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Cloud–Aerosol Interactions from the Micro to the Cloud Scale

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Abstract

The effect of suspended particles (aerosol) on clouds and precipitation from the micro to the cloud scale has been well studied through laboratory, *in-situ*, and remote-sensing data but many uncertainties remain. In particular, there is scant observational evidence of aerosol effects on surface precipitation. Clouds and precipitation modify the amount of aerosol through both physical and chemical processes so that a three-way interactive feedback between aerosol, cloud microphysics, and cloud dynamics must be considered. The fundamental cloud microphysical properties are driven by dynamics; vertical motions and mixing processes between the cloud and its environment determine the concentration of cloud water, a key parameter for both climate and precipitation. However, aerosol particles can significantly affect the microphysics and dynamics of clouds by changing the size distribution of drops, their ability to grow to raindrops, their rates of evaporation, and their mixing with the environment. The physical system is strongly coupled and attempting to separate aerosol effects has only been done using some simple constructs, some of which will be shown to be of dubious utility. Both observations and modeling suggest that not only the magnitude, but perhaps also the sign of these effects, depends on the larger-scale meteorological context in which aerosol–cloud interactions are embedded. Some alternate approaches are considered, as we explore the possibility of self-regulation processes that may act to limit the range over which aerosol significantly affects clouds.

Historical Perspective

Historical evidence of the effects of aerosol on atmospheric processes abounds, as seen in ancient texts all the way through to modern times. Common human

experience tells us that less sunlight reaches the surface under cloudy skies and that aerosol laden skies (e.g., from biomass burning or volcanic eruptions) produce hazy skies and colorful sunsets. In 1783, Benjamin Franklin experienced a particularly cool summer in Europe and attributed it to "fog" associated with a volcanic eruption in Iceland, earlier in the year, that visibly dimmed the sunlight. Evidence that aerosol particles are complicit in cloud droplet formation came in the late 19th century, when P.-J. Coulier (1875), J. Aitken (1880), and C. T. R. Wilson (1897) showed that aerosol particles are necessary for cloud droplet formation. Subsequent work by Aitken (1923) and Köhler (1936) strengthened further the link between aerosol and cloud droplet formation. Houghton (1938) discussed the importance of collision and coalescence of cloud droplets for rain formation and even recognized the role of giant hygroscopic nuclei, a subject that is of great interest to this day. Howell (1949) developed a numerical model of the growth of a population of droplets in a rising parcel of air and laid the foundations of similar tools used today. Analytical solutions to these equations (Twomey 1959) are still in common use. Thus, although the early literature did not focus on aerosol as a means of *perturbing* cloud microphysical processes, the link between aerosol and clouds has existed in the scientific literature for over a century.

Pollution and Climate Change

A more direct link between pollution particles and climate change was raised by Twomey (1974), who considered the fact that the positive correlation between aerosol number concentration, N_a, and drop number concentration, N_d, would result in smaller droplets and more reflective clouds *ceteris paribus*; that is, all else, particularly the amount of condensed water, being equal. This constitutes a shortwave radiative forcing since low-level clouds radiate in the longwave at temperatures close to the surface temperature. The concept is a hypothetical one since it is known that feedbacks also result in modifications to liquid water. It is applied in global climate models by modifying the reflectance of fixed cloud fields. The recently released IPCC (2007) report still regards this "cloud albedo effect" as the single biggest unknown in climate change predictions. It is also of note that although many aspects of the effect of aerosol particles on clouds are well-known, the global radiative (and therefore climate) implications are still poorly constrained. For example, most satellite remote-sensing studies quantify the change in cloud reflectance, or cloud drop size, in response to an aerosol perturbation without stratifying for similar meteorological conditions (e.g., cloud liquid water path, LWP). Thus "*ceteris paribus*" is often ignored, resulting in considerable ambiguity: Are clouds more reflective because of higher N_d and smaller drops, or because of higher liquid water content (LWC), or some combination of the two?

Pollution and Precipitation

One of the oldest problems in cloud microphysics is the inability to describe quantitatively broad drop size distributions that are observed (compared to the narrow, modeled size distributions that assume adiabatic growth) and the onset of precipitation. Broadening has been attributed to entrainment-mixing, aerosol effects (such as giant aerosol), and turbulence effects on collision–coalescence. The first experimental evidence of a link between pollution and precipitation processes known to us appeared in a paper by Gunn and Phillips (1957). They performed experiments in a giant cloud chamber and observed that when clean air was drawn into the chamber, large, precipitation-sized drops were produced; polluted air produced smaller drops and no precipitation. These observations were confirmed in natural clouds a decade later by Warner (1968), but subsequent analysis by Warner yielded inconclusive results. The experimental evidence for this link is still a subject of enormous debate, particularly for deep convective cloud systems (e.g., Levin and Cotton 2007; see also Cotton, this volume; Ayers and Levin, this volume).

The implications for climate change of aerosol effects on precipitation were considered by Albrecht (1989), who used a numerical model of a cloudy boundary layer to show that an increase in N_d results in clouds with less collision–coalescence, less precipitation, and therefore increased LWP and higher albedo. This constitutes a positive feedback to the albedo effect since Twomey (1974) had considered the radiative impacts for static clouds of the same LWP. In addition, Albrecht (1989) suggested that a suppressed precipitation process might increase the lifetime of a cloud. In the intervening decades, the precipitation suppression hypothesis has become known as the "Albrecht effect," or "lifetime effect." It has also come under increasing scrutiny in recent years as observational evidence for this phenomenon (*in-situ*, satellite remote sensing) has been pursued. Various aspects of this linkage between aerosol, precipitation, and climate had been qualitatively confirmed (e.g., the suppression of precipitation by aerosol in warm rain), but to date there is still considerable debate over the effects of aerosol on cloud fraction, cloud LWP, and cloud lifetime, and the quantitative relationship between aerosol and surface precipitation.

Aerosol Effects on Clouds: Deconstructing the Constructs

The Albedo Effect

There is very little argument about the existence of aerosol perturbations to cloud albedo, provided that clouds are stratified by similar amounts of condensed water. The classic example is of ship tracks, which manifest themselves as bright linear features in a cloudy background associated with aerosol particles emitted from ship stacks.

The primary discussion centers around the *degree* of this effect; that is, the extent to which pollution contributes to the particle number concentration at diameters greater than ~0.05 μm, and the extent to which N_d increases with increasing aerosol.

Parameters Affecting Activation: Number and Size versus Composition

There is some controversy over the relative importance of various aerosol parameters (number, size, composition) in determining N_d. Whereas inorganic sulfate particles were assumed to be the dominant aerosol type vis-à-vis cloud microphysics, research in the last decade has pointed to the prevalence of organics (both primary and secondary), and much work has addressed their influence on clouds relative to inorganics. It has been suggested that the surface tension-reducing properties of some organics may significantly enhance the number of activated droplets by reducing the Kelvin (curvature) effect. However, modeling attempts, which consider composition by taking into account a variety of (sometimes competing) factors (e.g., molecular weight, surface tension, van't Hoff factor, and solubility), have shown that the effects of composition are relatively small ($\leq 15\%$) compared to aerosol parameters such as N_a and size, or dynamical parameters such as updraft velocity (Ervens et al. 2005). These findings echo the words of Houghton (1938), who upon examining the form of the Köhler curves stated: "This would seem to indicate that hygroscopic nuclei are not much more effective than neutral [insoluble] nuclei of the same size." A number of exceptions to this may exist: the presence of film-forming compounds could act as a barrier to droplet growth. Composition, as manifested in an external mixture of both hydrophopic and hygroscopic particles, is also of great importance in determining N_d. This, and other composition issues, are discussed in much more depth by Kreidenweis et al. (this volume).

It is important to note that the fact that aerosol composition may significantly influence the equilibrium diameter of a particle does not equate to a concomitant effect on the number of activated drops. The system, comprising a population of particles growing in an updraft, is self-regulating to a degree. Composition effects that reduce (increase) condensation are compensated for by an increase (decrease) in available vapor for activation of smaller particles, and/or growth of existing particles. Therefore, calculations of N_d based on equilibrium calculations, rather than dynamic calculations, must always overestimate the importance of composition for N_d.

Perhaps the single, greatest uncertainty in droplet activation and the growth of a population of drops is that of the mass accommodation, α_c, defined as the fraction of water vapor molecules arriving at a droplet that condenses. Values ranging from 0.03 to 1 have been measured experimentally and proposed based on theoretical arguments. Some of the experimental variability may be associated with the purity of the water droplets, as well as temperature and pressure. The effects of α_c are significant; larger values (order 0.3–1) allow growing haze

droplets to grow much more efficiently, thus reducing the supersaturation and N_d significantly. Conversely, smaller values (on the order of 0.05) allow the supersaturation to build up, resulting in higher N_d. The effect is considerably larger than those associated with identified composition effects. Drop number concentration closure experiments suggest $\alpha_c \sim 0.06$ (Conant et al. 2004), but more direct measurements show that α_c may vary by more than a magnitude depending on location and time (Ruehl et al. 2008).

We would be remiss in not mentioning at least one case where an increase in N_a has been hypothesized to result in a *decrease* in N_d. Large, or giant particles (> a few μm in size), and in sufficient concentrations (order 1 cm^{-3}), have been shown in modeling studies to suppress the development of supersaturation and prevent the activation of smaller particles (O'Dowd et al. 1999). Again we stress the importance of calculating N_d as a dynamic process. Had one performed equilibrium calculations of N_d in the presence of giant nuclei, N_d would simply have increased by the number concentration of giant nuclei, rather than decreased.[1]

Finally, in the case of ice nucleation unlike that for water droplets, insoluble particles tend to be better ice nuclei than soluble particles. It is noteworthy that most parameterizations pose ice crystal concentration, N_i, as a function of temperature and sometimes ice supersaturation. The absence of explicit representation of the total aerosol population in which these ice nuclei reside may at least partially explain the very large dynamic range of observed N_i when compared to predictions from said parameterizations.

Albedo Susceptibility

An understanding of the increase in N_d for an incremental increase in aerosol is an essential component of the shortwave albedo effect but does not constitute an understanding of the radiative response. For the simple case of clouds, which exhibit a linear increase in LWC with increasing altitude (adiabatic or subadiabatic), cloud optical depth τ_c can be shown to scale with $N_d^{1/3}$ $LWP^{5/6}$ (e.g., Boers and Mitchell 1994). Cloud optical depth is therefore two-and-a-half times more sensitive to LWP than to N_d. For thin clouds, cloud albedo is linearly dependent on τ_c and exhibits therefore similar sensitivities. This points to the importance of stratification by LWP if a pure aerosol effect on cloud optical depth is sought. Alternatively, in a dynamic system with variable LWP, the extent of variability in LWP must be considered alongside the variability in the aerosol (and N_d) perturbation. A useful approach has been to consider the cloud susceptibility S_0 (Platnick and Twomey 1994), defined as the change in cloud albedo A for an incremental increase in N_d. For a plane-parallel cloud, at constant LWP and fixed distribution breadth (constant "micro"),

[1] Giant nuclei, in concentrations as low as 10^{-3} cm^{-3}, also expedite the formation of rain (Houghton 1938), as will be discussed later.

$$S_0 = \left.\frac{dA}{dN_d}\right|_{\text{micro}} = \frac{A(1-A)}{3N_d}, \tag{14.1}$$

or

$$S_0' = \left.\frac{d\ln A}{d\ln N_d}\right|_{\text{micro}} = \frac{(1-A)}{3}. \tag{14.2}$$

S_0' decreases monotonically with A and has the advantage of being independent of N_d.[2] Relaxing the assumptions of constant LWP and breadth yields

$$S_0'' = S_0'\left[1 + \frac{5}{2}\frac{d\ln \text{LWP}}{d\ln N_d} + \frac{d\ln k}{d\ln N_d}\right], \tag{14.3}$$

where k is a parameter inversely proportional to drop distribution breadth, and the double prime implies that microphysical assumptions have been relaxed. We note the strong dependence of susceptibility on LWP changes. When spectral broadening (smaller k) is associated with increasing N_d (because of competition for water vapor in the relatively polluted, condensation-dominated regime), S'' is diminished, whereas when broadening is associated with a reduction in N_d (the cleaner, coalescence-dominated regime), S'' is enhanced.

A complete description of the radiative response of a cloud system to an aerosol perturbation requires 3-D radiative transfer modeling based on a description of both microphysical properties (drop size distributions) and macroscale properties, such as the spatial distribution of cloud optical depth, LWP, cloud fraction, cloud morphology, and distances between clouds. Moreover, the regions of aerosol haze surrounding clouds can also have a substantial radiative impact (Koren et al. 2007; Charlson et al. 2007). Zuidema et al. (2008) have demonstrated that cloud microphysical and macrophysical responses to perturbations in aerosol can work in unison or counter one another to produce either stronger or weaker albedo response to a change in aerosol.

Higher-order Aerosol Perturbations to Clouds

The effect of aerosol on clouds without stratification by LWP is a far more complex construct because it addresses an evolving system which includes multiple feedbacks over the life cycle of a cloud. Parts of this construct, such as the aerosol suppression of warm rain, are fairly well established, at least from a qualitative point of view. Other aspects, such as the increase in LWP, cloud fraction, and cloud lifetime are highly uncertain and either completely lack observational support (e.g., cloud lifetime) or are plagued by measurement difficulties, artifacts (e.g., satellite measurement of aerosol effects on cloud

[2] These susceptibility calculations are posed in terms of N_d perturbations. Similar relationships could be developed in terms of N_a perturbations by linking N_d to N_a and other parameters; a nontrivial problem.

fraction), and the fact that observed correlations are not necessarily indicative of causal relations. In the case of cold clouds, there is even greater uncertainty because of significantly increased complexity and a dearth of measurements, or measurement difficulties.

Unlike the case of the albedo effect, there is no underlying normalization by LWP in the "lifetime effect" because it attempts to address the LWP response. Separation of dynamical influences on LWP and other cloud properties, as opposed to aerosol effects, is therefore the primary challenge that has eluded us for decades. The fact that so many coupled processes act in unison suggests that new approaches need to be considered.

Increase in Cloud Fraction and Liquid Water Path

A central tenet of the "lifetime effect" is that both LWP and cloud fraction increase in response to an increase in (scattering) aerosol attributable to suppression of collision–coalescence and precipitation. (The special case of absorbing aerosol will be discussed separately below.) The ACE-2 field campaign illustrated the pitfall of assuming that polluted clouds generate clouds with greater LWP. There, pollution events were associated with drier and warmer air (Brenguier et al. 2003) and, consequently, lower LWP, which was most likely further reduced by free tropospheric entrainment. Thus, it is not surprising that satellite studies have produced mixed messages. These studies are correlative in nature, and causality is much harder to establish. Satellite remote sensing is also not without measurement challenges: aerosol and cloud measurements are not collocated; the closer one gets to a cloud, the more difficult it is to distinguish cloud from hydrated aerosol ("cloud contamination"). Some of these problems are alleviated by surface and/or aircraft-based remote sensing.

Modeling studies have established causal links between both negative and positive responses of cloud LWP to aerosol but we await direct observational support. We will dwell on this point since it is of central importance. When dry air is entrained into positively buoyant clouds, evaporative cooling can cause buoyancy reversal. Negatively buoyant parcels produce more turbulence kinetic energy, which enhances entrainment and mixing. In the case of stratocumulus clouds, this process has been hypothesized to break up clouds (cloud-top entrainment instability).

Some recent modeling studies have begun to challenge the generality of the assumption that LWP increases with increasing aerosol. Wang et al. (2003) used large eddy simulations (LES) of stratocumulus clouds to show that LWP decreased in response to an aerosol perturbation for idealized non-precipitating clouds. This decrease was linked to the fact that polluted clouds have smaller drops and thus a shorter evaporative timescale than clean clouds. The result is a stronger entrainment rate and a decrease in cloud water. We wish to highlight the fact that bulk microphysical schemes assume instantaneous condensation/evaporation and therefore (a) cannot simulate this feedback and

(b) overestimate entrainment rates. This same result was also demonstrated for shallow cumulus (Xue and Feingold 2006). Ackerman et al. (2004) simulated precipitating and non-precipitating stratocumulus clouds for a variety of environmental soundings and reinforced the importance of evaporation rates at cloud top. They showed that only under very low N_d or humid conditions above cloud top did an increase in aerosol result in an increase in LWP. Under more polluted conditions and/or drier free tropospheric air, entrainment dominated and cloud LWP decreased with increasing aerosol (thus opposing the Twomey effect).

Can Aerosol Influence the Behavior of Small-scale Mixing in Clouds?

Above, we discussed the fact that aerosol can influence cloud dynamics via modification to the condensation/evaporation timescale (Wang et al. 2003). The tightly coupled nature of the cloud droplet/dynamical system at these small scales (order centimeters to meters) requires some discussion of mixing mechanisms. When a cloudy air parcel mixes with ambient subsaturated air, two bounding scenarios have been considered:

1. Homogeneous mixing: where turbulent mixing is much faster than the rate at which droplets can react to their new environment. Under these conditions all droplets experience approximately the same thermodynamic conditions of the newly mixed air parcel followed by evaporation.
2. Inhomogeneous mixing: where turbulent mixing is comparably slow and the boundary regions between entrained and cloudy air exist long enough to ensure that the droplets in the interfacial regions evaporate while other regions remain unaffected by mixing.

These mixing scenarios have different influences on droplet size distributions. For homogeneous mixing, the mean drop radius, $\bar{r}$, is shifted to smaller sizes as all drops experience the same subsaturation. For inhomogeneous mixing, $\bar{r}$ is maintained but N_d is reduced by the number of evaporated droplets. The reduction in N_d means that the mixed air parcel may subsequently experience higher supersaturations compared to parcels that have not experienced mixing, and produce larger droplets than those associated with adiabatic growth. Entrainment of unactivated nuclei is more likely to result in activation of smaller particles. Both processes suggest that inhomogeneous mixing will generate broader droplet spectra than those resulting from homogeneous mixing.

The relevant number for examining these processes is the Damköhler number, D_a, defined as the ratio between the timescale typical for the mixing process τ_m, and a timescale for the reaction of the droplets to their new (subsaturated) thermodynamic environment τ_e. For a given air parcel with typical dimension l and energy dissipation rate ε describing the turbulence intensity, the time τ_m for complete mixing of this parcel is given by classical turbulence

theory, $\tau_m = (l^2/\varepsilon)^{1/3}$. Although it is quite straightforward to estimate ε from cloud measurements, it is not clear what the governing length scales, l, are. In fact, l will likely change during the course of a cloud's life cycle.

Definition of an appropriate timescale for reaction (evaporation) is also not straightforward. A typical timescale can be defined as the evaporation time τ_e of a droplet with mean radius $\bar{r}$ in an environment with ambient subsaturation $S - 1$ and can be derived from $r\frac{dr}{dt} \propto \frac{S-1}{r}$ to yield $\tau_e \propto \frac{\bar{r}^2}{|S-1|}$. However, as evaporation proceeds, the humidity increases, and if saturation is reached before all droplets have evaporated, then a phase relaxation timescale $\tau_p \propto (\int rn(r)dr)^{-1} \sim (N_d\bar{r})^{-1}$ is more appropriate. It has been proposed that one use the minimum value of τ_e and τ_p to avoid unrealistically large values of τ_p in strongly subsaturated regions or very large values of τ_e close to saturation. Although τ_p and τ_e appear to have very different dependences on $\bar{r}$ ($\bar{r}^{-1}$ and $\bar{r}^{-2}$, respectively), calculation of τ_e and τ_p for drop populations with the same LWC results in similar values for both (since $N_d \sim \bar{r}^{-3}$ for constant LWC).

A decrease in the evaporation timescale will lead to a shift in the mixing process towards the inhomogeneous extreme (*ceteris paribus*) with attendant reduction in N_d, broadening of the droplet size distribution, and an increased chance of producing larger droplets. This behavior tends to reduce the albedo response to an increase in aerosol number concentration. Furthermore, the production of a few large droplets can initiate a more effective coalescence process which could lead to the onset of precipitation and counter the conventional higher-order effects as posed by Albrecht (1989).

The suggested linkage between τ_e and τ_m raises the question of how D_a ($= \tau_m/\tau_e$) might respond to an aerosol perturbation. This is represented schematically in Figure 14.1. An aerosol perturbation reduces τ_e. The shorter τ_e will likely generate stronger mixing as a result of a more efficient evaporation process. How this affects τ_m and D_a will depend on how ε and/or the characteristic size of the entrained blob of air l are affected by evaporation (vertical arrows). Large eddy simulations of non-precipitating cumulus (Xue and Feingold 2006; Jiang et al. 2006) have shown that turbulence kinetic energy increases in response to increasing aerosol because enhanced evaporation rates generate more negative buoyancy and stronger horizontal buoyancy gradients, which generates stronger vorticity. This suggests a shorter τ_m (downward arrow) and modulation of increases in D_a resulting from reduced τ_e.

Increase in Cloud Lifetime

There is no observational support for the claim that an increase in aerosol increases cloud lifetime. In stratiform clouds, such as stratocumulus, it is impractical to define a cloud lifetime since the existence of a cloud for many hours does not mean that individual parcels of cloudy air exist for many hours; rather,

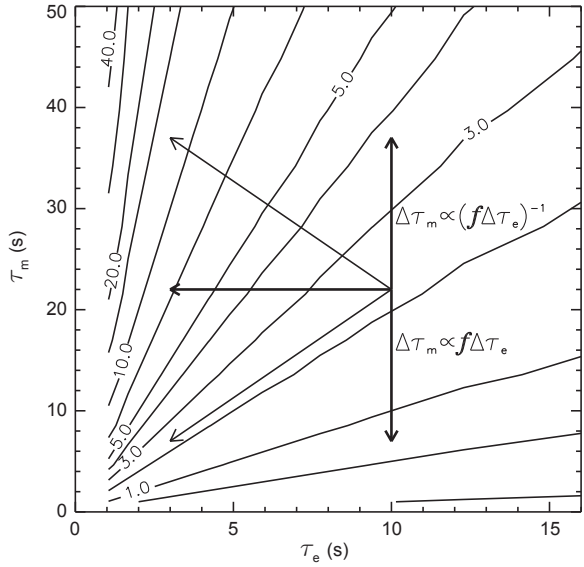

Figure 14.1 Contours of Damköhler number D_a and possible effects of aerosol on D_a. Calculations are based on modified gamma drop size distributions for a range of N_d and fixed breadth parameter. An aerosol perturbation of 300 cm^{-3} versus 50 cm^{-3} results in a reduction in τ_e (left arrow). The end point in D_a space (thin solid arrows) depends on whether τ_m increases (up arrow) or decreases (down arrow) as a result of the perturbation.

these parcels cycle in and out of clouds, and cloud elements constantly form and decay.

In the case of convective cumulus clouds, it is more straightforward to calculate the lifetime of a cloud cell. Radar has been used to track the lifetime of precipitating cells; however, this is very different from the lifetime of the cloud. Anecdotal evidence of clouds in very clean conditions precipitating almost as soon as they form has been noted, but we are unaware of any systematic observations of aerosol effects on cloud lifetime. Modeling work has shown no effect of aerosol perturbations on the lifetime of populations of cumulus clouds (Jiang et al. 2006). There are also indications that aerosol effects may depend on the size of the perturbed clouds and that small clouds may respond differently from large clouds.

The "lifetime" concept is one that climate modelers have sustained because it is relatively easy to simulate. In a climate model, study of the "lifetime effect" is equivalent to changing the rate at which cloud water converts to rain and has little to do with cloud lifetime. It does, however, artificially force water to rain out earlier or later on timescales on the order of half an hour (the typical timestep of a global climate model). Since this is a significant fraction of the lifetime of a real cloud, lifetime studies in climate models are ill-posed because they attempt to simulate many unresolved processes.

The "Semi-direct" Effect

The so-called "semi-direct" effect is a poignant example of how aerosol might affect clouds "indirectly"[3] by influencing cloud dynamics over a range of spatial scales.[4] Light-absorbing aerosol generate local heating, and their presence at the appropriate concentrations may modify the atmospheric stability and suppress vertical motion and cloud formation (Hansen et al. 1997). However, in general, absorbing aerosol layers may either stabilize or destabilize the boundary layer depending on where the aerosol is located, so that this factor alone may not explain observed reductions in cloudiness.

We expand our definition of the semi-direct effect to include the fact that the presence of aerosol, particularly light-absorbing aerosol, is very effective at reducing the downwelling solar radiation at the surface. Therefore, balance in the net surface radiation requires that the upwelling surface sensible and latent heat fluxes are reduced. Over land, where surface heating is a primary driver for convective clouds, this can have a significant impact on cloud fraction and cloud depth. This process links aerosol–cloud interactions to land surface type, another large-scale control that cannot be ignored. Together these mechanisms provide a means for aerosol to reduce the cloud fraction of surface-forced clouds (Koren et al. 2004; Feingold et al. 2005).

Cloud Effects on Aerosol

Clouds are not only affected by aerosol, they also exert significant effects on aerosol. These effects, sometimes lumped into a category of "cloud processing of aerosol," comprise a number of different processes such as *washout* (the removal of aerosol by rain falling to the surface), *convective redistribution* (the vertical transport of aerosol by clouds), *coalescence processing* (modification in the number and size of aerosol particles resulting from repeated drop coalescence events), *chemical processing* (the formation of nonvolatile mass attributable to aqueous chemical reactions, discussed briefly below), and *new particle formation around clouds* (addressed in somewhat greater detail). Since most cloud droplets evaporate and removal by precipitation is a relatively rare occurrence, aerosol processing by clouds is a major factor in the aerosol particle life history.

In terms of *chemical processing*, more than half of global sulfate is produced in clouds via aqueous reactions (e.g., Scott and Hobbs 1967); clouds may also be a significant source of secondary organic aerosol (Ervens et al. 2008). Aqueous production of aerosol is manifested in a bimodal size distribution: the first (smaller) mode comprises unactivated aerosol particles whereas

[3] We use "indirectly" here to convey that we are not referring to one of the classical aerosol indirect effects.

[4] Other examples include the self-organization of cloud structures at the mesoscale.

the second is made up of activated particles, upon which additional nonvolatile mass has formed. The implications range from significant increases in light scattering to modification in activation in subsequent cloud cycles.

New particle formation events can produce number concentrations that exceed background concentrations by orders of magnitude (Hegg et al. 1990; Perry et al. 1994; Clarke et al. 1998). These freshly formed particles can grow by condensation to Aitken mode particles (20 nm < diameter < 100 nm) over the course of many hours, and thus serve as cloud condensation nuclei (CCN) or influence the radiative properties directly.

Most explanations of increased ultrafine number concentrations are based on binary homogeneous nucleation of sulfuric acid (H_2SO_4) and water vapor (H_2O); however, ternary nucleation ($H_2SO_4 - NH_3 - H_2O$) or H_2SO_4 ion clusters may also play a role. For all of these mechanisms, an elevated concentration of sulfuric acid is essential, and the high relative humidity in the vicinity of clouds promotes further particle formation. The sulfuric acid can be produced by oxidation of SO_2 with photochemically produced OH, the latter requiring high actinic fluxes. The sulfuric acid vapor has to be supersaturated before condensation can occur, so low temperatures and high humidity are ideal. In addition, clean (vis-à-vis aerosol) conditions are favored, otherwise gases will simply condense onto existing particles.

Cloud edges with high actinic fluxes, high relative humidity, and relatively low temperatures, combined with convection and therefore vertical transport of precursor gases, are thought to be an ideal environment for new particle formation. However, many details of these processes are still not well understood; discrepancies between observed and modeled nucleation rates exist, with models typically underestimating these rates significantly. Many open issues remain, including:

1. Ammonia is usually not measured in clouds and thus ternary nucleation is poorly understood.
2. What possible influence do ion clusters have in the vicinity of clouds?
3. What is the exact location of the nucleation process? Most airborne particle measurements have a spatial resolution of only 50–100 m. This is of the same order of magnitude as the detrainment zone itself, which has been identified as one favored region for new-particle formation.
4. Drop and/or ice particle shattering on inlets of fast-flying aircraft may explain the observed high number concentrations of small particles (Weber et al. 1998).

Reconstructing the Constructs and Possible Ways Forward

A Proposed New Construct for Warm Clouds

Current constructs of aerosol effects on clouds are useful provided that they can be related to observations that prove causality. We propose here a simple construct that may assist in addressing aerosol effects on precipitation. Consider an analog to the albedo susceptibility S'_0, which we will term "*precipitation susceptibility*," R'_0, defined as:

$$R'_0 = -\frac{d \ln R}{d \ln N_d}. \tag{14.4}$$

R'_0 expresses the relative change in precipitation R for a relative increase in N_d in warm clouds. The minus sign is applied so that a positive R'_0 will reflect the conventional wisdom that for warm rain, R and N_d are negatively correlated. (Cold clouds, and the ensemble of results from a population of interacting clouds, may respond quite differently; e.g., Orville and Chen 1982.) To begin, we have performed some simple calculations with a cloud parcel model that includes warm microphysical processes (activation, condensation, collision–coalescence), solved using size-resolved methods, to test the usefulness of this approach. The parcel rises at constant velocity and neglects entrainment. It is run for a range of LWP ($50 < \text{LWP} < 1100$ g m^{-2}) and CCN concentrations ($25\ \text{cm}^{-3} < \text{CCN} < 500\ \text{cm}^{-3}$). Results suggest that R'_0 is a function of LWP (Figure 14.2). There appear to be three regimes:

1. A low LWP regime where R'_0 is small, primarily because low LWP limits the cloud's ability to generate precipitation.
2. A mid-LWP regime where R'_0 increases steadily with increasing LWP. Here the precipitation process is not limited by LWP and addition of N_d tends to suppress R.
3. A high LWP regime where R'_0 begins to decrease, most likely because there is sufficient condensed water for clouds to precipitate regardless of N_d.

These results should be considered qualitative and subjected to further scrutiny, given the limitations of the model (primarily the neglect of sedimentation and entrainment)—limitations that become increasingly severe as LWP increases. However, results from an ensemble of 500 trajectories from two LES simulations of stratocumulus show agreement in the low LWP regime (horizontal line in Figure 14.2).[5] More importantly, remarkable new results from CloudSat radar analyses (M. D. Lebsock and G. L. Stephens, pers. comm.) show that Figure 14.2 is qualitatively reasonable. Their data show that deeper,

[5] Our analysis of 500 trajectories derived from LES of ASTEX and FIRE cases (LWP ~ 100–200 g m^{-2}) yields a maximum rainrate of $R \sim \text{LWP}^{1.6} / N_d^{0.67}$, i.e., $R'_0 = 0.67$.

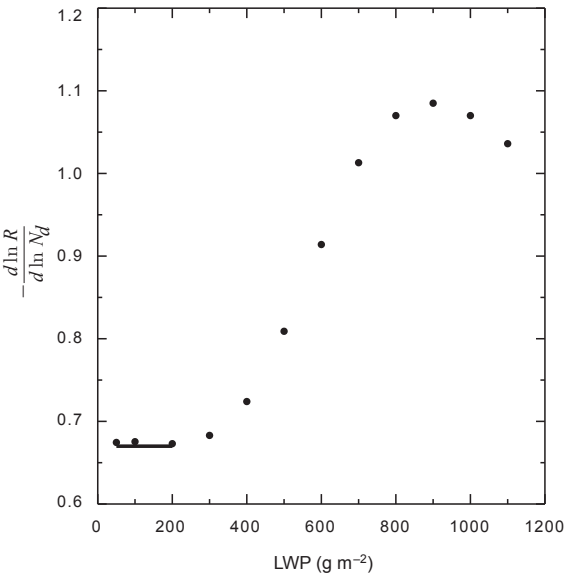

Figure 14.2 Precipitation susceptibility as a function of the liquid water path (LWP). Note the existence of three distinct regimes: (a) low R'_0 at low LWP; (b) steadily increasing R'_0 at intermediate LWP; (c) decrease in R'_0 at high LWP. The solid horizontal line at $R'_0 = 0.67$ is derived from analysis of large eddy simulations trajectories for ASTEX and FIRE-I stratocumulus modeling studies.

warm clouds tend to exhibit a stronger reduction in rain with increasing aerosol than do shallow clouds.

This seems to be a construct worth pursuing, regardless of the functional form of Equation 14.4, since it provides guidance as to where efforts might be invested in exploring the sensitivity of precipitation to aerosol perturbations. Other parameters which should be considered include aerosol size distribution parameters and perhaps dynamic parameters such as turbulence kinetic energy and/or cloud-top heights (to the extent that the latter are not reflected in LWP). In cold convective clouds, one should include total condensate and perhaps even separate terms for ice and water content.

Equation 14.4 is amenable to testing with both *in-situ* and remote-sensing (satellite, as described above, and surface-based) observations. Its logarithmic form should alleviate requirements of accurate precipitation and drop concentration measurements. From aircraft, ACE-2 and DYCOMS-II field experiments have already derived expressions for cloud base precipitation as a function of cloud depth H and N_d of the form $R \sim H^3/N_d$ (e.g., van Zanten et al. 2005) or $R \sim \mathrm{LWP}^{1.5}/N_d$ (for linearly stratified clouds), consistent with $R'_0 = 1$. Moreover, these relationships have been tied to scavenging rates (Wood 2006), providing further motivation for their study. From space, microwave radiometers and radar provide measures of precipitation, as well as LWP, and retrievals of

column-integrated N_d can be derived under various assumptions. From the surface, R, LWP, and N_d can be derived using similar remote-sensing techniques.

Self-Regulation Mechanisms and Non-Monotonic Behavior in Aerosol–Warm Cloud Systems

The previous discussion was peppered with examples of multiple feedbacks, some of which work in unison and some that counter one another. We outline a number of these below and consider the implications. Note that these effects are somewhat artificially separated in this listing and that more complex interactions are also possible.

1. Aerosol effects on LWP and cloud fraction: An increase in aerosol simultaneously acts to slow collision–coalescence and to enhance evaporation and reduce drop fall velocities. Xue et al. (2008) consider the possibility of non-monotonic responses of LWP to increasing aerosol perturbations in cumulus and stratocumulus clouds and the existence of two regimes: (a) at low aerosol concentrations, aerosol and LWP are positively correlated; (b) at high aerosol concentrations, aerosol and LWP are negatively correlated. The suggestion is that there is a limit to the degree of LWP buildup resulting from an aerosol perturbation. This may have some support in the satellite study of Han et al. (2002), who showed that ~1/3 of cases followed the first regime, ~1/3 followed the second regime, and ~1/3 showed no clear response.
2. Albedo susceptibility: Associated with the above regimes are corresponding albedo susceptibility responses. Clouds with low A have high S_0'; an increase in aerosol (and associated increase in N_d) produces monotonic increases in A but decreases in S_0'. However, if one relaxes the assumption of constant LWP (Equation 14.3), it is clear that macroscale cloud properties may significantly affect susceptibility. Although albedo is likely to respond monotonically with increasing aerosol, S_0'' may not, because of LWP and cloud fraction responses (Zuidema et al. 2008).[6]
3. The "semi-direct" effect: Under relatively clean conditions, an increase in aerosol containing an absorbing component will generate brighter clouds because many of these particles are hygroscopic enough to act as CCN. With further increases in aerosol, however, the absorption will be strong enough to suppress surface fluxes and stabilize the atmosphere, with concomitant reduction in τ_c, LWP, and cloud fraction (Jiang and Feingold 2006).

[6] Note that to the extent that the R and LWP responses to N_d are related, there may be an interesting link between R_0' and S_0''.

4. Deep convective clouds: In our modeling studies (S. Tessendorf and G. Feingold, pers. comm.), we have found non-monotonic responses of precipitation to aerosol in single deep convective, mixed-phase clouds. Warm clouds tend to have an active warm-rain process that facilitates the production of surface precipitation. On the other hand, polluted clouds enjoy the benefits of a higher freezing level and invigoration due to latent heat of freezing, resulting in an increase in the mass of condensed or frozen hydrometeors, which assists in generating precipitation via accretion processes. Clouds that are affected by moderate levels of pollution seem not to enjoy either of these benefits and may precipitate less than clean or polluted clouds.
5. Aerosol effects on clouds in the longwave: Thin clouds in the wintertime high latitudes experience an aerosol effect in the longwave that has not yet been discussed. Clouds with LWP$<$25 g m^{-2} act as gray bodies with emissivity $\varepsilon<1$ (e.g., Garrett and Zhao 2006). An aerosol perturbation to these clouds will increase the downwelling longwave radiation and warm the surface. An analog to the shortwave albedo susceptibility developed by Garrett ($S_{LW}=-(1-\varepsilon)\ \ln(1-\varepsilon)/3N_d$) 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.

These examples are all suggestive of subtleties that preclude simple assessment of aerosol perturbations to clouds as expressed by the albedo and lifetime effects. They also point to the possibility that in terms of both radiative forcing and precipitation, neither the cleanest nor the most polluted clouds are particularly susceptible to perturbations, but that some intermediate regime may be where efforts should be expended. This "intermediate regime" will depend on the problem at hand and the meteorological context, so that we hesitate to venture further. An interesting case in point is that of giant nuclei, which are known to expedite the collision–coalescence process. Their effect has been shown to be negligible in clean clouds which already have an active warm-rain process, progressively more significant (in a relative sense) in polluted clouds, but most important (in an absolute sense) in moderately polluted conditions.

It is important to distinguish between absolute effects and relative measures such as susceptibility (either albedo or precipitation). For example, the fact that albedo susceptibility is low does not imply that cloud radiative forcing is low, rather that aerosol perturbations will have little impact. One illustration of this is the stark difference in reflectance between relatively polluted, non-precipitating stratocumulus (high albedo, low S_0'), and cleaner, precipitating stratocumulus (low albedo, high S_0') which sometimes manifest themselves as closed cell convection and pockets of (precipitating) open cells, respectively (e.g., Xue et al. 2008).

Statistical Methods

The complexity of the aerosol–cloud–precipitation system is such that it may render the reductionist approach impractical. Statistical methods may need to be employed alongside *in-situ* and remote observations and numerical modeling. These would assess the primary factors driving aerosol effects on radiative forcing and precipitation using methodologies such as principal components analysis, factor separation, data assimilation, and others.

Summary

In conclusion, we provide a synthesis of the implications for aerosol effects on clouds and precipitation and offer a new framework in which the effects of aerosol on precipitation can be considered.

1. *Aerosol number concentration and size:* The available body of evidence suggests a dominant role for aerosol number concentration and size in determining drop concentrations in warm clouds. Composition is of secondary importance, except perhaps in very polluted conditions and for weak updrafts. Composition effects that are important include the mixing state of the particles (e.g., an externally mixed, hydrophobic mode) and the mass accommodation coefficient relevant to atmospheric conditions.

   For ice clouds, it is worth noting that characteristics of the total aerosol population, of which ice nuclei are a subset, do not appear in common ice nucleation parameterizations. This may explain the very wide range of predicted ice crystal concentrations.
2. *The coupled aerosol–cloud–dynamical system: Self regulation in response to aerosol perturbations?* Clouds respond to aerosol in a manner that is strongly dependent on meteorological context. Clouds are dynamic entities that are influenced by meteorological factors which control convection. Aerosol particles, a necessary ingredient for cloud droplet formation, are not the primary driver for clouds. Rather, they can modify cloud microphysics in sometimes subtle ways to generate feedbacks that may amplify or dampen their influence. These feedbacks have the potential to make aerosol effects on clouds of major climatic and hydrological importance. The extent to which this is true is the challenge of this community.

   The simple constructs, now decades old, referred to as the "albedo" and "lifetime" effects, have been shown to be of somewhat limited use. This is especially true of the lifetime effect, which requires separation of meteorological and aerosol influences in a tightly coupled system. Even for a given meteorological context, the response of a cloud to an aerosol perturbation may not be monotonic. Based on modeling

studies, we suggest the possible existence of two regimes: a cleaner regime where aerosol suppresses precipitation and results in increased cloudiness, and a more polluted regime where aerosol reduces cloud fraction because of more efficient evaporation. A similar non-monotonic response has been suggested for cloud response to absorbing aerosol: under clean conditions, clouds become brighter with increasing aerosol, but above some threshold aerosol absorption dominates, convection is suppressed, and cloud optical depth is reduced.

We also raise the possibility that aerosol may influence the nature of entrainment-mixing through effects on evaporation timescales and feedbacks of the aforementioned to the timescales of mixing (Figure 14.1). As yet, no firm conclusions have been drawn.

3. *A proposed new construct for investigating aerosol effects on precipitation*: We propose investigating the utility of the "precipitation susceptibility,"

$$R_0' = \frac{-d \ln R}{d \ln N_d}, \tag{14.5}$$

where R is rainrate, an analog of the albedo susceptibility. Some very simple calculations suggest that clouds in some intermediate LWP regime exhibit increasing susceptiblity to aerosol perturbations (Figure 14.2). There may be a maximum in R_0' at high LWP, beyond which clouds tend to precipitate regardless. There is new observational support for this hypothesis from spaceborne cloud radar. It is proposed that we identify parameters other than LWP that affect R_0' through statistical analyses of large observational datasets (satellite and surface remote sensing, *in-situ*) and model outputs. More generally, an increased use of statistical analyses of this kind may be required alongside the reductionist approach to expedite progress in the field.

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