Chapter 22

ARM’s Aerosol–Cloud–Precipitation Research (Aerosol Indirect Effects)

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1. Introduction/historical perspective

The conception of the Department of Energy’s (DOE’s) Atmospheric Radiation Measurement (ARM) Program over 20 years ago demonstrated prescience on the part of a number of astute scientists, many of whose words fill the pages of this monograph. The early years focused on a handful of cloud and radiation measurements and activities of relatively limited scope. The intervening decades have seen these efforts expanded to some of the finest instrumentation in the world to measure aerosol, clouds, radiation, and precipitation, accompanied by a substantial modeling effort. Together these have allowed the United States and international communities to tackle one of the thorniest problems associated with climate change, namely the influence of aerosol particles on cloud microphysics, precipitation, and radiation forcing, by definition, the indirect effect is the radiative forcing associated with these interactions. We will therefore introduce the term aerosol–cloud interactions (ACI) when we refer primarily to the microphysical/dynamical aspects of the problem and reserve “indirect effects” for the radiative forcing. ARM’s early focus on atmospheric radiation measurements, followed by years of refinement of microphysical retrievals, has placed it in an excellent position to address both ACI and the associated indirect effects.

ACI or aerosol indirect effects are often used to convey a few underlying microphysical processes. The first is the “albedo effect” (Twomey 1977), which states that an increase in the number of aerosol particles results in more cloud condensation nuclei (CCN), a higher droplet concentration, and, all else being equal (particularly liquid water content), smaller drops and a more reflective cloud.1 It is a fundamental expression of the ability of aerosol particles to generate a larger drop surface area to volume ratio. The second, the “lifetime effect” (Albrecht 1989), proposes that aerosol suppression

1 An exception to the rule is the addition of giant CCN, the presence of which might lead to more collision/coalescence, fewer larger drops, and a less reflective cloud.

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of collision-coalescence will suppress precipitation and allow clouds to sustain higher liquid water path (LWP) for a longer duration and therefore enhance cloud albedo. In mixed-phase clouds and ice clouds, the physics becomes more complex. For example, mixed-phase clouds could be expected to be more reflective because the reduction in droplet size resulting from an increase in aerosol reduces snow riming rates (Borys et al. 2003) and ice loss through precipitation (the riming effect). On the other hand, an increase in the aerosol might result in an increase in ice nuclei (IN), more efficient precipitation, lower cloud cover, and more solar absorption (the glaciation effect; Lohmann 2002). Finally, in deep convective clouds, there is evidence of an association between the aerosol and cloud-top height, updraft velocity, and lightning activity (the invigoration effect; Koren et al. 2005). Of all of these effects, the albedo effect has the strongest theoretical underpinnings; when the same amount of liquid water is divided among more drops, the cloud albedo must increase. The problem is that many other cloud processes act to obscure detection and quantification, rendering the magnitude uncertain. The others are less certain, mostly because they allow for realism in the form of the existence of multiple, simultaneous microphysical–dynamical interactions that occur while the system adjusts to the aerosol perturbation.

In addition to documenting ARM’s history of ACI and indirect effect studies, a secondary goal of this chapter is to shift the community’s thinking away from a linear superposition of these physical processes, and to encourage the reader to think more broadly about how all of these microphysical processes interact and adjust within the cloud system. This then places more emphasis on understanding how cloud systems work, and the role of meteorology in shaping cloud system evolution. It is in this spirit that the Intergovernmental Panel on Climate Change (IPCC 2013, p. 578) adopted the terminology ERFaci (effective radiative forcing due to aerosol–cloud interactions) to replace albedo and lifetime effects. This is a more inclusive term for the radiative forcing that occurs when the aerosol–cloud system is allowed to adjust to its environment, thereby allowing for changes in cloud fraction ($f_c$) and LWP or ice water path (IWP).

2. Development of crucial components that enabled ACI studies

a. Measurements

Shupe et al. (2016, chapter 19) and McComiskey and Ferrare (2016, chapter 21) describe more specifically the relevant measurement and retrieval capabilities; therefore we focus on the geophysical measurements themselves, with infrequent reference to instrument and measurement issues. This chapter is biased toward warm clouds (liquid water only), primarily because of the importance of shallow, warm convective clouds for the shortwave forcing of the climate system (Bony and Dufresne 2005), but also because of the inherent difficulties in quantifying cloud microphysical properties of the mixed phase (ice + liquid). It is also biased toward surface remote sensing of clouds, arguably one of the major strengths of ARM, whose early architects recognized the value of long-term datasets as an anchor to in situ aircraft sampling, and less flexible satellite remote sensing. While airborne instruments were developed earlier for direct measurement of microphysical properties, many surface remote sensing measurements still required refinement for quantitative ACI work. The emphasis on surface remote sensing does not detract from ARM’s significant collaborative efforts with other agencies in the field of satellite remote sensing (e.g., Minnis et al. 1992; Greenwald et al. 1999), and efforts in airborne campaigns in various parts of the world that have contributed greatly to the field. The 2009 RACORO$^1$ campaign (Vogelmann et al. 2012), conducted over the course of nearly six months at the Southern Great Plains (SGP) site, deserves special mention as an example of long-term in situ measurements addressing, among other topics, ACI and indirect effects.

Central to the study of ACI was capacity building of three primary measurement components:

1) Cloud microphysical properties such as drop effective radius $r_e$, drop concentration $N_d$, and cloud optical depth $\tau_c$ (Frisch et al. 1995; Dong et al. 1997; Chiu et al. 2007; 2010). Of these, $\tau_c$ is related closely to cloud reflectance, or cloud albedo, and is thus particularly relevant for ACI studies. Drop size retrievals typically separate into those that are averaged over height (e.g., derived from optical depth and liquid water path; Min and Harrison 1996; Kim et al. 2003; Chiu et al. 2012) or those that provide $r_e$ profiles (e.g., from radar reflectivity and liquid water path; Frisch et al. 1995). Tradeoffs include the fact that optical measurements are more relevant to radiation than radar reflectivity (sixth moment of the drop size distribution) but can only be measured during daylight, whereas radar reflectivity is measurable at any time as the surface so that there is little longwave (LW) compensation for shortwave (SW) forcing. An exception is high-latitude wintertime clouds, where LW effects can be important, as discussed later.

2 Shallow clouds radiate at approximately the same temperature as the surface so that there is little longwave (LW) compensation for shortwave (SW) forcing. An exception is high-latitude wintertime clouds, where LW effects can be important, as discussed later.

3 RACORO: Routine AAF CLOWD Optical Radiative Observations; CLOWD: Clouds with Low Optical Water Depths.
Aerosol microphysical properties such as aerosol number concentration (typically at diameters \(>60\) nm), CCN concentrations at specific supersaturation set points, aerosol extinction, total scatter, aerosol optical depth \(\tau_a\), backscatter, and aerosol index (Ångström exponent \(\alpha\) and \(\beta\)) have all been used as measures of aerosol influence on clouds. Surface aerosol measurements may not always be representative of the aerosol entering the cloud; corrections can be made (e.g., Ghan and Collins 2004) or lidar measurements can be used to retrieve the aerosol properties below cloud base at a level where it is more likely that the aerosol is entering the cloud (Feingold et al. 2003). Aerosol chemical composition is being measured increasingly at ARM sites (McComiskey and Ferrare 2016, chapter 21). Although less important than aerosol number and/or size vis-à-vis influence on cloud microphysics (e.g., Feingold 2003; McFiggans et al. 2006), composition provides important insight into aerosol formation, growth, and removal processes, as well as the total aerosol budget. Aerosol composition is related closely to hygroscopicity, which is particularly relevant to the ability of an aerosol particle to act as a CCN, and to scatter light. The broader perspective of aerosol radiative forcing therefore requires consideration of aerosol number concentration, size, and composition. Moreover, aerosol indirect effects can be hard to separate from direct effects (McComiskey and Ferrare 2016, chapter 21) in the vicinity of clouds because the delineation between cloudy air and cloud-free air is often difficult to draw, and because the aerosol–cloud mix presents a different radiative forcing from the sum of the independent aerosol and cloud components (e.g., Schmidt et al. 2009). This means that key aerosol optical properties such as extinction (\(\alpha\)), single scattering albedo, and asymmetry parameter are also important to the total radiative forcing (e.g., McComiskey et al. 2008).

**LWP measurements** are derived from microwave radiometers (Liljegren et al. 2001; Turner et al. 2007; Cadeddu et al. 2009) and for low LWP conditions, the Atmospheric Emitted Radiance Interferometer (AERI; Turner et al. 2007; 2016, chapter 13). Satellite sensors such as the Moderate Resolution Imaging Spectroradiometer (MODIS) also provide LWP measurements through combination of visible and near-IR measurements. Essential to all ACI and indirect effect evaluations, LWP is a bulk property that provides some reference against which to evaluate the influence of the aerosol on the cloud microphysical and/or radiative properties. Twomey (1977) likely had this in mind when formulating the principle of the albedo effect, which postulates that an increase in the number concentration of aerosol particles results in higher \(N_a\), and all else being equal (i.e., LWP), smaller \(\tau_a\). It is worth noting that this is not an assumption of constant LWP but rather a logical and necessary way to stratify the data within the context of Twomey’s albedo effect. Also central to this discussion is the concept of cloud albedo susceptibility \(S_o = \partial A/\partial N_a|_{\text{LWP}}\) where \(A\) = cloud albedo (Platnick and Twomey 1994; Platnick and Oreopoulos 2008; Oreopoulos and Platnick, 2008), which assesses the magnitude of a cloud albedo response to an increase in \(N_a\) and, by inference, aerosol concentration (\(N_a\)) at constant LWP. Below we will also discuss the idea of precipitation susceptibility \(S_{o,r} = -\partial \ln R/\partial \ln N_a|_{\text{LWP}}\) (Feingold and Siebert 2009; Sorooshian et al. 2009), which attempts to quantify the extent to which aerosol, via its influence on \(N_a\), can modify rain rate \(R\). Again, LWP is used as a reference. Thus both \(S_o\), and \(S_{o,r}\) represent the potential rather than the actual influence of the aerosol, and in both cases LWP is an essential determinant of the possible effect.

While these three components form the backbone of ACI efforts, cloud-base vertical velocity is another key measurement relevant to ACI. Drop concentration is a function not only of aerosol properties, but also of the ability of a cloud to generate supersaturation, which is in turn a function of updraft velocity \(w\). Doppler radars (in cloud) and lidars (below cloud) have made great strides in providing these measurements (Miller and Albrecht 1995; Clothiaux et al. 2000; Kollias and Albrecht 2000; Ghate et al. 2010).

**b. Modeling of ACI**

In parallel with deployment, testing, and refinement of aerosol and cloud measurement capabilities, development of cloud models appropriate to the study of ACI have been a significant ARM/ASR effort. Large-eddy simulation (LES) as a tool for studying ACI experienced significant development in the early stages of the ARM Program. LES is attractive in that it resolves the spatiotemporal scales relevant to aerosol activation and cloud turbulence. By coupling LES to detailed cloud microphysical models, several groups began to assess the ability of their models to simulate observed cloud structure and to provide a framework in which to test hypotheses, as well as being a simulation world to test retrieval methods (Kogan et al. 1994; Feingold et al. 1994; Ackerman et al. 2004; Stevens et al. 1996; Duda et al. 1996). The number of groups performing LES with
microphysical models of varying complexity has grown significantly over the ARM/ASR lifetime and has facilitated the rigorous intercomparison of models to assess robustness of responses (e.g., Ackerman et al. 2009). Significant efforts also have been expended on developing cloud-resolving models (CRMs; Krueger et al. 2016, chapter 25) that simulate aerosol–precipitation interaction in deep clouds (Morrison and Grabowski 2011; van den Heever et al. 2011; Grabowski 2006; Fan et al. 2009).

3. Warm clouds

a. Demonstrations of surface remote sensing of aerosol–cloud interactions

Early demonstrations of ACI were performed at SGP using two different approaches, each with its own merits and drawbacks. The first approach (Feingold et al. 2003) considered a cloudy column of air and combined subcloud aerosol extinction (Raman lidar), a profile of $r_c$ (cloud radar and microwave radiometer), $w$ (Doppler cloud radar), and LWP (microwave radiometer) to produce plots of a cloud average $r_c$ versus subcloud aerosol extinction $\alpha$. A small sample of seven cases was analyzed as a proof of concept. Theoretical considerations indicate that if, as is observed, $N_d \propto N^b_a$ ($0 < b < 1$), then, for constant LWP and drop-size dispersion, $r_c \propto N^{b/3}$. It then follows, assuming that $\alpha$ is a good proxy for the aerosol influencing the cloud, that $r_c \propto \alpha^b$. This power-law dependence was confirmed by the linear dependence of $\log r_c$ on $\log \alpha$. The data were sorted into different LWP bins in accord with the precept of the albedo effect. Values of $b$ were calculated at different $w$ and shown to be dependent on $w$, as expected from theory.

Simultaneously a second group (Kim et al. 2003) took a different approach using observations from SGP and derived a cloud-mean $r_c$ from cloud optical depth [multifilter rotating shadowband radiometer (MFRSR)] and LWP (microwave radiometer) and used surface aerosol measurements of light scattering as a proxy for the aerosol concentration affecting the cloud. They too quantified $b$ and showed the strong correlation between LWP and $r_c$ for a much larger sample size; on average more than 60% of the variance in $r_c$ was due to variance in LWP. Garrett et al. (2004) used similar instrumentation to quantify $b$ at the North Slope of Alaska (NSA), arguing that the aged aerosol particles reaching the Arctic are likely to be better CCN and therefore have a stronger ACI than at midlatitude locations such as SGP. Figure 22-1 shows some early examples of ACI measurements at SGP and NSA from the aforementioned studies.

The parameter $b$ represents the magnitude of the microphysical response to an aerosol perturbation that is the basis of the albedo effect. Values tend to be $\sim 0.2$ to 0.6 (i.e., somewhat lower than one might expect due to aerosol activation alone). Both the magnitude and the variability suggest the presence of multiple cloud processes including collision/coalescence, entrainment, and wet scavenging (McComiskey et al. 2009). These values may be biased low when surface aerosol measurements are not representative of the aerosol entering cloud base.

b. Surface, satellite, and aircraft in situ approaches

Both approaches described above are attempts to achieve, from a single fixed location on the surface, what stacked aircraft can achieve by flying below, within, and above cloud (e.g., Brenguier et al. 2000; Wilcox et al. 2006; Roberts et al. 2008) or what is routinely attempted with space-based remote sensing. Logistically, surface remote sensing is a much simpler approach than stacked aircraft; however, it cannot measure the cloud radiance from above. Surface-based remote sensing provides a more controlled environment for measurement of ACI than does space-based remote sensing; sample volumes are much smaller, the temporal resolution is much higher, and meteorology can be better characterized. It is also amenable to collocated aerosol and cloud measurements; however, it lacks the huge global sampling advantage of satellite remote sensing. When using measurements from space, the aerosol in cloud-free pixels adjacent to cloudy pixels is assumed to be representative of the aerosol feeding into the base of the target clouds. There are several concerns with this approach, particularly because it is not trivial to identify cloud-free air (e.g., Koren et al. 2007; Charlson et al. 2007) and because humidification near clouds enhances $\tau_n$, the typical proxy for subcloud CCN, in “cloud-free” pixels. The problem is exacerbated when addressing layer clouds of large spatial extent, since the cloud-free pixel may be a significant distance from the target clouds and at some point not particularly relevant to ACI. Even if one could identify purely cloud-free pixels, horizontal
light scattering from adjacent clouds has also been shown to enhance $\tau_a$ (Wen et al. 2007; Kassianov et al. 2009), which would exacerbate quantification of ACI.

Figure 22-2 provides a schematic showing some of the key differences in the approaches of satellite versus surface remote sensing to measurement of ACI.

c. Quantification of ACI in warm clouds

The various approaches to assessing ACI have differing strengths and weaknesses that have been explored in follow-up studies (e.g., McComiskey et al. 2009; Kim et al. 2008; Berg et al. 2011). Primary lessons are the sensitivity of $b$ to binning data by LWP; the positive correlation between $b$ and adiabaticity (i.e., how close the LWP is to the adiabatic value; Kim et al. 2008); use of subcloud aerosol as a more relevant CCN proxy to surface aerosol in decoupled boundary layers; the sensitivity of $b$ to updraft $w$ (Feingold et al. 2003); and the large influence of spatiotemporal scale on quantification of $b$ (McComiskey and Feingold 2012). In many studies, $b$ represents the full diversity of cloud microphysical processes—activation, collision/coalescence, entrainment/mixing, scavenging, and sedimentation—rather than simply the activation. This is particularly relevant in the context of climate models, which sometimes use $b$ (or equivalent) as a representation of drop activation alone, thus biasing radiative forcing estimates. For example, $b$ values are sometimes derived from remote sensing without sorting the data by LWP and used to represent the albedo effect in climate models. While this is clearly not a measure of the ACI associated with the albedo effect, it is more representative of effective ACI and associated forcing (ERFaci).

d. ARM aircraft campaigns

While various early field campaigns to some extent addressed ACI, we discuss several field campaigns that had a particularly strong focus on ACI. In chronological order these are MASE (2005), CLASIC/CHAPS (2007), VOCALS (2008), and RACORO (2009).6

MASE took place in July (2005) to overlap with the first ARM Mobile Facility (AMF) deployment at Point Reyes, California. The DOE G-1 flew aerosol and cloud microphysical payloads for addressing ACI in stratocumulus. DOE also partnered with the California Institute of Technology (CalTech), who operated the Center for...
Interdisciplinary Remotely-Piloted Aircraft Studies (CIRPAS) Twin Otter with a similar aerosol/cloud payload. The G-1 data acquired proved useful for testing theoretical formulations of the self-collection of cloud droplets to drizzle (autoconversion; Liu et al. 2005) and for identification of drizzle initiation at cloud top where LWC is highest. A notable aspect was the analysis of the droplet dispersion and its dependence on aerosol and drop concentrations (Lu et al. 2007), which influences cloud albedo susceptibility $S_o$. Some earlier studies showed dispersion decreasing with increasing drop concentration (Miles et al. 2000), which translates to an increase in $S_o$ (Feingold et al. 1997). Others showed dispersion increasing with increasing aerosol concentration (Liu and Daum 2002) and therefore a decrease in $S_o$. The enhancement in $S_o$ is expected in the coalescence-dominated growth regime, whereas the reduction should exist for condensation-dominated growth. Using Twin Otter MASE observations, Lu et al. (2007) showed decreasing dispersion with increasing...
but increasing dispersion with increasing $N_d$ and therefore did not completely resolve this issue. Both coalescence- and condensation-dominated regimes exist, so conclusions are hard to draw about the influence of aerosol concentration on $S_a$.

CHAPS (Berg et al. 2009) is interesting because it used the DOE G-1 to address cloud processing of aerosol and gases, a closely related topic to ACI since the mutual effects of these two elements cannot be separated from one another. It was also one of several new studies to adopt use of (insoluble) CO as a proxy for CCN (Berg et al. 2011) and thus avoid the difficulty of characterizing the aerosol close to cloud, or in situations where wet removal may influence aerosol measurements. By performing flight legs downwind of source regions, CHAPS verified that even relatively small cities like Oklahoma City have a detectable ACI signal.

The DOE G-1 deployed to the VOCALS Regional Experiment (VOCALS-Rex) field campaign in Chile in the austral spring of 2008 for a large international deployment that included five aircraft, ground stations, and a number of ships targeting the stratocumulus regime in the southeast Pacific. ACI was an important goal of VOCALS-Rex; however, a large component of the campaign was to understand the meteorological environment of this stratocumulus regime. Because of the enormous scope of VOCALS, we touch briefly on the DOE G-1 activities and refer the reader to Wood et al. (2011) for an overview of the experiment. The G-1 flew repeated legs along 20°S to collect statistics of aerosol and cloud properties over the course of a month; these will undoubtedly contribute to future documentation of the cloud field properties and boundary layer structure (Kleinman et al. 2012). G-1 data also have been used to evaluate satellite remote sensing (Min et al. 2012).

ARM undertook an ambitious task through deployment of the CIRPAS Twin Otter for RACORO—a long-duration (>5 month) field program in 2009 addressing shallow convective clouds (Vogelmann et al. 2012) and their radiative influences. The long duration and range of conditions encountered produced a dataset that is more statistically representative than the typical one-month intensive deployment. The deployment also provided a valuable test of surface remote sensing retrievals of aerosol and cloud retrievals, and important information on the relevance of surface CCN measurements to the clouds sampled. Lu et al. (2012) studied a large sample (568) of shallow cumulus and found a positive relationship between updraft velocity and $N_d$. They also found a negative correlation between $N_d$ and relative droplet dispersion, supporting similar results from stratocumulus (Miles et al. 2000; Lu et al. 2007). Continuing analyses of ACI and indirect effects are currently in progress through a combination of LES and measurements of RACORO cases, as part of the Fast-physics System Testbed and Research (FASTER) project.

e. Modeling of ACI in warm clouds

Modeling of ACI in warm clouds has seen significant effort, and at a range of scales. Given our interest in microphysical processes and the capability of ARM to represent these properties at these scales, we focus on results from LES and CRMs coupled to microphysical models with either two-moment or bin microphysics schemes (i.e., schemes that carry information on drop size and therefore the potential for the aerosol to influence cloud system evolution). Early modeling efforts (Feingold et al. 1994; Kogan et al. 1994; Stevens et al. 1996) attempted to demonstrate the original constructs of “more aerosol particles lead to more/smaller droplets (all else equal)” (Twomey 1977) and “more aerosol particles lead to suppression of collision/coalescence and reduced rainfall” (Albrecht 1989). Perhaps the most important lesson learned from these studies is that, although the underlying “Twomey” and “Albrecht” constructs are physically sound, the cloud system often behaves in a much more nuanced manner because these aerosol perturbations occur within a dynamical framework that responds to, and sometimes buffers, the aerosol perturbation (e.g., Wang et al. 2003; Jiang et al. 2002; Ackerman et al. 2004; Xue et al. 2008; Stevens and Feingold 2009). Cancellation of aerosol effects has also been demonstrated in mixed-layer models (Wood 2007) lending more confidence to these ideas. Nevertheless, the cloudy boundary layer also may exhibit strong sensitivity to the aerosol (e.g., Ackerman et al. 2003; Christensen and Stephens 2011); in severely CCN-limited situations, boundary layers can even collapse because cloud dissipates as fast as it is generated. It therefore becomes important to identify conditions in which the cloudy boundary layer is either sensitive or insensitive to the aerosol. The general picture that has emerged is that, under clean conditions that are apt to generate precipitation, increases in the aerosol do indeed increase cloud fraction and LWP, but in nonprecipitating conditions clouds tend to thin in response to increasing aerosol through a combination of droplet sedimentation (Bretherton et al. 2007) and evaporation/entrainment adjustments (e.g., Ackerman et al. 2004; Hill et al. 2009). These results are supported by satellite studies of ship tracks (Christensen and Stephens 2011).

f. Organization in shallow convection

Since the dawn of the satellite era in the 1960s, images of cloud field organization have become increasingly
accessible. In addition to organizational issues with precipitating deep convective cloud systems, shallow cloud fields also have been shown to organize into characteristic patterns (e.g., Agee 1984). Interagency field experiments such as the Eastern Pacific Investigation of Climate (EPIC; Bretherton et al. 2004), the Dynamics and Chemistry of Marine Stratocumulus Phase II (DYCOMS-II; Stevens et al. 2005), and VOCALS (Wood et al. 2011) have shown that marine stratocumulus cloud fields tend to prefer one of two states: the closed cellular state and the open cellular state. Closed cells have high cloud fraction and high albedo and tend to be nonprecipitating. Open cells are characterized by low cloud fraction, low albedo, and precipitation. Given the role of the aerosol in regulating precipitation, there is a clear pathway for the aerosol to select which of these is the preferred state, and therefore the albedo of the system. ARM has been a strong supporter of both observational (Sharon et al. 2006) and modeling studies (Xue et al. 2008; Wang and Feingold 2009a,b; Wang et al. 2010) that have clearly identified drizzle, whether a result of low CCN concentrations or of thick clouds, as playing a key role in the formation of open cells. Kazil et al. (2011) also identified aerosol replenishment mechanisms (nucleation of new particles, surface production of aerosol in cold pools, and entrainment of free tropospheric aerosol) that help make the open-cell system resilient. Transitions between closed- and open-cell states continue to be a topic of great interest because of the strong influence on cloud and planetary albedo. The importance of marine boundary layer clouds led ARM to support the first marine-based deployment of the AMF to the Pacific Ocean in 2012 [Marine ARM GCSS Pacific Cross-Section Intercomparison (GPCI) Investigation of Clouds (MAGIC)] to make regular observations of the stratocumulus to trade cumulus transition.

g. Precipitation susceptibility ($S_{o,R}$)

The precipitation susceptibility (Feingold and Siebert 2009) was proposed as a means to quantify the sensitivity of rain rate to aerosol perturbations. It has primarily been applied to warm clouds, but also to mixed-phase convection (Seifert et al. 2012). In warm clouds, raindrops are generated by two processes: 1) the self-collection of cloud droplets (autoconversion), which generates the first raindrop embryos, and 2) the collection of cloud droplets by rain drops (accretion). Autoconversion is a function of droplet (and cloud liquid water) concentration, whereas accretion is only a function of cloud and rain liquid water concentration. Therefore the potential for the aerosol to influence rain formation is via autoconversion, and the magnitude of $S_{o,R}$ is an indication of the relative balance of autoconversion and accretion.

Several ARM scientists have analyzed $S_{o,R}$ with parcel models (Feingold and Siebert 2009), heuristic one-dimensional models (Wood et al. 2009), LES (Jiang et al. 2010), and climate models (Wang et al. 2012). Observations have targeted warm clouds using satellite remote sensing (Sorooshian et al. 2009) and in situ aircraft data from VOCALS (Terai et al. 2012). LWP has been identified as having a strong control over $S_{o,R}$.

Two primary responses have been identified:

1) Parcel models, LES, and satellite remote sensing suggest that at low LWP $S_{o,R}$ increases with increasing LWP, reaches a maximum, and then decreases with further increases in LWP. The location and magnitude of the maximum depend on spatiotemporal averaging but also appears to depend on the background aerosol conditions, among other factors.

2) Heuristic models and in situ aircraft observations suggest a monotonic decrease in $S_{o,R}$ with increasing LWP. Ongoing efforts are attempting to explain these differences.

In addition to $S_{o,R}$ being useful for understanding rainforming processes, it also provides an important way to evaluate the balance of these processes in global climate models (GCMs); the larger the value of $S_{o,R}$, the longer the cloud lifetime and the stronger the effective radiative forcing ERFaci. Quaas et al. (2009) evaluated a large number of GCMs and highlighted the sensitivity of indirect effects to parameters such as $b$ and an equivalent parameter to $S_{o,R}$. As expected, models with larger $b$ or $S_{o,R}$ have stronger indirect forcing. It is noteworthy that AMF Point Reyes data played an important role in this GCM evaluation study.

4. Arctic mixed-phase clouds

Arctic mixed-phase clouds are another particularly important component of the climate system. They also have been the focal point of several large ARM field campaigns such as M-PACE (2004) and ISDAC (2008). They will be discussed relatively briefly, in part, because aerosol influences on these clouds are relatively poorly known.

The nucleation of ice is a key unknown in the maintenance of mixed-phase Arctic stratus. Numerical models are being confronted increasingly by aircraft measurements such as those from M-PACE and ISDAC.

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7 M-PACE: Mixed-Phase Arctic Clouds Experiment; ISDAC: Indirect and Semi-Direct Aerosol Campaign.
(McFarquhar et al. 2011) and ground-based observations such as those at NSA (van Diedenhoven et al. 2009) or Eureka (de Boer et al. 2009). Key issues with simulating mixed-phase clouds are 1) understanding ice-nucleating mechanisms (e.g., Fridlind et al. 2007); 2) resolving disparities between observed (low) ice nuclei and (higher) ice crystal concentrations (Fridlind et al. 2007); and 3) simulating the observed relative amounts of liquid and ice in clouds of different thickness (Klein et al. 2009; Morrison et al. 2009).

To this point, the discussion has centered on microphysical processes that are likely to change the shortwave forcing of the planet. However, in the wintertime, investigations at NSA show that the aerosol has an influence on the longwave (LW) emissivity of Arctic clouds. In thin clouds with low LWP, an increase in the aerosol results in an increase in LW emissivity and a warming of the surface (Lubin and Vogelmann 2006; Garrett and Zhao 2006). An analog to the shortwave albedo susceptibility developed by Garrett and Zhao (2006) \[ S_{LW} = -(1 - \varepsilon) \ln(1 - \varepsilon)/3N_d \] shows that, in the longwave, the maximum susceptibility is at \( \varepsilon = 0.6 \) and low \( N_d \). Thicker clouds act as black bodies (\( \varepsilon = 1 \)), whereas thinner clouds are simply too thin to have much of an effect.

The Arctic boundary layer often presents one of two states: an opaque cloudy state and a cloud-free state. The mixed-phase Arctic stratus cloud appears to be a particularly robust state, often persisting for many days at a time, in spite of the inherently unstable mixture of ice and water. Lidar and radar imagery at NSA show ice precipitating from the base of these clouds and yet the clouds are not consumed. Why is this so?

Work based on many years of observations and modeling studies (Morrison et al. 2012; Fridlind et al. 2012a) suggests that the following factors, singly or in combination, might all play a role in maintaining the cloudy state:

- relatively low ambient IN concentrations (on the order of 1 per liter),
- a self-regulating ice crystal concentration,
- longwave cooling of the water layer driving sufficiently large turbulence to sustain the liquid cloud layer (but not strong enough to remix ice and water),
- moist inversions above cloud top, and
- long-range transport of water vapor.

The disappearance of the cloudy state seems to be caused by changes in the large-scale meteorology (Stramler et al. 2011)—particularly when accompanied by higher surface pressure. The Arctic boundary layer therefore appears to follow a slow manifold (a slowly evolving surface in phase space) with faster microphysical processes slaving the system to the slow manifold. Just how much microphysical versus dynamical processes contribute to the switch between these slow manifolds is unclear. Regardless, the Arctic cloudy boundary layer tends to be resilient, and when transitions between states do occur they seem to be controlled to some extent by changes in meteorology. Addressing questions like these requires significant effort. Continuous observations in the Arctic are sparse, and ARM continues to commit resources to extensive, continuous monitoring in this region from the ground and the air.

5. Deep convective clouds

a. Background

As with Arctic mixed-phase clouds, the influence of aerosol on deep convective clouds is a far more complex topic than it is for warm clouds, and although much work has been done over the past two decades, there is still a significant amount of disagreement on the extent to which the aerosol influences various cloud field properties. The complexity emanates from the multitude of liquid and ice microphysical pathways that become possible in the presence of ice. Indeed, a variety of possible responses probably exist depending, among other things, on environmental humidity (Khain et al. 2008) and shear (Fan et al. 2009). It has been hypothesized that, by delaying the onset of freezing to colder temperatures, the latent heat of freezing will be released at higher altitudes, thereby generating clouds with higher vertical velocities and higher cloud tops. This chain of events is often termed “aerosol invigoration of convection” (Koren et al. 2005). Invigoration tends to be used as a “catch-all” to describe various aspects of the aerosol influence, such as stronger convection, more rain, heavier rain, stronger cold pool outflow (Khain et al. 2005; Lee et al. 2008), and higher cloud tops. Given the complexity of the system it is possible that some of these responses occur in different cloud systems, and at different stages of cloud system evolution. Satellite studies seem to confirm this picture by showing a correlation between cloud-top height and aerosol loading (e.g., Koren et al. 2005). Concerns about potential “false correlation” have been raised because higher cloud tops often occur in the presence of higher instability and higher moisture, and therefore higher \( \tau_d \) (e.g., Quaas et al. 2010). Efforts to address this issue (e.g., Koren et al. 2010) have used both chemical transport models and reanalysis data to place the observations in the context of meteorology. The reader is referred to Tao et al. (2012) for a broad review of this topic, which also
covers work done at ARM sites (e.g., Li et al. 2011; see below).

b. Process studies

The physical processes behind “invigoration” describe a plausible response of an individual convective entity to an aerosol perturbation. However, given the myriad microphysical and dynamical internal feedbacks, what is more important is how a larger cloud system will respond to the aerosol perturbation and at time scales much longer than the lifetime of individual convective cells. For example, modeling by van den Heever et al. (2006) illustrated how the response of surface precipitation to aerosol perturbations may change during the course of a 12-h simulation. More recent work extending simulations to two weeks (Morrison and Grabowski 2011) and to multimonth radiative convective equilibrium (Grabowski 2006; van den Heever et al. 2011) underscores the fact that conclusions regarding aerosol influences on radiation at the top of the atmosphere cannot be drawn from short simulations. Whether the interest is climate-related or associated with an individual high aerosol event will dictate the required length of the simulation. This requires consideration of the duration and magnitude of the aerosol perturbation. For example, radiative convective equilibrium simulations with exceptionally high aerosol loadings may not be relevant for climate studies since aerosol perturbations typically subside as aerosol is removed by wet and dry deposition (e.g., Lee and Feingold 2010) or simply replaced by cleaner air through advection. Sometimes responses ascribed to invigoration may not necessarily be part of the originally hypothesized chain of events. For example, Morrison and Grabowski (2011) showed that, in spite of a small aerosol-induced weakening in convection, cloud tops are higher because the resulting smaller ice particles have smaller fall velocities and can be lofted higher. Parsing out the various potential responses attributed to the aerosol, and their etiologies, would appear to be an important direction for research. The GCSS Tropical Warm Pool International Cloud Experiment (TWP-ICE) modeling study (Fridlind et al. 2012b), based on ARM measurements collected at the tropical western Pacific Darwin site in early 2006, has taken this essential step toward understanding sensitivity of results to microphysical complexity. Extensive environmental, cloud, and precipitation measurements to address convective processes, along with aerosol measurements, provide the necessary linkages for aerosol–deep convective cloud studies.

c. Surface-based observations

In addition to studies mentioned briefly above, surface-based measurements over a 10-yr period at SGP have also suggested elements of invigoration (Li et al. 2011). Using a combination of surface aerosol measurements (condensation nuclei, which include the very numerous nanometer sized particles) and ground-based remote sensors the authors concluded that, for mixed-phase clouds, aerosol perturbations cause increases in cloud depth and cloud-top height if the cloud base is warm. In contrast they found no such effect in clouds with cold bases and no ice. The authors also found evidence of aerosol influences on rain frequency and amount. To bolster conclusions, future studies will benefit from a more extensive set of precipitation measurements from radars at SGP, a closer focus on meteorological controls on convection, and CCN as opposed to condensation nuclei (which often do not participate as CCN) to represent the aerosol influencing the cloud.

6. Summary

As ARM moves toward the next 20 years, it is worth taking stock of some key successes and challenges that have arisen over the past 20 years. First, by developing and deploying a first-class array of aerosol and cloud instrumentation, the program has been able to demonstrate the ability of surface-based remote sensing to detect some expected correlations between aerosol and cloud microphysical properties. It has, together with other agencies, supported model development that has been crucial for understanding how clouds of various types respond to aerosol perturbations. Along with surface-based remote sensing data, ARM’s airborne experiments have produced datasets that will for years to come provide data against which to evaluate models, at a range of scales. One of the most difficult aspects of ACI studies is separating an observed correlation between aerosol and cloud parameters from a causal relationship between the two. That is, has the aerosol caused a change in the cloud properties, or are we merely observing a correlation between the two that might be related to each being correlated with the meteorological conditions? While models have played an important role in helping resolve this ambiguity, it is difficult to contend with the complexity of the models and the large number of internal feedbacks. As noted at the outset, the emphasis has continued and should continue to shift from identifying correlations to understanding the meteorological controls on cloud systems and the ways that the aerosol might perturb the system. From an observational perspective, characterizing the meteorological context of ACI is essential and will help determine whether the cloud system is resilient to aerosol perturbation or not. Refinement in variational analysis methodologies for
model forcing and development of best estimate products for various ARM sites (e.g., Xie et al. 2010) are proving particularly useful. Methodologies for identifying drivers of ACI also are increasingly being applied.

In assessing the achievements, one does notice that more effort has been placed on detecting ACI compared to the radiative manifestation of ACI (i.e., indirect effects). Some important progress has been made. An understanding of the relationship between the parameter $b$ ($N_d \approx N_v^0$) and radiative forcing by a cloud of known LWP has been established for plane-parallel clouds (McComiskey and Feingold 2008). ARM/ASR has advanced the study of 3D radiative transfer in a field of clouds (e.g., Barker and Marshak 2001); however, the forcing of the “aerosol-cloud soup” has received less attention. One approach is that taken by Schmidt et al. (2009), who compared spectral measurements of downwelling irradiance in a polluted cumulus cloud field with calculations of the spectral irradiance based on LES modeled cloud fields. In doing so the authors identified the importance of adequate characterization of aerosol properties such as hygroscopicity and absorption, alongside cloud optical depth, cloud size distribution, and other cloud-field properties. Improved understanding of how a cloud field influences aerosol radiative properties (Wen et al. 2007), together with the simultaneous influences of the aerosol on both microphysics and macrophysics, would be worthy of more attention. This is, after all, the essence of the “indirect effect.” To this end ARM continues to develop spectral radiation measurement capabilities. New retrievals of cloud fraction and cloud albedo from surface broadband radiation measurements (Liu et al. 2011) will also be of great value for linking ACI to radiative effects.

Finally, with the recognition that the decades-long record of observations of clouds, aerosol, and radiation at megasites such as SGP or NSA could be used more fruitfully for model evaluation, ARM is creating a framework to further facilitate comparison between models and observations. Modelers tend to focus on a few choice case studies because of the difficulties of evaluating their models for a broad range of conditions. ARM and ASR are together engaging in plans for regular, high-resolution observations. Model output from regular high-resolution modeling to complement regular, high-resolution observations. The benefits would include rigorous testing of models under different cloud conditions, comparison with observations at the appropriate scales, and testing, improvement, and development of new parameterizations for climate models (e.g., Neggers et al. 2012). The observationally constrained model output from regular high-resolution modeling will generate datasets that can be used for a variety of applications, including aerosol–cloud interactions. Efforts such as these will support efforts to quantify aerosol indirect effects and enhance ARM’s legacy in the field.

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