between 0.8 and 1 (Ref[26]), with lower values indicating a tendency towards a warming effect. The critical SSA, below which the overall aerosol effect will shift from negative to positive, is generally between 0.85 and 0.95 at 550 nm[13,49], and is typically higher over brighter surfaces or under cloudy sky conditions[15,50].

Absorbing and purely scattering aerosols interacting with each other produce additional complications. One scenario is the internal or core-shell mixing of BC with scattering aerosols that are often found in fossil fuel and biomass burning emissions; this internal mixing enhances BC absorption by 1-2.5 times, depending on the structure and morphology of the mixture[51-54]. Another scenario is through the vertical distribution; when scattering aerosols are concentrated below absorbing aerosols, the net aerosol absorption can be strengthened, and vice versa, analogous to the absorbing aerosol-over-cloud case. This vertical superposition of aerosol layers results in a combined direct forcing that might be different from the sum of the direct forcing of individual aerosol components. This nonlinear effect accounts for 14% of the total clear sky aerosol direct forcing globally, but can reach 100% regionally[55].

Aerosol–cloud interaction

Aerosols can serve as CCN and change cloud microphysical properties. Water soluble aerosols, such as sulfate, nitrate, sea salt, and secondary organic aerosols are more efficient CCNs than insoluble species (mainly dust and organic aerosols having high BC content), making them major

players in ACI[56]. The aerosol impact on warm clouds is complicated and involves several processes. The immediate effect of an increase in aerosol number concentration is to increase the number of cloud droplets and thus cloud reflectivity, known as the Twomey effect[57]. The Twomey effect typically cools the climate, as more cloud droplets tend to reflect more radiation back to space. Subsequently, cloud fraction and liquid water content (LWP) adjust in response to the increase of cloud droplets, which further impact the radiative effects of aerosol-perturbed clouds[9]. On the one hand, aerosol-perturbation tends to produce more cloud droplets with smaller size that take longer to precipitate, thereby increasing LWP and resulting in a cooling effect. On the other hand, these smaller droplets are easier to evaporate, which also enhances the mixing of clouds with ambient dry air. This effect reduces LWP and thus causes a warming effect[9]. Other factors, such as meteorological conditions, also complicate the LWP adjustment[58]. As a result, various relationships between cloud droplet number and LWP have been observed[59-62]. Globally, it is likely that the above two competing effects offset each other, so the overall effect of LWP adjustment in response to increased aerosols is weak[63].

Aerosols can also act as INPs and impact both ice and mixed-phase clouds, especially for the intermediate absorbing species of dust and organic aerosols[64,65]. BC is more absorptive and could also act as an INP, but this potential function is controversial and might be negligible relative to background INPs[66]. The aerosol–ice cloud interaction processes are more complicated and less well understood than warm cloud interactions, especially considering the competing effects of homogeneous freezing from liquid phase particles and heterogenous freezing by INPs. If INPs are added to an ice cloud already dominated by heterogenous freezing, they will lead to more ice crystals and possibly a warming effect. Alternatively, if the cloud process is dominated by homogeneous freezing, adding INPs will decrease ice cloud optical depth and likely induce a cooling effect because ice clouds (mostly cirrus) have a positive radiative effect[64].

Absorbing aerosols can interact with clouds through the semi-direct effect, which refers to the heating of the ambient atmosphere, changing the temperature profile[13,34]. The sign of this effect depends on the relative height of the aerosol and cloud layer, as well as cloud type. BC near low clouds causes the most warming, whereas BC below clouds or near high clouds can lead to a cooling effect[34,67]. Overall, the majority of the latest climate models show a cooling effect by

cloud adjustment in response to BC forcing. This cooling is due to the heating in the upper troposphere by BC, which decreases upper troposphere stability, decreasing high level clouds and increasing low level clouds[67-69]. BC internally mixed with sulfate or organic aerosols might decrease or prevent the activation of these particles to form cloud droplets[70].

Measurement of aerosol properties

Measurements are fundamental in investigating the role of aerosols in the climate system. Since the year 2000, aerosol observations from instruments on the surface and in space have greatly expanded. The observations have provided crucial data to understand aerosol optical and microphysical properties, space-time variability and climate effects[71-74].

Aerosol Measurement Techniques

Aerosol observation can be generally divided into *in situ*, surface-based remote sensing and space-borne remote sensing. *In situ* observations that sample the ambient air can accurately measure the mass concentration, scattering and/or absorbing properties, and more detailed information such as chemical composition, shape, mixing states, hygroscopicity, and particle size distribution that determines CCN concentration[75-77]. When instruments are deployed on aircraft or tethered balloons, the vertical distribution of these properties can also be measured[78]. Due to their high accuracy and comprehensiveness, *in situ* measurements often serve as benchmarks for remote sensing observation and model simulations[7].

Remote sensing instruments, which measure the transmitted and/or scattered radiances that contain the scattering and absorption characteristics of aerosols, are relatively easy to operate, albeit at the cost of decreasing the measurement accuracy and properties retrieved compared to *in situ* measurements. From the surface, the total column loading of aerosols (aerosol optical depth (AOD), **Table 1**), can be inferred from the attenuation of direct solar radiation from the top of the atmosphere to the surface. Combined with diffuse radiation measured at multiple angles and spectral bands, different inversion algorithms have been developed to retrieve column averaged aerosol SSA, phase function and size distribution (**Table 1**)[79-81] at moderate accuracy (for

example, ±0.03 for SSA), provided there is sufficient aerosol loading (typically AOD at 440nm > 0.4, but also depending on surface brightness and variability)[82]. NASA has operated a global surface network using sky scanning photometers configured in this fashion – the Aerosol Robotic Network (AERONET)[71], which has now grown to over 800 sites covering many aerosol source regions. The climatologies of aerosol retrievals from AERONET[26] provide important quantitative insights into the magnitude and spectral variability of aerosol SSA and particle size distribution for different types of ambient aerosol. There are also similar regional sunphotometer networks, such as the SKYNET in Asia and Europe[83] and CARSNET in China[84], that offer detailed information of regional aerosol optical properties.

Aerosols can also have complicated vertical distributions, with various profiles or layered structures of different types and sizes[85,86] (Supplementary Figure 1). Lidars that emit a laser beam and measure the backscattered light can obtain such vertical information. For the simplest backscatter lidar, the extinction profile can be retrieved only by assuming an extinction-to-backscatter ratio[87]. Profiles of aerosol scattering/absorption and some particle size information can be retrieved using more advanced approaches, such as the multi-wavelength Raman scattering or high-spectral-resolution techniques, or a combination of backscattering lidar and sunphotometers[88]. Examples of established lidar networks include MPLNET, which operates over worldwide locations[89], and EARLINET[90]and LALINET[91], which operate in Europe and Latin America, respectively. A more comprehensive understanding of aerosol properties within a single column can be estimated by combining passive sun photometers and active lidar[92].

Knowledge of global aerosol variability generally relies on the analysis of global satellite measurements in combination with constraints provided by global aerosol models. For passive satellite sensors that measure solar radiation scattered back to space by the Earth-atmosphere system, the channels are typically located in the visible-near-infrared spectrum, from 400 to 900 nm, to optimize the detection of aerosols. After accounting for surface reflectance, molecular scattering and gas absorption, the angular spectral radiation received by a satellite sensor is a function of column AOD, column-averaged SSA and column-effective particle single-scattering phase function[93]. For single-view sensors, AOD is the primary parameter to be retrieved, and SSA and phase function are usually prescribed based on some prior knowledge of the global

distribution of aerosol types[72]. Some particle size information, primarily fine and coarse mode fraction, can be inferred from the spectral dependence of AOD, but is only reliable over the ocean[72].

Attempts have also been made to retrieve SSA from satellites. One approach is to use UV observations, based on the theory that the underlying surface is sufficiently dark, and that aerosol absorption can measurably change the spectral dependence of upwelling Rayleigh scattering by atmospheric gas[94]. This method has been applied to the Total Ozone Mapping Spectrometer and Ozone Monitoring Instrument data, and has achieved qualitative agreement with surface observations[95]. However, the UV technique cannot detect aerosols near the surface[96], and is highly sensitive to aerosol vertical distribution[97]. Another approach is through multi-angle viewing geometry, represented by the Multi-angle Imaging SpectroRadiometer, which can constrain SSA by separating the directional reflectance of the surface and aerosols[98]. This technique, if combined with polarization, can increase the sensitivity of SSA retrieval[29,99,100] . This advantage has been demonstrated in the analysis of SSA retrieved by the Earth's first multi-angle, polarization imager in space, the Polarization and Directionality of the Earth's Reflectance (POLDER)[101,102], and by the Airborne Multiangle SpectroPolarimetric Imager[103].

The vertical distribution of aerosols is difficult to retrieve from passive sensors, and is usually prescribed in retrieval algorithms. The height of the absorbing aerosol layer can be detected using UV[104,105] radiance, near-UV polarimetry[106], or oxygen A or B band absorption[107-109] techniques under favorable retrieval conditions. The height of near-source wildfire, volcano, and dust aerosol plumes can be derived geometrically and mapped from the parallax of plume contrast features as observed in multi-angle imagery[110]. However, active space borne lidar is the only reliable method to obtain global information of aerosol vertical profiles on large scales, albeit with limited areal coverage. The only current space aerosol lidar in operation, Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP), is a two-channel backscattering lidar. By assuming the lidar ratio, it has the capability to derive aerosol extinction profiles from backscatter measurements, and some information about aerosol type based on depolarization sensitivity, surface type, and prior knowledge of aerosol type from surface measurements[73]. The

previous Cloud-Aerosol Transport System lidar[111] that was deployed on the international space station demonstrated the capability of diurnal sampling of clouds and aerosols[112].

Limitations of current aerosol observations

For each type of observation, there is a trade-off between accuracy, comprehensiveness and spatial representation. To constrain ARI, AOD, spectral SSA, the phase function and their vertical distribution are needed. In particular, both AOD and SSA need to be measured at an accuracy of ~0.01 to yield a global mean aerosol direct forcing uncertainty comparable to that of GHGs[11,31,113]. More detailed information is needed to constrain ACI. Model estimations require particle hygroscopicity and high-resolution size distribution (**Table 1**) down to sizes that cannot be distinguished with remote sensing alone[114]. For observational-based estimates, some proxies for aerosol size, such as aerosol index (AOD multiplied by the Ångström Exponent) have been considered[115] .

In situ observations are the only means to measure the complete set of parameters at the required accuracy to constrain aerosol forcing. For example, *in situ* instruments can measure aerosol scattering at an accuracy below 10%[116] and absorption at an accuracy below 20%[117,118]. For the information needed to estimate ACI, surface and aircraft platforms must be jointly deployed so that the horizontal and vertical distributions of aerosol size distribution, chemical composition, and cloud microphysical parameters are accurately measured. However, some large particles and INPs are still difficult to measure with the current in situ technique[119]. Notably, *in situ* measurements typically sample only over few meters in the vicinity of the instrument. Column integrated or averaged values of some aerosol parameters can be relatively well retrieved under clear sky conditions; however, certain assumptions must be made, and only the AOD retrieval can meet the required accuracy (~0.01). Knowledge of the aerosol vertical distribution offered even by active remote sensing is mostly limited to extinction, with weaker constraints on scattering and absorption properties. Furthermore, the most obvious drawback of in situ or surface remote sensing is the lack of global coverage, particularly for quantities that vary on large spatial scales. Also, the maintenance and calibration of instruments differs by site, creating problems in integrated usage of observations from different instruments.

Although satellite remote sensing can overcome the spatial coverage and calibration limitations, it has additional sources of uncertainty compared to surface observations. The most reliable parameter retrieved quantitatively is AOD; however, existing sensors can only achieve an accuracy of ~±0.02±20% of the AERONET AOD for a large-scale average[72], insufficient by itself to constrain aerosol forcing. SSA is rather difficult to retrieve to the accuracy needed to constrain aerosol forcing due to its high sensitivity to surface noise, even with current multi-angle, polarization designs. The accuracy of the aerosol extinction profile retrieved from space lidar depends on the assumed lidar ratio, which is prescribed according to limited surface and *in situ* observations and is significantly impacted by aerosol absorption[73]. The vertical distribution of absorbing aerosols and SSA cannot be retrieved quantitatively with satellite remote sensing.

In short, on a global scale, current remote-sensing observational techniques can only retrieve aerosol loading properties, such as AOD and the extinction profile, with moderate confidence, and the accuracy of aerosol microphysical properties retrieved by remote sensing alone is not adequate to constrain aerosol forcing to a level comparable to GHGs. The most critical factors, especially quantitative spectral SSA and its vertical distribution, are only available through limited *in situ* aircraft observations.

Climatology and temporal changes

Because of the short aerosol lifetime, the spatial distribution of different aerosol types is highly heterogeneous. Aerosol concentration over different regions also exhibits short- or long-term variability, produced by temporal changes of emissions, meteorological and climate conditions, and some extreme events.

Climatology of aerosol distribution

According to the CMIP6 model results, most aerosols are concentrated near their sources, which results in their highly variable horizontal and vertical distribution (**Fig. 1**). Sulfate aerosols are concentrated over East and South Asia, where anthropogenic emissions are the most intense.

These two regions also exhibit high BC and OC concentrations, together with South America, Southern Africa and Southeast Asia where biomass burning related BC and OC emissions can be intense[120]. Dust distributions emanate from deserts in North Africa, the Middle East and Central Asia, and extend to their surrounding areas. Sea salt is concentrated over the oceans. The SSAs at mid visible wavelength for dust, OC, and sulfate are typically around 0.97 at most wavelengths; sea salt has SSA essentially equal to 1. Although the simulated SSA for BC is below 0.2 (Fig. 1b, f, j, n, r), such low SSA cannot be observed as BC is typically mixed with the other species. These aerosol species combined yield relatively low SSA values found over East and South Asia, southern African, southeast Asia and southern South America, compared to other regions (Fig. 1v). The vertical distribution is expressed as the extinction weighted height (EWH, **Table 1**), which represents the layer that contributes the most to column AOD. Overall, EWH is lower near sources than remotely, which is reasonable as elevated aerosols typically travel longer distances. EWH at emission is the lowest for sea salt, and the highest for BC and OC over biomass burning regions and for volcanic emissions. Nonetheless, uncertainties in the above parameters can be considerable. For example, over high-aerosol-concentration regions such as North Africa and East Asia, the spread in AOD can be as large as 50% (supplementary Figure 2).

Temporal changes of aerosol scattering and absorption properties

To understand role of aerosols in climate change, it is necessary to monitor long-term changes in aerosol amount and optical properties. Statistically significant AOD trends since the late 1990s have been found in different parts of the world based on satellite retrievals, with declines of ~0.01 y^{-1} over eastern US, western Europe, and the tropical North Atlantic, strong increases of 0.02 y^{-1} over the Arabian Peninsula, and weak increases of ~0.01 y^{-1} over East China and the Indian subcontinent[121,122]. Since ~2006-2008, AOD over East Asia also began to decrease[123], leaving the Indian subcontinent and Arabian Peninsula as the only regions with significant upward AOD trends (Fig. 1d).

However, fewer results are available for SSA trends due to the lack of consistent, global observations. Limited SSA retrievals from surface sun photometers indicate that SSA has

systematically increased by as much as 0.03 decade^{-1} at sites in North America, Europe, and Northeast Asia from 2000 to 2013[50,124]. Updated records from AERONET still indicate statistically significant SSA increases over most European, North American, African and Asian sites (Fig. 1p). These SSA changes are of great importance for both the global radiation balance and satellite retrievals. In particular, a trend in aerosol SSA will affect the accuracy of any trend in AOD retrieved if SSA is assumed constant[125,126], which is the case in many passive satellite aerosol retrieval algorithms.

Insights on the change of aerosol properties during the COVID-19 lockdown

The lockdown induced by COVID-19 pandemic serves as a valuable opportunity to examine the effect of strict emission control measures that could take place in the future[127]. During the pandemic, extensive lockdown was implemented worldwide, which dramatically reduced the emission of anthropogenic aerosols and weakened their direct and indirect radiative forcing[128,129]. Over East Asia, anthropogenic aerosol emission decreased by 32%, resulting in an increase of surface shortwave radiation of 1.3 W m^{-2} (Ref[130]). Along with the decreased total aerosol loading, changes in aerosol composition, and thus their optical properties and radiative effects, have also been observed. In particular, absorbing carbonaceous aerosols experienced the most significant reduction in Asia (by 48%-70%)[145] and Europe (by 20%-40%)[131], whereas secondary aerosols that are mostly scattering even increased due to the increase in tropospheric O_3 associated with the reduction of NO_x emissions. Specifically, a 65% reduction of NO_x during the lockdown has been observed in East China, making the atmospheric oxidizing capacity at the peak of secondary aerosol formation and increasing O_3 production by as much as 100%[132]. Specifically, the combined effect was an increase in the reflectivity of aerosols, or increased SSA[133]. Although the aerosol forcing induced by these short-term perturbations is minor compared to the baseline condition[134], this extreme case provides insights into future changes of aerosol scattering and absorption under different economic growth scenarios.

Future projections of the change of aerosol properties

Global and regional aerosol concentrations and their optical properties might substantially change in the future. On one hand, emission-intense countries, such as China and India, have implemented or will likely implement stricter air quality control regulations in an effort to alleviate air pollution, resulting in an overall decrease in AOD (Fig. 1h). Anthropogenic aerosols have already decreased markedly in China since 2008, with a 0.2 decade^{-1} downward AOD trend[135], a decrease that will likely continue. Aerosols in South Asia are projected to decrease at least around 2050, and could be reduced by as much as 50% by 2100[136]. On the other hand, emissions might shift to currently under-developed regions of Africa, Central and South America, and Southeast Asia, especially under the shared social-economic pathway (SSP) "regional rivalry" scenario (SSP3)[136]. Emission of natural aerosols, such as dust, biogenic aerosols formed from biogenic organic vapors, and black and organic carbon aerosols from wildfires, seem also likely to increase due to the increase of dry and hot extremes and the increase of wind speed under global warming[137,138,139]. Sea salt aerosol emission tends to increase with a warmer ocean surface[140,141], although the amount might be small and varies in different models[4,142,143].

More importantly, changing anthropogenic and natural emissions alter not only total aerosol loading, but also aerosol optical properties, such as SSA. For example, with the adoption of clean energy as predicted by SSP[144] experiments, the fraction of absorbing aerosols is likely to decrease, which means an increase of SSA (Fig. 1). However, increased wildfire emissions[145] means that more light-absorbing aerosols might be released, leading to a darkening aerosol effect regionally[138].

In the future, it has been projected that aerosols might induce a negative radiative forcing up of to -0.09 W m^{-2} due to increased aerosol concentration caused by reduced precipitation[139]. Dust aerosol alone can account for -0.04-0.02 W m^{-2} (Ref[146]). Strict emission controls in polluted areas such as China imply a possibly weakened projected forcing by anthropogenic aerosols, which then positively contributes to future warming because well-mixed GHGs will continue to increase[147,148]. However, there is little confidence about how SSA and its impact on the projected forcing will change in the future, leaving a large uncertainty for climate prediction.

Aerosol radiative and climate effects

There have been many attempts to quantify the different effects of aerosols, as well as their impacts on global and regional climate[8]. This section summarizes progresses in these areas, and discusses the remaining uncertainties.

Estimating aerosol radiative forcing

Aerosol radiative forcing (ARF) can be estimated readily using interactive chemistry-climate models by comparing the radiative fluxes calculated using present day and pre-industrial aerosol emissions (Box 1). Many projects have been carried out to intercompare the ARF estimated by different models, such as the CMIP[149], AeroCom[150] and ACCMIP[151] projects. The inter-model ARF spread is often presented as its uncertainty[4], although it actually represents model diversity, which is generally a lower bound on uncertainty.

Global ARF has also been estimated purely from satellite observations. The direct forcing is easier to estimate, which is based on constructing the relationship between anthropogenic AOD (sometimes taken as fine particles, though this does not account for natural biomass burning, sulfate, secondary organic, and biogenic particles, nor for the fine-particle part of the dust and marine particle size spectrum) and radiative flux, using collocated radiation and aerosol retrievals by satellites[152]. However, this estimate is confined to clear sky conditions, which leads to an overestimate of the overall negative forcing, and also neglects the positive forcing by absorbing aerosols above clouds. Estimation of cloudy-sky direct forcing requires the vertical profiles of both aerosol extinction and SSA, which is challenging. To estimate the forcing from aerosol-cloud interaction, the relationship between retrieved cloud properties (such as cloud reflectance, effective radius and liquid water content) and AOD[153] or more precise aerosol proxies[115,154] needs to be established. However, the cloud and aerosol properties are typically from nearby but different pixels, as cloud and AOD retrievals cannot be strictly collocated, which creates uncertainty in the results. The effective aerosol forcing (ERF) includes aerosol-radiation interaction (ERFari) and aerosol-cloud interaction (ERFaci) effects, and allows shorter time-scale atmospheric elements to adjust to equilibrium. Observation-based ERFari is slightly

larger than model-based, whereas the ERFaci estimated by the two approaches largely converge (IPCC AR6). In particular, applying the decomposition method that separates ERFaci into instantaneous forcing and rapid adjustments appears to improve the convergence of model and observation-based ERFaci[155].

Aerosol radiative forcing and its uncertainties

The strongest aerosol forcing is found in the northern hemisphere, primarily in the mid-latitudes where most anthropogenic aerosol emissions are concentrated (Fig. 3). The largest negative forcing, reaching below -5 W m^{-2}, is found over East Asia. Positive forcing mostly occurs over the desert regions, including northwest China, the Sahara and the Middle East, due to the high surface albedo. The forcing of BC and sulfate is representative of most absorbing and scattering aerosols, respectively, except where iron oxide absorption from mineral dust particles dominates. The sign of the forcing is mostly positive for BC but negative for sulfate, especially over land. The all-sky forcing is nearly three times as much as the clear-sky forcing based on these simulations. Considering that all sky ERFari is typically smaller than that under clear sky, the more negative all sky forcing indicates a significant contribution from ERFaci. However, the BC forcing is more negative for all-sky than clear sky conditions over the ocean, implying negative forcing from the cloud adjustments to BC forcing. Note that the spread in the ERF among different models can be considerable (Supplementary Figure 3), indicating large uncertainty.

Over the past 20 years, numerous attempts have been made to estimate different components of aerosol forcing based on observation, modeling or both (Fig. 4 and supplementary Tables 1&2). Earlier observation-based estimates gave more negative direct forcing under clear-sky conditions but reached consistency with model estimates at ~-0.7 W m^{-2} after about 2007, driven largely by global, monthly satellite AOD products. All-sky direct forcing is estimated at about half as large as clear-sky, as cloud scattering significantly masks the scattering of aerosols. The estimated magnitude stays relatively stable after 1995, varying between -0.5–0 W m^{-2}, with four estimates after 2020 arriving at ~0.25 W m^{-2} (Fig. 4b and Supplementary Table 1). The indirect forcing estimates exhibit greater uncertainty and more fluctuation over time, although the smaller effects in earlier estimates are associated with calculations of only the albedo effect. The latest model-

554 based estimation indicates an indirect forcing of ~-0.75 W m^{-2}, which is slightly weaker than the
555 IPCC AR6 result that combines model and observations[4]. There are far fewer estimates of the
556 semi-direct effect of absorbing aerosols than the direct and indirect effects. The mean values of
557 different reported estimates fluctuate between -0.5 W m^{-2} and +0.2 W m^{-2}, with large error bars.
558 This effect, however, is likely a minor contribution based on current understanding[4].
559
560 An encouraging result is the decrease in uncertainty estimates of the direct aerosol forcing over
561 time (Fig. 4a&b), especially for the all-sky direct radiative forcing, which tends to stabilize with
562 an uncertainty of ~±(0.1-0.4) W m^{-2}. The indirect forcing uncertainty is still large, reaching or
563 exceeding 100%. Large uncertainty is also present in estimates of the semi-direct effect (Fig. 4e
564 and Supplementary Table 2). Limited global estimates indicate an averaged negative forcing in
565 the range of – (0.4-0.1 W m^{-2}). Yet the uncertainty can well exceed the mean and change the
566 forcing to weakly positive.
567
568 Different methods are used to estimate the uncertainty of different forcing components, meaning
569 that reported uncertainties might not be directly comparable. In particular, many model-based
570 estimates consider the multi-model spread (or the model diversity) as the uncertainty, which is
571 likely to underestimate model uncertainty, especially as model inter-comparison projects
572 encourage modelers to adopt similar assumptions. Overall, consensus about the total aerosol
573 forcing uncertainty shows some decrease over time[9]. However, substantial differences still exist,
574 especially for the aerosol-cloud interactions.
575
576 Sensitivity experiments indicate that uncertainties in SSA and the vertical distribution of
577 absorbing aerosols contribute the most to the uncertainty of direct forcing. In particular, a ±0.03
578 uncertainty in SSA, which is already the lower uncertainty limit of surface remote sensing, can
579 contribute 50% of the total direct forcing uncertainty[11]. The vertical distribution of absorbing
580 aerosols can also contribute to ~10-20% of the direct forcing uncertainty globally[60]. With respect
581 to modeling, the assumed aerosol mass extinction and absorption efficiencies, which are needed
582 to convert the simulated aerosol mass into optical properties, show large variations among
583 different models but are poorly constrained by observations[7].
584

Uncertainty in the aerosol indirect effect can be associated with both the lack of understanding of aerosol and cloud microphysical processes (such as precursor emissions, chemical reactions, nucleation and growth and hygroscopicity), incorrect/coarse representation of these in models, and the role of transport and transformation processes. For example, models disagree in characterizing the differences in the aerosol size distributions between present-day and pre-industrial conditions, and mixing states of absorbing and scattering aerosols, which leads to different AOD, CCN concentrations and cloud optical properties. Aerosol vertical distribution determines the involvement of aerosol in cloud processes but is poorly constrained. Moreover, microphysical processes in ice or mixed-phase clouds are very unclear. The role of different INP types, in particular, BC as an INP, is debated but not yet well constrained by observations. Competition between homogeneous freezing and heterogeneous nucleation before adding aerosol particles might lead to opposite final forcing estimates[64].

The uncertainty in semi-direct aerosol forcing arises mainly from the vertical distribution of BC, and sometimes the cloud water content. A negative forcing is produced below a relatively high BC layer, which causes a cooling and an increase of low-level clouds. However, the BC loading in the upper troposphere in models might be biased high compared to observations[156], and so could be responsible for the simulated negative forcing.

Knowledge of preindustrial aerosols is also critical in estimating anthropogenic contributions to aerosol forcing. Different estimates of preindustrial aerosol emissions can lead to 15-60% variability in aerosol forcing estimate[157], and contribute to ~45% of the total aerosol forcing uncertainty[158]. However, preindustrial aerosol concentration cannot be measured and is therefore typically inferred from measurements in remote areas[157,159], contributing further to forcing uncertainty[160]. Other external factors, such as in radiative transfer calculations, absorbing gas profiles, surface albedo and dynamical background also contribute to the model-estimated forcing uncertainty[146,161,162].

Global climate impacts

Although the largest concentrations of aerosols and their instantaneous forcing occur near their sources, their impact on the energy balance and climate can be more geographically extensive and profound. They introduce perturbations in the radiative energy balance at the top-of-atmosphere and surface, both of which are spatially inhomogeneous. The heterogeneous energy perturbation by aerosols includes a pronounced interhemispheric asymmetry that has particular implications for the climate in both hemispheres[163-165]. They directly change the atmospheric and oceanic circulation by altering the vertical and horizontal thermal structure on hemispheric scales. By inducing an SST response similar in pattern but opposite in sign to that of GHGs[166], aerosols might have contributed to the global warming slowdown from 2000-2015[167]. The aerosol-induced SST changes further result in ocean circulation anomalies and can impact the Atlantic Meridional Overturning Circulation[168]. In contrast to the greenhouse gas effects on climate, which tend to strengthen the hydrologic cycle, aerosols tend to weaken it[148,169-171].

Aerosols also contribute to some large-scale climate anomalies such as Arctic warming[172], extreme Northern Hemisphere winters[173], precipitation reduction in the Northern Hemisphere[174], as well as hemispherically asymmetric rainfall trends[175]. In these effects, absorbing and scattering aerosols typically induce distinct changes in the global SST pattern[176], energy budget[177], hydrological cycle[15] and climate responses[178].

Regional climate impacts

The local cooling or warming induced by aerosols can form a temperature gradient between polluted regions and the surrounding areas, which leads to a perturbation or shift in regional circulation. The most representative case is the monsoon scenario, especially in South and East Asia. Aerosol cooling at the surface reduces the temperature contrast between land and ocean, weakening the South and East Asian monsoons[171,179-181]. The fraction of absorbing aerosols plays a critical role in the monsoon shifts[182]. BC mixed with organic carbon forms the so-called Atmospheric Brown Clouds (ABC), first discovered over the Indian Ocean in air transported from the Indian subcontinent[169]. These aerosols heat the atmosphere above the ocean, reducing the temperature gradient between ocean and land, and weakening the South Asian monsoon. BC can also substantially contribute to the weakening of the African and South American

monsoons[183,184]. Both local and remote aerosols can affect regional changes in the hydrologic cycle in Asia[185].

On even smaller scales, aerosol cools the surface and increases the lower atmospheric stability. Absorbing aerosols are more efficient in this process, although the overall effect is usually larger for scattering aerosols due to their higher loading[186]. This process might decrease the planetary boundary layer height, resulting in the so-called aerosol-boundary layer feedback[187], a process thought to significantly contribute to severe pollution events in China[37].

Summary and Future Prospects

Aerosols are known to have an important role in the regional and global climate system. Tremendous efforts have been made, in both observation and modelling, to advance understanding of the mechanisms by which aerosols impact the climate system, and to increase the accuracy of aerosol forcing estimates. Many physical processes through which aerosols impact the global atmospheric and oceanic circulation, and regional climate anomalies, have been identified. In terms of forcing estimation, some convergence has been achieved between model-based (bottom up) and observation-based (top down) results. Unfortunately, the discrepancies among different models are still considerable. The latest models still suggest an inter-model forcing spread of ~50%[188], and the actual model uncertainties are probably larger. When the models are tuned to fit the observed historical increases in temperature, this large uncertainty also impacts climate sensitivity estimates, which translates into a large uncertainty in climate projections. As such, confidence is still low in quantifying the role of aerosols in the climate system.

There is now compelling evidence that human influence has warmed the atmosphere, ocean and land. Drastic reductions in global GHG emissions are required to keep the surface temperature increase within 2°C by the end of the 21st century[4]. It is imperative to understand the aerosol component of climate change better so that the required constraints on GHG concentrations can be defined accurately. There is thus urgent need to improve understanding of the role of aerosols in the climate system.

The fraction of aerosol absorption relative to scattering, expressed as the SSA parameter, is one the of most critical factors in quantifying the role of aerosols in the climate system. Uncertainties in SSA as well as its vertical distribution contribute substantially to uncertainties in ARI. The fraction of scattering and absorbing aerosols also impacts ACI estimates. However, there is no consistent global monitoring of SSA, leaving this parameter poorly constrained in climate models, which leads to further uncertainties in aerosol forcing estimates. In addition, aerosol size distribution, or a proxy of it, is critical in quantifying ACI. Better constraining pre-industrial and present-day aerosol SSA and size distribution from observations, and better representation of these in climate models, are thus warranted to quantify aerosol forcing.

Future observation needs

Future aerosol observation strategies focus on expanding the space of parameters that can be retrieved by satellite sensors, and improving the measurement accuracy. For this purpose, high accuracy, multi-angle polarization measurements could be a promising technique. The NASA's Glory mission[189] carrying the first high accuracy polarimeter with the potential to produce a more nuanced picture of aerosols, unfortunately failed at launch. China launched the country's first multi-angle polarimeter – Directional Polarimetric Camera (DPC) in 2018, with similar settings as POLDER, but it stopped service in 2020. AOD and the fine mode fraction over land have been retrieved from DPC with reasonable accuracy[86]. Two other DPCs on different platforms will be launched between 2022 and 2026. NASA's HARP multi-angle, multi-spectral polarimeter imager was launched on a CubeSat in April 2020[190], and an updated version HARP2 will be launched on NASA's PACE space mission[20]. Two more advanced multi-angle polarimeters, the 3MI sensor by European Space Agency[18] and the SPEXone sensor on PACE, are both scheduled to orbit in the next two years. The increasing number of polarimeters and rapidly growing volume of polarimetric data, especially from orbital instruments, along with sustained advances in forward modeling, retrieval methodologies, and algorithms, serve as a compelling reason to envision multi-angle polarimetry as the main tool for global aerosol monitoring and characterization[19].

To derive the vertical distribution of more parameters, especially SSA, a multi-wavelength, hyperspectral resolution technique that isolates molecular scattering from aerosol scattering has been proposed, which can provide information on the profiles of aerosol extinction, absorption and size distribution[191]. The lidar-based technique has limited spatial sampling compared to imagers, but it promises to be implemented on satellite platforms, and many aircraft-based experiments have already been carried out[21,181].

A combination of the above-mentioned multi-angle, polarized passive sensor and HSRL lidar, and possible multi-channel spectrometers, can yield invaluable insights into how different types of aerosols interact with radiation, clouds and the climate system. Multiple campaigns under NASA's Aerosol-Cloud-Ecosystem mission demonstrated the advantage of such combinations of sensors in retrieving horizontal and vertical variability of key aerosol parameters[192]. Several satellite platforms in development offer this configuration, including NASA's PACE mission[193], Aerosol-Clouds, Convective-Precipitation mission[194] and Europe's EarthCARE mission[195].

However, satellite remote sensing must be combined with *in situ* observations to reach the required accuracy of different parameters for constraining aerosol forcing, and to obtain key parameters that cannot be retrieved from remote sensing alone. Accurate measurement of key aerosol parameters likely requires the establishment of an international network with a routinely operating, relatively small aircraft *in situ* sampling program that is designed to provide the key particle microphysical property information unobtainable or inadequately constrained by remote sensing, for the major aerosol air masses identified by chemical transport models[17]. Such data would improve both model and satellite-retrieval assumptions about particle size distributions and CCN properties, hygroscopicity, SSA, as well as the mass extinction efficiencies used to relate satellite-derived AOD to aerosol mass in climate and air quality models. Information optimization techniques can be used to select sites where the measurements can contribute the most to reducing forcing uncertainty[187]. Urgently needed are highly accurate and efficient retrieval algorithms designed in line with future satellite sensors, and data synergy techniques that maximize the information of multi-sensor datasets and *in situ* observations.

Given the difficulty of simulating all aerosol-cloud interactions, reliable determination of the ERFaci requires global monitoring of aerosol and cloud properties. Planetary missions have demonstrated the ability of a high accuracy (~0.1%) polarimeter to obtain accurate microphysical data such as refractive index and size distribution on aerosols and cloud-top particles. A second instrument – a Michelson interferometer that simultaneously measures the spectrum of heat radiation emitted by Earth at the same spatial resolution as the polarimeter – can provide water vapor, ozone and temperature in several layers of the atmosphere, as well as cloud-top temperature and cloud properties to a greater depth than reached by the polarimeter. Along with a simple high-resolution camera, these two instruments on a small satellite would provide a monitoring capability crucial to help understand aerosol cloud interactions[196]. Such remote sensing must be complemented with intensive aircraft field campaigns aimed at better characterizing the microphysical processes involved in aerosol-cloud interactions, so that parameterizations of these processes in models can achieve greater accuracy.

Future modeling needs

Constraining ARI in models requires accurate simulation of the emissions (including primary aerosols and the precursor gases for secondary aerosols), chemical reactions, particle growth, transport, and removal, as well as the optical and microphysical properties of different aerosols species. It is necessary to take advantage of different types of observations, especially the advanced sets of satellite and *in situ* observations envisioned in the previous subsection, to improve the representation of aerosol extinction, absorption, size distribution, mixing and other related processes in climate models. Data assimilation techniques have been extensively used that not only assimilate AOD[197] retrievals, but also aerosol absorption[198] and SSA retrieved by POLDER[199], and aerosol extinction profiles retrieved by CALIOP[200,201]. These practices effectively improved simulation results, yielding several global aerosol reanalysis datasets[202,203]. Nonetheless, the retrieved products themselves rely on model assumptions and their uncertainties will negatively affect the assimilation results. A comparison of the radiances derived with assimilated aerosol properties and those observed by satellites can test some of the assumptions of aerosol microphysical properties between the model and the retrieval algorithm for inconsistency. Such practices will facilitate the interpretation of the measurements, and possibly

achieve a closed assimilation-retrieval system that could be beneficial both for remote sensing and climate modeling.

Constraining ACI in climate models is more challenging. More detailed aerosol schemes have been implemented in many climate models, including explicit consideration of soluble and insoluble species, secondary organic aerosols, the parameterization of aerosol nucleation schemes and CCN activation, for example[204-206]. Cloud microphysics parameterizations have also been refined and ice or mixed phase cloud schemes have been included[207,208]. Although these improvements can lead to better agreement with observations, they might not lead to improved climate projections, given the large uncertainties in particle microphysical properties and in the processes involved in ACI. For example, the increased cloud feedback in the latest climate models, partly related to the updated aerosol-cloud interaction schemes, leads to unrealistically high climate sensitivity[5,209], highlighting both the complexities and insufficient understanding of ARI and ACI processes, of which the ice cloud microphysics remains the least understood. The targeted in situ observations discussed here must be implemented and carefully analyzed to further clarify the roles of dust, BC, and organic aerosols as INPs, upon which ice cloud microphysical schemes can be improved. As computational resources become yet more abundant, implementing cloud resolving simulations globally would become possible, which can avoid many parameterizations and provide better constraints on ACI and climate sensitivity, though the mechanisms themselves must also be better understood to be modeled accurately.

With a series of planned space and surface observation missions globally, a large increase in the number of observations is anticipated. Combining with systematic characterization of particle microphysical properties, particle aging and ACI processes by *in situ* measurement, and improved integration of satellite and *in situ* measurements with modeling, there is hope that the uncertainties in aerosol forcing and climate effects that have persisted for decades will greatly improve in the near future.

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Author contributions

J. Li, B. E. Carlson, A. A. Lacis and Y. Yung led the Review. J. L. wrote the initial draft and prepared Figure 2, Box 1 and Supplementary Figure 1. L. Zhang prepared Figure 1, Figure 4, and Supplementary Figures 2 and 3. Y. Dong prepared Figure 3 and Table 1. All authors contributed to the manuscript preparation, interpretation, discussion, and writing.

Competing interests

The authors declare no competing interests.

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Data availability

Coupled Model Intercomparison Project Phase 6 (CMIP6) data used in Figs. 1 and 3 are from the Earth System Grid Federation, available at https://esgf-node.llnl.gov/projects/cmip6 . AOD and SSA data used in Fig. 1 are from Aerosol Robotic Network (AERONET), available at https://aeronet.gsfc.nasa.gov/.

1354

1355 **Table 1.** Definitions, meaning and radiative effects of some basic aerosol optical and

1356 microphysical properties

Variable	Definition	Physical Meaning	Relevant radiative effect
AOD (Aerosol Optical Depth, τ)	The integration of aerosol extinction coefficient (β_e) from top of atmosphere (TOA) to the surface.	Quantity of direct solar radiation prevented from reaching the ground by the total column aerosol loadings.	Direct effect.
SSA (spectral Single Scattering Albedo, ω)	The ratio of scattering coefficient (β_s) to β_e.	How absorptive and reflective aerosols are.	Direct effect and semi direct effect.
Phase function ($P(\cos(\Theta))$)	The radiance intensity at a certain direction Θ relative to the integral scattered radiance at all angles.	The dependence of scattered radiance on scattering angle (Θ), representing which direction aerosols scatter the light.	Direct effect.
Polarized phase function ($q_a(\Theta)$)	The parameter in the scattering phase matrix to describe the change of polarization of light being scattered.	The polarization properties of scattering light, which is sensitive to aerosol size distribution and complex refractive index.	Direct effect and indirect effect.
asymmetry factor (g)	The integration of $P(\cos(\Theta))$.	The proportion of forward scattering.	Direct effect.
Size distribution	A function that describes the relative amount of particles in each bin of size.	Scattered radiance intensity is directly affected by particle sizes.	Indirect effect.

Effective radius (r_{eff})	A weighted mean of the size distribution of aerosol particles.	Simplifies the expression of aerosol distributions, which is useful for inter-comparisons of different aerosols and their retrieval.	Indirect effect.
AE (extinction Angstrom Exponent, α)	A indicator describing how extinction AOD depends on the wavelength of the light	Aqualitative indicator of aerosol particle size distribution. Typically, larger particles induce lower α. Also for direct forcing spectral dependence	Direct and Indirect effect.
Scale height (H_p)	The distance over which aerosol concentrations decrease exponentially.	The vertical distribution is also important for assessing aerosol transports and material fluxes.	Direct effect and indirect effect.
Hygroscopic growth factor (f_D(RH))	The ratio of the wet particle diameter $D(RH)$ at a certain relative humidity to the corresponding dry diameter $D(dry)$.	Determines the hygroscopic properties of aerosol particles and their influences, needed for CCN behavior and to interpret ambient AOD measurements.	Direct and Indirect effect.
Extinction Weighted Aerosol Layer Hight (EWH)	A weighted mean of aerosol layer height using the extinction of each layer as the weight.	It is a proxy of aerosol vertical distribution/profile.	Direct, semi-direct and indirect effect.

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Figure. 1. Climatologies and trends of aerosol parameters. **a|** Left column: climatological spatial distribution of aerosol optical depth (AOD) calculated over 2010-2014 for black carbon, dust, organic carbon, sulfate, sea salt and all aerosols combined. The numbers in the top right indicate the global mean value. Middle column: As in the left column, but for single scattering albedo (SSA)[71]. Right column: as in the left, but for extinction weighted aerosol layer height, calculated as a weighted average of the height of all layers using the total extinction of each layer as the weight (Table 1). **b|** top: linear AOD trends calculated over 2002-2016 (left), and changes in AOD by 2100 relative to 2014 for SSP 126 (middle) and SSP 370 (right) using the average of 14 CMIP6 models[210]. Values in the top right of each panel indicate the globally-averaged trend or change. Bottom: as in top but for SSA. These maps provide a comprehensive understanding of the sources, distributions, optical properties, and trends of aerosols.

Figure 2. The radiative effects of aerosols. Schematic of radiative effects of absorbing and scattering aerosols, as well as their interactive effects. Dark dots indicate absorbing aerosols and gray dots indicate scattering aerosols. Scattering aerosols induce negative forcing by directly reflecting sunlight and interacting with clouds; absorbing aerosols in general have warming effect, although its interaction with clouds might produce slight cooling. The interaction between scattering and absorbing aerosols enhances the absorption and thus the warming effect.

Figure 3. Top row: all sky effective radiative forcing (ERF) for all aerosols (left) black carbon aerosols (middle) and sulfate aerosols (right). ERF is calculated by averaging the results of 14 CMIP6 models, as the difference in net top of atmosphere radiative flux between the aerosol forcing run for the year 2014 and preindustrial control run. The numbers in the upper right corner represent the multi-model mean and standard deviation. Middle row: as in top, but for clear sky ERF. Bottom row: as in top, but for cloud sky ERF. The climate forcing for scattering and absorbing aerosols are opposite over most regions, and that for absorbing aerosols is highly uncertainty.

Figure 4. Aerosol radiative forcing and its uncertainty. a| Published estimates of clear sky direct radiative forcing (DRF) since 1995 (Supplementary table 1**)**. b| As in a, but for all sky DRF. c| As in a, but for indirect radiative forcing (IRF). d| As in a, but for total radiative forcing

(TRF) which includes aerosol-radiation interactions and aerosol-cloud interactions. e| As in a, but for semi-direct forcing.

Box 1: Modelling of Aerosol Radiative and Climate Effects

Zero-D Models

The most straightforward, intuitive effect of aerosols on the climate system is a change in the planetary albedo, whose first order effect can be estimated using the zero-D model by considering the Earth as a spherical blackbody as a whole.

The impact of aerosols is reflected in the change of planetary albedo (A). For clear sky, aerosols can induce positive or negative changes in A that correspond to negative or positive changes in Earth's temperature, respectively, depending on aerosol single scattering albedo and surface reflectance. Under a cloudy sky, the aerosol impact on cloud albedo also needs to be estimated. The impact is usually positive, but can be negative for regions with large fraction of absorbing aerosols or low SSA. However, the simple zero-D model cannot provide estimates of surface climate or reflect the dependence of aerosol forcing on its vertical distribution.

1-D Models

Because of the limitations of zero-D models, they have been extended to column radiative transfer models, in which the aerosol parameters, including AOD, SSA, phase function or *g* (Table 1) are prescribed for each layer. By comparing the radiative flux at TOA or the surface with and without aerosols, ARF can be calculated. The effect of clouds can also be accounted for by parameterizing their optical properties. These models work well for sensitivity experiments, but are not suitable for global analyses.

General Circulation Models

Aerosol modules are thus incorporated into General Circulation Models (GCMs) to investigate their impact on global climate. Early GCMs usually adopted an offline approach, whereby the aerosol mass density fields were simulated using a chemical transport model (CTM). Then by assuming complex refractive indices, size distributions and hygroscopic growth factors for each species, their masses were converted into optical properties that participate in the radiative transfer calculation. The CTM models the emission, chemical reaction and gas-to-particle conversion, transport, and dry and wet deposition processes of different aerosol species, and can

be driven by the GCM simulated meteorology fields or by reanalysis meteorology. This configuration allows aerosols to influence radiation and dynamics in the model, but the aerosol processes are not interactive with the dynamics.

New generation GCMs are mostly two-way coupled, in which the chemistry module is driven by the simulated meteorology and the calculated aerosols feedback to the radiation and dynamics for each model step. Both the impact of aerosols on climate and the impact of climate on aerosols are simulated in this way. The global ARF is typically estimated as the difference between the TOA or tropopause net flux simulated using present day aerosol emissions and that using preindustrial aerosol emissions (usually for the year 1750).

Earth System Models

The newest global models—global earth system models (ESMs)—incorporate many real-world processes beyond the general circulation, such as those associated with vegetation, sea ice, and land ice. Aerosols are mostly also interactive in the ESMs. More realistic ARF estimations can be achieved using these models, as well as estimations of the impact of aerosols on many regional and global physical processes.