Aerosol interactions with deep convective clouds

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Summary. Deep convective clouds (DCCs) are associated with the vertical ascent of air from the lower to the upper atmosphere. They appear in various forms such as thunderstorms, supercells, and squall lines. These convective systems play important roles in the hydrological cycle, Earth's radiative budget, and the general circulation of the atmosphere. Changes in aerosol (both cloud condensation nuclei and ice-nucleating particles) affect cloud microphysics and dynamics, and thereby influence convective intensity, precipitation, and the radiative effects of deep clouds and their cirrus anvils. However, the very complex dynamics and cloud microphysics of DCCs means that many of these processes are not yet accurately quantified in observations and models. This chapter outlines the main ways in which changes in aerosol affect the microphysical, dynamical, and radiative properties of DCCs.

Aerosol interactions with DCCs depend on aerosol properties, storm dynamics, and meteorological conditions. When aerosol particles are light-absorbing, such as soot from industry or biomass burning, the aerosol radiative effects can alter the meteorological conditions under which DCCs form. These radiative effects modify temperature profiles and planetary boundary layer heights, thus changing atmospheric stability and circulation, and affecting the onset and development of DCCs. These large-scale effects, such as the effect of anthropogenic aerosol on the East and South Asian monsoons, can be simulated in coarse-resolution models. These processes are described in Chapter 13.

This chapter is concerned with aerosol interactions with DCC systems ranging from individual clouds to mesoscale convective systems. Increases in cloud condensation nuclei (CCN) can enhance cloud droplet number concentrations and decrease droplet sizes, thereby narrowing the droplet size spectrum. For DCCs, a narrowed droplet size spectrum suppresses warm rain formation (rain derived from non-ice-phase processes), allowing the transport of more, smaller droplets to altitudes below 0°C. This may result in (i) freezing of more supercooled water, thereby enhancing latent heating from ice-related microphysical processes and invigorating storms (ice-phase invigoration); (ii) modification of ice-related microphysical processes, which changes cold pools, precipitation rates, and hailstone frequency and size; (iii) expansion of the mixed-phase zone and decreases in the cloud glaciation temperature; and (iv) slowing down of cloud dissipation, resulting in larger cloud cover and cloud depth in the stratiform and anvil regions due to numerous smaller ice particles. The increased cloud cover and cloud depth constitute an influence of aerosol on the cloud radiative effect. Reduced diurnal temperature variation has been observed and simulated as a result of enhanced daytime cooling and nighttime warming by expanded anvil cloud area in polluted environments. However, the global radiative effect of aerosol interactions with DCCs remains to be quantified.

An increase in CCN also enhances droplet condensational growth and latent heat release, which may invigorate the storm intensity (water-phase invigoration). This effect can be particularly significant for storms developing in warm, humid, and low background aerosol environments. Convective intensity can be further enhanced by high concentrations of ultrafine aerosol particles (less than 50 nm diameter) that can be activated inside convective cores where high supersaturation exists (so-called secondary nucleation) through the water-phase invigoration. The numerous droplets formed from secondary nucleation also affect ice-related processes and may amplify the various effects listed above.

Aerosol particles that can effectively serve as ice-nucleating particles (INPs), such as dust, marine organics, and biological particles, can increase ice crystal number concentrations. In DCCs, the response of ice particle size to increased ice crystal number concentration is complicated, depending on conditions such as convective intensity and relative humidity. Another important INP effect on DCCs is the increase in cloud glaciation temperature, which affects the cloud radiative effect and precipitation. Our understanding of INP and ice nucleation mechanisms in DCCs is still limited; therefore the effects of INP have large uncertainties.

As well as being affected by changes in aerosol, DCCs can also strongly affect aerosol properties. In mechanisms known collectively as "cloud processing" DCCs transport aerosol particles and trace gases from the lower to the upper troposphere, droplet nucleation and precipitation deplete aerosol, chemical reactions in the droplets affect aerosol, and droplet evaporation and ice sublimation regenerate aerosol. Very small particles (less than about 10 nm diameter) can be effectively transported to the upper troposphere and become involved in ice nucleation, thereby affecting anvil and cirrus cloud properties and radiative balance. Insoluble gases and nonhygroscopic aerosol particles are not easily removed; therefore convective transport redistributes them in the vertical. For partially soluble trace gases it is very challenging to estimate convective transport because of the complexity of aqueous-phase chemistry and cloud processing. Aqueous-phase chemistry increases the amount of sulfate, and DCC transport and processing of organic compounds may help explain high aerosol particle concentrations at high altitudes over Amazon. However, our qualitative understanding of DCC transport and processing of aerosol particles and their precursor gases, particularly related to organic compounds, is still limited.

Increasing observational evidence of aerosol–DCC interactions has emerged from intensive studies around the world in the last decade or so. Responses to increases in aerosol include systematic increases in the thickness and horizontal coverage of DCC anvils whose warming effect in the terrestrial spectrum may offset the cooling effect in the solar spectrum, enhancement and delay of heavy rain, decreased frequency of light rain and increased frequency of heavy rain, and enlarged cloud coverage of hurricanes. Nevertheless, it has proven difficult to establish causality from observation-based studies, which requires well-constrained measurements of meteorology, cloud, and aerosol properties. Long-term systematic observations from ground sites and satellites would help to isolate the aerosol effects.

Modeling aerosol-cloud interactions has very large uncertainties in coarse-resolution models because DCCs cannot be resolved. Convective parameterizations have limitations when representing various forms of DCCs and usually give little consideration to cloud microphysics and aerosol-cloud interactions. With a grid spacing of 4km and finer, model simulations can better resolve DCCs and aerosol interactions than coarse-resolution models. However, it is computationally challenging to perform such high-resolution simulations globally, and the uncertainties in turbulence and cloud microphysics parameterizations remain major limitations. Studies with different representations of aerosol and cloud microphysics do not show consistent results even for the same cloud case.

Despite the tremendous progress made in the last decade or so, there are still many observational and modeling challenges to reducing the large uncertainty in aerosol interactions with DCCs. As computer

power further increases, global cloud-resolving model simulations could lead to a leap in understanding of aerosol impacts on weather and climate, in combination with ample and more-advanced observations of storm dynamics, thermodynamics, and microphysics at various locations around the world.

14.1 Fundamental aspects of deep convective clouds

Deep convective clouds (DCCs) play critical roles in the water cycle, energy budget, and global circulation. They are prominent in both the midlatitudes and the tropics. The greatest frequency of such clouds is in the equatorial belt, where DCCs are constantly releasing latent heat into Earth's atmosphere (Yuan and Li, 2010; Houze, 2014).

Deep convection refers to the vertical ascent of air parcels from the lower to the upper atmosphere. Such vertical movement is generally associated with environmental instability. In the tropical oceanic region, the instability is driven by strong solar heating and warm oceans, and the high tropopause allows the clouds to be especially deep (Houze, 2014). Over the continental midlatitudes, convective initiation is often associated with complex dynamical processes, such as orography, drylines, and bores (Markowski and Richardson, 2010). A key difference between tropical and midlatitude DCCs, important for the interaction with aerosol, is that cloud bases at midlatitudes are higher, and the freezing level is lower relative to DCCs in the tropics. The difference in the warm cloud depth (above 0°C) can lead to different responses to changes in aerosol, as discussed in Section 14.2.

DCCs manifest themselves in various forms like thunderstorms, supercells, and squall lines depending on the dynamical conditions (Houze, 2014; Markowski and Richardson, 2010). Besides the complex dynamical conditions, DCCs have complicated cloud microphysics and processes (Fig. 14.1), ranging from the warm-cloud regime (liquid only) to the mixed-phase regime (liquid and ice) to the pure ice-cloud regime. Although we have a good understanding of some fundamental microphysical processes such as droplet nucleation, condensation, evaporation, deposition, and sublimation, our understanding of convective microphysics is incomplete in several respects, especially for processes involving ice.

DCCs can be influenced by aerosol particles (both CCN and INP), which can change droplet and ice number concentrations and their size distributions, altering subsequent microphysical processes and the feedback between microphysics and dynamics. Aerosol can also modify the meteorological conditions under which DCCs form through the aerosol radiative effect on surface and atmospheric temperatures. Through these pathways aerosol can affect cloud cover, cloud-top height, precipitation rate, lightning, and hail.

14.1.1 Deep convective cloud dynamics

For an introduction to deep convective cloud dynamics that extends the brief description below, the reader is referred to Houze (2014) and Markowski and Richardson (2010).

14.1.1.1 Convective environment, buoyancy, and vertical motions

Convection occurs where the surface heats up very rapidly or more than neighboring regions. The reduction in air density causes air to rise as a warm thermal or updraft. Cold thermals, generated by latent heat of evaporation, are colder than the environmental air and sink due to their negative buoyancy, producing downdrafts and, at the surface, "cold pools."

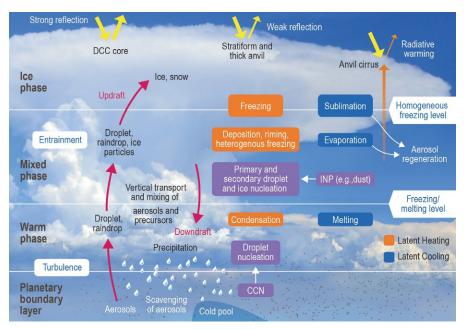


FIG. 14.1

Schematic view of major deep convective cloud microphysical processes and their interactions with dynamic processes, as well as how cloud condensation nuclei (CCN) and ice-nucleating particles (INPs) may interact with DCCs.

Buoyant convection begins at the level of free convection, above which an air parcel may ascend with positive buoyancy. Parcel buoyancy turns negative at the equilibrium level, but vertical momentum may carry it to the maximum parcel level where the negative buoyancy decelerates the parcel to a stop. Integrating the buoyancy force over the parcel's vertical displacement yields convective available potential energy (CAPE), the number of joules of energy available per kilogram of potentially buoyant air. CAPE is the upper limit for an ideal undiluted parcel, and the square root of twice the CAPE is sometimes called the thermodynamic speed limit for updrafts, based on the simple kinetic energy equation $E = \frac{1}{2} \text{m v}^2$. In reality, the net buoyancy, which is determined by thermal buoyancy, water vapor buoyancy, and condensate loading, as well as the vertical pressure perturbation gradient, influence the updraft speed of a DCC. The condensate loading effect is an opposite force countering buoyancy and is often called a "drag."

14.1.1.2 Entrainment and detrainment

Entrainment is the mixing of environmental air into the cloud. It dilutes the parcel and reduces buoyancy. Detrainment is the opposite effect, when the air from a convective cloud, usually toward the top of the cloud, is injected into the environment. Detrainment is also called outflow in the case of DCCs. The amount of outflow, as well as detrained hydrometeor properties, including type, mass, size, and number concentrations, strongly influences stratiform precipitation and the stratiform/anvil area

(Han et al., 2019). These properties in turn determine the radiative properties of DCCs, which are often dominated by the detrained cloud mass. Changes in aerosol can also alter the amount of outflow and hydrometeor properties, influencing the stratiform and anvil properties and area, and hence radiative energy fluxes (Fan et al., 2013).

14.1.1.3 Pressure perturbation fields

Buoyancy in DCCs cannot exist without a simultaneous disruption of the pressure field. Based on the continuity equation of air motion, when the horizontal gradient of pressure accelerates environmental air to replace the buoyant air, a larger vertical pressure perturbation gradient (VPPG) is produced and the updraft is accelerated. This is the buoyancy-sourced VPPG. Another source of VPPG is the dynamic source (Houze, 2014). In most DCCs, the pressure perturbation is buoyancy-source-dominated. However, in severe convective storms, such as supercells, the dynamic VPPG is significant. Changes in aerosol can affect the updraft velocity by influencing both the buoyancy-source and dynamic-source vertical pressure perturbation gradients (Chen et al., 2020).

14.1.2 Deep convective cloud microphysics

Cloud microphysical processes exchange latent heat with the atmosphere, produce precipitation (a key water-cycle process), and influence cloud macrophysical properties such as cloud cover and thickness. These changes alter the radiation budget and influence atmospheric circulation. Microphysical processes, and the ways in which these alter the cloud dynamics (and are also altered by the dynamics), are the most sophisticated and least understood among all common cloud types (Fig. 14.1). By serving as CCN and INP, aerosol particles can directly alter cloud microphysical processes, which can then feed back to cloud dynamics, thereby altering buoyancy and cold pools, thus influencing precipitation, radiation, and circulation (Tao et al., 2012).

14.1.2.1 Warm-phase microphysics

The main microphysical processes in the warm phase (above 0° C) of a DCC are droplet nucleation, condensation, evaporation, rain formation through droplet–droplet collision, ice particle melting, rain accretional growth (by collecting droplets), and breakup of drops.

Droplet nucleation (or aerosol activation) can be quite different in a DCC compared to a shallow cloud (Chapter 12). In particular, particles as small as 50 nm diameter can be activated because of the large updraft speeds, which lead to high supersaturations. Supersaturation can be further enhanced by strong removal of droplets via rain formation, which reduces the water vapor condensation sink. Therefore, secondary droplet nucleation (i.e., away from cloud base) can be stronger in DCCs than in shallow clouds. Strong transport and entrainment of aerosol can also contribute to secondary droplet nucleation. Secondary activation of particles as small as 15 nm has been observed in the humid Amazon (Fan et al., 2018), which is typical of the broad oceanic environment in the tropics.

Condensation, collisional growth, and melting are important for the generation of precipitation in DCCs. Tropical DCCs have low-altitude bases and high-altitude freezing levels that provide enough time for droplets to grow in updrafts through condensation and collisional growth. Because of removal by efficient warm rain, droplet number concentrations inside DCC updraft cores can be very low, particularly in clean conditions where larger droplets increase the production of raindrops. These unique features can lead to strong CCN effects on tropical DCCs. Over the continental midlatitudes, DCCs

often have a relatively high cloud base and low freezing level. Droplets may not have sufficient time to grow into rain before they are lifted to the freezing level, particularly when droplets are small under polluted conditions. The aerosol effects on midlatitude DCCs, therefore, tend to be weaker.

Strong turbulence in DCCs is another unique feature, making the turbulence effect on drop collisions more important than in shallow warm clouds. Quantitative studies of this effect are difficult, and very few microphysics schemes represent it. Aerosol effects on rain in DCCs can be modulated by strong turbulence (Khain and Pinsky, 2018).

Rain collisional breakup and spontaneous breakup are important processes for large raindrops (Paukert et al., 2019). Since DCCs generally produce heavy rain, these processes play more important roles in affecting the rain rate frequency distribution on the ground than in shallow clouds.

14.1.2.2 Mixed-phase and ice-phase microphysics

Precipitation in DCCs is strongly associated with mixed-phase and ice-cloud processes. Precipitation in stratiform regions of DCCs is mainly through the melting of precipitating ice particles from the ice cloud above the melting level. The formation of precipitation below 0°C is known as "cold rain."

Phase transition processes in the mixed-phase region include condensation, evaporation, deposition, sublimation, riming, freezing, and melting. Collisions occur between liquid particles, between ice particles, and between liquid and ice particles. There are also other processes, such as primary and secondary droplet and ice nucleation, and ice multiplication. There is a lack of theoretical understanding of many of the processes in the mixed-phase cloud regime, such as ice nucleation and multiplication and collisions involving nonspherical ice particles. Many of the aerosol effects on DCCs involve changes to these processes, which introduces some fundamental uncertainties in assessments of aerosol-cloud-climate effects.

The anvil and cirrus portions of DCCs consist of ice particles only. These clouds affect Earth's radiation budget by often imposing a warming effect at the top of the atmosphere (Box 14.1). They form convective detrainment of hydrometeors and water vapor. Changes in aerosol can significantly affect anvil properties, such as cloud cover and thickness, through increasing CCN and INP (e.g., Yan et al., 2014; Fan et al. 2010, 2013).

Gaining an understanding of mixed-phase and ice-phase clouds in the convective regions of DCCs is more difficult compared with other types of cloud due to the unique strong-updraft-velocity environment, which makes aircraft or laboratory-based measurements difficult. Nevertheless, we next

BOX 14.1 Deep convective cloud radiative effects

DCCs are composed of a deep convective core, a thick stratiform anvil, and a thinner anvil cirrus (top part of Fig. 14.1). The deep convective core has a large cloud optical depth and therefore strongly reflects incoming solar radiation, but it usually makes up a small fraction of the cloud's overall area. The thick anvil also has a substantial cloud optical depth and therefore also a high reflectance. However, the thin anvil cirrus, which may cover a broad area, only weakly reflects incoming solar radiation, but it strongly absorbs outgoing terrestrial longwave radiation from the warm surface and atmosphere and emits longwave radiation at the local (low) temperature of the upper troposphere. Thin anvil cirrus therefore causes a net positive top-of-atmosphere *cloud radiative effect*—that is, a warming effect. Increases in aerosol cause an increase in the DCC stratiform and anvil area, but the net climatic effect of aerosol–DCC interactions is unclear even qualitatively.

summarize some basic understanding of the major processes. Pruppacher and Klett (1997) and Khain and Pinsky (2018) provide more detailed descriptions.

Primary ice nucleation occurs in mixed-phase and ice-phase clouds via heterogeneous nucleation and homogeneous freezing (see Chapter 15), which can be strongly affected by aerosol (Rosenfeld and Woodley, 2000; Heymsfield et al., 2009; Khain et al., 2012; Fan et al., 2014, 2017a).

Heterogeneous ice nucleation between 0° C and about -38° C can occur in three ways involving various insoluble or partially insoluble INPs: (1) the deposition mode in which water vapor deposition on the surface of INP forms ice crystals, (2) the immersion/condensation mode in which freezing occurs on the surfaces of INPs immersed within the droplets, and (3) the contact mode in which droplets freeze when their surfaces are in contact with INP from either inside or outside of droplets. More details can be found in Chapter 15, Hoose and Möhler (2012) and Murray et al. (2012). Heterogeneous ice nucleation produces ice crystals with concentrations a few orders of magnitude lower than droplet concentrations; thus liquid and ice coexist between 0° C and about -38° C. There is large uncertainty in heterogeneous ice nucleation rates due to a relative lack of laboratory and field observations (compared to CCN). Unlike for CCN, there is also no well-defined theory to predict INP effectiveness.

Homogeneous freezing, without involving INPs, generally occurs at temperatures below about -35° C (Herbert et al., 2015) through the freezing of water drops and hygroscopic aerosol particles when the humidity with respect to ice is larger than threshold values (Ren and Mackenzie, 2005). The latent heat of freezing leads to an increase in buoyancy, which provides a mechanism for CCN and INP to alter cloud dynamics. Numerous small ice crystals in the anvils of DCCs and in cirrus clouds are mainly formed from homogeneous freezing of droplets (Khain et al., 2012).

Ice multiplication, or secondary ice production (SIP), is the production of additional ice particles from an initial population of primary ice particles (Field et al., 2017). Measured ice crystal concentrations above -10° C are often orders of magnitude larger than the INP number concentration. It has thus long been proposed that a secondary ice production process must exist, capable of rapidly enhancing the number concentration following primary ice nucleation. Secondary ice production is important for the production of ice crystal concentration and the subsequent evolution of clouds, but the physical basis of the process is not understood, and the production rates are not well constrained.

Ice particle growth and evolution are complicated in the mixed-phase regime. Because the saturation vapor pressure over ice is less than that over water, ice can grow at the expense of liquid (Korolev and Mazin, 2003; Korolev, 2007), causing droplets to evaporate. This process is known as the Wegener–Bergeron–Findeisen (WBF) mechanism (Wegener, 1911; Bergeron, 1935; Findeisen, 1938). WBF is a major ice growth mechanism in some mixed-phase clouds, particularly when updrafts are weak and supersaturation with respect to water is not high (Fan et al., 2011). However, in DCCs and even in mixed-phase, stratiform clouds with strong updrafts and high water supersaturations both liquid and ice particles may grow simultaneously. Changes in CCN and INP can alter the WBF process by altering droplet and ice concentrations (Fan et al., 2017a). This process affects the supercooled liquid fraction in mixed-phase clouds, thus significantly affecting climate sensitivity in global climate models (Tan et al., 2016) (Chapter 15).

Another important ice particle growth and evolution pathway is through collisions between ice particles and between ice and liquid particles. The collision between ice crystals may form snow. Snow can further grow to large sizes by collecting ice and snow particles, so-called snow aggregation. Ice and snow particles can grow into rimed particles such as graupel and hail by collecting liquid particles, which is the so-called riming process. All collision-related processes are sensitive to the hydrometeor

size distribution and are therefore sensitive to changes in aerosol. Many studies have shown an effect of changes in aerosol on graupel and hail (e.g., Van Den Heever and Cotton, 2007; Carrio and Cotton, 2011; Khain et al., 2011; Li et al., 2021; Wellmann et al., 2018).

14.2 Aerosol effects on deep convective clouds

Changes in aerosol can alter cloud microphysical properties, such as hydrometeor size distributions, as well as cloud morphology such as cloud height and coverage. Changes in cloud microphysical properties affect latent heating, precipitation, and the cloud radiative effect (the net effect of the existence of the cloud—see Box 14.1), which feed back to cloud dynamics and cloud structure (Fig. 14.1).

14.2.1 Effects on cloud microphysics

Changes in aerosol can alter the number, size, and phase of cloud particles by acting as CCN or INP, or both. The ability of aerosol particles to act as CCN is determined by the particle size, composition, and mixing state, which is described in Chapter 5. For a given cloud water amount and updraft speed, the cloud particle size is strongly determined by CCN and INP concentrations due to their competition for water vapor, and thus an increase in CCN concentration leads to a higher concentration of smaller cloud particles (Chapter 12).

Increases in CCN suppress warm rain and enlarge anvil cloud cover. Increasing CCN increases droplet number and reduces droplet size, suppressing the collision-coalescence process for warm rain (or effectively delays it to higher in the cloud). This allows more, smaller droplets to be transported to altitudes where temperatures are below 0°C. Freezing of a greater number of smaller droplets increases the ice crystal number concentration but reduces their size, resulting in larger stratiform and anvil area (up to 100% increase) because of slower sedimentation of ice particles (e.g., Fan et al., 2010, 2013; Yan et al., 2014; Morrison and Grabowski, 2011; Chen et al., 2017, 2020). This microphysical effect has an important implication for how changes in aerosol affect cloud radiative properties (Section 14.2.4). Increases in supercooled droplet number and mass expand the mixed-phase zone and decrease the cloud glaciation temperature (Khain et al., 2001; Rosenfeld et al., 2011; Fan et al., 2017a). The increase in droplet number enhances the condensation rate, which releases more latent heat. The latent heat can also be enhanced in the cold cloud phase by freezing more liquid. The feedback of latent heat to cloud dynamics can invigorate convection (see details in Section 14.2.2). Increases in supercooled liquid may affect hail frequency and size. Explicit model simulations show that increases in CCN concentration result in more hail and increased hailstone sizes induced from more supercooled droplets (Khain et al., 2011; Loftus and Cotton, 2014; Ilotoviz et al., 2016).

Observations from space-borne and ground-based instruments clearly show the effect of variations in aerosol on the microphysical and macrophysical properties of DCCs. Increased aerosol loadings are correlated with increased cloud fraction (Koren et al., 2005, 2008, 2010), cloud thickness, and frequency of occurrence (Li et al., 2011; Niu and Li, 2012; Storer et al., 2014; Wall et al., 2014; Sarangi et al., 2017, 2018; Zhang et al., 2007b). These "cloud invigoration" effects are accompanied by systematic changes in cloud microphysics, especially in ice particle size and ice water path (Peng et al., 2016; Jiang et al., 2018), as well as in radar reflectivity which depends on particle size and ice water content (Min et al., 2009; Storer et al., 2014; Chen et al., 2016; Massie et al., 2016; Jiang et al.,

2018). However, the changes depend on several factors including aerosol type, cloud phase, atmospheric thermodynamics, as emphasized in the studies listed above.

Increases in INP enhance ice number in the mixed-phase regime but have varied effects on ice particle size depending on cloud depth. Observations show that the ice particle effective radius (see Chapter 12) near the cloud top decreases with increasing aerosol loading in strong convective systems, but the radius increases in moderate convection (Fig. 14.2) (Zhao et al., 2019; Peng et al., 2016). This is because ice formation in deep convection is dominated by homogeneous freezing of cloud droplets, so increases in aerosol loading lead to more, smaller droplets and smaller ice particles. In less-deep convection (mixed-phase), ice is mainly formed from heterogeneous ice nucleation, so increases in INP

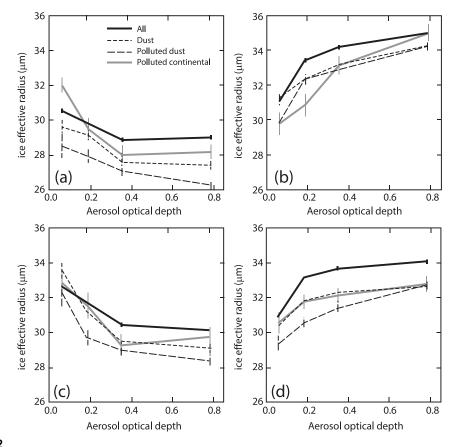


FIG. 14.2

Relationships between column aerosol optical depth and ice effective radius (Rei) of cold-top convective clouds and anvil cirrus with different ranges of cloud-top height (CTH) or convective available potential energy (CAPE). (a) >67% percentile of CTH, (b) <33% percentile of CTH, (c) >67% percentile of CAPE, and (d) <33% percentile of CAPE. For each CTH/CAPE group, the aerosol optical depth of each aerosol type is divided into four bins.

lead to enhanced ice particle growth through the WBF process. Modeling studies show that the INP effects on cloud microphysics, convection, and cloud radiative effects may depend on the nature of the convective cloud (Fan et al., 2010) and the ice nucleation parameterization employed (Hawker et al., 2021).

For the pure ice-cloud zone, the relationship between INP and ice particle size depends on relative humidity, with a positive relationship under dry conditions and a negative relationship under moist conditions (Zhao et al., 2018). Since secondary ice production mechanisms are not well understood or constrained (Field et al., 2017), it is not clear how it is affected by changes in aerosol. Recent studies show that the role of secondary ice production in the cloud radiative effect depends on INP properties (Hawker et al., 2021) and the secondary ice production is enhanced as CCN concentrations increase (Gayatri et al., 2022). Another important effect of INP is the warmer cloud glaciation temperatures, which decreases the mixed-phase cloud fraction but increases the ice cloud fraction (e.g., Zhang et al., 2015; Fan et al., 2017a). Glaciation in clouds affected by dust and aerosol pollution was observed to occur at relatively high temperatures near -20° C based on satellite measurements (Yuan et al., 2010; Rosenfeld et al., 2011, 2014).

Observational limitations complicate studies of aerosol effects on cloud microphysics. CCN and INP retrievals from satellite observations are not straightforward for DCCs. In general, aerosol optical depth (τ_a) and the aerosol index retrievals have been used as proxies for CCN (Kaufman and Nakajima, 1993; Nakajima et al., 2001, 2003; Yuan et al., 2008), but the uncertainty is large (Li et al., 2009b) and the relationship with CCN is not robust (Andreae, 2009; Liu and Li, 2014). A key problem is that τ_a can be enhanced by high humidity near clouds (Jeong and Li, 2010). Neglecting this effect led to a systematic underestimation of the aerosol microphysical effect on DCCs (Liu and Li, 2018). For a given aerosol sample, the conversion from aerosol particles to cloud particles depends critically on updrafts that are now retrievable from satellite for some particular cloud regimes (Zheng et al., 2015, 2016), and so are CCN (Rosenfeld et al., 2016). Having these two quantities at cloud base is essential for differentiating between the aerosol microphysical effect and the dynamic effect. However, the retrieval may not work well for DCCs, so more studies are needed.

14.2.2 Effects on cloud dynamics

Aerosol effects on cloud dynamics occur through the feedback of microphysical processes to dynamics. The convective intensity of DCCs can be enhanced, and a few mechanisms have been proposed.

Water-phase invigoration can occur because more-numerous, smaller droplets results in an increase in condensation thus latent heat release at the lower part of DCCs, which scales approximately with the product of droplet number and mean droplet size (Korolev and Mazin, 2003). The increased convective intensity resulting from condensational heating, which can occur in the liquid-only and mixed-phase zones of DCCs, has been extensively reported (e.g., Fan et al., 2007a, 2018; Sheffield et al., 2015; Chen et al., 2017; Lebo, 2018). The water-phase invigoration can be particularly important in DCCs because of strong updraft speeds and low droplet concentrations as a result of efficient warm rain formation and growth, both of which lead to very high supersaturation, particularly in clean conditions. Very high supersaturation activates ultrafine aerosol particles (UAP; diameter 50 nm or smaller), which usually have very high number concentrations in polluted conditions, amplifying aerosol effects (Khain et al., 2012). The strong invigoration by UAP (nearly doubled for the mean of top 10 percentile updraft speeds) has been demonstrated near an industrial city in the central Amazon region

(Fan et al., 2018). Here the air is very humid, the background aerosol loading is very low (pristine conditions), and the ratio of UAP to large aerosol particles is very large in the pollution plume. The droplets formed at cloud bases are low in number; thus they quickly grow into raindrops, reducing the droplet surface area available for condensation and increasing in-cloud supersaturation, which thereby allows a greater number of UAP to be activated (Fig. 14.3). The UAP effect is of particular interest to aerosol-climate science since such particles were thought to be too small to be activated in clouds. The UAP effect has not been widely explored yet but has been reported in several recent studies (Chen et al., 2017, 2020; Zhang et al., 2021; Grabowski and Morrison, 2021). The role of this mechanism in altering the cloud radiative effect remains to be established.

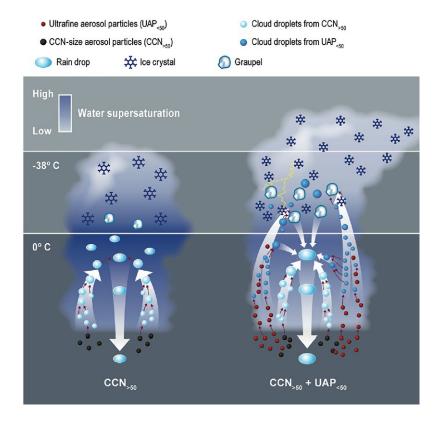


FIG. 14.3

The effect of ultrafine aerosol particles on deep convective clouds. In clouds that lack ultrafine aerosol particles smaller than 50 nm (UAP $_{<50}$; *left*), the clouds are highly supersaturated as a result of fast drop coalescence that forms warm rain and reduces the integrated droplet surface area available for condensation. With added UAP $_{<50}$ (*right, red dots*), an additional number of cloud droplets are nucleated above the cloud base, which lowers supersaturation drastically by enhanced condensation, releasing additional latent heat at low and middle levels, thus intensifying convection (i.e., water-phase invigoration).

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Ice-phase invigoration occurs because an increase in CCN suppresses warm rain and allows more cloud water to be lifted to altitudes where temperatures are below 0°C. Freezing of the extra liquid and the subsequent enhancement of ice processes such as deposition and riming lead to larger latent heat release in the mixed and ice phases, which invigorates convection (e.g., Andreae et al., 2004; Koren et al., 2005, 2008; Khain et al., 2005; Rosenfeld et al., 2008). At the same time, lifting more cloud water to higher levels increases the loading effect. The extent to which loading offsets invigoration is debated (Fan and Khain, 2021; Grabowski and Morrison, 2016, 2021). Several studies with detailed cloud microphysics schemes showed that the loading effect is not large enough to completely offset the latent heating effect (e.g., Khain, 2009; Lebo et al., 2012; Tao and Li, 2016; Chen et al., 2020). Parcel model estimates with the hydrometeor loading effect considered showed this mechanism can increase convective energy by 50% because of the off-loading of hydrometeors by precipitation (Rosenfeld et al., 2008). Ice-phase invigoration was found to be the major mechanism invigorating midlatitude warm cloud-based DCCs because cloud droplets formed at cloud bases can be readily transported to altitudes where temperatures are below 0°C (e.g., Khain et al., 2005; Fan et al., 2012b). But it is shown to play a much less significant role than the water-phase invigoration in a few recent studies (Fan et al., 2018; Lebo, 2018; Zhang et al., 2021).

Changes in aerosol can modify cloud dynamics by influencing cold pools. A cold pool is a region of evaporatively cooled air that is transported to the surface in convective downdrafts. Several studies show that increases in aerosol weaken the intensity of cold pools (Storer et al., 2010; Morrison, 2012; Lebo and Morrison, 2014; Chen et al., 2020), while other studies show that it enhances them (Morrison, 2012; Tao et al., 2007). The response depends on wind shear conditions (Lee et al. 2008; Chen et al., 2020) and the altitude of the dry layer (Grant and Van Den Heever, 2015). Also, how cold pool intensity affects convection is complicated. For example, for squall line systems, both Lebo and Morrison (2014) and Chen et al. (2020) found that the weakened cold pool intensity in the polluted case invigorated convection because of a more optimal balance between the cold pool and the environmental shear for more upright convection (the Rotunno-Klemp-Weisman theory; Rotunno et al., 1988). However, strengthening of cold pool intensity by an increase in aerosol leading to enhanced low-level convergence and convection has also been reported for squall lines (Tao et al., 2007) as well as other convective systems (Morrison, 2012; Chen et al., 2020).

Convective invigoration by moistening the environment has been recently reported for aerosol impacts on DCCs (Abbott and Cronin, 2021; Chua and Ming, 2020). Enhanced droplet evaporation in a polluted condition may increase environmental humidity, favoring stronger convection over a long time scale. The two studies of this phenomenon used fixed droplet concentrations and a two-moment cloud microphysics scheme, both of which may lead to overestimation of droplet evaporation, which could exaggerate the effect. Besides, the water-phase invigoration, which was shown to play an important role in tropical environments, was not considered in both studies due to the use of saturation adjustment for condensation. The idealized lateral boundary condition setup can be another potential source of the large increase in moisture by aerosols shown in those studies. Therefore, more studies are needed to gain a clear understanding of the significance of this mechanism.

Overall, CCN effects on cloud dynamics operate mainly through microphysics-dynamics feedback, which depend on environmental conditions, including aerosol properties, wind shear, relative humidity, and CAPE (e.g., Khain et al., 2008; Lee et al., 2008; Khain, 2009; Fan et al., 2007a, 2008, 2009; Storer et al., 2010; Tao et al., 2012; Li et al., 2017, 2019). Convective invigoration tends to occur in DCCs with warm cloud bases, weak wind shear, and humid conditions. The presence of absorbing

aerosol or strong wind shear conditions could suppress the invigoration by increased aerosol (e.g., Fan et al., 2008, 2009; Jiang et al., 2018; Koren et al., 2008).

14.2.3 Effects on precipitation

Precipitation can be affected by aerosol through various pathways involving changes in cloud dynamics and microphysics. The sign and magnitude of aerosol effects on precipitation vary widely from -93% to 700% in cloud-resolving model studies (as summarized in Tao et al., 2012) and in measurements (Li et al., 2016, 2019), although absolute changes may not always be very large—see Fig. 14.4.

In the initial stage of DCC development, when clouds are in the warm phase, increasing the number of CCN often suppresses precipitation. As the clouds deepen, CCN can enhance convection through the mechanisms listed in Section 14.2.2, which may increase precipitation. Aerosol is found to increase precipitation in many studies but an optimal CCN exists (e.g., Li et al., 2008; Cui et al., 2011).

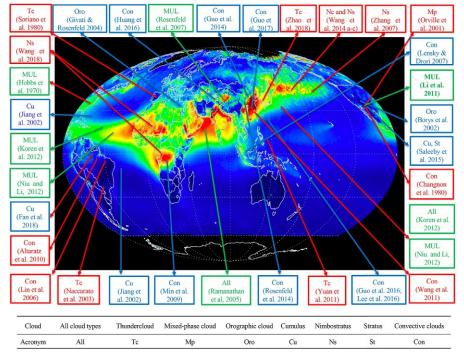


FIG. 14.4

A survey of observation-based studies of the effects of changes in aerosol on precipitation around the world sorted by location and cloud/precipitation regimes (Tc, Con, Mp, Oro, Cu, Ns, St, Con; acronyms defined in the plot). *Colored text* denotes enhanced (*red*), suppressed (*blue*) and dual effects (*green*) on precipitation by increases in aerosol.

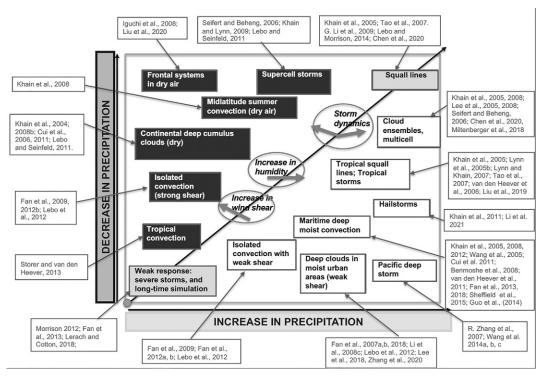


FIG. 14.5

A summary of the aerosol effects on deep convective clouds under distinct meteorological conditions by acting as CCN, as simulated by cloud-resolving models.

Updated based on Khain (2009).

However, precipitation is very complicated and many other processes besides convective intensity could modulate it.

Precipitation responses to changes in aerosol depend on environmental conditions, including environmental relative humidity (Khain et al., 2004, 2008; Fan et al., 2007a, 2009; Tao et al., 2007; Yuan et al., 2008), wind shear (Leo et al., 2008, 2012; Khain, 2009), and CAPE (Lee et al., 2008; Storer et al., 2010; Liu et al., 2016)—Fig. 14.5. In general, for isolated DCCs, precipitation tends to increase when CCN increase under moist and weak wind shear conditions and decrease under dry and strong wind shear conditions. However, for mesoscale convective systems, increases in CCN enhance precipitation by 10%–30% under different wind shear conditions (Chen et al., 2020). Variations in meteorological conditions cause large uncertainty than variations in aerosol concentration, particularly for cloud dynamics and precipitation. When there is covariability between aerosol properties and meteorological conditions, aerosol effects on precipitation may not be as significant as those induced by the meteorological changes (Morrison, 2012; Grabowski, 2018). In observations, it can be very difficult to isolate aerosol effects due to their covariability with other factors, and very long-term measurements may be needed in the regions where meteorological conditions vary significantly, such as the U.S.

Southern Great Plains (Varble, 2018). However, recent studies using ensemble methods that perturb both aerosol and meteorological conditions showed aerosol-induced changes in precipitation are still statistically significant (Miltenberger et al., 2018; Li et al., 2021). In addition, the uncertainties in aerosol effects can be more affected by cloud microphysical parameters than the meteorological variables (Morrison, 2012; Wellmann et al., 2020).

The effect on precipitation also depends on the light-absorbing properties of aerosol particles. When the particles are strongly absorbing, like black carbon, they can heat some part of the atmosphere, depending on their location (horizontally and vertically), which changes atmospheric stability and circulation, leading to complicated responses of DCCs and precipitation to aerosol loading (Lau et al., 2006; Bollasina et al., 2011; Wang et al., 2013; Yang et al., 2013a,b, 2016; Fan et al., 2008, 2015).

Giant CCN (with radii in the micrometer range or larger) may have a strong influence on precipitation because they can be activated directly into rain embryos and readily initiate the warm-rain process (e.g., Feingold et al., 1999; Lasher-Trapp et al., 2001). In DCCs, studies show that giant CCN enhance precipitation in continental clouds (where the background concentration of small CCN is high) because of enhanced coalescence (Yin et al., 2000; Van Den Heever et al., 2006).

Often the total precipitation over a domain may not be sensitive to the aerosol perturbation because of the compensation and buffering effects from cloud microphysics (Fan et al., 2013), cloud dynamics (Stevens and Feingold, 2009), and the convective—radiative quasi-equilibrium relationship (Grabowski and Morrison, 2011). However, changes in aerosol can significantly alter the probability distribution of precipitation rates. Many observational and modeling studies have reported the reduced frequency of light rain but increased frequency of heavy rain (e.g., Qian et al., 2009; Li et al., 2011; Wang et al., 2011c; Fan et al., 2012a, 2013; Tao et al., 2012; Koren et al., 2012; Guo et al., 2014; Jiang et al., 2016). It has been attributed to suppression of warm rain and invigoration of convective intensity as described in Section 14.2.2.

Changes in aerosol may affect precipitation through the INP effect. Increasing the number of INP generally enhances precipitation in mixed-phase and deep clouds as a result of a stronger WBF process where ice particles grow at the expense of evaporated water droplets, and enhanced riming (e.g., Van Den Heever et al., 2006; Ekman et al., 2007; Fan et al., 2014, 2017a). Fan et al. (2014, 2017a) have shown that the INP effect on mixed-phase cloud precipitation is generally larger than the CCN effect.

14.2.4 Effects on cloud macrophysical properties and radiation

Increased cloud fraction, cloud-top height, and cloud thickness are associated with increases in aerosol. These associations are seen in analyses of field experiments (Andreae et al., 2004), long-term (10 years) measurements at a fixed site in the U.S. Southern Great Plains (Li et al., 2011; Yan et al., 2014), and satellite-retrieved data regionally (Koren et al., 2005, 2010; Sarangi et al., 2017, 2018; Storer et al., 2014; Wall et al., 2014) and globally (Koren et al., 2008; Niu and Li, 2012; Peng et al., 2016). Many modeling studies have shown qualitatively the same results (e.g., Fan et al., 2010, 2013; Grabowski and Morrison, 2016; Chen et al., 2017, 2020), although the magnitudes vary greatly with the study period, location, and models employed. A contrary effect can occur when light-absorbing aerosol heats and stabilizes the lower atmosphere leading to weaker convection and reduced cloud fractions and cloud-top heights (e.g., Fan et al., 2008; Jiang et al., 2018). Observation-based studies of the effects of aerosol on cloud properties are more challenging to interpret in terms of causal relationships than model-based studies. Varble et al. (2011), for example, found that the relationship

between cloud top height and aerosol number concentration covaries with CAPE. While this may undermine, to some extent, the causal relationships found in Li et al. (2011), the aerosol–cloud interaction itself can alter the CAPE especially if the CAPE was determined after clouds developed.

An increase in aerosol can increase cloud fractions and cloud-top heights (thicknesses) through invigorated convective intensity and reduced fall velocities of smaller ice particles that result in slower dissipation of anvil clouds (Fan et al., 2013; Grabowski and Morrison, 2016). In addition, suppressed warm rain in the lower part of DCCs allows more cloud mass to be detrained into the stratiform area in polluted conditions. According to monthly simulations of summer convection at cloud-resolving scales over the tropical west Pacific, southeast China, and the US Southern Great Plains, the reduced ice fall velocity because of smaller ice particle size is mainly responsible for the increased cloud fraction and thicker anvil clouds (Fan et al., 2013).

DCC cores (the deepest part of DCCs) have a strong radiative cooling effect because of their thick optical depth. In the tropical land, the cloud radiative effect (the net effect of the cloud) for DCC cores can be up to $-544 \,\mathrm{W\,m^{-2}}$ (Peng et al., 2016). Thick stratiform/anvil areas of DCCs also have a cooling effect, while thin anvils can have a warming effect (Box 14.1). Increases in cloud-top height and expansion of the anvil coverage caused by an increase in aerosol would modulate the cloud radiative effect. The effects of aerosol on anvil radiative properties are not yet well understood and the global radiative effect has not been quantified. A recent study of tropical convection estimated a global effect of order 0.2–0.5 W m⁻² cooling for a doubling of aerosol (Nishant et al., 2019). Observational studies based on satellite and ground data have shown a net daily mean positive radiative effect of 29.3 W m⁻² at the TOA and 22.2 W m⁻² at the surface at the Southern Great Plains site from the clean to polluted conditions for days with DCCs (Yan et al., 2014). Accounting for the frequency of occurrence of DCCs and aerosol loading, the long-term (10 years) diurnal mean forcing of DCCs induced by aerosol amounts to $0.5 \,\mathrm{W\,m^{-2}}$ warming at the TOA. However, over the tropics, a negative radiative effect by aerosol at the TOA up to $-76 \,\mathrm{W\,m^{-2}}$ was seen because shortwave cooling overwhelmed the longwave warming (Peng et al., 2016; Sarangi et al., 2018). Although there is no qualitative agreement in the net forcing, the increased cloud fraction caused by an increase in aerosol consistently enhances surface cooling in the daytime and warming in the nighttime, leading to a reduced diurnal temperature variation (\sim 0.5–0.6 K), as observed (Sarangi et al., 2017) and simulated (Fan et al., 2013). A solid understanding of aerosol radiative forcing through DCCs requires quantification of the changes of optically thick and thin anvil clouds.

At very high aerosol loading there is evidence that the sign of the aerosol effect could reverse. Peng et al. (2016) and Koren et al. (2008) observed a transition from a cooling to a warming cloud radiative effect as aerosol optical depth increased. The hypothesis is that when aerosol optical depth exceeds a certain value, the aerosol radiative effect dominates and suppresses DCC development.

An increase in INPs reduces the convective anvil extent as found in several studies (e.g., Hawker et al., 2021; Gasparini et al., 2020; Kärcher and Lohmann, 2003; Lohmann and Gasparini, 2017), caused by increased liquid consumption in the mixed-phase regime due to heterogeneous freezing. However, increased anvil extent was also reported in cloud-resolving model simulations because of higher ice number and reduced ice particle size (Fan et al., 2010). Hawker et al. (2021) noted that the reduction in the anvil extent caused by INPs are somewhat offset by the overall increases in the cloud fraction across the domain. They showed that the presence of INPs increases domain-mean daylight TOA outgoing radiation by between 2.6 and 20.8 Wm⁻², depending on the choice of INP parameterization.

Under anthropogenic climate change, DCCs will change in response to changes in greenhouse gases (Prein et al., 2017) and aerosol. Currently, there is no study comparing the aerosol-induced radiative effect with the anthropogenic warming-induced effect on DCC, mainly because global models are currently too coarse to simulate DCCs let alone aerosol-cloud interactions effects. Qualitatively, we know that the increased anvil cover caused by increased aerosol should to some extent compensate the reduced anvil cloud cover caused by climate warming and the increased stability of the upper atmosphere and increased convection aggregation (Bony et al., 2016). As concluded in Kreidenweis et al. (2019), the resulting radiative forcing on the climate system due to aerosol–cloud interactions remains a large uncertainty in future climate projections.

14.3 Aerosol effects on severe storms

Severe storms refer to storms that can render property damages and/or loss of life by producing hazardous weather, such as flash floods, hail and tornados, strong winds, and lightning. They are primarily driven by large-scale and synoptic-scale dynamic and thermodynamic conditions. Aerosol can affect these storms by modulating small-scale microphysics, thermodynamics, and circulation, which might offset or amplify the effect from greenhouse gases. As aerosol properties may covary with meteorological quantities, the true effects of changes in aerosol are not readily observable in the real atmosphere. Given the large uncertainty, below, we will just briefly summarize the relevant work.

14.3.1 Effects on tropical cyclones and mid-latitude cyclones

A tropical cyclone is a large rotating storm system characterized by a low-pressure center, closed low-level circulation, strong winds, and a spiral arrangement of thunderstorms that produce heavy rain or squalls (Lupo, 2016). Depending on its location and strength, a tropical cyclone is referred to by different names, such as a hurricane, a typhoon, or a tropical storm. Experimental studies on the effects of aerosol on typhoons date back to 1946–47 when the US Navy and Air Force conducted Hurricane Hunter missions over the Pacific and Atlantic Oceans, "seeding" hurricanes over the Atlantic to test the hypothesis that it would help initiate rainfall but reduce its intensity. The latest study by means of both observation analysis and modeling indicates that an increase in aerosol can enhance precipitation and lightning by a factor of 2 for Hurricane Harvey in the urban area of Houston in 2017 (Pan et al., 2020).

Maximum wind speeds of cyclones ingested with increased CCN (varying from 2 to 15 times in different studies) are reduced due to the suppression of coalescence and the subsequent invigoration of the outer rainbands. The invigorated outer bands prevent air from flowing into the typhoon eye and weaken it, increasing the eye diameter and reducing the wind speed (Rosenfeld et al., 2007, 2012; Zhang et al., 2007a, 2009; Khain et al., 2010; Carrio and Cotton, 2011). Increases in aerosol would offset the global warming effect, which is expected to intensify tropical cyclones (Knutson et al., 2020). The mechanisms of aerosol effects on hurricanes are summarized by Rosenfeld et al. (2012).

Tropical cyclone frequency could be influenced by the aerosol radiative effect. Studies show that dust over the Atlantic could significantly reduce the occurrence of hurricanes (Lau and Kim, 2007; Sun et al., 2008) by reducing sea surface temperatures. This cooling effect on sea-surface temperature works against the global warming effect, as most studies have projected an increased tropical cyclone

intensity (globally averaged) with a 2°C warming based on the recent review (Knutson et al., 2020). Dunstone et al. (2013) have shown that long-term historical changes in anthropogenic aerosol played a key role in the decadal variability of hurricanes over the North Atlantic in the twentieth century through aerosol-induced shifts in the Hadley circulation.

Anthropogenic aerosol from Asia affects typhoons in the Western Pacific (Zhang et al., 2009; Wang et al., 2014c). Increased typhoon rainfall rate and rainfall area were observed based on data from 2000 to 2015 (Zhao et al., 2018). Volcanic eruptions can also affect the activities of tropical cyclones. Strong volcanic eruptions cause strong asymmetric cooling in the Southern or Northern Hemispheres and the intertropical convergence zone. This, in turn, leads to changes in the distribution of tropical cyclones. Its impact can last for at least 4 years (Pausata and Camargo, 2019) and is different at different latitudes of oceans (Yan et al., 2018). However, there is no evidence showing that volcanic aerosol affected the number of tropical cyclones based on observations, reanalyses, and models (Camargo and Polvani, 2019). Aerosol cooling reduces tropical cyclone "potential intensity" more strongly, by about a factor of 2 per degree change in sea surface temperature, than greenhouse gas warming increases it (Sobel et al. 2019).

For mid-latitude cyclones, observations and modeling studies show that air pollution from Asia has altered mid-latitude cyclones and the storm track in the North Pacific in the past three decades (Wang et al., 2014b, 2014a; Zhang et al., 2007b), leading to increased cloud amount and precipitation.

14.3.2 Effects on mesoscale convective systems (MCSs)

MCSs commonly occur in the tropics and mid-latitudes and are responsible for the majority of extreme rainfall and much of other weather hazards such as hail and tornadoes (Houze, 2018). They also have significant effects on radiation and circulation. Thus, how MCSs are influenced by aerosol is of significant interest. MCSs exist in various forms depending on how the system is organized. Squall lines are linearly organized MCSs and they cause heavy precipitation, hail, frequent lightning, strong straight-line winds, and possibly tornadoes. A unique feature of the squall-line organization and maintenance is the balance between the cold-pool strength and the low-level environmental wind shear, that is, the Rotunno-Klemp-Weisman theory (e.g., Rotunno et al., 1988). Aerosol can affect squall-line dynamics and organization by altering evaporative cooling and hence the cold-pool strength. Some modeling studies have shown that an increase in CCN concentration reduces the cold-pool intensity (Lebo and Morrison, 2014; Chen et al., 2020). Modeling results regarding CCN effects on squall-line precipitation vary depending on the model and microphysics parameterizations employed (Khain et al., 2015; Li et al., 2009a). Precipitation in the stratiform area of a squall line is increased by increasing CCN (Chen et al., 2020), and the anvil cover of a squall line is increased (Saleeby et al., 2016).

A recent study of various types of MCSs found that an increase in CCN enhanced vertical mass fluxes, increased precipitation (by 12%–30%), and increased cloud anvil area (Chen et al., 2020). The aerosol radiative effect also enhanced an MCS-induced flooding event in China in a modeling study (Fan et al., 2015). Due to the significance of MCSs in precipitation, radiation, and circulation, extensive studies of aerosol impacts on them are needed. With anthropogenic warming, North American summertime MCSs (including squall lines) are projected to occur more frequently, with a 15%–40% increase in maximum precipitation rates (Prein et al., 2017). There is a lack of studies comparing aerosol effects with anthropogenic warming effects on MCS precipitation.

14.3.3 Effects on hail and tornadoes

Hail and tornadoes are generally produced by severe convective storms. Together, they are responsible for more than 60% of annual average economic losses in the United States (Gunturi and Tippett, 2017). Changes in aerosol could affect hail embryo formation and growth by changing the supercooled water droplet number and mass and updraft speeds. Modeling studies have shown that an increase in aerosol in a DCC can increase the likelihood of producing large hailstones, but the mechanisms are not consistent (Loftus and Cotton, 2014; Khain et al., 2011; Ilotoviz et al., 2016). Observations have shown that hail and tornado frequencies have a weekly cycle that correlates with aerosol pollution (Rosenfeld and Bell, 2011) but the causal relationship is difficult to establish (Yuter et al., 2013). Biomass-burning aerosol from Central America has been linked with the increase in severe weather events (hail and tornadoes) over the central U.S. (Wang et al., 2009; Saide et al., 2015, 2016). The interannual variability of hail at the SGP site was found to correlate with variability in aerosol from northern Mexico (Jeong et al., 2020).

By serving as INP, aerosol can suppress hail by creating a large number of ice crystals competing with each other for supercooled cloud water and not becoming hailstones. This is the underlying hypothesis of cloud seeding with silver iodide INP for hail suppression.

Studies of both aerosol effects and anthropogenic warming effects on hail and tornadoes have been preliminary, with large uncertainties (Allen, 2017; Li et al., 2021). No meaningful comparison can be derived yet.

14.3.4 Effects on lightning

Lightning occurs in thunderstorms due to the release of cloud electrification when graupel collides with ice crystals in the presence of supercooled cloud droplets and strong updrafts (Williams, 2005). Increasing the amount of aerosol in clouds suppresses coalescence and leads to an enhanced amount of supercooled water, along with stronger updrafts from more latent heating, which enhances cloud electrification (Andreae et al., 2004; Rosenfeld et al., 2008; Wang et al., 2011c).

The effect of aerosol is among the factors contributing to the contrasting lightning occurrence between oceans and land, between cities and rural areas, and between regions/seasons governed by different types of aerosol and meteorological conditions (Rosenfeld and Lensky, 1998; Orville et al., 2001; Williams and Stanfill, 2002; Kar and Liou, 2014; Proestakis et al., 2016; Yair, 2018; Tinmaker et al., 2019). The aerosol effect is usually more significant under clean background conditions, as seen from the enhancement of lightning activity along major shipping lanes (Thornton et al., 2017).

The competing effects of the CCN effect and the aerosol-radiation effect (Section 14.2.4) can also be seen in the lighting response (Altaratz et al., 2010; Stallins et al., 2013; Li et al., 2018). This is vividly illustrated in Fig. 14.6, showing the relationship between lightning frequency and aerosol optical depth in dust-dominated northern Africa and smoke-dominated central and southern Africa (Wang et al., 2018). Despite the distinct differences in aerosol type and meteorology, there is a common threshold at the aerosol optical depth value of \sim 0.3, below which lightning increases with increasing aerosol optical depth (due to the CCN effect) and then decreases (due to aerosol-radiation effects). Although these studies are long-term analyses based on observations, the covariability between aerosol and meteorology might still contribute to the uncertainty of the analysis.

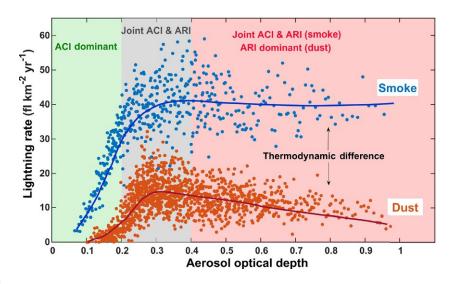


FIG. 14.6

Lightning flash rate as a function of aerosol optical depth in regions dominated by dust (*orange points*) and smoke (*blue points*). Turning points in the curves occur around an aerosol optical depth of 0.3. Aerosol–cloud interactions play a dominant role in lightning activity under relatively clean conditions. When aerosol optical depth exceeds 0.3, both aerosol–cloud and aerosol–radiation interaction effects come into play with different magnitudes. For dust aerosol, aerosol–cloud and aerosol–radiation interactions have the same effect of suppressing convection in the dry environment, favorable for evaporating cloud droplets. The moist environment of central Africa strengthens aerosol invigoration, offsetting the suppression due to aerosol–radiation interaction, leading to a nearly flat line when aerosol optical depth is larger than 0.2.

From Wang et al. (2018).

The relative importance of the CCN effect and the aerosol-radiation effect has also been observed in China. In central China, aerosol absorption of solar radiation has increased the stability of the lower atmosphere, reducing thunderstorm activity by 50% from the 1960s to the end of the last century (Yang et al., 2013a; Li et al., 2016), while in the south and southeast China increases in hygroscopic aerosol have significantly stimulated lightning activities over recent decades (Wang et al., 2011c; Yang and Li, 2014).

14.4 Effects of deep convective clouds on aerosol particles and precursor gases

DCCs affect aerosol and trace gases through three pathways (Fig. 14.1): (1) transport from the lower to the upper troposphere; (2) scavenging by clouds and precipitation; and (3) cloud processing through aqueous chemistry and regeneration of aerosol through droplet evaporation and ice sublimation.

14.4.1 Vertical transport and scavenging

Many field campaign observations and modeling studies have shown that convective storms affect nitrogen oxides, ozone, and aerosol in the upper troposphere (Dickerson et al., 1987; Dye et al., 2000; Li et al., 2005; Allen et al., 2010; Barth et al., 2012, 2015). Soluble gases and aerosol can be effectively removed by in-cloud scavenging and rain-out, so vertical transport might not be an important process. For example, the wet scavenging efficiency (i.e., the removal by cloud and precipitation) for gaseous nitric acid is 87% and over 80% for sulfate and ammonium particles, based on field campaign data for DCCs (Yang et al., 2015; Bela et al., 2016). However, ultrafine aerosol particles smaller than 50 nm diameter require very high supersaturations to activate so they are not efficiently removed by in-cloud scavenging, and they are too small to be removed by precipitation. Thus they may be transported to the upper troposphere and become involved in ice nucleation and affect high-level cloud properties (Rosenfeld and Woodley, 2000; Heymsfield et al., 2009).

Insoluble gases and aerosol particles are not easily removed; therefore convective transport can redistribute them vertically and horizontally, thereby affecting the composition of the upper troposphere and regions downwind (Dickerson et al., 1987; Dye et al., 2000). Convective transport has been shown to enhance upper tropospheric aerosol particle number and mass concentrations by factors of 2–3 and 3–4, respectively (Yin et al., 2005), and aerosol originating from the boundary layer can be more efficiently transported upward aerosol from the midtroposphere (Yin et al., 2012). For partially soluble trace gases (e.g., formaldehyde, hydrogen peroxide, and methyl hydroperoxide), which are important precursors for hydroxide and ozone, it is very challenging to estimate their convective transport (Barth et al., 2015). The Deep Convective Clouds and Chemistry (DC3) field campaign over the United States (Barth et al., 2015) measured gas and aerosol vertical profiles during convective storms, showing that the mixing ratios of insoluble gases in the upper troposphere convective outflow consisted of 47% low-level (<3km) inflow air, 32% entrained mid-tropospheric air, and 21% upper tropospheric air (Yang et al., 2015). The wet scavenging efficiencies were 80%–84% for submicron aerosol organic, sulfate, and ammonium. Secondary droplet nucleation was found to be very important to aerosol wet removal in deep convective storms (Yang et al., 2015).

14.4.2 Cloud processing and aerosol regeneration

Particle mass can be produced by clouds through aqueous-phase chemical reactions, and can be regenerated by drop evaporation and ice sublimation. There is a relatively robust understanding of sulfate formation in clouds (Walcek and Taylor, 1986; Barth et al., 2000; Rasch et al., 2000), and there is an increasing understanding of secondary organic aerosol formation through reactions in clouds and fog (Lim et al., 2010; Shrivastava et al., 2017). In cloudy environments containing sufficient oxidants, sulfur dioxide can be rapidly oxidized by hydrogen peroxide and ozone (Kreidenweis et al., 2003), see Chapter 5. Parameterization of sulfate formation in clouds is usually considered in regional and global climate models because this process affects the aerosol budget significantly, but it is highly uncertain. For DCCs, sulfur dioxide can be transported to the upper troposphere and enhance aerosol nucleation there, so DCCs can serve as an aerosol source (Engström et al., 2008).

DCCs can produce a substantial number of new particles in the upper troposphere (a few orders of magnitude) by transporting inorganic and organic compounds that subsequently undergo

gas-to-particle conversion (e.g., Clarke, 1993; Clarke, et al. 1998; Twohy et al., 2002; Murphy et al., 2015; Kulmala et al., 2006; Andreae et al., 2018).

Hydrometeor evaporation and sublimation recycle aerosol particles that have been activated to become droplets and ice particles. The regeneration of aerosol by complete droplet evaporation is an important source of aerosol that can affect cloud properties in warm clouds (Wurzler et al., 2000; Xue et al., 2010) and DCCs (e.g., Yin et al., 2005; Engström et al., 2008; Ekman et al., 2011; Shpund et al., 2019). Measurements of INP regeneration in DCCs are lacking. However, INP regeneration has been considered in modeling studies (Engström et al., 2008), which show that it can increase aerosol particle concentrations by $600 \, \mathrm{cm}^{-3}$ at $6-10 \, \mathrm{km}$ altitude, explaining the observed high particle concentrations in the middle troposphere (De Reus et al., 2001; Fridlind et al., 2004).

14.5 Modeling

Modeling DCCs is challenging because the dynamical processes are multiscale, and small-scale dynamical processes are difficult to represent in a model. Cloud-resolving model (CRM) simulations with a grid scale of 0.25–3 km are often used to simulate DCCs, resolving most of the dynamical processes. These studies have greatly advanced our understanding of DCCs over a spectrum of scales (from individual thunderstorms to MCSs that can extend thousands of kilometers). These studies have also enhanced parameterization development for large-scale models. However, simulations of the same DCC case with different CRMs produce different results (Fridlind et al., 2012; Zhu et al., 2012; Varble et al., 2011, 2014; Fan et al., 2017b; Han et al., 2019). A recent model intercomparison study for aerosol effects on DCCs showed that models vary significantly in simulating aerosol effects on updraft speeds and mean updraft intensity above 8 km (Marinescu et al., 2021). CRM simulations also cannot fully resolve turbulence and microphysics, which could contribute to model uncertainties.

Large-eddy simulations (LES) with a grid scale of <250 m explicitly calculate turbulence and are often employed for process-level understanding studies of shallow cumulus or stratiform clouds. For DCCs, simulations at the LES scale are too expensive, so it has not been often used for 3-D simulation studies on aerosol interactions with DCCs. Global models use convective parameterizations, in which aerosol–DCC interactions are generally not considered or crudely considered, as detailed in Section 14.5.2.

14.5.1 Modeling aerosol interactions with deep convection at cloud-resolving scales

14.5.1.1 Features of models at cloud-resolving scales

Simulations at a cloud-resolving scale (0.25–3 km grid spacing) have been widely used in recent decades to simulate aerosol interactions with DCCs. Grid spacings of 2–4 km are also often called the convection-permitting model (CPM). CRMs/CPMs are designed to explicitly resolve deep-convective motion (Guichard and Couvreux, 2017), although they cannot fully resolve turbulent motions as LES models can. They are therefore in the "gray zone" regarding turbulence parameterizations (e.g., Wyngaard, 2004). Since deep-convective motion is assumed to be resolved, a convective parameterization for deep cumulus clouds (as used in large-scale models) is generally not employed. A nonhydrostatic dynamic core is employed (meaning that the vertical momentum equation is solved) and the

interactions of dynamics with physical processes (microphysics, radiation, and surface) are explicitly represented.

There are two common approaches in simulating clouds at a cloud-resolving scale: (1) idealized CRM simulations are initialized with sounding data (vertical profiles), forced by horizontally homogeneous large-scale and surface flux forcing, and often supplied with periodic lateral boundary conditions; and (2) real-case simulations in which the initial and lateral boundary conditions are provided by global/regional analysis/reanalysis or forecasts with open boundary conditions and realistic land surface conditions. It is often called a "limited area model (LAM)" simulation (Zhu et al., 2012). Idealized CRMs are simple, with some interactions often turned off, such as land–atmosphere interactions, which is beneficial for understanding cloud behavior under controlled environmental conditions. Real-case simulations likely provide a more realistic simulation of observed DCCs but interactions may complicate the role of a particular process.

14.5.1.2 Bulk and bin microphysics schemes

Cloud microphysics processes in atmospheric models are commonly represented using a bulk or bin approach (see Chapter 6).

In bulk schemes hydrometeor size distributions are defined based on the predicted (average) mass (one-moment schemes) or number and mass (two-moment schemes) of hydrometeors and an assumed size distribution function (spectral shape, e.g., an exponential or gamma function—see Chapter 6). Triple-moment schemes have been developed to predict shapes of hydrometeor size distributions to better represent important size-dependent processes such as sedimentation (Milbrandt and Yau, 2005; Naumann and Seifert, 2016; Shipway and Hill, 2012; Paukert et al., 2019). The bulk approach is computationally efficient. Kessler (1969) developed the first bulk parameterization scheme for warm-cloud simulations in numerical models. In recent decades, a great number of bulk parameterization schemes have been developed (currently over 20 in the WRF model), as summarized by Khain et al. (2015).

Bulk schemes are required to calculate size-dependent microphysical processes according to the defined size distribution function (which can still evolve during the simulation) rather than explicitly for each size bin of a bin scheme, which has implications for simulations of aerosol effects. In many models condensation and evaporation are parameterized with the saturation adjustment approach (e.g., Morrison et al., 2005), and rain formation is parameterized with an autoconversion formula (e.g., Khairoutdinov and Kogan, 2000). With saturation adjustment, water-saturated conditions are maintained by condensing the excess water vapor or evaporating the liquid condensate. This approximation leads to the adiabatic profile of liquid water content, which removes the effects of aerosol on condensation and supersaturation (Pinsky et al., 2013, 2014; Khain and Pinsky, 2018). Another approach is to employ the quasi-steady approximation, which uses the equilibrium supersaturation to represent the true supersaturation. This approximation makes condensation depend on vertical velocity but not droplet number and size, and thus aerosol (Khain et al., 2000; Pinsky et al., 2013; Fan and Khain, 2021). Thus, both the saturation adjustment and the equilibrium-steady assumption are not appropriate for aerosol-DCC interaction studies because the former mutes the water-phase invigoration mechanism (Zhang et al., 2021) and the latter weakens aerosol effects on droplet diffusional growth (Fan and Khain, 2021).

In bin schemes the size distributions of aerosol and cloud particles are discretized into a number of size bins and predicted. The size distributions are calculated by solving explicit microphysical

equations for each bin. For example, in the spectral-bin microphysics scheme, the particle size distribution is calculated on a finite-difference doubling mass grid containing several tens to several hundred mass bins. Therefore, size-dependent microphysical processes such as condensation/evaporation, deposition/sublimation, hydrometeor collision, and sedimentation are explicitly calculated rather than being based on an assumed size distribution function as in bulk schemes. The bin approach is very computationally expensive, generally requiring 10–100 times more computer time than bulk schemes, but the approach has been very useful in improving understanding of aerosol–DCC interactions (e.g., Khain et al., 2005, 2015; Fan et al., 2009, 2013, 2018).

Bin-emulating and the super-droplet methods are also used. In the bin-emulating approach the particle size distribution is still represented by a prescribed size distribution (i.e., the bulk approach), but the rates of microphysical processes are precalculated for a wide range of atmospheric conditions using a cloud parcel model with a bin scheme. The example model for this approach is the Regional Atmospheric Modeling System (e.g., Meyers et al., 1997; Cotton et al., 2003; Saleeby and Cotton, 2004; Van Den Heever and Cotton, 2004, 2007; Van Den Heever et al., 2006). The super-droplet method is a Lagrangian approach in which each super droplet represents a large number of real droplets of equal size (Morrison et al., 2018; Grabowski et al., 2019). It allows for detailed simulations of droplet size distributions in turbulent flows (Shima et al., 2009; Khain et al., 2015) and realistic tracking of soluble components (Grabowski et al., 2019). However, the accuracy of the approach depends on realistically simulating the turbulent structure of the atmosphere. This potentially powerful approach is computationally more expensive than bin schemes and is currently still at the development stage.

14.5.1.3 Significant findings

Significant progress has been made in the recent two decades or so in modeling DCCs and aerosol interactions with them at the cloud-resolving scale mainly because of the fast development of computer power, making the use of 3D models and bin schemes affordable.

Model developments include 3-moment bulk schemes and development in ice particle evolution (e.g., Milbrandt and Yau, 2005; Naumann and Seifert, 2016; Paukert et al., 2019; Morrison and Milbrandt, 2015). INP effects have been considered by linking aerosol particles with ice nucleation (e.g., Thompson and Eidhammer, 2014; Demott et al., 2015). Microphysical schemes have been improved to treat prognostic aerosol coupled with chemistry (Gao et al., 2016; Iguchi et al., 2008, 2020). In addition, CRM simulations of DCCs have been run over long periods to obtain statistical results (e.g., Van Den Heever et al., 2011).

Scientific results include the convective invigoration mechanisms discussed in Section 14.2.2. Another important understanding is that meteorological factors, such as wind shear, relative humidity, and DCC type, can affect the magnitude and sign of aerosol effects on convective intensity and precipitation. Aerosol effects can be significant for DCCs developed in warm, humid, and clean background environments. Consistent results are found concerning aerosol microphysical effects on stratiform/anvil areas of DCCs, namely, increasing cloud cover and thickness (Section 14.2.4). UAP can also play an important role in enhancing convection and precipitation. Finally, accurately modeling aerosol effects on DCCs requires microphysical schemes with the physical calculation of diffusional growth and prognostic size distributions of aerosol particles and hydrometeors. Bin schemes satisfy these requirements, but the majority of current bulk schemes do not.

The majority of modeling studies of aerosol effects on DCCs focus on aerosol concentration changes, but aerosol composition and size distribution effects have also been simulated. Aerosol

particle composition affects hygroscopicity, therefore droplet activation. Fan et al. (2007b) showed that slightly soluble organics decrease the droplet number, leading to weaker storms and less precipitation. Besides affecting hygroscopicity, with the addition of absorbing aerosol such as black carbon and brown carbon, aerosol radiative effects can dominate over cloud microphysical effects (Fan et al., 2008). Similarly, Wang et al. (2014c) showed that both radiative and microphysical effects of anthropogenic aerosol play an important role in the weakened minimal surface pressure and maximal wind speed near the eyewall of midlatitude cyclones and the enlarged rainbands and increased total precipitation. Aerosol radiative effects on regional and large-scale circulation and precipitation have been studied using relatively coarse-resolution models (Chapter 13), which usually cannot simulate aerosol effects on DCCs because of parameterized convection, as discussed in detail in the next section.

14.5.2 Modeling aerosol interactions with deep convection using coarse model grids

14.5.2.1 Features of coarse-resolution models

Coarse-resolution models (grid spacing larger than 10 km) generally assume that the atmosphere is in hydrostatic equilibrium. DCCs exist below the grid resolution of the model therefore must be parameterized (Chapter 6). How to accurately parameterize such subgrid clouds at various resolutions has been a major issue in regional and global models. Subgrid-cloud parameterizations do not explicitly calculate cloud-scale processes, and aerosol–cloud interactions are usually not considered except in a few studies as listed in the paragraph below. Current estimates of the effective radiative forcing due to aerosol–cloud interaction from global climate models are therefore limited to the effects on resolved large-scale stratiform clouds.

Convective parameterizations are generally based on the bulk mass flux approach, representing subgrid-scale convection by one "average" cloud with an estimation of the vertical updraft velocity (e.g., Tiedtke, 1989; Zhang and Mcfarlane, 1995; Bechtold et al., 2001; Kim and Kang, 2012). Such schemes have difficulties representing either the heterogeneity of convective clouds within a grid column or the detailed microphysics through which aerosol-cloud interaction effects operate. Despite these limitations, there have been several attempts to represent aerosol-cloud interaction in regional and global models by incorporating one-moment or two-moment cloud microphysics schemes into the convective parameterizations (Song and Zhang, 2011; Song et al., 2012; Grell and Freitas, 2014; Lim et al., 2014; Berg et al., 2015; Glotfelty et al., 2019; Kipling et al., 2020). The Subgrid Importance Latin Hypercube Sampler (SILHS) (Larson and Schanen, 2013) allows aerosol-cloud interactions to be treated at the subgrid scale, but it has not been often used yet.

Super-parameterization involves embedding a CRM (typically 2D) inside each column of the global model. Because this approach allows for the explicit calculation of convective clouds, it is well suited to representing aerosol—cloud interaction (e.g., Wang et al., 2011a,b). However, there are some associated problems, including the high computational cost, the lack of nonlocal interactions (i.e., no physics interactions between the CRMs in the columns), and the limited ability of a 2D CRM to capture the range of convective types.

Even for aerosol interactions with resolved clouds (i.e., large-scale stratiform clouds) in coarse-resolution models, large uncertainties can exist because of the following reasons: (1) aerosol activation is parameterized without explicit vertical velocity and supersaturation (e.g., Abdul-Razzak and Ghan, 2000; Gordon et al., 2020), and (2) major microphysical processes, such as condensation, evaporation,

and the WBF process, cannot be physically calculated because in-cloud updrafts and supersaturation are not resolved.

14.5.2.2 Significant findings

There have been studies incorporating full two-moment microphysics into the mass-flux convective parameterizations to consider aerosol interactions with cumulus clouds (e.g., Lohmann, 2008; Song and Zhang, 2011; Song et al., 2012), although in a bulk flux approach aerosol interactions with individual clouds can be not considered. Song et al. (2012) showed that aerosol—convection interaction improved simulations of droplets, ice, and precipitation over the western Pacific as well as midand low-level cloud fractions over the ITCZ—southern Pacific convergence zone and subtropical oceans. In a regional climate model, Lim et al. (2014) showed a markedly improved simulation of the summertime monsoon rainband in East Asia. Another regional climate model study that considered aerosol interactions with parameterized cumulus clouds showed increased cloud-top heights, reduced precipitation, and increased the shortwave cloud radiative effect by the increased aerosol (Glotfelty et al., 2019).

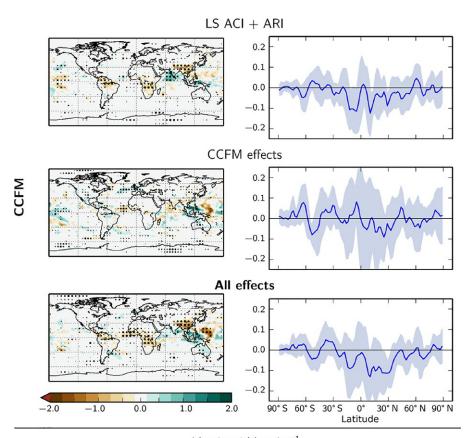
A recent global model study (Kipling et al., 2020) employed a convective scheme representing a spectrum of convective clouds within a grid cell, with the number of each cloud type dynamically determined by the environment. The scheme incorporated one-moment cloud microphysics but with a simple treatment of the evolution of cloud droplet concentration within the rising parcel, as in Labbouz et al. (2018). Their results show that aerosol effects on precipitation from parameterized deep clouds are comparable to the effects on large-scale clouds (Fig. 14.7). Trade cumulus over the Caribbean was found to be particularly sensitive to aerosol-induced invigoration, which is consistent with idealized CRM studies showing that warm and humid cloud regimes with weaker forcing may be more susceptible to aerosol-induced invigoration than strongly forced deep convection (e.g., Lee et al., 2008; Fan et al., 2009, 2013; Storer et al., 2010).

14.6 Knowledge gaps, future challenges, and research directions 14.6.1 Knowledge gaps and challenges

Much of our understanding is qualitative and large uncertainties arising from observational and model limitations hinder our quantitative understanding.

Observations. The importance of observational data in reducing DCC-aerosol uncertainties in weather and climate prediction lies in the following aspects: (1) they reveal statistical relationships that lead to hypotheses, (2) they improve understanding of fundamental physical processes to derive model parameterizations, (3) they provide data for evaluating and improving models, and (4) they provide observation-based estimates of the magnitudes of aerosol effects under real atmospheric conditions. Here, we summarize the challenges.

• It is difficult to single out aerosol effects from meteorological variability when covariability of aerosol properties with meteorological conditions occurs. Very long-term measurements of key variables, such as CCN below cloud base, updraft velocity, and droplet concentrations, are needed for sorting out a large variety of meteorological conditions including those that covary with aerosol such as those at the SGP (Li et al., 2011; Varble, 2018). To tackle covariability and its effect on



 $\Delta(\text{total precip})/\text{mm day}^{-1}$

FIG. 14.7

Annual mean precipitation response (present-day minus pre-industrial) in the ECHAM–HAM GCM with aerosol effects on both parameterized convective clouds and large-scale clouds considered (*bottom*). The *top* and *middle panels* show the decomposition into aerosol effects through large-scale and parameterized convective clouds, respectively. The plots on the *right-hand side* show the zonal mean of the maps on the *left*. (PD: present-day aerosol; PI: pre-industrial aerosol; stippling indicates areas where the PD–PI difference is statistically significant at the 95% level, while the corresponding confidence interval is *shaded* on the zonal mean plots).

Revised from Kipling et al. (2020).

aerosol-cloud interaction, we need to not only identify the covariability but also determine if it is a manifestation of, or affected to some extent by, the aerosol effects. To this end, field campaign measurements are needed for observing the entire process of cloud development and all pertinent meteorological variables (e.g., Andreae et al., 2004; Fan et al., 2018). Unfortunately, sampling periods in observation campaigns are generally too short for robust statistical analyses. Some fixed-site, single-point measurements are longer term, but they are generally lack the profile measurements of aerosol, CCN, and cloud microphysics, and thus it is difficult to establish any

- casual relationships between atmospheric stability and aerosol effects. In addition, such long-term measurement sites are too few globally.
- There is a lack of concurrent measurements of cloud dynamics, microphysics, and aerosol
 properties with sufficient temporal resolution in DCCs. In particular there is a lack of measurements
 of meteorological variables at the high temporal resolution at the altitudes where aerosol—cloud
 interactions take place This prevents us from gaining a good understanding of convective
 microphysics and how they are connected with changes in aerosol.
- There are challenges in measuring kinematics, thermodynamics, and microphysics in mixed-phase
 and ice-phase parts of DCCs, particularly in terms of separating liquid from ice particle properties.
 Ice microphysics has been a major issue leading to model differences in simulating DCCs at CRM
 scales (e.g., Fan et al., 2017b), particularly ice nucleation and ice particle growth and conversions.
 Our understanding of ice-related processes in the convective cores of DCCs is rudimentary.
- There are large data uncertainties in the aerosol, cloud, and precipitation measurements, particularly retrieved data from spaceborne and ground-based remoting sensing instruments that may lead to systematic biases in the aerosol-cloud interaction studies (e.g., Liu and Li, 2018). Observational data are often used as the "truth" for model evaluation and parameterizations. However, measurements suffer from all kinds of uncertainties. Therefore, reducing the measurement or retrieval uncertainty and providing consistent datasets across relevant variables is a major issue.

Modeling. There have been two significant modeling challenges concerning aerosol interactions with DCCs: model resolution and physics parameterization. The model resolution is limited by computing power. Global and regional climate models generally use coarse resolutions where DCCs are not resolved. As described in Section 14.5.2, convective parameterizations generally do not consider much cloud microphysics; therefore the climatic effects of aerosol interactions with DCCs are not taken into account in those models. Convective parameterizations also struggle to represent various convective types and associated heavy precipitation rates as well as the diurnal cycle of precipitation. These issues make studying the effects of changes in aerosol on convective clouds at the global scale challenging. Currently, there are some short-term global cloud-resolving simulations (e.g., Satoh et al., 2008; Miyakawa et al., 2014). However, these models are not yet coupled with aerosol and chemistry processes for aerosol—cloud interaction studies and the single-moment or two-moment schemes employed in these models would limit the realism of aerosol-DCC effects.

Simulations with 4-km grid spacing or smaller are often limited to regional domains or case studies. Problems associated with limited-domain simulations include biases in initial and boundary conditions and the difficulty in accounting for feedback between large-scale and local processes. The potential large-scale adjustment and buffering (Stevens and Feingold, 2009) cannot be studied well without a global model configuration. Case studies lead to difficulty in obtaining statistical results.

Another significant challenge concerns the accuracy of aerosol, turbulence, and microphysics parameterizations. Varying the cloud microphysics scheme only in modeling a DCC can lead to differences in cloud dynamical and microphysical properties (Fan et al., 2012a, 2017b) that exceed the magnitude of aerosol effects (White et al., 2017). It is known that commonly used two-moment bulk schemes have significant limitations in representing aerosol–DCC interaction processes (e.g., saturation adjustment for condensation/evaporation, excessive size sorting, etc.), as detailed by Khain et al. (2015). However, even among bin schemes (Xue et al., 2017), a wide spread of microphysical and

dynamic characteristics of a squall line was seen, due to different algorithms and numerical representations of microphysical processes, and assumptions made regarding hydrometeor processes and properties. Both Fan et al. (2017b) and Xue et al. (2017) pointed to ice microphysics parameterizations as the reason for the large model spread. The improvement in ice microphysics parameterizations is limited by our fundamental understanding. For turbulence parameterization, CRM scales are still in the "gray zone" (the so-called Terra Incognita) (e.g., Wyngaard, 2004). Appropriately accounting for turbulence is important for DCC development but very difficult at CRM scales because it is very challenging to separate the resolved and unresolved portions. To resolve turbulence and accurately simulate entrainments, both of which may be important aerosol—cloud interactions, the LES scale is needed, which is computationally prohibitable for global simulations.

14.6.2 Research directions

The remaining large uncertainties in aerosol–DCC interactions and effects on climate could be narrowed through developments in observations and models.

Observations. Long-term measurements of atmospheric states, aerosol properties, clouds, precipitation, and radiation at a few permanent sites supported by the ARM Program have provided useful data in detecting anthropogenic influences on radiation and precipitation (e.g., Yan et al., 2014; Feldman et al., 2015). Further progress could be made by expanding such observations to different climate regimes to robustly reveal the signals of aerosol effects on DCCs. For understanding convection and precipitation, it would be particularly helpful to obtain long-term observations under warm, humid locations where large aerosol variations occur but meteorological variations are small, such as in the Amazon or shipping lanes in the Indian Ocean. The Green Ocean Amazon is a good example of such a setting (Martin et al., 2017; Fan et al., 2018). Observations of cloud optical depth, hydrometeor size distributions, fall speeds, supersaturation, and vertical velocities over wide stratiform/anvil regions would help to improve our understanding of how changes in aerosol affect anvil clouds and radiation.

Another important observational advance would be the development or improvement of satellite retrievals and instrument technologies to reliably measure vertical velocity, supersaturation, and cloud microphysical properties in convective core areas. Without reliable vertical velocity information and concurrent aerosol/CCN measurements inside convective cores, the convective invigoration and significance are difficult to be directly evaluated with observations. Concerning cloud microphysical properties in mixed-phase clouds, developing more reliable methods to distinguish liquid and ice particle properties is important to improving our understanding. Concerning aerosol, reliable vertical profile measurements to determine the aerosol particles entering DCCs are key measurements, but it is difficult to attain reliable aerosol vertical profiles in the case of DCCs. CCN instruments also have a limitation for high supersaturation (generally not exceeding 1%), which is too low to understand the activation of ultrafine aerosol particles and their role in DCC invigoration (Fan et al., 2018).

Modeling. To improve the fidelity of aerosol–DCC simulations at CRM/CPM scales, model requirements include: (1) a prognostic supersaturation is needed for secondary aerosol activation, condensation, evaporation, deposition, and sublimation calculations; (2) hydrometeor size distributions need to be prognostic to physically simulate the responses of microphysical processes to CCN; and (3) aerosol particle size distributions need to be prognostic since fixed aerosol particle concentrations result in unrealistic effects on convective intensity (Fan et al., 2012a) and fixed size distribution limits the nucleation of small aerosol particles. Improving cloud microphysics parameterizations is certainly

important for aerosol–DCC simulations, but it is currently limited by our physical understanding in ice-related microphysics. Although bin schemes have many uncertainties and are computationally expensive (Section 1.5.1.2), particularly in representing ice-related processes, the response of microphysical processes to aerosol changes is more physically calculated compared with bulk schemes (e.g., the response of collision-related rain formation and accretion processes to CCN changes), so their wider use would help to advance our understanding.

In addition to exploiting the best-available models, progress will rely on (1) selecting well-observed cases and extensively evaluating the baseline simulation before conducting sensitivity tests to examine the aerosol effect; (2) coupling of DCC processes with chemistry and aerosol simulations over large regional domains to capture the important effects of spatial heterogeneity of aerosol (Lee et al., 2018); (3) performing ensemble simulations or long-time (monthly, seasonally, or even longer) simulations to help suppress the uncertainty caused by natural variability; (4) understanding causes of model uncertainty and spread, for example through perturbed parameter ensembles (Johnson et al., 2015; Wellmann et al., 2018, 2020); (5) incorporating the interactions of local processes with regional or global circulations, as well as studying aerosol effects on mesoscale convective systems owing to their importance in global precipitation and radiation budgets (Nesbitt et al., 2006).

For coarse-resolution modeling, with the current computing power, it is still not practical to run global simulations at convection-permitting scales with chemistry and aerosol components included for extensive research. Improving cumulus parameterizations to represent various types of DCCs and their interactions with aerosol, for example, scale-aware and aerosol-aware convective parameterizations, would help in this regard. Scale-aware parameterizations are particularly concerned with variable-resolution simulations. Until we have the computer power to run convection-permitting simulations over the entire globe for extensive research, variable resolutions with fine grids of 3 km or less or the multiscale modeling framework are intermediate approaches to better our understanding (Wang et al., 2011a,b; Schwartz, 2019). When computational advances permit us to run CRM simulations globally, reducing uncertainties in physical parameterizations of aerosol, turbulence, and cloud microphysics will be a primary focus.

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