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Aerosols and Their Impact on Radiation, Clouds, Precipitation, and Severe Weather Events

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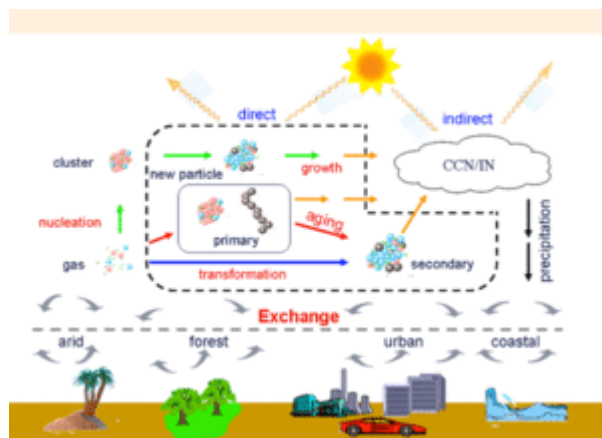
Summary and Keywords

Aerosols (tiny solid or liquid particles suspended in the atmosphere) have been in the forefront of environmental and climate change sciences as the primary atmospheric pollutant and external force affecting Earth's weather and climate. There are two dominant mechanisms by which aerosols affect weather and climate: aerosol-radiation interactions (ARIs) and aerosol-cloud interactions (ACIs). ARIs arise from aerosol scattering and absorption, which alter the radiation budgets of the atmosphere and surface, while ACIs are connected to the fact that aerosols serve as cloud condensation nuclei and ice nuclei. Both ARIs and ACIs are coupled with atmospheric dynamics to produce a chain of complex interactions with a large range of meteorological variables that influence both weather and climate. Elaborated here are the impacts of aerosols on the radiation budget, clouds (microphysics, structure, and lifetime), precipitation, and severe weather events (lightning, thunderstorms, hail, and tornadoes). Depending on environmental variables and aerosol properties, the effects can be both positive and negative, posing the largest uncertainties in the external forcing of the climate system. This has considerably hindered the ability to project future climate changes and make accurate numerical weather predictions.

Keywords: aerosol, radiation, cloud, precipitation, severe weather, lightning, thunderstorm, hurricane, typhoon, tornado

Introduction

Aerosols are suspended solid or liquid particles in the atmosphere. Ubiquitous aerosols have long been recognized as potential agents for altering the Earth's climate through their interactions with radiation, cloud composition and structure, water and air quality, and weather and climate on regional and global scales. Knowledge of aerosol formation and growth processes, and thus their physical, chemical, and radiative properties, is essential to study these interactions. Aerosols can be directly emitted (primary) or formed through the gas-to-particle conversion process in the atmosphere (secondary) (Zhang, Khalizov, Wang, Hu, & Xu, 2012; Zhang et al., 2015; also see Fig. 1). Primary aerosol sources include emissions from sea spray, volcanoes, combustion, road or wind-blown dust, and plants, while secondary formation processes include nucleation and growth by multiphase chemical processes. There are several aerosol nucleation mechanisms: binary nucleation (Weber, McMurry, Eisele, & Tanner, 1995), ternary nucleation (Korhonen et al., 1999), and modified ternary nucleation (Kulmala, Pirjola, & Mäkelä, 2000). After nucleation, particles may grow in size via several mechanisms, as summarized by Kulmala (2003). Currently, aerosol nucleation and growth processes are not fully understood. Primary and secondary particles undergo chemical and physical transformations and are subject to cloud processing and transport in the atmosphere.



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Figure 1. Formation of atmospheric aerosol particles and their impact on climate due to their direct and indirect effects through ARI and ACI (from Zhang et al., 2015).

By their origins, aerosols are also classified as natural or anthropogenic. Natural ones include dust, sea salt, smoke from forest fires, and biogenic and biological particles, while anthropogenic aerosols (AAs) include sulfate, nitrate, organics, and soot, which is also known as *black carbon (BC)*. BC aerosols result from the incomplete combustion of hydrocarbons (e.g., from internal combustion

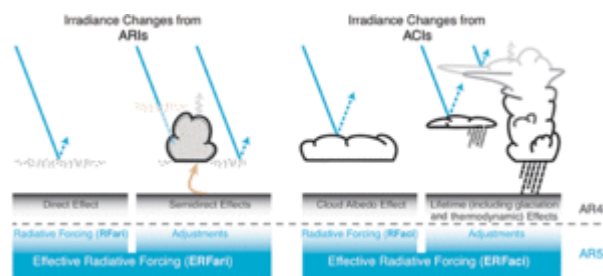
engines, coal-firing power plants, burning of agricultural residues, and home cooking). BC aerosols are in increasing abundance in Asia as the demand for energy from a rapidly growing population and economy is growing. By their radiative properties, aerosols range from strongly absorbing (e.g., BC) to nonabsorbing or conservative scattering (e.g., sulfate). By their hydrophilicity, aerosols may be hygroscopic or hydrophobic. This

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determines their ability to serve as cloud condensation nuclei (CCN) or ice nuclei (IN). The effects of aerosols on weather and climate depend critically on these properties.

Aerosol-Radiation Interactions (ARIs)

Aerosols alter the radiation budget by scattering, absorption, or both, which reduces the amount of solar energy reaching the ground and increases the radiation absorbed in the atmosphere and reflected back to space. The effects of aerosols on radiation are generally referred to as *aerosol-radiation interactions (ARIs)* (IPCC, 2013).



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Figure 2. Schematic of aerosol radiative effects due to both aerosol direct radiative effects via scattering and absorbing radiation (left) and by altering cloud properties via serving as cloud condensation nuclei (right); from IPCC (2013).

This term refers to any changes associated with aerosols' radiative effects, including traditionally defined aerosol direct and semidirect effects (Schwartz, 1996; Ackerman et al., 2000). Strongly scattered radiation creates the appearance of a white-gray sky that is often referred to as *haze*, which

prevails over urban areas and megametropolises, especially in the South and East Asian regions (Ramanathan et al., 2007; Li et al., 2016).

Aerosol Radiative Properties

ARIs are governed by aerosol optical properties—namely, aerosol optical depth (AOD), single-scattering albedo (SSA) and asymmetry factor, as well as surface albedo (Chylek & Coakley, 1974). Aerosol optical properties are determined by the size distribution, the mixing state, and chemical composition, although in many aerosol-climate models, particularly earlier ones, only the aerosol mass mixing ratio was predicted (Haywood & Boucher, 2000; Forster et al., 2007). Aerosol radiative properties can change with the mixing states of different chemical species in the same particle (internal mixing) or as separate particles (external mixing). The mixing state of aerosols contributes not only to the magnitude, but also to the sign of the radiative forcing of aerosols. The aerosol mixing state is determined by complex chemistry and physical processes, making it difficult to understand (and even more difficult to predict) in models. Recent studies have shown that significant errors in scattering, absorption, and hygroscopicity are produced by

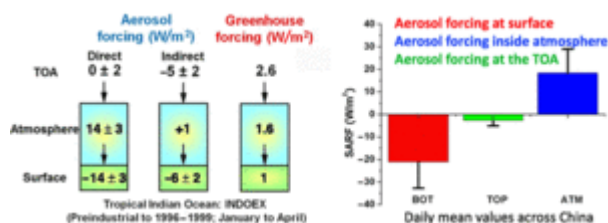
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inadequate representations of the aerosol mixing state (Ching, Zaveri, Easter, Riemer, & Fast, 2016).

Aerosol Radiative Effects

Aerosol direct forcing, also known as *aerosol radiative forcing*, is defined as the difference in the radiation budget of a system between aerosol-free and aerosol-laden conditions. The system usually refers to the atmosphere-surface, the top of the atmosphere (TOA), or the atmospheric column. As aerosols can both scatter and absorb solar radiation, aerosol radiative forcing is always negative at the surface, is zero to positive in the atmosphere, and can be either negative or positive at the TOA depending on a combination of AOD, surface albedo, and back-scattering fraction (Chylek & Coakley, 1974; Chylek & Wong, 1995).

Aerosol direct forcing is much larger under clear conditions than under cloudy conditions. In tropical regions prone to biomass burning (such as Africa, the Amazon, and Southeast Asia), the monthly mean aerosol direct radiative forcing can be more than 100 W/m^2 (Li, 1998). For conservative scattering (no absorption), direct aerosol radiative forcing is the same at the TOA and at the surface, but for absorbing aerosols, they may differ considerably (i.e., the forcing may be much larger at the surface than at the TOA). Over China and India, for example, the TOA forcing is close to zero, but the atmosphere and surface are warmed and cooled, respectively, by aerosols in nearly equal amounts, but with opposite signs (about 15 and 20 W/m^2 in magnitude, respectively) (Ramanathan, Crutzen, Kiehl, & Rosenfeld, 2001; Li et al., 2010), as shown in Fig. 3.



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Figure 3. Mean values of aerosol radiative forcing at the top and bottom of, as well as inside, the atmosphere observed over the tropical Indian Ocean (left; from Ramanathan et al., 2001) and across China (right; from Li et al., 2010).

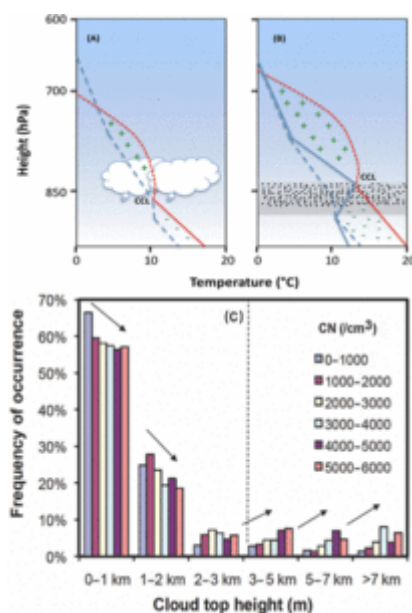
Aerosol Semidirect Effect (SDE)

Absorbing aerosols have a strong influence on the atmospheric thermodynamic state (temperature, and therefore relative humidity) due to its adiabatic heating, which can be computed under clear-sky conditions if aerosol vertical extinction profiles and the SSA are known (e.g., Liu, Zheng, Li, Flynn, & Cribb, 2012). Adiabatic heating modifies the atmospheric thermal state, and thus stability and circulations, on small to large scales

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(e.g., Fan, Zhang, Tao, & Mohr, 2008, Fan et al., 2015; Bollasina, Ming, & Ramaswamy, 2013). This aerosol thermodynamic effect has also been referred to as the aerosol *semidirect effect (SDE)* (Hansen, Sato, & Ruedy, 1997; Johnson, Shine, & Forster, 2004), which arises from the aerosol direct radiative effect and is therefore classified as a category of ARI based on IPCC (2013).

Most aerosols reside in the planetary boundary layer (PBL), an atmospheric layer in which the atmosphere-surface exchanges of energy and water take place. For absorbing aerosols, radiative heating may alter the PBL in numerous ways, as comprehensively reviewed by Li et al. (2017). First, aerosols reduce surface radiative energy, and thus sensible heat fluxes that drive the evolution of the PBL. Second, absorbing aerosols warm the air in the PBL, thus altering the atmospheric thermodynamic structure. Third, aerosols and the PBL likely interact and induce a positive feedback process that exacerbates the initial radiative effect (Yu, Liu, & Dickinson, 2002). As such, Wang, Khalizov, Levy, and Zhang (2013) hypothesized that the convective available potential energy (CAPE) is increased above the PBL and decreased inside the PBL, which seems consistent with the finding that low clouds tend to occur less frequently under polluted conditions than under cleaner conditions (Koren, Kaufman, Rosenfeld, Remer, & Rudich, 2005; Li et al., 2011). Figure 4 illustrates this hypothesis and shows two observed phenomena. Note that the latter may not be construed as sole casual evidence due to other possible reasons for them, such as the aerosol SDE or covariability between aerosol and meteorological conditions.



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Figure 4. Upper panels: A hypothesis of the impact of absorbing aerosols on atmospheric stability: (A) without and (B) with the presence of absorbing aerosols in the PBL. The dashed and solid blue lines correspond to the vertical temperature profiles in the absence and presence of the absorbing aerosol layer,

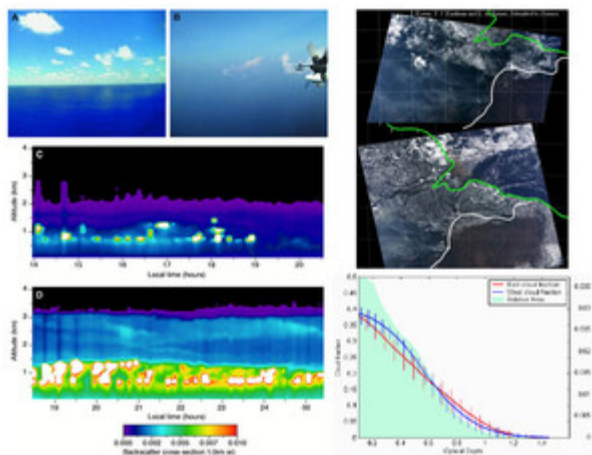
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respectively, and the solid and dashed red lines denote the dry and moist adiabats, respectively (Wang et al., 2013).

Lower panel: Observed cloud frequency of occurrence as a function of aerosol number concentration (CN) for convective clouds with different cloud-top heights derived from 10 years of ARM measurements made at the U.S. southern Great Plains site. Clouds are classified according to cloud top height, with the dashed line separating water (left) and ice (right) clouds. Within each cloud height group, they are further sorted by the concentration of aerosol number (CN). They were based on 10 years of measurements at the U.S. southern Great Plains (Li et al., 2011).

Another category of SDE has to do with the reduction in cloud fraction as a result of the warming of clouds due to the absorption of aerosols inside cloud droplets (Ackerman et al., 2000; Koren, Kaufman, Remer, & Martins, 2004) (Fig. 5), the so-called cloud burning effect. Jacobson (2002)

showed that the BC effect may be enhanced due to a low-cloud positive feedback loop in which the cloud loss leads to an increased opportunity for BC absorption. An enhanced BC layer decreases the cloud liquid water path, lowers the cloud top, and elevates the cloud base (Hill & Dobbie, 2008). In addition, embedding BC can reduce the cloud droplet SSA, increase the absorption of solar radiation, and expedite cloud evaporation, thus affecting the atmospheric heating profile.



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Figure 5. Comparison of cloud scenes under polluted and clean conditions, suggesting the semidirect effect of absorbing aerosols burning off clouds.

Left panels: (A) and (B) are images of clouds within clean and dirty marine boundary layers obtained from a research aircraft during the INDOEX in February 1999 over the southern Indian Ocean. (C) and (D) are images of time-height contours of particle backscatter cross section measured by micropulse lidar (MPL) on board a research vessel, the National Oceanic and Atmospheric Administration's *R.H. Brown* (Ackerman et al., 2000).

Right panels: Top images: Terra and Aqua satellite images of the east Amazon basin, August 11, 2002. (A) The clouds (Terra, 10:00 local time) are beginning to form. (B) The clouds (Aqua, 13:00 local time) are fully developed and cover the whole Amazon forest except for the smoke area. The boundary between forest and Cerrado region is marked in white on both images, and the seashore is marked in green. Bottom image: Cloud fraction as a function of aerosol optical depth derived over the Amazon (Koren et al., 2005).

While these two types of aerosol SDEs are both sound in principle, there have been very few studies carried out to differentiate them. In other words, the phenomena presented in Figures 4 and 5 could result from either effect. For example, the finding of decreasing cloud fraction in smoky regions (Fig. 5) could stem from a stabilized boundary layer that does not generate or suppress any would-be convection to prevent clouds from forming in the first place (i.e., the first

type of aerosol SDE).

Aerosol-Cloud Interactions (ACIs)

By serving as CCN and IN, aerosols alter cloud radiative forcing and precipitation by directly changing cloud microphysical processes (Seinfeld & Pandis, 2006; Tao, Chen, Li, Wang, & Zhang, 2012; Fan, Wang, Rosenfeld, & Liu, 2016). This is referred to as *aerosol-cloud interactions (ACIs)* in IPCC (2013).

ACIs alter cloud properties, which induces changes in cloud radiative forcing. This used to be referred to as *aerosol indirect radiative forcing*, or more precisely, aerosol-mediated changes in cloud radiative forcing. Its magnitude can be substantial for certain types of clouds, such as deep convective clouds (DCCs; Fan et al., 2013; Peng, Li, Zhang, Liu, & Cribb, 2016). The estimation of both direct and indirect aerosol radiative forcing is still fraught with great uncertainty (Yu et al., 2006; IPCC, 2013; Rosenfeld et al., 2014).

Aerosols can alter cloud microphysics, which is concerned with the amount, phase, and size of cloud droplets and ice crystals. When aerosol particles are hydrophilic, they are generally good CCN and can be activated at certain supersaturation rates to form cloud droplets. The CCN behavior of an aerosol particle is determined by its composition and size, and by aerosol activation theory as detailed by Seinfeld and Pandis (2006) and Tao, Chen, Li, Wang, and Zhang (2012). The aerosol mixing state also affects particle hygroscopic properties significantly, and therefore the CCN capability (Wang, Cubison,

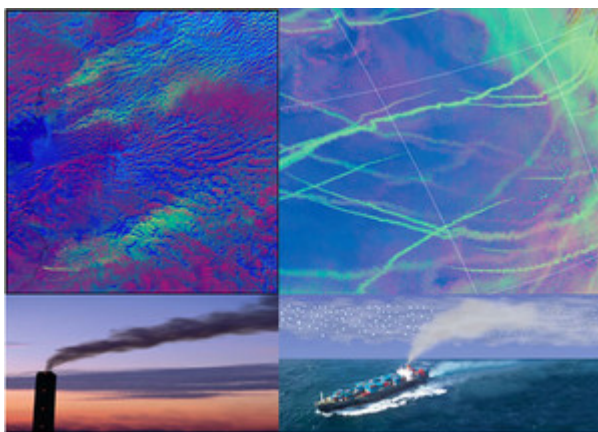
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Aiken, Jimenez, & Collins, 2010). There is ample evidence of the aerosol effect on cloud microphysics that is revealed from a variety of observational platforms, such as in situ aircraft observations and ground-based and space-borne remote sensing.

Aerosol Effect on Cloud Microphysics

Cloud droplets and ice crystals would not be formed in nature without CCN/IN because that would require a water vapor pressure level too high to achieve naturally. On the other hand, too many CCN would lead to too many droplets competing for water vapor, producing numerous smaller cloud droplets for a given fixed amount of cloud water. Such smaller cloud droplets are more efficient at scattering solar radiation than fewer but larger droplets; the scattering efficiency is proportional to the cross section of all droplets. As a result, the total amount of solar energy reflected back to space is increased by increasing the aerosol loading or the CCN concentration. This helps cool the planet and is referred to as the *Twomey effect* (Twomey, 1977).

There have been many observational instances of the Twomey effect. Ship tracks generated by emissions from fuel consumption were arguably the earliest and most vivid demonstration of it (Radke, Coakley, & King, 1989; Nakajima & King, 1990; Durkee et al., 2000; Platnick et al., 2000; Rosenfeld & Woodley, 2001; Platnick, 2007). Under certain meteorological conditions, exhaust from either power plants (Fig. 6A) or ships (Fig. 6B) serve as CCN to produce far more cloud droplets than clouds in nearby neighboring regions, causing plumes of brighter clouds due to reduced cloud particle sizes.



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Figure 6. Tracks originating from emissions from a refinery, coal power plants, and a smelter over land in South Australia on October 21, 1998, at 0444 GMT (left panels) and ships over the North Pacific Ocean on June 22, 2000, at 0300 GMT (right panels). Both images were composited from satellite measurements made by the Advanced Very High Resolution Radiometer. All are adopted from Rosenfeld and Woodley (2001), except for the picture

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of a sailing ship in the lower right panel (from Platnick, 2007).

Using atmospheric radiation measurement (ARM) data acquired over

the U.S. southern Great Plains, several studies have revealed the impact of aerosols on cloud particle size, cloud optical depth, and cloud reflection and emission (Feingold, 2003, 2006; Kim, Miller, Schwartz, Liu, & Min, 2008; Yan, Li, Huang, Cribb, & Liu, 2014).

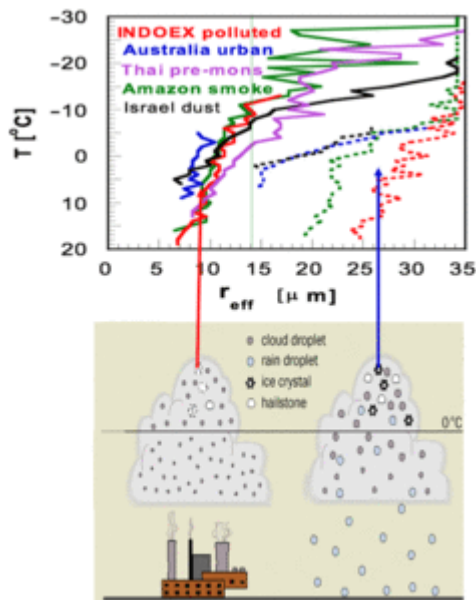
Contingent upon aerosol properties and meteorological conditions, the magnitude was found to differ drastically (Feingold et al., 2006). Various aerosol impact mechanisms tend to be mediated or buffered by interactions across scales (Stevens & Feingold, 2009; Liu, Li, & Cribb, 2016). Satellite retrievals of AOD and its derivative, the aerosol index, have also been used as a proxy of the CCN to study the impact of aerosol on cloud microphysics (Kaufman & Nakajima, 1993; Nakajima, Higurashi, Kawamoto, & Penner, 2001, Nakajima et al., 2003). While satellite-based studies generally lead to more evidence of the Twomey effect, some exceptions were also noted, such as the positive correlation between AOD and cloud particle size, or the so-called *anti-Twomey phenomenon* (Yuan, Li, Zhang, & Fan, 2008) and systematic different magnitudes of the ACIs estimated from space-borne and ground-based observations (Liu et al., 2016). There are numerous reasons for these seemingly inconsistent findings, such as uncertainty in satellite retrievals of AOD (Li, Wang, & Zhang, 2008), the analysis method (Rosenfeld & Feingold, 2003), AOD-CCN conversion (Andreae, 2009, Liu & Li, 2014), or lack of accounting for aerosol chemical composition and the ensuing swelling effect (Liu & Li, 2017). In general, such studies suffer from a general bottleneck difficulty of separating the microphysical effects of aerosol from the dynamic effects of the atmosphere and surface. New methods of retrieving the CCN (Rosenfeld et al., 2016) and updraft speed (Zheng, Rosenfeld, & Li, 2015, 2016; Zheng & Rosenfeld, 2015) at cloud base from satellite will pave the way to disentangling the two effects in studying ACIs, particularly for low convective and stratus clouds.

By acting as IN, aerosols help form ice crystals at temperatures below 0°C. Therefore, increasing IN could lead to the so-called glaciation indirect effect for mixed-phase clouds (i.e., turning supercooled liquid into ice clouds that have a much lower particle number density and reduced cloud optical depth and that settle relatively rapidly, thereby reducing the cloud lifetime) (Harrington, Reisin, Cotton, & Kreidenweis, 1999; Morrison et al., 2011; Ovchinnikov, Korolev, & Fan, 2011). A number of heterogeneous ice nucleation mechanisms have been proposed (Seinfeld & Pandis, 2006). In the upper troposphere, water vapor can deposit directly onto solid particles (so-called *deposition freezing*). In clouds warmer than about -37°C, where liquid water can persist in a supercooled state, IN immersed inside droplets can trigger droplets to freeze (i.e., immersion freezing). Contact nucleation can occur if an ice nucleus collides with a supercooled droplet. In the absence of ice-nucleating particles, pure water droplets can persist in a supercooled state to temperatures approaching -37°C, where they freeze homogeneously. Aerosol particles

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that can be efficient IN include mineral dust, soot, organic matter, and biological aerosols (Christner, Morris, Foreman, Cai, & Sands, 2008).

The impact of aerosols on the profiles of cloud microphysics over the entire column of convective clouds composed of both liquid droplets and ice crystals, as well as mixed-phase clouds, has been observed in several field experiments, as summarized by Ramanathan et al. (2001) and shown in Fig. 7. The trends and underlying cloud processes can vary for different aerosol types, which can significantly alter precipitation.



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Figure 7. The impact of aerosols on cloud droplet size: smaller sizes in polluted regions than in cleaner regions (from Ramanathan et al., 2001). The top panel shows satellite-retrieved vertical profiles of cloud particle effective radius under pristine conditions (dashed lines) and under polluted conditions, where clouds ingest various types of aerosols (solid lines).

Note that IN form a very small fraction of aerosol particles, and their concentrations are typically over five orders of magnitude less than CCN concentrations. Studies have shown that the IN effect on supercooled water and cloud phase is much more significant than the CCN effect (Fan, Leung, Rosenfeld, & DeMott, 2017). For cloud anvil sizes and lifetimes of DCC, the CCN effect is more significant (Fan et al., 2010).

Aerosol Effect on Cloud Lifetime

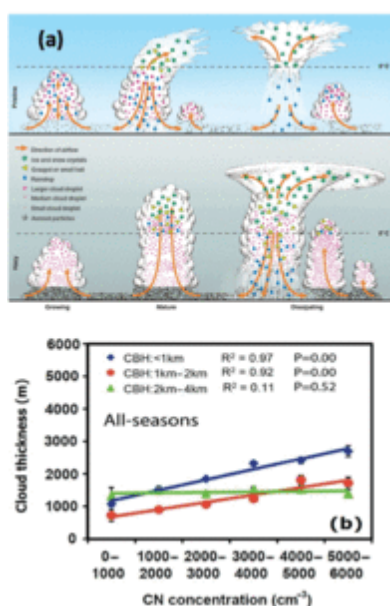
Having smaller droplets in a polluted environment means that it takes longer for cloud droplets (in sizes ranging from a few micrometers to tens of micrometers) to reach the size of raindrops (tens to hundreds of micrometers) needed to initiate precipitation (Albrecht, 1989). In DCC, the freezing of smaller droplets leads to smaller ice particles. Smaller sizes mean lower fall speeds, so they take longer to fall from a cloud. Clouds then dissipate more slowly (Fan et al., 2013). Such an effect on cloud lifetime is referred to as the *cloud lifetime effect*. The cloud lifetime effect in low-level clouds is much more complicated. The life cycles of clouds are controlled by an intimate interplay between meteorology and aerosol/cloud microphysics, including complex feedback processes. It has thus been difficult to identify and quantify the lifetime effect, especially by observation. In shallow cumulus clouds, cloud lifetime is not necessarily increased, even

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though precipitation is suppressed (Jiang, Xue, Teller, Feingold, & Levin, 2006). An explanation is that smaller cloud droplets are more susceptible to evaporation, inducing evaporative cooling, and thus reduce the buoyancy of a cloud parcel. This suppresses cloud growth because the cloud parcel cools and becomes heavier than the surrounding air (Lee, Feingold, & Chuang, 2012). The reversal in thermal contrast generates an increased vorticity around cloud boundaries, which speeds up the convective turnover and thus shortens the cloud lifetime. The cloud lifetime effect is thus the net consequence of aerosol-induced changes in cloud microphysics, atmospheric dynamics and thermodynamics. These effects are also driven by large-scale meteorological conditions.

Aerosol Effect on Cloud Height and Thickness

By altering cloud microphysics, which feeds back into cloud thermodynamics and dynamics, aerosols can modulate cloud macrophysics, especially cloud vertical and horizontal extents. The suppression of warm rain by aerosols causes most condensates to ascend as cloud water, freeze, and then grow through deposition, in which latent heat is released to invigorate convection, as described by Khain, Pokrovsky, Pinsky, Seifert, and Phillips (2004) and Rosenfeld et al. (2008) (Fig. 8A). This is referred to as the *aerosol invigoration effect*. It is most significant for DCC with warm cloud bases and relatively weak wind shear (Fan et al., 2009, 2012A, 2012B, 2013). Under the conditions of strong wind shear, dry air, or cold cloud bases, aerosols could strongly suppress convection due to strong evaporative cooling of the small cloud droplets, less efficient ice-growing processes, or both (Khain, 2009; Fan et al., 2009, 2012B; Lebo & Seinfeld, 2011; Morrison, 2012). The most prominent manifestations of the invigoration effect are increases in cloud-top height, thickness, or both—a finding that has been revealed by analyzing short-term field experimental data (Andreae et al., 2004), long-term (10-year) ARM data (Li et al., 2011) (Fig. 8B), and regional and global satellite products (Koren, 2005; Niu & Li, 2012; Peng et al., 2016).



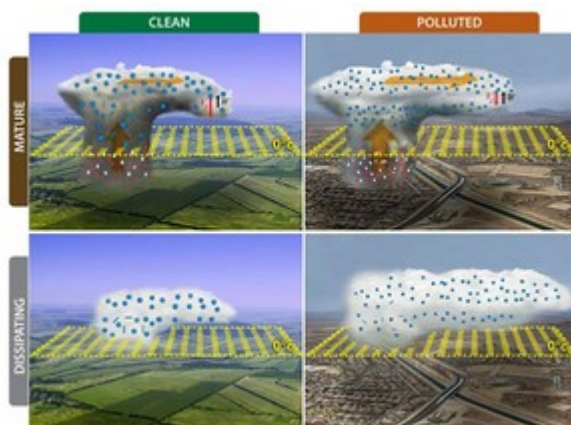
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Figure 8. Top panel: The impact of aerosols on the height and thickness of convective clouds; i.e., the hypothesis of the aerosol invigoration effect (Rosenfeld et al., 2008). Bottom panel: Observational evidence found from 10 years of ground-based observations made at the U.S. southern Great Plains site (Li et al., 2011).

Aerosol Effect on Cloud Fraction

In general, aerosols can influence the horizontal extent of clouds in many ways. Cloud fraction is controlled by atmospheric dynamics (vertical velocity in particular) and thermodynamic (humidity and temperature) conditions. The aerosol effect on cloud fraction is complex, and both positive and negative effects have been reported due to various mechanisms for different types and heights of clouds. The positive relationship between maritime stratocumulus cloud cover and aerosol optical depth (Koren et al., 2005; Myhre et al., 2007) may originate from aerosol microphysical and lifetime effects because smaller droplets resulting from more aerosols and CCN are less likely to rain and more likely to grow in space and time. On the other hand, smaller droplets are more vulnerable to evaporation, thus reducing the cloud fraction. Moreover, the thermodynamic effects of aerosols (e.g., stabilizing the lower atmosphere, radiative warming, or both by absorbing solar radiation) inhibits the formation and growth of clouds, reducing the cloud fraction or the cloud frequency of occurrence (Koren, Vanderlei Martins, Remer, & Afargan, 2008; Li et al., 2011). For DCC, the aerosol microphysical effect increases the anvil cloud cover significantly in a polluted environment, as observed by Yan et al. (2014) and modeled by Fan et al. (2013). The freezing of a larger number of smaller droplets produces more numerous but much smaller ice particles in the stratiform regime of polluted clouds, which leads to much reduced fall velocities of ice particles and slows the dissipation of stratiform and anvil clouds significantly, as illustrated in Fig. 9.



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Figure 9. Schematic drawing of the differences in cloud-top height, cloud fraction, and cloud thickness

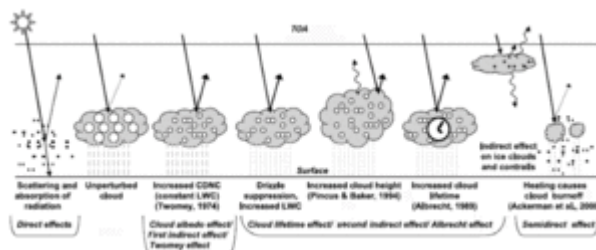
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for storms in clean and polluted environments. Red dots represent cloud droplets, light blue dots represent raindrops, and blue shapes are ice particles (from Fan et al., 2013).

Since high clouds have a warming effect, the increased fraction of anvils and cirrus clouds offsets

the cooling effect induced by an increase in cloud fraction for low clouds (Yan et al., 2014). This is likely a leading cause for the long-standing gap in estimates of aerosol-induced changes in cloud radiative forcing because global climate models cannot simulate aerosol effects on DCC or the aerosol indirect effect on the radiation budget well (Anderson et al., 2003). The uncertain response of cloud fraction to aerosols is the largest and most sensitive factor dictating aerosol-induced changes in cloud radiative forcing (Penner, Dong, & Chen, 2004; Rosenfeld et al., 2014).

Figure 10 summarizes all the aforementioned effects of aerosols on various cloud properties (IPCC, 2007), which is virtually a chain of effects originated from ARIs and ACIs (IPCC, 2013).



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Figure 10. Schematic of a chain of effects of aerosols on warm cloud properties leading to the suppression of precipitation (IPCC, 2013). For the same amount of water, more aerosols lead to more, smaller cloud droplets. The total cross-sectional area of these droplets is larger and reflects more solar radiation than droplets in clouds formed under cleaner conditions. Clouds with smaller particle sizes are less likely to rain out, leading to longer lifetimes and greater thicknesses.

Aerosol Effects on Precipitation

Warm Rain from Shallow, Warm Clouds

In principle, aerosol radiative effects (stabilizing the atmosphere and warming cloud droplets) and the microphysical effect (reducing cloud droplet sizes) are unfavorable for the growth of cloud droplets, and thus tend to suppress rain. The aerosol rain suppression effect is most significant for warm clouds. Aerosols also make the collision and coalescence process, a key mechanism for obtaining precipitation from warm clouds, less efficient because they narrow the size distribution of cloud droplets. It has been observed and simulated that drizzle is suppressed and the onset of precipitation is delayed by aerosols. This has been found in regions with heavy industrial pollution and biomass burning (Warner & Twomey, 1967; Rosenfeld, 2000; Andreae et al., 2004; Li et al., 2011; Yang, Ferrat, & Li, 2013A) and desert dust (Rosenfeld, Rudich, & Lahav, 2001; Mahowald & Kiehl, 2003).

The suppression effectively elevates the height of the initiation of warm rain to greater heights above the cloud base (Prabha et al., 2011; Konwar et al., 2012; Freud & Rosenfeld, 2012). Satellite retrievals of the vertical evolution of cloud drop effective radius with height have shown the suppression effect over China (Rosenfeld et al., 2014), where an increase in AOD of 1.0 can lead to an increase in the rain-initiation height by 5.5 km (Zhu, Rosenfeld, Yu, & Li, 2015). Precipitation from shallow and short-lived clouds is most susceptible to aerosols because the time in forming precipitation is limited. This is most evident over mountain barriers where orographic precipitation decreases significantly with increasing atmospheric pollution (Rosenfeld et al., 2007A; X. Yang et al., 2013A; Guo et al., 2014; Yang et al., 2016).

Precipitation from Deep Clouds

For mixed-phase deep clouds, the effect of aerosols on precipitation depends on numerous factors, such as cloud-base height, wind shear, and cloud systems. If the cloud base is high, increasing CCN often suppresses precipitation because smaller droplets reduce riming, which is especially the case for stratiform mixed-phase clouds (Fan et al., 2012A). For orographic mixed-phase clouds, increasing CCN generally suppresses precipitation (Jirak & Cotton, 2006; Rosenfeld & Givati, 2006; Lynn, Khain, Rosenfeld, & Woodley, 2007), but the reverse could happen under extremely polluted conditions due to an invigoration mechanism for orographic mixed-phase clouds (Fan et al., 2017). Other studies show that CCN may not have a significant effect on the total precipitation; rather, they shift precipitation from the windward to leeward slope, a so-called spillover effect (Lynn et al., 2007; Saleeby, Cotton, & Fuller, 2011; Saleeby, Cotton, Lowenthal, & Messina, 2013).

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For DCC with low cloud bases and weak wind shear, precipitation is intensified by increasing aerosols due to the aerosol invigoration effect (Rosenfeld et al., 2008; Fan et al., 2009, 2012A; Li et al., 2011; Yang & Li, 2014). Studies have shown differential responses of precipitation to aerosols depending on environmental factors, as summarized by Khain (2009) and also depending on, for example, aerosol-induced intensification of gust fronts, which is important for organized convective systems (Lee, 2011; Lee & Feingold, 2013). Relative humidity is another major influential factor (Khain et al., 2004, Khain, Rosenfeld, BenMoshe, & Pokrovsky, 2008D; Fan, Zhang, Li, & Tao, 2007B; Fan et al., 2009; Tao et al., 2007; Yuan et al., 2008).

Aerosols were found to suppress convection for isolated clouds formed under relatively dry conditions and to invigorate convection in convective systems in a moist environment (Khain, 2009). Wind shear also determines whether aerosols suppress or enhance convective precipitation (Lee, Donner, Phillips, & Ming, 2008B; Fan et al., 2009; Khain, 2009). The impacts of aerosols on convective clouds and precipitation vary as a function of CAPE, with a stronger influence observed in environments with reduced CAPE (Lee et al., 2008B; Storer & van den Heever, 2013; Liu et al., 2016). Overall, the effects of aerosols on precipitation depend on meteorological conditions, cloud regimes, aerosol properties, and other elements, as illustrated in Figure 11, which summarizes various studies classified according to these factors.



[Click to view larger](#)

Figure 11. A summary of the aerosol effects on deep convective clouds under distinct meteorological conditions, as simulated by cloud-resolving models (modified based on Khain, 2009).

Increasing IN enhances precipitation from mixed-phase clouds resulting from the stronger Wegener-Bergeron-Findeisen process, in which ice particles grow at the expense of evaporated water droplets and riming processes (Korolev, 2006; Ovchinnikov et al., 2011; Fan et al., 2014). For DCC cases, some studies have shown that increasing IN enhances cloud vertical

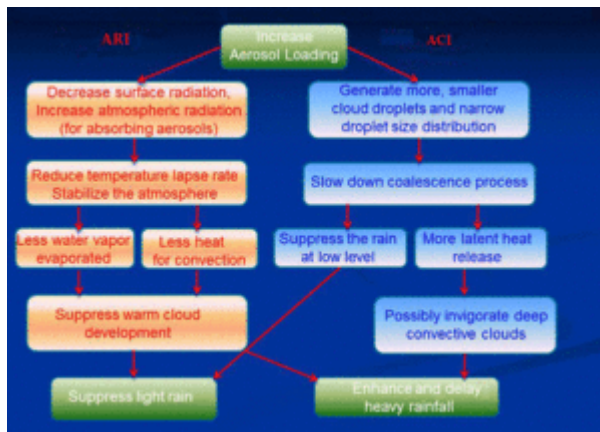
development and surface precipitation (van den Heever, Carrio, Cotton, DeMott, & Prenni, 2006; Ekman, Engström, & Wang, 2007).

Aerosols could also change the probability distribution function of precipitation intensity, as well as local spatial patterns of precipitation, even when the total precipitation is insensitive to aerosols. By suppressing light precipitation, an increase in CCN may enhance heavy precipitation resulting from enhanced mixed-phase and ice cloud microphysics. Such an aerosol-induced distribution shift in precipitation intensity,

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resulting in more extreme precipitation events, has been widely reported in observational and modeling studies (e.g., Qian et al., 2009; Li et al., 2011; Y. Wang et al., 2011; Tao et al., 2012; Fan et al., 2013; Guo et al., 2014).

As a summary of the impact of aerosol on precipitation, Figure 12 illustrates the joint impact of two primary aerosol effects on precipitation and various processes leading to the effects.



Click to view larger

Figure 12. A schematic diagram of the joint impact of ARIs and ACIs on precipitation by altering various processes leading to rainfall.

Aerosol Effects on Severe Weather Events

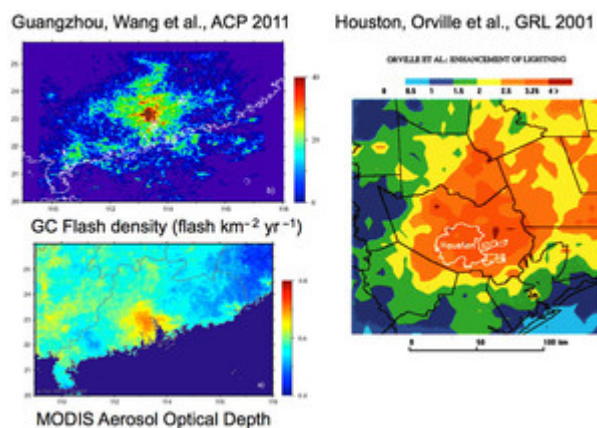
Severe weather events are primarily driven by atmospheric dynamic and thermodynamic conditions, which are largely dictated by regional and global circulation. Aerosols could affect severe weather events by modulating the thermodynamic state of the atmosphere. Because storms are sensitive to any small change in atmospheric thermodynamics and dynamics, differentiating the effects of aerosols requires fixing other factors. As other factors may covary with aerosol quantities, the true effects of aerosols are not readily identifiable. As such, in the following review of published studies, emphasis is placed on the underlying physical principles.

Aerosol Effects on Lightning and Thunderstorms

Cloud electrification occurs when graupel (ice hydrometeors that are formed by the collection and freezing of supercooled water) collide with ice crystals in the presence of supercooled cloud droplets and strong updrafts. DCC that develop in a very clean atmosphere are composed of a small number of large cloud droplets that quickly coalesce into raindrops that fall before reaching the freezing level. The remaining water freezes very quickly because of the large size of the drops. With a lack of supercooled water, cloud electrification is insufficient for producing lightning. It has long been noted that a

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striking difference in the frequency of lightning occurrence between continents and oceans exists. This finding had been attributed to thermodynamic differences until Rosenfeld and Lensky (1998), Orville et al. (2001), and Williams et al. (2002) proposed that aerosols may be involved. Adding aerosols to clouds suppresses coalescence and leads to an enhanced amount of supercooled water, along with cloud invigoration that enhances the updrafts, which in turn enhances the cloud electrification (Andreae et al., 2004; Rosenfeld et al., 2008; Wang et al., 2011). Lightning is enhanced by other thermodynamic factors, such as enhanced instability and shorter distance between the cloud base and the freezing level. The role of aerosols and thermodynamic properties of clouds have comparable importance in explaining the variability in lightning frequency (Stolz, Rutledge, & Pierce, 2015). A vivid illustration of the potential link between aerosols and lightning in southern China is given in Fig. 13 (Wang et al., 2011).



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Figure 13. Enhanced aerosol loading and lightning over the metropolitan area of the Pearl Delta region in southern China (left panels; Wang et al., 2011) and a similar enhancement over the Houston metropolitan area in the southern United States (right panel: Orville et al., 2001).

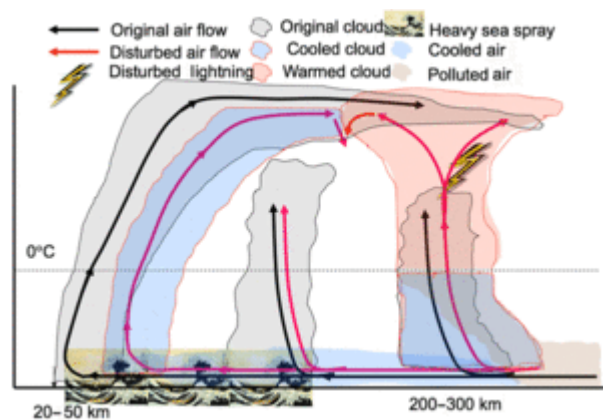
In general, lightning is associated with strong convective cloud systems that produce severe thunderstorms. Besides the numerous factors dictating the development of convection stated previously, aerosol type is a key factor influencing thunderstorms. Due to ARIs, absorbing aerosols tend to suppress thunderstorms, whereas hygroscopic aerosols invigorate thunderstorms under suitable

environmental conditions due to ACIs. Such effects have been illustrated in China, where aerosol loading of virtually all types of aerosols has been increasing for decades (Li et al., 2016). In regions dominated by absorbing aerosols (such as central China), the occurrence of thunderstorms has reduced by 50% over the past half century (Yang, Yao, Li, & Fan, 2013B, 2016). An increasing trend was seen in southeast China, where weakly absorbing but strongly hydrophilic aerosols dominate (Yang & Li, 2014). Visibility in both regions is worst on Fridays, presumably due to accumulated pollution during the week when thunderstorms occurred most and least frequently in the two regions, respectively. The ARI effect also leads to a delay in thunderstorms during the day, but the intensity is strengthened by ACIs (Guo et al., 2016; Lee, Guo, J., & Li, 2016).

Aerosol Effects on Cyclones and Hurricanes/Typhoons

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Cyclones, especially tropical cyclones (TCs), are the primary producer of heavy rainfall. Hurricanes or typhoons are strong types of cyclones, with severe consequences due to extremely high wind speeds and storm surges, as well as heavy rainfall. The maximum wind speeds of cyclones that ingest aerosols are reduced due to the suppression of coalescence and the subsequent invigoration of the outer rain bands. The invigorated outer bands compete with the eye wall and weaken it. As a result, the eye diameter increases, the maximum wind speed decreases, and the central pressure increases. The invigoration of the outer bands by air pollution was observed in a typhoon that approached southern China (Rosenfeld et al., 2012). This effect has been simulated (Rosenfeld, Khain, Lynn, & Woodley, 2007B; Zhang, Li, Fan, Wu, & Molina, 2007; Zhang, McFarquhar, Cotton, & Deng, 2009; Khain, Lynn, & Dudhia, 2010). It was shown that the amount of aerosols ingested into hurricanes could partially explain the deviations in storms' observed maximum wind speeds from their predicted values (Rosenfeld, Clavner, & Nirel, 2011; also see Fig. 14).



[Click to view larger](#)

Figure 14. Schematic diagram showing the influences of aerosols on hurricanes: a vertical radial cross section in clouds of a tropical cyclone. The unperturbed reference clouds and flows are shown in gray and black, respectively. When polluted air is ingested into the outer cloud band of the storm, it invigorates it via the mechanism shown in Fig. 8, at the expense of the converging air to the eye wall. This redistributes the energy from the center to the periphery of the storm, thus widening the eye and decreasing the maximum wind speeds around it (Rosenfeld et al., 2011).

The sea surface temperature (SST) is a key factor influencing the size of TCs (Lin, Zhao, & Zhang, 2015; Chavas, Lin, Dong, & Lin, 2016). Latent heat release from the sea into the atmosphere is the primary source of energy driving TCs. Dust activities across the Atlantic generated from the Sahara Desert could significantly reduce the occurrence of hurricanes (Lau & Kim, 2007; Sun, Lau, & Kafatos, 2008) because they reduce both SSTs and surface heat fluxes. On the other hand, serving as CCN/IN, dust or

aerosols in general influence storm development and intensity by modifying the diabatic heating and thermodynamic structure of the atmosphere (R. Zhang et al., 2007; H. Zhang et al., 2009). Diabatic heating anomalies resulting from Asian pollution can increase transient eddy meridional heat fluxes by 9%, which helps explain the increases in winter storms over the North Pacific (Wang, Zhang, & Saravanan, 2014). Model simulations of the roles of both natural and various external factors in TCs revealed that long-term historical changes in AAs played a key role in the decadal variability of TCs over the North Atlantic

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in the 20th century through aerosol-induced shifts in the Hadley circulation (Dunstone, Smith, Booth, Hermanson, & Eade, 2013).

Aerosol Effects on Hail Formation

Hail forms when an ice precipitation particle grows by accreting supercooled cloud drops at such a rate that the latent heat of freezing warms its surface to the melting temperature. This requires very high supercooled cloud water contents, along with strong updraft speeds. Adding aerosols to DCCs increases their likelihood of producing hail for the following reasons:

- Added aerosols create a larger number of smaller cloud droplets that suppress drop coalescence and rain formation. This prevents the loss of cloud water into rain before the cloud develops to heights where its water becomes supercooled and available for the growth of hail.
- Raindrops freeze relatively quickly in updrafts between -10°C and -15°C and then become potential embryos of hailstones. When there are too many such embryos, the competition for the limited amount of supercooled cloud water prevents the growth of large hailstones. Aerosols suppressing rain formation therefore also greatly reduce the number of hail embryos, and thus allow little competition for the available cloud water and fast growth into large hailstones.

This theory was supported by cloud model simulations (Iltoviz, Khain, Benmoshe, Phillips, & Ryzhkov, 2016). Observations showed that the weekly cycle of hail frequency in the southeastern United States was linked to the weekly cycle of air pollution. The hail frequency peaked by the polluted midweek and dropped to a minimum during the weekend (Rosenfeld & Bell, 2011).

IN aerosols can have a similar effect on suppressing hail by creating a large number of ice crystals, which become hail embryos competing with each other for supercooled cloud water and do not become hailstones. This is the underlying hypothesis of cloud seeding with silver iodide IN for hail suppression.

Aerosol Effects on Tornadoes

Tornadoes are typically produced by supercell storms. While in multicell storms, the growth stage of clouds is short-lived and updrafts are replaced by downdrafts, in supercell storms, the updraft continues for very long periods while the downdraft and precipitation shaft are shifted by strong wind shear aloft. The longevity of the updraft allows for the spin-up of tornadoes within the converging air into it. Adding aerosols can push a multicell storm across the threshold to become a supercell storm in two ways:

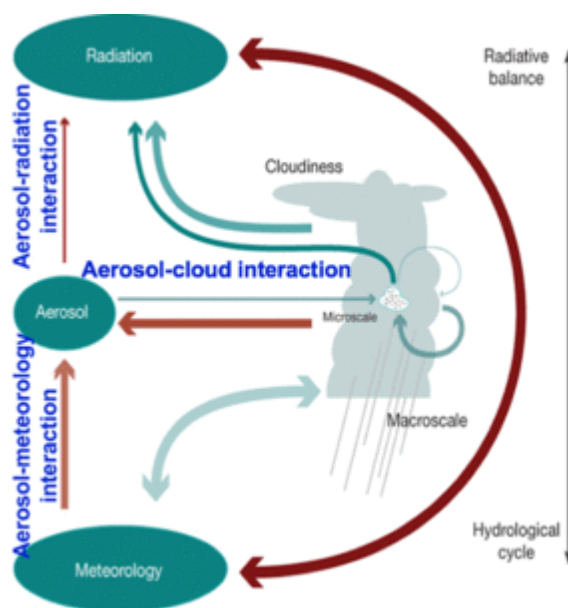
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- Aerosols delaying the initiation of precipitation in the updraft may cause the precipitation shaft to fall farther downwind of the updraft. Therefore, the cold air that descends with the downdraft has to travel a longer distance upwind until it can undercut the updraft and end it.
- Added aerosols suppress coalescence and thus produce a smaller number of precipitation particles. With less competition for cloud water, they become larger and induce larger hail. Having the same precipitation intensity but involving a smaller number of larger hydrometeors decreases their overall surface area, and thus the evaporation amount and the resulting cooling.

Air with less negative buoyancy has a lesser tendency to propagate upwind and undercut the updraft, which leads to its termination. This mechanism of aerosols triggering tornadoes was simulated by Snook and Xue (2008).

Concluding Remarks: Limitations and Recommendations

Despite the rapid and extensive development in this area, our understanding of aerosols and their impact on radiation, clouds, precipitation, and severe weather events is still fraught with great uncertainty. It should be pointed out that the various effects described in this article aim at illustrating the principles of the potential effects of aerosols on various meteorological variables. They by no means testify to the effects with the kind of scientific rigor found in the cited papers. In nearly all circumstances, the aerosol effects are tangled with meteorological variations that are connected via three interactions: ARIs, ACIs, and aerosol-meteorology interactions, as shown in Fig. 15.



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Figure 15. The entangled effects of aerosols with meteorology, clouds, and radiation that jointly affect the Earth's energy and water cycles (revised based on Stevens & Feingold, 2009).

There have been controversies surrounding many of the effects discussed here, with

central goals to single out any causal relationships and to quantify the effects. Resolving the problems would require a drastic advancement in observation and modeling.

For observation studies, the following key variables are essential but are still lacking:

- Profiles of cloud dynamics, microphysics, and aerosols in convective cores of DCCs, as well as the time evolution of environment measurements, including aerosol properties near the system
- Evolution of a single-cell cloud throughout its entire life cycle characterized by changes in the number concentrations of CCN, IN, and cloud droplets, as well as the size distributions of hydrometeor sizes, updrafts, and downdrafts
- Large spatial-domain measurements of aerosol properties, CCN, cloud microphysics, and updrafts to study aerosol-mesoscale convective system interactions
- Concurrent long-term measurements of aerosol properties (size distribution and composition) and meteorological fields to cope with the covariability of aerosol, dynamic, and thermodynamic quantities

For modeling studies, the following challenges need to be confronted:

- Accurate simulation of updraft intensity and cloud properties in large-eddy simulation/cloud-resolving models.
- Separation of true aerosol impacts from covariability due to the sensitivity to the initial small perturbation.
- Bulk microphysics schemes are limited to studying ACIs, whereas bin microphysics schemes are too computationally intensive. The representation of many microphysical processes suffers from lack of understanding.
- Severe parameterization problems involving ice nucleation, cloud microphysical processes, subgrid cloud variability, scale-aware, and aerosol-aware problems.

For research concerning aerosol effects, it is recommended to do the following:

- Conduct field experiments measuring aerosol and cloud properties simultaneously at different spatial and temporal scales in diverse climate regimes.
- Conduct ensemble model simulations to single out aerosol impacts from natural variability.
- Understand the evolution of hydrometer size distribution, conversions of hydrometeor types, and their fall speeds.

- Improve parameterizations of ice nucleation and microphysical processes for better simulations of mixed-phase and ice cloud properties.

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