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Aerosol cloud-mediated radiative forcing: highly uncertain and opposite effects from shallow and deep clouds

Daniel Rosenfeld¹, Robert Wood², Leo Donner³, Steven Sherwood⁴

1. Institute of Earth Sciences, The Hebrew University of Jerusalem, Israel,
daniel.rosenfeld@huji.ac.il
2. Atmospheric Sciences, University of Washington, Seattle, WA, USA,
robwood@atmos.washington.edu
3. Geophysical Fluid Dynamics Laboratory/NOAA, Princeton University, Princeton,
NJ, USA, Leo.J.Donner@noaa.gov
4. Climate Change Research Centre and ARC Centre of Excellence for Climate
System Science, University of New South Wales, Sydney, NSW, Australia,
s.sherwood@unsw.edu.au

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Abstract

Aerosol cloud-mediated radiative forcing, commonly known as the aerosol indirect effect (AIE), dominates the uncertainty in our ability to quantify anthropogenic climate forcing and respectively the climate sensitivity. This uncertainty can be appreciated based on the state of our understanding as presented in this paper.

Adding aerosols to low clouds generally causes negative radiative forcing by three main mechanisms: redistributing the same cloud water in larger number of smaller drops, adding more cloud water, and increasing the cloud cover. Aerosols affect these components sometimes in harmony but more often in opposite ways. These processes can be highly non-linear, especially in precipitating clouds in which added aerosol can inhibit rain. There is probably little overall sensitivity in most clouds but hyper sensitivity in some, where the processes become highly nonlinear with positive feedbacks, causing changes of cloud regimes in marine stratocumulus under anticyclones. This leads to a complicated and uneven AIE. Process models at high resolution (LES) have reached the stage that they can capture much of this complicated behavior of shallow clouds. The implementation of the processes of cloud aerosol interactions into GCMs is rudimentary due to severe computational limitations and the current state of cloud and aerosol parameterizations, but intense research efforts aimed at improving the realism of cloud-aerosol interaction in GCMs are underway.

Aerosols added to deep clouds generally produce an additional component of positive radiative forcing due to cloud top cooling, expanding, and detraining vapor to the upper troposphere and lower stratosphere. The level of scientific understanding of the AIE on deep clouds is even lower than for the shallow clouds, as mixed phase and ice processes play an important role. Respectively, the parameterization of these processes for GCMs is further away than for the low clouds.

Crucially, the AIE of both shallow and deep clouds must be considered for quantifying anthropogenic climate forcing and inferring climate sensitivity from observations.

While our objective is reducing the uncertainty, it appears that the recently acquired additional knowledge actually increased the uncertainty range of the AIE, as we learn of additional effects that should be quantified.

1. Introduction

Aerosols are thought to have exerted a net cooling effect on earth's climate that will have grown over the last century or two due to aerosol added by anthropogenic activities, influencing climate. This negative radiative forcing must have offset some of the warming that would otherwise have occurred due to greenhouse gases. The magnitude of this however remains highly uncertain; indeed aerosols represent the most uncertain climate forcing over the last 150 years (IPCC 2007), due to the complex ways aerosols can directly and indirectly affect radiation.

First, aerosols scatter sunlight to space that would otherwise have been absorbed, causing a so-called direct radiative forcing especially for aerosols over dark surfaces (oceans and forests). This negative forcing is offset somewhat by the absorption of outgoing infrared radiation (e.g., Myhre, 2009) and by the absorption of sunlight by dark (primarily carbonaceous) aerosols, both of which cause net warming, though nearly all studies find the cooling effects of the non-absorbing aerosols to be larger. This article will not discuss direct radiative forcing in detail, but papers elsewhere in this volume touch on some of the issues (Sherwood et al., 2012; Shepherd et al., 2012).

Second, aerosols serve as the nuclei (CCN or ice nuclei IN) for cloud droplets and can alter the albedo of clouds. As this component contributes the greatest uncertainty to our knowledge of Earth energy budget, it is the focus of our article. Adding CCN typically produces more droplets in a cloud, although this depends on details of the aerosols. Indeed the opposite can occur if the added particles are large enough compared to those already present, for example if sea salt is introduced into polluted continental air (Rosenfeld et al., 2002), although anthropogenic particles are generally too small for this to happen. All other things being equal (in particular, the cloud's size and condensed water content), more numerous droplets result in a so-called "Twomey" or droplet radius effect whereby the increased droplet surface area increases the cloud albedo, producing a negative indirect radiative forcing by the added CCN (Twomey, 1977).

All other things are however not generally equal: aerosols can also alter the subsequent fate of condensed water, and can drive circulations that alter the formation of clouds. These impacts lead to "adjusted" aerosol forcings analogous to those following the stratospheric adjustment to added greenhouse gases (e.g., Hansen et al., 2005). Both direct (radiative) and indirect (CCN-based) pathways produce such adjustments. For

example, heating of the air by absorbing aerosols can alter local stability and/or drive circulations that alter local or remote cloud amounts, producing a “semi-direct forcing” on regional or global radiative balances (e.g., Allen and Sherwood 2010). Smaller droplets may cause a cloud to dissipate either more quickly (by reducing fall speeds and increasing cloud break-up by increasing evaporative and radiatively driven entrainment) or more slowly (by decreasing droplet lifetimes in subsaturated air and the rate at which cloud is depleted by precipitation) – so called “lifetime” or “cloud amount effects” (Albrecht 1989). They also typically delay the formation of precipitation, which alters the latent heat release and therefore the dynamics of the cloud. Impacts can include invigoration and deepening of already deep clouds that would have rained anyway (e.g., Rosenfeld et al., 2008), or the suppression of rain in weaker, shallower and more susceptible cloud systems (e.g., Rosenfeld, 2000). Either implies changes to cloud water content, hence albedo; to cloud top height, hence greenhouse effect; to cloud amount, which affects both of these; and to net rainfall, hence the larger-scale circulation. It is in these “adjustments” where most of the uncertainty lies in quantifying the net climate forcing due to anthropogenic aerosols. Understanding of these has been sufficiently poor that the IPCC has not attempted to assess them up until now, but will do so to a limited degree in the upcoming AR5 report.

Model calculations of the aerosol indirect effect (AIE) have yielded radiative forcings of about -0.5 to -2.0 Wm^{-2} (e.g., Forster et al., 2007); these values overlap recent estimates based on satellite observations, which range from -0.2 to -1.2 Wm^{-2} (Quaas et al., 2009). Quaas et al. (2009) argued that models overestimate the AIE compared to satellite observations in present-day climate, while Penner et al. (2011) argue that flawed assumptions used in interpreting satellite data can cause severalfold understimation of AIE between pre-industrial and present-day climate. Another possible reason for the discrepancy could be that additional effects not yet included in models offset the Twomey effect. Such an effect might be positive radiative forcing due to aerosol impacts on deep convective clouds.

Since other anthropogenic radiative forcings are known better than the AIE, and since temperature changes over the last century or so are relatively well-measured, the total net forcing due to aerosols (including also any semi-direct effects of greenhouse gases) can be constrained based on the energetics of recent global climate, yielding a so-called “inverse” or “top-down estimate.” Anderson et al. (2003) compiled similar inverse calculations and concluded that total (direct and indirect) aerosol forcing near -1.0 Wm^{-2} but without taking the ocean heat uptake into account. Murphy et al. (2009) obtained a

68% range of -1.5 to -0.7 Wm^{-2} based purely on observations since 1950, but with no direct estimate of contributions from cloud and other feedbacks. Forest et al. (2006) obtained a 90% range of -0.74 to -0.14 Wm^{-2} by fitting a simple climate model (including feedbacks and ocean heat storage) to the spatiotemporal distribution of observed 20th-century temperature changes.

Stronger (more negative) aerosol forcings correspond to higher climate sensitivity (Kiehl, 2007). Values stronger than -1.5 Wm^{-2} would negate the impact of CO_2 since 1850, as a lag of the oceans of even -1.0 Wm^{-2} would imply implausibly high climate sensitivities (Forest et al. 2006). Since these estimates include the direct effect of aerosols, which is already about -0.6 to -0.1 Wm^{-2} , the Forest et al. (2006) numbers imply an AIE near zero while the Murphy et al. (2009) numbers would leave room for an AIE of weaker than -1.0 Wm^{-2} . These numbers are hard to reconcile with the estimates from GCMs. General circulation models (GCMs) began to estimate AIE in the middle 1990s. Early estimates ranged from about -0.5 W m^{-2} to nearly -4.0 W m^{-2} , but more recently constructed GCMs do not cool more than about -2.6 W m^{-2} (Isaksen et al., 2009; Quaas et al., 2009). Quaas et al. (2009) used satellite observations, which generally indicate weaker interactions between clouds and aerosols than GCMs, to scale GCM estimates, finding that an average AIE estimate from ten GCMs of -1.1 W m^{-2} was reduced to -0.7 W m^{-2} when scaled by satellite observations. These lower numbers are presented in the radiative forcing chart of Isaksen et al. (2009), shown here as Figure 1. When considering the high uncertainty range, especially for the cloud lifetime effect, a net forcing of zero or even negative values are included in the range of possibilities. Net zero or negative forcings are unlikely, of course, because it is hard to understand how the climate has warmed with zero or negative overall forcing, and this situation exemplifies the difficulty in estimating forcing due to cloud-aerosol interactions.

The metrics of the effect of the aerosols on cloud properties are often defined in logarithmic formulations (e.g., McKominsky and Feingold, 2008; Koren et al., 2008). This means that the clouds respond to the fractional change in CCN concentrations. This means, in turn, that large impacts can be expected when small amounts of aerosols are added to pristine air. Therefore, the background to which the aerosols are emitted is at least as important as the amount of emissions.

This paper addresses the main sources of uncertainty in AIE in the various kinds of clouds and aerosols, the way that they might be working together or at opposite directions, and suggests possible ways to address these questions. Section 1 (this section)

introduces the uncertainty of the AIE and the motivation for its reduction. Section 2 addresses the processes that determine the AIE from low clouds, whereas Sections 3 and 4 do the same for deep clouds and for supercooled layer clouds, respectively. Section 5 contrasts the mostly negative radiative forcing caused by the AIE of low clouds to the mostly positive forcing due to the deep and supercooled layer clouds. It also discusses the implications with respect to GCMs. In Section 6 we provide some recommendations for ways to address the formidable challenges that were discussed in this paper. An overall summary is provided in Section 7.

2. Aerosol-induced radiative forcing by boundary-layer warm clouds

2.1 The fundamental physical processes

The CCN supersaturation activation spectrum, $CCN(S)$, along with the updraft at cloud base, determines the maximum super saturation at cloud base, S , and hence the number of activated cloud drops, N_d . In a rising adiabatic non-precipitating cloud parcel the liquid water content, LWC , is determined exclusively by thermodynamic considerations and is highly linear with the vertical distance z above cloud base. In general, however, mixing processes (lateral and cloud top entrainment) cause the liquid water profile to be subadiabatic. Under most circumstances, mixing is predominantly inhomogeneous and causes the observed growth of the mean volume radius r_v with z in boundary layer clouds to follow closely the theoretical value of an adiabatic cloud parcel (Brenquier et al., 2000; Freud et al., 2011). It follows that, at any given height, r_v is inversely proportional to $N_d^{1/3}$, as long as the development of the cloud drop size distribution is dominated by diffusional growth, i.e., before drop coalescence advances and initiates warm rain, unless rain is already falling from above into the cloud. The same applies to the cloud drop effective radius, r_e , as it is very highly linearly correlated with r_v , where $r_e = 1.08 r_v$ (Freud and Rosenfeld, 2012). The r_e is a useful measure because it can be directly retrieved from satellite observations (Arking and Childs, 1985). We can write the aerosol indirect effect as the sensitivity of the albedo \langle to changes in N_d as

$$\frac{d\alpha}{dN_d} = \left(\frac{\partial\alpha}{\partial N_d} \right)_C + \sum_i \left(\frac{\partial\alpha}{\partial C_i} \right) \left(\frac{\partial C_i}{\partial N_d} \right) \quad [1]$$

where C_i are radiatively important cloud macrophysical properties (e.g. liquid water path, cloud thickness, cloud cover, etc.). The first term on the RHS of [1] represents the change in albedo caused only by changes in microphysics, in the absence of changes in cloud macrophysical properties. This is generally referred to as the Twomey effect, or the first aerosol indirect effect. The second term on the RHS represents the changes in albedo associated with aerosol-induced changes in cloud macrophysical properties. Equation [1] is very general since C_i can represent *any* changes to the system induced by aerosols. Examples for such properties are cloud liquid water path, precipitation content, geometrical depth, cloud top height, cloud cover and organization. The Twomey term is called the albedo susceptibility (Platnick and Twomey 1994), and is well-approximated (e.g. Twomey, 1991) by

$$\left(\frac{\partial\alpha}{\partial N_d} \right)_C \approx \frac{\alpha(1-\alpha)}{N_d} \quad [2]$$

Equation [2] indicates that aerosol-induced cloud albedo increases are greatest for clouds with low initial N_d . Further compounding the impact of aerosols on the albedo of clean clouds with low N_d is the fact that in this aerosol-limited cloud regime, almost all accumulation mode aerosols are activated to form cloud drops, i.e. $N_d \approx N_a$. As aerosol concentrations increase, the limiting factor on N_d increasingly becomes the updraft speed (updraft limited regime), and $N_d < N_a$, leading to much weaker sensitivity of albedo to aerosol increases (Pöschl et al., Science 2010).

In addition to the Twomey effect, observations and modeling results indicate that, in this aerosol-limited regime, cloud macrophysical properties (i.e. the second term on the RHS in [1]) are also particularly sensitive to aerosols. Cloud macrophysical responses to aerosols are more challenging to understand than the purely microphysical effect and are mediated via changes in the precipitation, sedimentation and evaporation of hydrometeors. These changes induce macrophysical responses in turbulent dynamics, entrainment rate, and, in some cases, mesoscale reorganization. Many of these processes remain poorly understood (Wood, 2012). This issue will be discussed later where it will be shown that when CCN is decreased below a certain concentration a full cloud cover can no longer be sustained.

Aerosol effects on the microphysical properties of boundary layer clouds (i.e., cloud drop size distribution and precipitation forming processes) may affect the macrophysical properties of the same clouds (i.e., cloud LWP, geometrical depth, cover and organization). The microphysical impacts of aerosol changes on boundary layer cloud macrophysical properties can be partitioned into precipitation/sedimentation mediated impacts and those that do not involve precipitation changes. Precipitation impacts are non linear due to internal mechanisms of feedbacks (some positive and some negative), which under some circumstances may lead to changes in cloud regime (e.g. closed to open cells, or stratocumulus-to-cumulus transition) that are associated with drastic jumps in the cloud cover and the respective radiative effect (Ackerman et al., 1995; Rosenfeld et al., 2006a; Wang et al., 2009, 2010 and 2011). Because precipitation can play an important role in these transitions, it is critical to understand the processes controlling transitions between lightly or non-precipitating marine stratocumulus (MSC) and heavily drizzling MSC.

Marine stratocumulus that form in stable atmosphere and maintained by radiative cooling of their tops persist under anticyclones and subtropical highs over the ocean, and occupy nearly 25% of the ocean surface. The radiative properties of these clouds represent large sensitivity to CCN concentrations, and might have a substantial global impact. While having a globally important cloud radiative effect, the overall actual radiative forcing from these clouds is a subject of intense debate due to complicated feedback mechanisms that are positive in some cases, mostly in precipitating MSC, and negative in others, mostly in non precipitating MSC.

Rain intensity in stratocumulus depends on N_d and cloud thickness h (Figure 2). Aircraft measurements (Van Zanten et al., 2005), supported by physical considerations (Kostinski, 2008), showed that cloud base rain rate $R \sim h^3 / N_d$. Since effective radius $r_e^3 \sim LWC / N_d \sim h / N_d$, then $R \sim h^2 r_e^3$. This was also reproduced by the simulations of Wang et al. (2009), but only for clouds with $N_d < 100 \text{ mg}^{-1}$, and h of about 600 m. For clouds with similar h but $N_d \approx 150 \text{ mg}^{-1}$ the surface rain rate was zero. This implies cloud top r_e of about 15 μm . Wang et al. (2009 and 2011) found similar results of complete suppression of surface precipitation at high N_d and respectively small r_e . The relation of $R \sim h^3 / N_d$ depends on the existence of rain embryos, but their scarcity in clouds with very small drops, as expressed by cloud top significantly smaller than 15 μm , causes R to become practically zero for any h and N_d . The dependence of R on liquid water path (LWP) and h was replicated by bulk microphysics models (Kubar et al., 2009; Wood et al., 2009), but they could not capture the complete suppression of R at high N_d and low r_e that was simulated with the explicit bin

microphysics models. Aircraft measurements in MSC (Gerber, 1996) showed that when r_e exceeds 16 μm most cloud water already resides in the drizzle mode, and that this can occur due to diffusional growth in the convective elements when N_d is sufficiently small. Interestingly, this height for onset of heavy drizzle increases linearly with N_d . A similar linear relationship between N_d and cloud depth for initiation of rain was observed by Freud and Rosenfeld (2012) in convective clouds over land. The validity of this threshold cloud top r_e as separating between the logarithmic response of the Twomey effect (Equation [2]) and the highly non-linear response to aerosols by regime change is supported by satellite observations, which show consistently that a cloud top r_e of 16 μm separates the closed and open cell regimes (Rosenfeld et al., 2006). Aircraft-measurements show that this change in r_e is also manifested in changes of N_d . An average N_d of 21 cm^{-3} was measured near cloud base of the open cells and 70 cm^{-3} in the closed cells (Wood et al., 2011).

2.2 Aerosol effects on non-precipitating and modestly precipitating clouds

The aerosol indirect effect on cloud albedo was introduced by Twomey (1977) and Equation [2] expresses its dependence upon cloud albedo and droplet concentration N_d . However, changes in aerosols rarely affect only N_d without changing cloud macrophysical properties such as cloud thickness and LWP . One might expect LWP to increase with CCN because less water is lost to precipitation (Albrecht, 1989). This is true for some meteorological conditions (Ackerman et al. 2004, Wood 2007). Certainly, there is good modeling and observational evidence that added aerosols can suppress precipitation (Ackerman et al., 2004; Lu and Seinfeld, 2005; Sandu et al., 2008; Feingold and Seibert 2009, Sorooshian et al. 2009,2010, Wang et al., 2010 and 2011; Chen et al., 2011; Terai et al., 2011). However, besides influencing the moisture budget of the clouds, precipitation also impacts the turbulent mixing, which can alter the moisture and energy budget of the boundary layer by changing entrainment (Ackerman et al. 2004, Wood 2007). Aerosol-suppressed precipitation results in increased cloud top entrainment that can warm and dry the boundary layer and thin the cloud, an effect that works in the opposite direction to the effects of precipitation on the surface moisture budget (Wood 2007). The overall effect on LWP therefore depends upon the ratio of the surface moistening (suppression of precipitation) compared with the entrainment drying/warming. When significant precipitation reaches the surface (usually heavily drizzling cases), or when the free-troposphere is relatively moist, precipitation suppression tends to increase LWP . In weakly

precipitating cases, where there is little surface precipitation, the entrainment drying may dominate, leading to aerosol-induced reductions in *LWP* (Chen et al. 2011). Indeed, many ship track cases appear to show such a response (Coakley and Walsh, 2002; Christensen and Stephens, 2011).

Increasing N_d can also enhance mixing due to faster evaporation of the smaller drops at the border of the clouds and resultant enhanced mixing with the dry ambient air (Wang et al., 2003; Lu and Seinfeld, 2005; Hill et al., 2008 and 2009; Chen et al., 2011, Small et al. 2009). Increased N_d also reduces the sedimentation of cloud droplets which can increase entrainment rate (Bretherton et al. 2007). Large eddy modeling shows that increases in CCN shorten the life time and reduce the size of small trade wind cumuli (Jiang et al., 2009a).

Overall, the macrophysical responses to aerosols in weakly precipitating and non precipitating clouds appear to reduce their solar reflecting capabilities, which counteracts the brightening associated with the Twomey effect itself.

2.3 Aerosol effects on the transition to precipitating clouds

The dependence of precipitation rate in marine stratocumulus clouds on N_d and h is shown in Figure 2. The strong dependence on aerosols is evident by the dependence of N_d on CCN. The relationship between CCN and N_d is approximately linear at the low concentrations characterizing the aerosol-limited regime (Martin et al. 1994, Hegg et al., 2011), where the transition from heavy to lightly or not drizzling clouds occurs (Figure 2). Deeper clouds transition at greater N_d .

Upon the transition to heavy drizzle the fast loss of cloud water can no longer be compensated by evaporation, and a net loss of cloud water from the domain occurs. The precipitation also scavenges efficiently the aerosols (Feingold et al. 1996, Wood 2006), hence reducing CCN and N_d even more, increasing r_e and causing even faster coalescence and precipitation in a positive feedback loop. In the extreme this process progresses all the way until there are insufficient CCN for sustaining the growth of new clouds. Because of the essential role of the clouds in determining the lapse rate of the marine boundary layer, the suppression of their formation due to dearth of CCN suppresses also the vertical mixing of air from sea surface to very shallow heights, thus in fact causing the collapse of the marine boundary layer to a thin layer of sea fog composed

of drizzle drops. The precipitation scavenging feedback leads in some cases to the collapse of the cloudy boundary layer (Ackerman et al., 1993) and in other cases to a deep boundary layer with open cellular convection.

This runaway feedback effect is a basis for a situation of bi-static stability (Baker and Charlson, 1990; Gerber, 1996), where once the atmosphere has reached a very clean situation the highly efficient rainout mechanisms keeps it clean until it will be overwhelmed by a strong aerosol source such as anthropogenic emissions.

The full cloud cover of closed cells is maintained by the strong radiative cooling from the cloud tops that causes top-down convection and entrainment of air from the free troposphere just above the clouds (Agee et al., 1973). This replenishes the CCN that may have lost by the cloud processes (Randall, 1980; Clarke et al., 1997; Jiang et al., 2002; Stevens et al., 2005).

A mechanism for the transition between the closed- and open-cell regimes was proposed by Rosenfeld et al. (2006). This mechanism is illustrated in Figure 3. Based on this mechanism, Rosenfeld et al. (2006) hypothesized that dynamically the closed cells are inverse Benard convection, where the cooling at the top causes polygons of sinking cool air with compensating rising air at the center of the polygons. The rising centers are manifested as patches of polygonal clouds, with narrow regions of dry downward moving air at the cell fringes (see Figures 3 and 4). The onset of heavy precipitation that occurs when the cloud top r_e exceeds $16 \mu\text{m}$, due to decrease in N_d and/or increase in h , breaks the full cloud cover by depleting the cloud water and by decoupling it from the surface due to the low level evaporation of the precipitation. With reduced cloud cover at the top of the boundary layer the radiative cooling there decreases respectively, and allows thermal radiation to be emitted upward from the vapor within the boundary layer and the lower cloud fragments. This reverses the driving of the convection, from inverse convection due to the radiative cooling at the top, to normal convection of Benard cells that is triggered by weak surface heating, where the air rises along the walls of the polygons and sinks in the centers. The rising polygons are manifested as the polygons of the clouds (see Figures 3 and 4). This picture is complicated by the evaporative cooling of the rain shafts, which form mini gust fronts at the surface that regenerate the convergence lines away from the rain cells, especially where several such fronts collide (Feingold et al., 2010). When the original rain cell decays new clouds and rain showers form at the convergence along the old gust fronts. This, in turn, produces new gust fronts and so on, leading to regular oscillations of

the locations of the low level convergence lines and the respective polygonal cloud and rain patterns.

The self organization of clouds into the three distinct regimes was described by Koren and Feingold (2011) by simple principles of prey (cloud water) and predator (rain process):

1. The non or weakly precipitating clouds, where the rain-forming process is too slow for large depletion of cloud water. This corresponds to the closed cells regime, with suppressed rain due to high aerosol concentration or a very shallow cloud with little *LWP*.
2. The heavily drizzling regime, where rain can deplete the cloud water, but the supply of new aerosols is able to replenish the cloud water after a while, so that cycles of clouds building and raining out occur. This corresponds to the regime of oscillating and raining open cells.
3. The heavily precipitating clouds, where all incipient cloud water effectively precipitates along with the aerosols on which it condensed, probably due to insufficient rate of replenishment of aerosol. This corresponds to the situation of the ultra-clean collapsed boundary layer.

The value in this highly simplified description is in elucidating these different cloud patterns as fundamentally different regimes. It is of particular importance on the background that internal processes can buffer the aerosol effects within the regimes (Stevens and Feingold, 2009), but not between the regimes. The buffering should not be regarded as a full compensation, but rather as a negative feedback that attenuates the results of the initial microphysical effect of the aerosols on the cloud microstructure. An example for the buffering in the closed cell regime is the opposite effects of aerosols increasing the cloud albedo for a given *LWP*, but decreasing the *LWP* at the same time. This is evident in the red rectangles in Figure 4, where half of the albedo changes due to N_d (Twomey effect) of -31 Wm^{-2} was offset by a decrease in *LWP* that incurred $+16 \text{ Wm}^{-2}$, leaving a net effect of -15 Wm^{-2} . An example for the buffering in the open cell precipitating regime is that an increase in aerosols would delay, but not completely shut off, the onset of rain in a convective cell, causing it to grow more, and when it eventually precipitates it would rain more.

Based on the above consideration, we have to consider the hypothesis that most of the cloud-mediated aerosol forcing is manifested by changes between cloud regimes. Such transitions are associated with change in cloud radiative effect (CRE) of the order of

100 Wm⁻², whereas the aerosol net effect within the cloud regimes are 1 – 2 orders of magnitude smaller.

It is difficult to ascribe the changes of CRE between regimes to aerosol cloud-mediated RF, because the aerosol amounts are interactive with the clouds, especially in the open and collapsed BL regimes, so that they are not independent of the cloud forms. Another major difficulty in ascribing satellite-measured aerosols to their effects on the clouds is the fact that the greatest effect occurs in the regime where $N_d < 100 \text{ cm}^{-3}$ (see Figure 2), where on average AOD is < 0.05 , which is at the low boundary of the measurement capability, and its conversion to CCN is highly uncertain (Andreae, 2009). Therefore, using the retrieved N_d instead of AOD as proxy for the CCN provides a more sensitive metric of the aerosol cloud-mediated effects on MSC. Therefore, it is argued here that assessment of the differential CRE between MSC regimes with respect to N_d captures an important element of the aerosol cloud-mediated radiative forcing. The remaining challenge will be quantifying the extents of the attribution of the regime changes to anthropogenic causes.

A case where the regime changes could be ascribed to anthropogenic aerosols from ship tracks is reproduced here from Figure 3 of Rosenfeld et al. (2006), with the added CRE, and presented here in Figure 4. It is shown for this case that the negative CRE over the closed cells is on average higher by well over 100 Wm⁻² than the adjacent open cells or collapsed boundary layer. The r_e in the closed cells of this example is smaller than 16 μm , very close to the heavy drizzle threshold of 15 μm (see Figure 2), whereas the r_e is considerably larger than 16 μm in the open cells. The appearance of heavy drizzle after cloud top r_e exceeds this threshold appears to be the main cause for opening the closed cells (e.g., Rosenfeld et al., 2006; Koren and Feingold, 2011). The cloud top r_e is determined mostly by N_d and h (Freud et al., 2011). Therefore, the combination of N_d and h is required for explaining the transitions from closed to open cells.

The ship tracks within the closed cells obviously did not incur a regime change and hence the associated change in CRE was about 10-15 Wm⁻², which is lower by an order of magnitude than the change associated with regime change.

A consistent picture emerges from the study of George and Wood (2010) who quantified the dependence of the variance in albedo over the southeastern Pacific Ocean on the variances in the controlling variables (i.e., cloud fraction, LWP and N_d). The

variability in cloud fraction, LWP and N_d explained on average roughly 1/2, 1/3 and 1/10 of the spatial variance of the area-mean albedo that was accounted for by these variables, respectively. It is interesting that despite a strong gradient in N_d within the analyzed region, N_d does not explain more than 10% in the variance of the area-mean albedo. Is it because h and hence LWP increases along with the decrease in N_d with distance from land? These results should be treated with caution, because part of this variability could be explained by meteorological factors that are correlated with the cloud fraction, LWP and N_d .

Does it mean that much, if most of the variability in the cloud RF in the southeastern Pacific is not contributed by MSC regime changes? It appears that this partition of the CRE components is not limited to areas where MSC regime changes occur frequently, because these results are in agreement with the previous global studies that separated the contributions of RF. Sekiguchi et al., (2003) showed based on AVHRR data that the N_d effect could not have contributed more than 25% of the total cloud RF over the global oceans. Kaufman et al. (2005) analyzed MODIS data over the Atlantic Ocean and showed that only 10 – 20 % of the enhanced cloud RF that was associated with increased τ_a was contributed from N_d . The dominance of cloud cover effect over ocean was also supported qualitatively by several other satellite studies (Matheson et al., 2006; Myhre et al., 2007; Menon et al., 2008). Lebsock et al. (2008) used CLOUDSAT for showing that the LWP effect dominated the Twomey effect, being positive with added τ_a in precipitating clouds and negative in non-precipitating clouds.

How much of the aerosol indirect effect on climate can be explained globally by regime changes, and how much by net radiative changes within regimes? It is possible that a large fraction occurs through the latter. Buffering (Stevens and Feingold, 2009) and cancellation (Wood, 2007) mechanisms have been shown to work within regimes, but between the regimes it is not so clear that this is the case (Koren and Feingold, 2011). A possible mechanism to communicate information that may cause some buffering between regimes pertains to the determination of the inversion height.

The two regimes have two different equilibrium states. Weakly precipitating closed cells have large inversion heights at the top of a well-mixed boundary layer and strong entrainment at the top of the inversion. Very pristine drizzling clouds or a thin layer of very low clouds in equilibrium state are topped by a very low inversion height, also defined as a "collapsed" boundary layer (Bretherton et al., 2010b). However, this does not result in a step change in PBL height at the boundary between the regions, but instead the inversion

tends to "homogenize" due to the strong buoyancy forcing at a scale in the order of at least 100 km, thus inducing a shallow secondary circulation above the PBL top (Berner et al., 2011) so that, in effect, open cell regions keep the adjacent closed cell region's PBL from deepening as fast as it would in the absence of the open cell region. From the other side, the closed cells regions keep the open cell PBL from collapsing in their vicinity. We don't yet know what the consequences of this interaction are for cloudiness, but they are likely to be important for determining AIEs associated with regime change in MSC.

These questions will have to be answered quantitatively by future research. In particular, an emphasis should be placed on the role that aerosols play in mediating regime changes in marine low clouds. This might require some experiments with controlled dispersion of aerosols into MSC.

2.4 The frequency of occurrence of aerosol-starved cloud regimes

The regime of open cells cannot inherently sustain full cloud cover, and water that does condense is depleted quickly by precipitation. Therefore, it is appropriate to describe this as a situation where scarcity of aerosol limits the cloud cover and *LWP*, i.e., an aerosol starved cloud regime (Van Zanten and Stevens, 2005; Petters et al., 2006; Sharon et al., 2006; Wood et al., 2008 and 2011). The regime of the collapsed boundary layer was not yet analyzed for its differential CRE with respect to the other regimes, but given the mechanism of its creation, it can be considered even more strongly as an aerosol starved cloud regime.

How frequent are these conditions where clouds are starved for aerosols, such that the depletion of aerosols can incur a regime change from closed to open cells with decreased radiative forcing in the order of -100 Wm^{-2} ? The addition of aerosols has been observed to close the open cells, at least in the regime of collapsed boundary layer (Christensen and Stephens, 2011). Simulations of added aerosols to open cells stopped their precipitation, but failed to convert them back to closed cells (Wang et al., 2011). The ability of aerosols to close relatively deep open cells requires additional research. Figure 5 presents global maps of the occurrence of mesoscale cellular convection, partitioned into closed cells, open cells that are organized in Benard convection, and disorganized open cellular convection. The organization of the first two regimes can be ascribed clearly to the aerosols and N_d as discussed above, but this is not obvious for the latter regime. These three regimes cover a large part of the eastern subtropical and tropical oceans. The

frequency of the open cells increases with the distance westward away from land. This occurs due to a combination of decreasing N_d (Figure 6) and increasing cloud thickness (see e.g. George and Wood 2010), the combination of which increases precipitation dramatically (Figure 6, see also Bretherton et al. 2010a). Open cells observed during the VOCALS Regional Experiment tended to be associated with aerosol-starved conditions (e.g. Wood et al. 2011), but it is not yet clear the extent to which this is the case for all open cell regions in the subtropics.

Open cells are also frequent in midlatitudes, but here they can occur due to cold advection of air (e.g. cold air outbreaks), which provide strong surface forcing in subsiding conditions which dominates the dynamics of open cells regardless of possible aerosol effects. The extent to which these open cell systems modulate their own microphysical state and become aerosol-starved is currently poorly known.

Some light can be shed on this question from the shape of the functional dependence of cloud cover on aerosol amounts, as represented by AOD. Globally, almost all of the increase in cloud cover f_c with AOD occurs at $\text{AOD} < 0.2$ (see Figure 7). For $\text{AOD} \leq 0.75$ the $\ln(f_c)/\ln(\text{AOD}) = 0.57$. This shows that the sensitivity of f_c to AOD is much greater than logarithmic at the lowest AOD, and that the behavior is consistent with the aerosol changes with the MSC regimes responsible for a large part of the dependence of f_c on AOD.

2.5 The attribution of the regime changes to anthropogenic aerosols

Open cellular convection is more frequent over the Southern Hemisphere subtropical and midlatitude oceans than over the corresponding regions of the Northern Hemisphere (Figure 5). It is interesting to ask the extent to which this might be attributable to anthropogenic aerosol influence. Mean N_d values for low clouds in polluted regions are higher than for clean regions (e.g. Quaas et al. 2009), and the ability of increased cloud droplet concentrations to keep large areas of MSC at the closed regime is evident in the observations of Goren and Rosenfeld (2012), where the large areas of closed cells are shown to have been shaped by old ship emissions. Other mechanisms that can transport aerosols from land to the remote ocean areas are pollution plumes in the free troposphere that subside in the anticyclones to the underlying MSC (Wilcox et al. 2006).

It is hypothesized that the greater amount of aerosols from the northern hemisphere continents are responsible for the hemispheric differences in open cell

frequency, but more understanding of factors controlling this frequency, including the large-scale meteorology, is required to test this hypothesis. If the reduction in open cells is a manifestation of the added anthropogenic aerosols it implies a huge negative radiative forcing, because the differential RF between the closed and open cells can exceed 100 Wm^{-2} (see Figure 4).

2.6 The possible underestimation of the radiative forcing via low clouds

As we have discussed in Section 2.3, it is possible that the Twomey effect is 1/4 the overall AIE from low clouds, or less (Sekiguchi et al., 2003; Kaufman et al., 2005; Lebsock et al., 2008). Yet, the IPCC AR4 found a cloud drop radius effect of -0.7 Wm^{-2} with the large uncertainty range of -0.3 to -1.8 Wm^{-2} . If indeed the cloud-cover effect is much larger than the clouddrop-radius effect, the AR4 range has to be increased by a large factor to account for other effects. Even if not all cloud types respond in the same way as our example of MSC, we face the possibility of a very large and highly uncertain net forcing from low clouds, especially once adjustments involving dynamics occur.

This should be contrasted with the inverse calculations showing that the overall net cloud-mediated RF should likely be even lower than the IPCC-estimated albedo effect alone (See Section 1). To resolve this apparent contradiction, there are two likely possibilities:

1. The aerosols that are involved in regime changes and the respective RF are predominantly natural, or,
2. Most of the strong negative RF is balanced by another similarly strong positive RF, particularly by anthropogenic aerosols interacting with deep and high clouds.

While at least part of the aerosols involved in the regime changes are natural, based on some of the evidence presented here, we cannot discard the second possibility, especially in view of its far-reaching consequences. The second possibility, that a strong negative RF from less-studied cloud types is the positive RF, is explored next.

3. Aerosol induced radiative forcing by deep convective clouds

If indeed the forcing of low-level cloud is large to the extent that the climate should have been cooling, the constraints described in Section 1 would be difficult if not impossible to satisfy without a similarly large positive radiative forcing to balance most of this cooling effect. We hypothesize here that such a positive forcing may indeed exist, in the form of aerosol effects on deep and/or high clouds, through several possible mechanisms that are presented in this section.

3.1 Aerosol invigoration of deep clouds in warm and moist atmosphere

Most of the condensed cloud water in deep tropical convective clouds in pristine air masses is precipitated as warm rain before reaching the freezing level. Adding CCN to the clouds causes N_d to increase, and respectively the height for onset of warm rain to increase as well. This effect was quantified in several aircraft field campaigns in the Amazon tropical clouds (Andreae et al., 2004), Argentina hail storms (Rosenfeld et al., 2006), California winter storms (Rosenfeld et al., 2008), Israel winter clouds and India summer monsoon clouds (Freud and Rosenfeld, 2012). As seen for the case of MSC (Figure 2), and for the same fundamental physical reasons, the number of activated cloud drops near cloud base scales linearly with the cloud depth required to grow droplets to the threshold r_e of $\sim 14 \mu\text{m}$ for rain initiation in deep convective clouds (Freud et al., 2011; Freud and Rosenfeld, 2012). Increasing the number of activated aerosols by 100 cm^{-3} increases h for the onset of rain by $\sim 280 \text{ m}$. Therefore, in deep tropical clouds with freezing level of 3-4 km above cloud base, an adiabatic concentration of nearly 1000 drops cm^{-3} would delay the onset of precipitation to above the freezing level, thus preventing warm rain formation. Observations from the Amazon (Andreae et al., 2004) and India (Freud and Rosenfeld, 2012; Konwar et al., 2012) support this conclusion.

It has been hypothesized (e.g., Khain et al. 2004, Rosenfeld et al., 2008) that delaying the precipitation to above the freezing level would cause the cloud water to freeze first onto ice hydrometeors and so release the latent heat of freezing, which would not have been realized had rain at lower levels not been prevented by the aerosols (see illustration at Figure 8). The released added latent heat adds buoyancy to the cloud, increases the updraft speed, and causes the cloud top to grow higher and the anvil to expand over a larger area. The melting of the ice hydrometeors while falling cools lower levels, with a net result of more low-level cooling and high-level warming for the same surface rainfall amount. This means consumption of more static gravitational energy and

its conversion into respectively more kinetic energy, which is the essence of the invigoration of the storm. The invigoration, along with enhanced ice precipitation processes, enhance also the cloud electrification (Molinie and Pontikis, 1995; Williams et al., 2002; Andreae et al., 2004; Rosenfeld et al., 2008). Set against this possibility, however, is the added gravitational loading of the retained condensate. The net result of these competing factors is not obvious a priori.

Cloud simulation studies have generally confirmed the net invigoration hypothesis for deep warm- base clouds with weak wind shear in moist environments. For other conditions no invigoration was obtained, and for cool-base clouds, dry environment and/or strong wind shear the precipitation amount was even decreased (Khain and Pokrovsky, 2004; Khain et al., 2004, 2005 and 2008a; Wang, 2005; Seifert and Beheng, 2006; van den Heever et al., 2006; Fan et al., 2007, 2009 and 2012). In some of the simulations, the greater low-level evaporative cooling of the enhanced rainfall produced stronger gust fronts that triggered more new clouds and invigorated them (Tao et al., 2007; Lee et al, 2010). Morrison and Grabowski (2011) argue that invigoration is counteracted in radiative-convective equilibrium by large-scale feedbacks, but clouds still become deeper.

Satellite observations using MODIS showed deeper and more expansive convective clouds associated with greater aerosol optical depth over the tropical Atlantic Ocean (Koren et al., 2005, 2010a and 2010b). The reality of such associations has been questioned due to possible errors in the retrieved aerosols due to cloud contamination and other artifacts that are caused by the proximity to the clouds, but Koren et al. (2010a) showed that this was not the cause in a study of the North Atlantic region, because the cloud invigoration was detected with a similar magnitude when comparing the retrieved cloud properties to the results of an aerosol transport model. They also partitioned their analysis to different meteorological conditions that control the depth of the convection, and still found the aerosol invigoration effect having a similar magnitude for the different meteorological partitions. However, the average measured cloud top height in the study of Koren et al. (2010a) was only about 3 km, which is well below the height of an anvil cloud. A more recent study that tested the aerosol invigoration only on the anvils (Massie et al., 2012) showed that the increase in height per unit AOD is 2 to 10 times smaller than compared to the previous calculations by Koren et al. (2005, 2010a and 2010b). This apparent discrepancy might be resolved when noting that the anvils are already capped from above by the tropopause, so that invigoration is not likely to make them much taller

and colder. The invigoration is more likely to increase the top height of the lower clouds, and/or expand the areas of the anvils.

The radiative effects of the aerosols reduce the solar radiation reaching the surface and therefore act to suppress the convection, working against the aerosol invigoration effect, at least on land. Therefore the aerosol effect is not monotonic, such that the invigoration effect was calculated by Rosenfeld et al. (2008) to reach a maximum at AOD of ~ 0.3 . This was confirmed observationally over the Amazon (Koren et al. 2008, Ten Hoeve et al. 2012). Satellite measurements reported for the Amazon region by Lin et al. (2006), also showed that total rainfall and cloud heights increased on average with AOD and that the effects weakened above an AOD near 0.3, although these observations did not specifically target deep convective clouds.

Anvil clouds associated with deep convection exert a substantial longwave and shortwave cloud forcing, and the longwave component dominates in clouds that are optically thin, including the cirrus clouds produced by the anvil. Aerosol-induced changes in anvil clouds associated with deep convection and more distant cirriform clouds whose ice is partly supplied by convective detrainment can therefore act as warming mechanisms. Lee et al. (2009) found in a deep-convection simulation that 28% of the increased shortwave cloud forcing (cooling) associated with higher aerosol concentrations was offset by increased longwave cloud forcing (warming). The corresponding offset for stratocumulus clouds was only 2-5%.

Critical supporting observational evidence to the validity of the invigoration hypothesis was obtained very recently, where volcanic aerosols, whose variability was completely independent on meteorology, were observed to invigorate deep convective clouds over the northwest Pacific Ocean and more than double the lightning activity (Yuan et al., 2011; Langenberg, 2011). This lends credibility to the suggestion of Zhang et al. (2007) that the trend of increasing emissions of air pollution from East Asia caused their observed trend of increasing deep convection and intensification of the storm track at the North Pacific Ocean.

The aerosol-induced invigoration on the peripheral clouds of tropical cyclones was hypothesized to occur at the expense of the converging air to the eye wall, and hence decrease maximum wind speeds (Rosenfeld et al., 2007). This aerosol effect was simulated extensively (Rosenfeld et al., 2007; Cotton et al., 2007; Khain et al., 2008b, 2010 and 2011; Zhang et al., 2007 and 2009). The variability in aerosols was also observed to explain about 8% of the variability in the intensity of Atlantic hurricanes

(Rosenfeld et al., 2011a). The aerosol effects on the microphysics and intensity of tropical cyclones are reviewed in Rosenfeld et al. (2012).

A weekly cycle in the anthropogenic aerosols, peaking during mid-week, was shown to be associated with a similar cycle in the rain intensity and cloud top heights (Bell et al., 2008), on the lightning frequency (Bell et al., 2009), and even on the probability of severe convective storms that produce large hail and tornadoes (Rosenfeld and Bell, 2011) in the eastern USA during summer. These findings are supported by a recent study analyzing 10 years of surface measurements of clouds and aerosols over the ARM site at the Southern Great Plains in Oklahoma, showing clearly the cloud invigoration effect, associated with decreasing probabilities of light rain matched by similar increasing probability for heavy rain (Li et al., 2011). It is important to note that the invigoration effect is anticipated mainly in the intensity of rain events or vigor of convection, and not necessarily in the average rainfall (e.g., Morrison et al. 2010), which may explain why studies examining total rainfall (e.g., Tuttle and Carbone, 2011) sometimes do not find it.

All these findings, and especially the long-term measurements of Li et al. (2011), suggest that the aerosol invigoration is a robust effect in the atmosphere. The aerosol invigoration of deep convective clouds could exert a cloud-modulated radiative forcing in several ways, as illustrated in Figure 9:

- Brightening of the clouds at a fixed cloud top, increasing their albedo and cooling effect. However, for already thick convective cloud, where the albedo effect is nearly saturated, the negative RF is expected to be rather small.
- Higher cloud tops, which emit less thermal radiation to space and hence induce a warming effect.
- More extensive anvils and/or more semi transparent ice clouds. Such cirrus clouds have small albedo in the visible, but still have large emissivity in the thermal IR, thus causing a strong positive RF. They could also reduce the radiative cooling and air subsidence rates in the upper troposphere, which would increase relative humidity and therefore the atmospheric greenhouse effect.
- Deep convection could more frequently reach the lower stratosphere adding more water vapor to the stratosphere which increases the greenhouse effect.

3.2 Aerosols enhancing detrainment of ice and vapor in the UTLS

Aerosols may well enhance the amount of ice contained in and detrained from anvils into the upper troposphere and lower stratosphere (UTLS), even without having any dynamic effects (e.g., invigoration) on the clouds. Analyzing deep convective clouds in satellite data, Sherwood (2002a) and Jiang et al. (2009b, 2011) found that biomass burning aerosols were associated with smaller ice particle r_e at the anvils of tropical deep convective clouds. These storms also were more intense, as indicated by their colder cloud tops, though these studies found that proxies for intensity were too small to explain the smaller r_e . The effect could be due to CCN nucleating small cloud drops that freeze homogeneously into respectively small ice particles (e.g., Phillips et al. 2002), or (in part) due to invigoration of the storms activating more aerosols aloft, or to meteorological factors not accounted for in those studies. The clouds with smaller ice particle r_e produce significantly more lightning, supporting the hypothesis that aerosols played a role in reducing the r_e of the ice particle (Sherwood et al., 2006). Satellite measurements of pyro-cumulonimbus showed that the extreme CCN concentrations in the dense smoke keep the cloud drops extremely small up to the homogeneous ice nucleation level, where they become similarly small ($r_e \sim 10 \mu\text{m}$) ice particles, whereas ice in the ambient clouds formed mostly by mixed-phase processes producing particles in the anvils with $r_e > 30 \mu\text{m}$ (Rosenfeld et al., 2007). Tracking the life cycle of such anvils showed that they lived twice as long as anvils from ambient clouds and expanded to much larger areas (Lindsey and Fromm, 2008). This is likely due to the smaller fall speeds and/or slower aggregation of the particles.

Aircraft measurements and model simulations show that aerosols from Africa indeed nucleate small cloud drops aloft that freeze homogeneously into small ice crystals in the anvils of clouds over southern Florida (Fridlind et al., 2004). In simulating this process, Jensen and Ackerman (2006) showed that the detrainment of small ice crystals was responsible for creating long-lived cirrus clouds. The simulations of deep tropical clouds by Fan et al. (2010) show that added CCN can lead to such enhancement of small ice particles in the anvils, and nearly double the extent of the resulting clouds; similar results were obtained by Morrison and Grabowski (2011).

A cloud-system resolving model simulation of the aerosol effect at a regional scale with bin microphysics for tropical and midlatitude summertime convective cloud situations (Fan et al., 2012) found invigoration in the tropical case with weak wind shear, but not with strong wind shear. However, the positive RF from the anvil expansion with added CCN dominated the negative RF due to cloud brightening in both cases, as shown in Figure

10a. In the temperate case the net RF were weaker and of opposite signs for the different wind shears (Figure 10b).

Another possible pathway for aerosol indirect effects is through altering stratospheric water vapour, a strong greenhouse gas. Solomon et al. (2010) found that decadal variability in lower stratospheric water vapor was contributing to decadal climate variability, following previous calculations showing that increases in stratospheric water vapor over the latter part of the 20th century contributed a radiative climate forcing of order 0.2 W m^{-2} (Forster and Shine, 1999 and 2002, Myhre et al. 2007). While the decadal humidity variations can largely be explained by those of tropopause temperature through a simple freeze-drying model (e.g., Fueglistaler et al. 2010), radiosonde data do not show a longer-term warming trend, and the source of the moistening trend is still unknown. The radiative forcing is significantly larger than accounted for by the IPCC in 2007, which only included the part attributable to methane oxidation.

Two plausible mechanisms have been suggested linking this trend to anthropogenic aerosols. First, smaller ice particles lofted in polluted storms could cause overshooting clouds to re-evaporate more quickly when mixing with dry stratospheric air, delivering more water vapor to levels where it can reach the lower stratosphere as shown by satellite and in-situ observations and simulated by models (Sherwood 2002b, Chen and Yin 2011, P. K. Wang et al. 2011, Nielsen et al. 2011). Back-of-the-envelope calculations suggest this mechanism, which is observed to affect stratospheric humidity independently of tropopause temperature, could account for the observed trend since 1950 even discounting any invigoration effect (Sherwood 2002b), but this has not been comprehensively modeled; isotopic data do not suggest any trend in ice re-evaporation since 1991 (Notholt et al. 2010) but most of the humidity trend occurred before 1991. A similar microphysical effect from ice nuclei could also occur for cirrus clouds formed near the tropopause (Notholt et al 2005). The second possibility is suggested by observations (Su et al., 2011; Wu et al., 2011) and models (Liu et al. 2009) indicating that pollution particles lofted in deep convection elevate cirrus cloud temperatures and water vapor mixing ratios, which would increase water transport into the stratosphere (Liu et al. 2009). Observations do not show a corresponding temperature trend since 1958, but this could be due to biased trends in the radiosondes which are difficult to correct (JS Wang et al. 2012).

In summary, aerosols probably exert a second indirect warming effect through lower stratospheric water vapor, and this could be of nontrivial magnitude.

4. Aerosol induced radiative forcing by supercooled layer clouds

New satellite remote sensing data, especially from active sensors, are revealing that supercooled layer clouds are more common than previously suspected (Hogan et al. 2004, Zhang et al. 2010, Hu et al. 2010, E. Morrison et al., 2011). Almost all clouds with tops warmer than -20°C contain supercooled liquid water (Hu et al. 2010), and supercooled liquid can exist in many stratiform clouds with temperatures down to -30°C (Hu et al. 2010, Shupe et al. 2007). In the midlatitude storm tracks and high latitudes supercooled liquid layers can occur 10-15% of the time (Zhang et al. 2010), making this a climatologically important category of clouds.

Often supercooled layer clouds, both at low and mid troposphere, are maintained by radiative cooling at their tops that induces inverse convection in a layer within a stable atmosphere, similar to the mechanism that sustains decks of marine stratocumulus. What is particularly remarkable about such clouds is their apparent sensitivity to small changes in ice nuclei (IN) ingested (e.g., Prenni et al. 2007, H. Morrison et al. 2011), with the result of increased IN being a rapid glaciation, the loss of liquid condensate and a suppression of longwave cooling that weakens the turbulent mixing sustaining the supercooled layer (H. Morrison et al. 2011). Thus, the hypothesis has been raised in recent studies that Arctic clouds exhibit bistability: they consist either of turbulent supercooled layers with minimal ice, or rarefied clouds containing only ice particles (Morrison et al. 2011, 2012). Because the ice-only clouds tend to be optically thin (perhaps even just diamond snow under some circumstances), such bistability could permit a particularly strong aerosol cloud-mediated radiative forcing. In addition to IN-mediated impacts, observational data indicate that a CCN-starved regime is often present in the Arctic over sea ice (Mauritsen et al. 2011). This change between the two stable regimes of supercooled water and ice clouds is illustrated in Figure 11.

Adding CCN to supercooled layer clouds may delay their transition to glaciated clouds for a given IN concentration. With more CCN cloud drops are smaller and less likely to accrete to ice crystals or coalesce and precipitate, leading to smaller loss of

cloud water. In such case, added CCN would have a negative forcing, whereas added IN have a positive forcing.

The remarkable sensitivity of supercooled layer clouds seems at odds with their frequent occurrence. This has led to the search for mechanisms limiting the formation of ice in these clouds. It appears that most of the IN in supercooled layer clouds, particularly in the Arctic, arrive via long-range transport in the freetroposphere. They enter the cloud by subsidence and are then entrained into the cloud. This process does not supply IN at a particularly high rate, with typical replenishment timescales of several days after entrainment removal (Fridlind et al. 2012). This results in a slower rate of IN activation than would occur if all the free-tropospheric IN were activated at once, and limits the desiccation of supercooled liquid clouds.

Similarly, the effect of aerosols on cirrus clouds could result in either a positive or negative forcing on climate. GCM simulations of the effect of added IN show a reduction in cirrus water content and optical thickness, and sometimes a reduction in relative humidity, producing a negative forcing of uncertain magnitude (Penner et al. 2009, Hendricks et al. 2011). Opposing this, the introduction of CCN aerosol can enhance cirrus coverage; the radiative forcing of this is fairly small for aircraft emissions (Burkhardt and Karcher, 2011) but could be larger when including all anthropogenic aerosol. As already stated, such calculations should be regarded as provisional at this time.

There are still major unresolved questions. Chief among these is that ice-nucleating mechanisms of aerosols are poorly understood. In some supercooled liquid clouds the concentration of ice crystals appears to be significantly greater than the IN concentration (Fridlind et al. 2007). We do not yet fully understand how pollution aerosols affect the IN concentration in the Arctic and midlatitude freetroposphere where supercooled clouds are common (McFarquhar et al. 2011). Aircraft observations suggest that high ice crystal concentrations in supercooled shallow cumulus in maritime polar airmasses tended to occur in the presence of large drizzle drops (Rangno and Hobbs 1991). This suggests a possibility whereby CCN availability might be important for ice formation. At this stage, it would be difficult to attribute even a sign to the potential aerosol cloud-mediated effects on supercooled water clouds, but there nevertheless exist important possibilities that warrant further exploration.

5. Discussion and Implications for GCMs

Based on the previous section, AIE on deep convective clouds appear to induce positive radiative forcing of yet unknown global magnitude by invigorating clouds, expanding their anvils, and enriching the lower stratosphere with water vapor. Air pollution aerosols were also observed to glaciate mid- and upper-tropospheric supercooled clouds (Rosenfeld et al., 2011b), and thus adding positive radiative forcing.

This compensates to an unknown extent the negative forcing due to the AIE on low clouds. Even if the net effect is very small on a global average, the cooling occurs mainly over the subtropical highs and migratory anticyclones over ocean, whereas the warming occurs mainly at the areas of deep tropical convection. The spatial separation can propel atmospheric circulation systems that would modify the weather patterns. GCMs do not yet treat AIEs in both deep convection and shallow clouds comprehensively enough to ascertain the nature of these changes, but studies focusing on direct effects of aerosols and/or indirect effects on shallow clouds suggest aerosol-induced circulation changes are possible in the tropical Atlantic climate (Chang et al., 2011), Sahel rainfall (Ackerley et al., 2011), south Asian monsoon circulations (Bollasina et al., 2011), the Hadley circulation (Ming and Ramaswamy, 2011; Allen et al., 2012), North Atlantic (Booth et al., 2012), and the boreal winter extra-tropical circulation (Ming et al., 2011).

As noted elsewhere in this paper, observational and process studies suggest that aerosols and clouds interact through a range of radiative, microphysical, thermodynamic, and dynamic mechanisms. With increasing aerosol concentrations, these mechanisms all recognize an initial response taking the form of smaller cloud particles, delayed precipitation formation, and larger water contents. The instantaneous radiative forcing is comprised of increased shortwave reflection (cooling) and increased longwave emission (possible warming from high clouds) and can be described as a radiative indirect effect. Several subsequent competing mechanisms resulting from smaller cloud particles, delayed precipitation formation, and larger water contents are possible. In the absence of mechanisms responding to larger water contents, cloud lifetimes and areas increase, enhancing the instantaneous radiative forcing (included in “adjusted” radiative forcing). Numerous counter-acting mechanisms have been identified. Increased water contents near cloud top enable evaporation resulting from entrained dry air to break up clouds, reducing water content, cloud lifetime, and cloud areas. The “adjusted” radiative forcing by

this mechanism opposes that described above. Increased water content near cloud top can enhance radiative cooling and generate instabilities, leading to a similar set of consequences. Increased water content can also lead to changes in the heights and thicknesses of clouds. Changes in the sizes of drizzle particles below cloud base can change evaporation and stability below cloud base. In some cases, aerosol-induced changes can alter the cloud regime, changing significantly cloud areas and lifetimes. Microphysical changes in deep convection can change distributions of latent heating and induce evaporatively driven downdrafts, increasing the intensity of convection. Effects related to ice nucleation are likely, and absorbing aerosols can heat the atmosphere around clouds, altering clouds in what is referred to as a semi-direct effect.

While observational and process studies suggest this wide range of cloud-aerosol interactions capable of both warming and cooling the earth-atmosphere system, scaling these interactions to global scale and inferring their impacts on climate and climate change requires synthesis provided by climate models. On the other hand, state-of-the-science atmospheric general circulation models (GCMs) treat processes relevant for cloud-aerosol interactions in a highly simplified manner, limiting the confidence with which conclusions can be drawn.

Quaas et al. (2009) compared ten GCMs which treat cloud-aerosol interactions with satellite observations. All of the GCMs in that study, as well as those summarized in Isaksen et al. (2009), are cooled by their cloud-aerosol interactions. To the extent underlying relationships between clouds and aerosols in GCMs can be evaluated using satellite observations, present-day positive relationships between aerosol optical depths and cloud liquid in GCMs seem to be too strong, while positive relationships between aerosols and drop number are comparatively well simulated (Quaas et al. 2009). Penner et al. (2011) note that GCMs suggest present-day relationships between cloud and aerosol properties may differ from their pre-industrial counterparts, with the latter stronger than the former. Quaas et al. (2009) had noted that present-day aerosol optical depths and their variations with cloud properties are related in GCMs to AIEs between pre-industrial and present-day climates in those GCMs. By replacing the modeled aerosol optical depths and their variations with cloud properties with the corresponding satellite observations, they infer GCM AIEs are larger than would be consistent with satellite observations. Quaas et al. (2009) also found most GCMs had difficulty simulating reductions in cloud-top temperature with increasing aerosol optical depth, especially over oceans, consistent with the absence of interactions between deep convection and aerosols in most GCMs.

The complexity with which GCMs treat aerosol processes varies widely, from empirical methods relating aerosol concentrations to drop number (e.g., Lin and Leaitch, 1997) to physically based methods using aerosol activation theory (e.g., Abdul-Razzak and Ghan, 2000; Ming et al., 2007). Aerosol size distributions are specified (e.g., in terms of aerosol concentration, Donner et al. (2011)) in some models but calculated from prognostic aerosol modal equations (e.g., Liu et al., 2011) in others.

The chief limitation in GCM representations of aerosol-cloud interactions arises from simplifications in their cloud macrophysics (the processes governing the environments for activating cloud liquid and ice particles and their subsequent microphysical evolution) and the absence of aerosol interactions with deep convection in most GCMs. GCM cloud macrophysics also dominates the interactions between radiation, microphysics, thermodynamics, and dynamics; these interactions are quite restricted in current GCM macrophysics relative to the interactions identified by process studies. As an example, in GFDL CM3, a normal distribution whose variance is related to large-scale eddy diffusivity is used to characterize the small-scale variations in vertical velocity, which is a major control on aerosol activation (Golaz et al. 2011). CM3 treats cloud-aerosol interactions only in stratiform and shallow cumulus clouds. CM3 macrophysics can straightforwardly capture microphysics interactions which increase cloud water paths as aerosol concentrations increase but is much less able to represent processes discussed in the preceding paragraph in which increasing aerosol concentrations could reduce water paths. Indeed, GFDL CM3 exhibits an annual global-mean temperature increase of 0.32°C between the period from 1980 to 2000 and the period from 1880 to 1920 (Donner et al., 2011). The corresponding increase for GFDL CM2.1, which does not include cloud-aerosol interactions, is 0.66°C (Knutson et al., 2006). Observed estimates of this difference from the Climate Research Unit (Brohan et al. 2006) and the Goddard Institute for Space Studies (<http://data.giss.nasa.gov/gistemp/tabledata/GLB.Ts+dSST.txt>) are 0.56°C and 0.52°C, respectively. Changes other than incorporation of cloud-aerosol interactions between CM2.1 and CM3 preclude attributing the change in temperature increase solely to these interactions. Six of the ten models analyzed in Quaas et al. (2009) impose lower limits on cloud drop number concentration, which arbitrarily restricts cooling by cloud-aerosol interactions. An important research priority is for GCMs to improve their parameterization of aspects of cloud-aerosol interactions which are poorly represented currently, many of which limit cooling by aerosols.

The simulation of temperature increases in climate models between pre-industrial and present times depends on their adjusted forcings, climate sensitivities, and transient climate responses. Since climate sensitivity is not known, the extent to which a climate model (e.g., CM3) simulates this temperature increase would not strongly constrain the adjusted forcing due to anthropogenic cloud-aerosol interactions, even if greenhouse gas forcing and aerosol direct forcing were known. Related to the latter, it is important that climate models simulate aerosol distributions and properties realistically. Global observation networks for aerosols and surface downward shortwave radiative fluxes are available for evaluating climate models, e.g., as in Donner et al. (2011).

Advanced cloud macrophysics parameterizations offer a prospect for improving representation of cloud-aerosol interactions in climate models. For example, Guo et al. (2010) show that a parameterization using multi-variate probability distribution functions for vertical velocity, liquid water potential temperature, and total water mixing ratio can capture a range of responses of liquid water path to increasing aerosol concentrations. Guo et al. (2011) find that a key mechanism in these responses is cloud entrainment, as discussed above and modeled by large-eddy simulation. These methods to date have been used successfully in simulating single columns in field experiments. Incorporating them in climate models is an ongoing activity, e.g., at GFDL and NCAR. Droplet activation and ice nucleation in deep convection depends on vertical velocities therein. Since most GCMs parameterize deep cumulus convection in terms of mass flux only, they are not able to represent the interactions between deep convection and aerosols described elsewhere in this paper. Examples of promising prospective developments include the use of deep cumulus parameterizations based on ensembles of cumulus clouds with vertical velocities in GFDL CM3 (Donner, 1993; Donner et al., 2001), the use of double-moment microphysics in deep convection in experimental versions of GFDL AM3 (Salzmann et al. 2010), the NCAR Community Atmosphere Model (Song and Zhang 2011), and the ECHAM5-HAM model (Lohmann, 2008).

In summary, assessing the role of cloud-aerosol interactions in the climate system requires studying these interactions in climate models to integrate them to global scales. Current macrophysical aspects of cloud-aerosol interactions in climate models remain rudimentary, however, with process studies suggesting a more nuanced picture of these processes than encompassed by current GCM parameterizations. In particular, a number of processes which may limit cooling by cloud-aerosol interactions are not well

parameterized at present. High priority should be given to addressing the challenge of more realistically representing cloud-aerosol interactions in climate models.

6. What should we do next?

A key obstacle to better understanding aerosol indirect effects is our poor ability to model cloud macrophysics. As noted in Section 5, high priority should be given to improving the realism with which cloud macrophysical processes governing cloud-aerosol interactions are represented in GCMs. Only recently have physically based approaches to aerosol activation been used in GCMs, and their usefulness is limited by incomplete representations of the full set of processes which govern cloud-aerosol interactions in GCMs and by the lack of resolution at the cloud scale. New approaches to parameterizing cloud macrophysics for both shallow and deep cloud systems are emerging. Evaluating and further developing these parameterizations will require extensive collaboration between GCM developers and scientists studying cloud macrophysics using process models, large-eddy and cloud-system simulation, and field observations. Satellite observations will also be critical in assessing cloud-aerosol interactions on a global scale.

More realistic physics has to be parameterized into both cloud resolving and global circulation models, and their results need to be validated against actual observations. A limiting factor in the present Earth observations is the ability to separate the aerosol from thermodynamic and meteorological effects. Doing so requires measuring of the CCN and cloud microphysical, thermodynamic and dynamic properties simultaneously from space at the necessary spatial and vertical resolution, which is in the order of 50 – 100 m. This requires a new generation of satellites with multi-spectral and multi-angle sensors. A way to do that is described by Rosenfeld et al. (2012) in a proposed satellite mission. High resolution multi-angle imager (as in MISR) will be able to map the topography of the cloud surfaces and their vertical motions. A multi-spectral imager can map the microstructure and temperature of the cloud surfaces at various heights above cloud base, which will allow retrieving N_a from the vertical evolution of N_d in convective elements (Freud et al., 2011). The vertical development of the cloud surface just above its base will provide a measure of cloud-base updraft, which when combined with N_a yields the supersaturation and the CCN concentrations. Multi-angular near-infrared observations can also provide

information on ice particle habit and microphysical history not obtainable at visible wavelengths (Sherwood, 2005). Such a mission does not represent a major technological challenge, but requires the recognition to be of high priority in addressing the large uncertainties in RF that are the subject of this paper.

Field campaigns are necessary for performing case studies of simultaneous measurements of the CCN and cloud microphysical, thermodynamic and dynamic properties in a way that will allow reaching closure of the aerosol, water and energy budgets, at a scale of a box of several hundred km on the side. This needs to be done both in the shallow and deep clouds, as much as possible in similar meteorological conditions but with very different aerosols. The outlines for such campaigns are given by Andreae et al. (2009).

7. Summary

The aerosol indirect effect on radiative forcing (AIE) is the main source of uncertainty in the overall anthropogenic climate forcing and climate sensitivity. The uncertainties are summarized in Table 1. The AIE can be generally divided into negative forcing from low clouds, which is at least partially countered by positive forcing from deep and high clouds and by the IN effects on glaciating supercooled water clouds. The quantification of both opposite and possibly large effects is highly uncertain, to the extent that even the sign of the overall net effect cannot be determined with any degree of certainty.

Aerosols added to low clouds generally incur negative radiative forcing, because they can cause cloud brightening by three main mechanisms: redistributing the water in larger number of smaller drops; adding more cloud water, and increasing the cloud cover.

Aerosols affect these components some times in harmony and quite often in opposite ways that cancel each other at least partially. These processes can be highly non-linear, especially in precipitating clouds that added aerosol can inhibit from raining. This amounts to behavior of little overall sensitivity in most of the clouds, and hyper-sensitivity in some of the clouds where the processes become highly non linear with positive feedbacks, leading to very complicated and uneven AIE. Present observations assume a logarithmic relation between aerosol amount and cloud response. This hides the physics of much more complicated behavior, whose state-of-the-art understanding is described in this paper. Key processes that are involved in the AIE are the precipitation-forming processes and the

response of the cloud properties to the precipitation, which have profound impacts on the clouds and their environment. Some of these impacts are the formation of downdrafts and cold pools that alter the dynamics of the clouds, change the vertical diabatic heating profiles and the atmospheric instability, and scavenging the aerosols that affect the clouds at the first place. Process models at high resolution (LES) have reached very recently to the development stage that they can capture much of this complicated behavior, but the implementation into a GCM has been rudimentary due to severe computational limitations and the present state of cloud and aerosol parameterizations in GCMs. The latter deficiencies are an active research area at present

Aerosols added to deep clouds generally incur positive radiative forcing, where to the effects that are operative in low clouds (cloud drop size, cloud water path and cloud cover) are added the effects of cloud top cooling, expanding, and detraining vapor to the upper troposphere and lower stratosphere. The latter three factors generally incur positive radiative forcing. The level of scientific understanding of the AIE on deep clouds is even lower than for the shallow clouds, as the deep clouds are much more complicated, where mixed phase and ice processes play an important role. Process models still have a major void in the knowledge in mixed-phase and ice processes, for both layer and deep convective clouds, both low and high level, in the arctic and lower latitudes. Respectively, the parameterization of these processes for GCMs is further away than for the low clouds.

Future efforts must address the AIE of both shallow and deep clouds for obtaining the net effect, which is required so much for quantifying the anthropogenic climate forcing, climate sensitivity and climate predictions. Furthermore, the cooling occurs mainly over the subtropical highs and migratory anticyclones over ocean, whereas the warming occurs mainly at the areas of deep tropical convection. The spatial separation can propel atmospheric circulation systems that would modify the weather patterns at all scales and the hydrological cycle. Therefore, the AIE must be quantified correctly not only for understanding climate, but also for improving weather and precipitation forecasts.

As a limiting factor in our understanding and quantification of the weather-forming processes and its integration into the climate system, we recommend coordinated field campaigns and satellite missions for addressing this problem, with the objective to describe and parameterize correctly these complex processes, and to measure these

processes from space and quantify their effects at a global coverage and climate time scales. Present day satellite missions (CLOUDSAT, CALIPSO, GPM) focus at measuring the precipitation and large cloud particles and aerosols, but lack the critical measurements of CCN and detailed cloud microstructure. An example of a proposed satellite mission that is being designed to address the issues described here is given by Rosenfeld et al. (2012). An example of the concept of field campaigns that are designed to address is issues is given by the Aerosols-Clouds-Precipitation-Climate initiative (Andreae et al., 2009), which provides the template for the design of a closure box experiment for quantifying all the energy and mass fluxes within a region of several hundred km on the side. The recommendations are summarized in Table 2.

This position paper can be summarized in the following points:

1. While many of the clouds have little sensitivity, some of the clouds are hyper-sensitive, especially when the mechanism of regime change is involved.
2. The sign of the effects are of opposite signs for different kinds of clouds and aerosols.
3. We have little quantitative knowledge on the AIE of any of these cloud and aerosol types.
4. We have even much less knowledge on the combined effect, even as far as its sign
5. We propose certain ways to address it.

Finally, we have shown here that the recently acquired additional knowledge actually increased the uncertainty bar in the chart of the radiative forcing, while everyone strives to reduce it. How large is this uncertainty? Do we know now all what we should know that we don't know yet? When we will be there the uncertainty range will peak, and start to be reduced from there on.

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Table 1: Aerosol Cloud-Mediated Radiative Forcing: Status of Current Understanding

Process	Current Understanding
Activation of liquid droplets	For aerosols with known solubility properties and size distributions, understanding is well-established.
Primary nucleation of ice crystals	Although some ice nuclei have been identified, significant uncertainty remains as to the nucleating abilities of black carbon, biogenics, and mixtures.
Aerosol size distributions for cloud condensation and ice nuclei	Size distributions can be modeled reasonably accurately in detailed process models, but considerable simplifications, the consequences of which are not fully understood, are required for computational efficiency in GCMs.
Aerosol-induced changes in cloud regimes and organization	Conceptual and numerical models have identified basic issues. Field and satellite observations have been limited and will remain so in the absence of simultaneous characterization of dynamics, microphysics, and aerosols, enabling closure of aerosol budgets. GCM parameterizations have not explicitly been developed, and capabilities of current GCM parameterizations to capture these changes are likely to be severely limited.
Aerosol-induced changes in cloud entrainment, dynamics, and microphysics	Large-eddy simulations with advanced microphysics have identified key issues. Observations have been limited. Current GCM parameterizations are very limited regarding these processes, but multi-variate probability distribution functions with dynamics have been able to capture entrainment-aerosol interactions.
Aerosol-induced changes in dynamics, radiation, and microphysics of deep convection	Cloud-system models have identified basic processes. Observations have been limited. Most GCM cumulus parameterizations lack the physical basis to simulate these processes, but GCM cumulus parameterizations with vertical velocities and advanced microphysics have recently been developed.

Table 2: Aerosol Cloud-Mediated Radiative Forcing: Key Uncertainties and Recommendations for Increased Understanding

Process	Issues and Uncertainties	Recommendations
Activation of liquid drops	Solubility properties of organics	Characterization and lab studies
Primary nucleation of ice Crystals	Identification of ice nuclei, especially roles of biogenics	Field and lab studies of nuclei candidates
Aerosol size distributions for cloud condensation and ice nuclei	Accurate, computationally efficient parameterizations, especially for GCMs	Interactions between process-level studies and GCM development
Aerosol-induced changes in cloud regimes and organization	Process-level understanding, parameterization for GCMs	Model development; analysis of satellite and field observations to evaluate conceptual models, numerical models, and parameterizations.
Aerosol-induced changes in cloud entrainment, dynamics, and microphysics	Improved process-level understanding, parameterization for GCMs	Interactions between process-level studies and GCM development; analysis of field and satellite observations.
Aerosol-induced changes in dynamics, radiation, and microphysics of deep convection	Convective invigoration, changes in UTL clouds and vapor, parameterization for GCMs	Interactions between process-level studies and GCM development; closure box experiment at a regional scale; analysis of field and satellite observations.

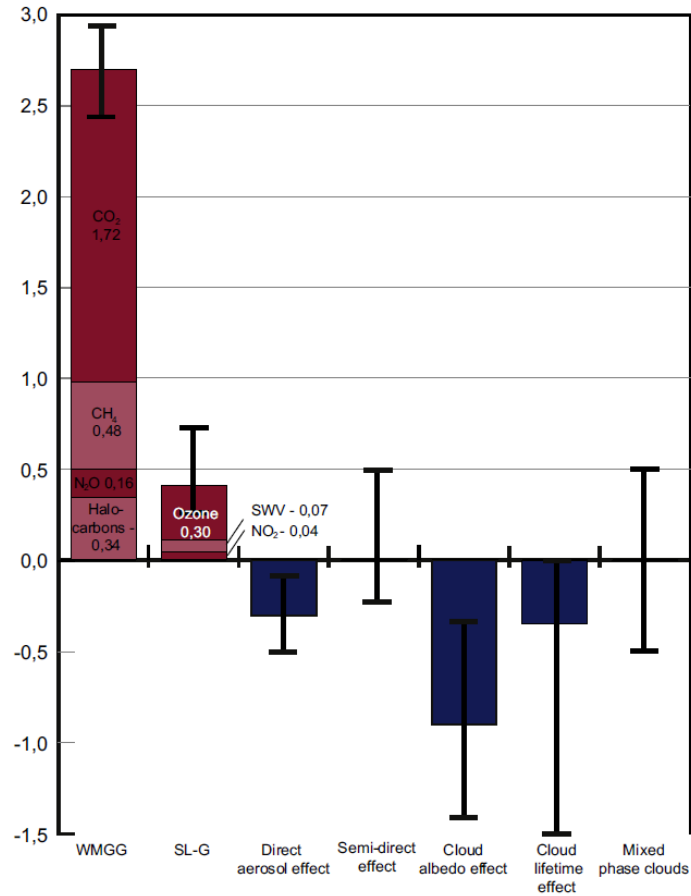


Figure 1. Radiative forcing estimates of atmospheric compounds from the pre-industrial period 1750 to 2007. From Isaksen et al., 2009.

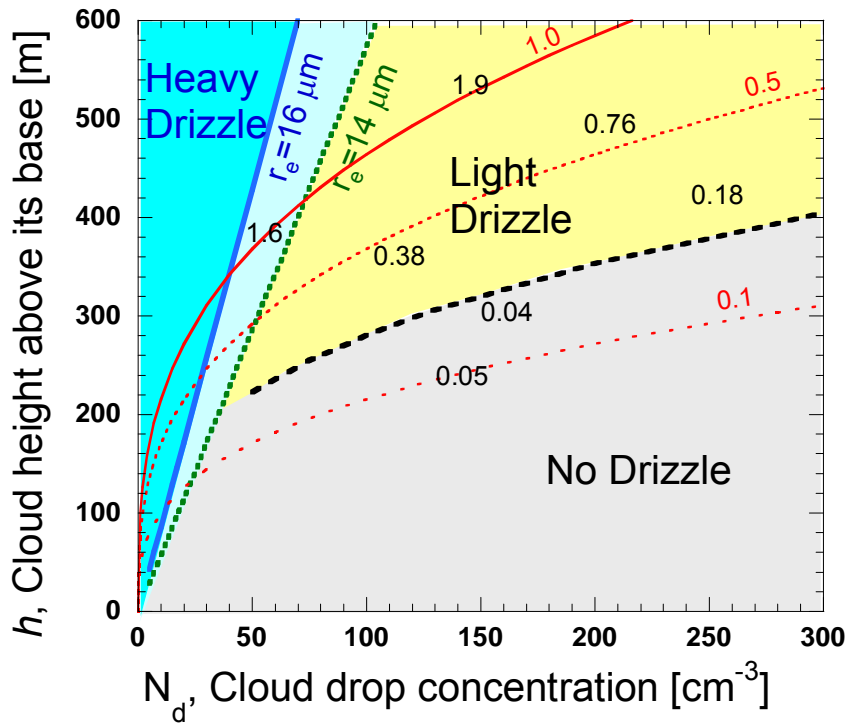


Figure 2. The dependence of drizzling regimes in marine stratocumulus clouds on drop number concentration and cloud depth. Heavy drizzle is defined where most water resides in the drizzle drops. Light drizzle is defined where most water resides in the cloud drops. The cloud drop effective radius of $r_e=16 \mu\text{m}$ was shown to be the minimal size for the heavy drizzle regime (Gerber, 1966). Transition to light drizzle occurs between r_e of 14 – 16 μm . The dashed line separates between negligible drizzle and light drizzle of $R > 0.2 \text{ mm day}^{-1}$ is based on DYCOMS-II observations. The red lines show the approximation of $R \sim h^3/N_d$, for R of 0.1, 0.5 and 1 mm/day. The individual points and their R values are posted (from Table 3 of vanZanten et al., 2005). After Rosenfeld et al. (ACP 2006).

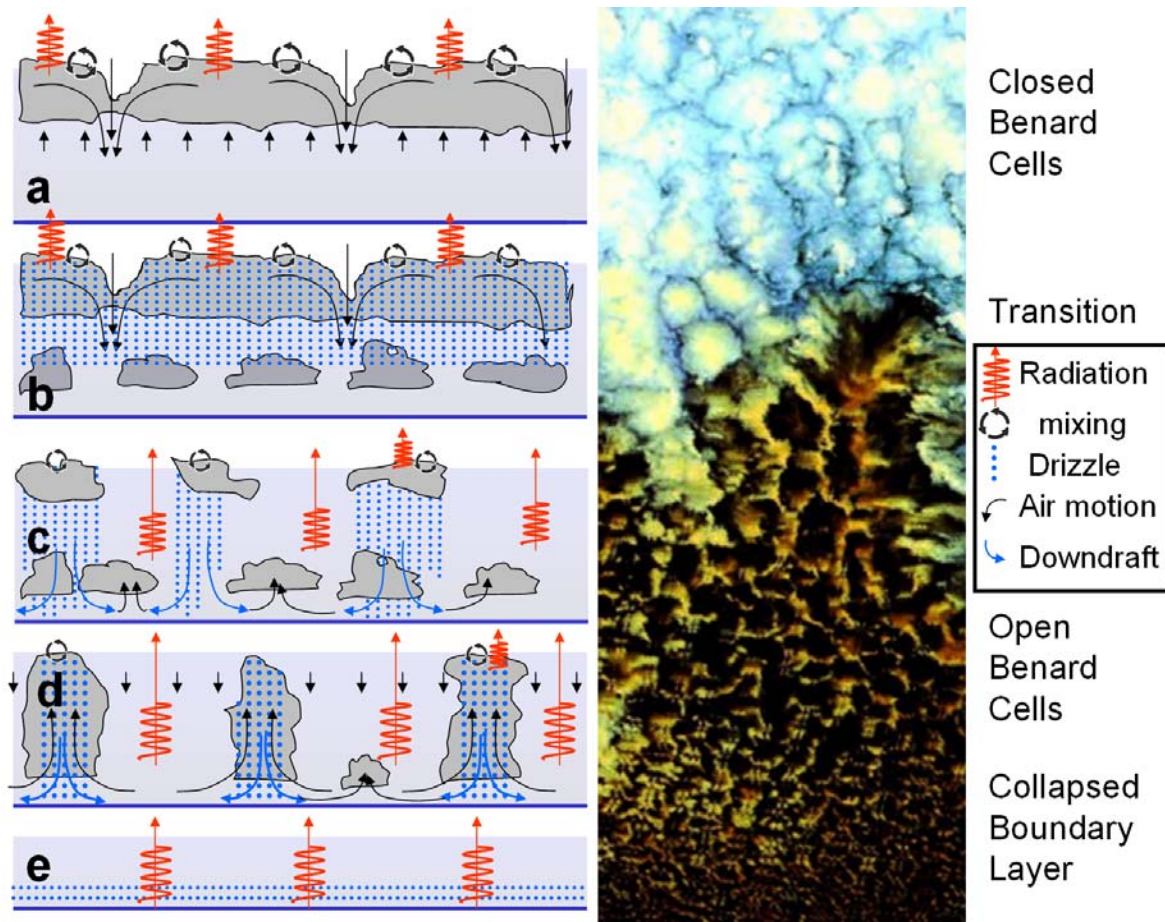


Figure 3: A schematic illustration of the mechanism for transition from non precipitating closed Benard cells to precipitating open cells and onward to nearly complete rainout and elimination of the clouds (After Rosenfeld et al., 2006). In the closed cells (a) the convection is propelled by thermal radiative cooling from the tops of the extensive deck of clouds with small drops. The clouds mix aerosols and vapor with the free troposphere from above. The onset of drizzle depletes the water from the cloud deck and cools the sub-cloud layer (b). This leads to decoupling of the cloud cover and to its subsequent breaking. The downdrafts due to the evaporational cooling starts triggering new convection (c). The propulsion of the convection undergoes transition from radiative cooling at the top of the fully cloudy MBL to surface heating at the bottom of the partly cloudy MBL. This causes a reversal of the convection from closed to open Benard cells, that develop, rainout and produce downdrafts that trigger new generations of such rain cells (d). The mixing with of aerosols with the free troposphere at cloud tops is much reduced. Therefore, the process can continue to a runaway effect of cleansing by the CCN and direct condensation into drizzle that directly precipitates and prevents the cloud formation altogether (e). The satellite strip is a 300 km long excerpt from Figure 34.

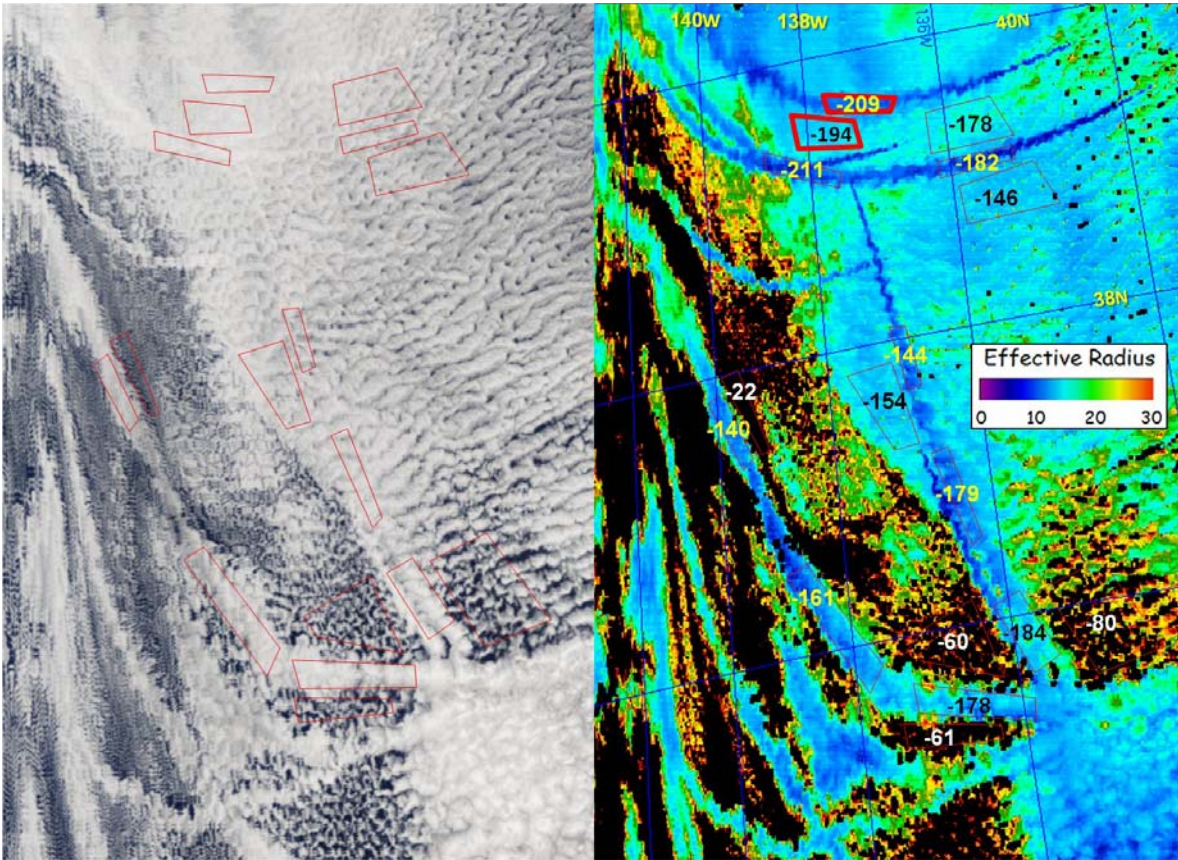


Figure 4. MODIS satellite image of open and closed cells in marine stratocumulus with ship tracks in an area lying between about 35-40 North and 134-142 West, off the coast of California on 26 July 2003 19:40 UTC. The left panel is a true color image, and the right panel is the MODIS-retrieved cloud top r_e . The ship tracks appear as a marked decrease in cloud drop effective radius (r_e in μm) on the right panel. The ship tracks are barely discernible in the true color image on the left panel, except for areas where $r_e > 16 \mu\text{m}$, above which significant drizzle occurs [Gerber, 1996] and open the closed cells. The cloud radiative effect (CRE, Wm^{-2}) is given for the marked rectangles. The difference in CRE between the open and ship track induced closed cells well exceeds 100 Wm^{-2} , whereas the RF of the ship tracks within the closed cells is an order of magnitude smaller. The image is the same as in from Rosenfeld et al. (2006) with added calculated CRE.

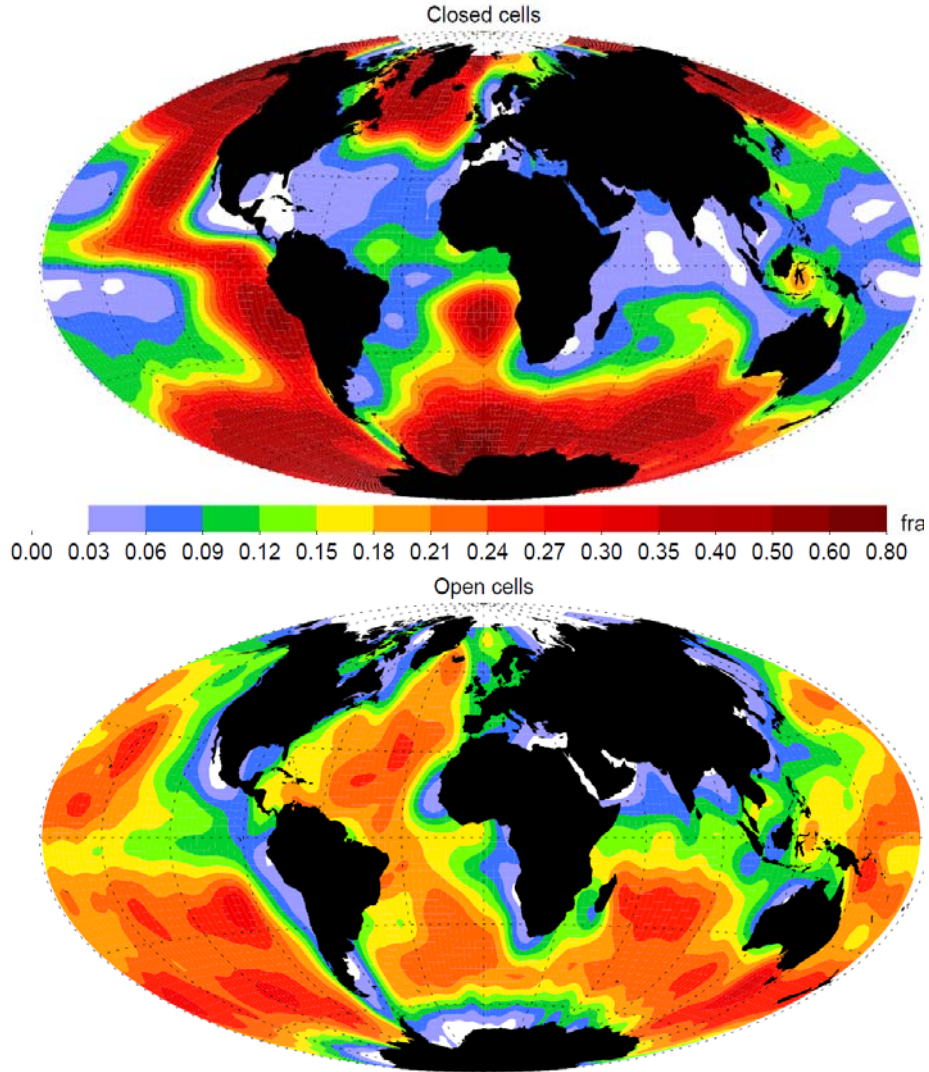
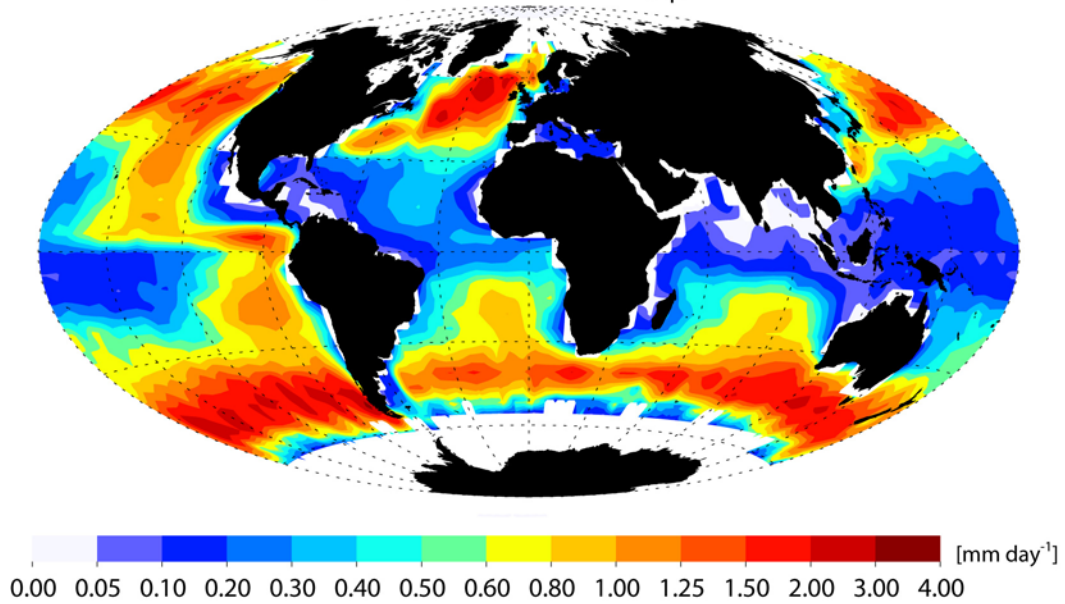


Figure 5. Frequency of occurrence of closed (top) and open (bottom) mesoscale cellular convection (MCC), based on all available MODIS data from 2008, using method of Wood and Hartmann (2006).

2C precipitation rate ($z_{\text{top}} < 3 \text{ km}$)



Cloud droplet concentration

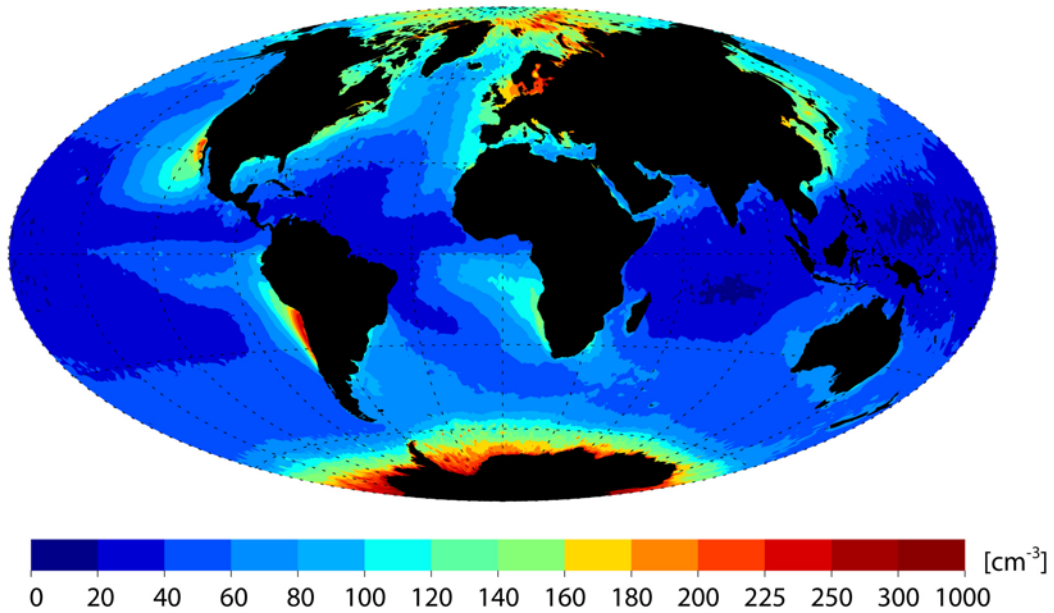


Figure 6. Top: Annual mean map of column maximum precipitation rate from clouds with tops below 3 km altitude, from the CloudSat Precipitation Radar (Lebsock et al. 2011). Bottom: Annual mean cloud droplet number concentration for horizontally extensive (instantaneous cloud cover exceeding 0.8 for 1x1 degree boxes) liquid clouds N_d , using data from MODIS, following the method of Bennartz et al. (2007).

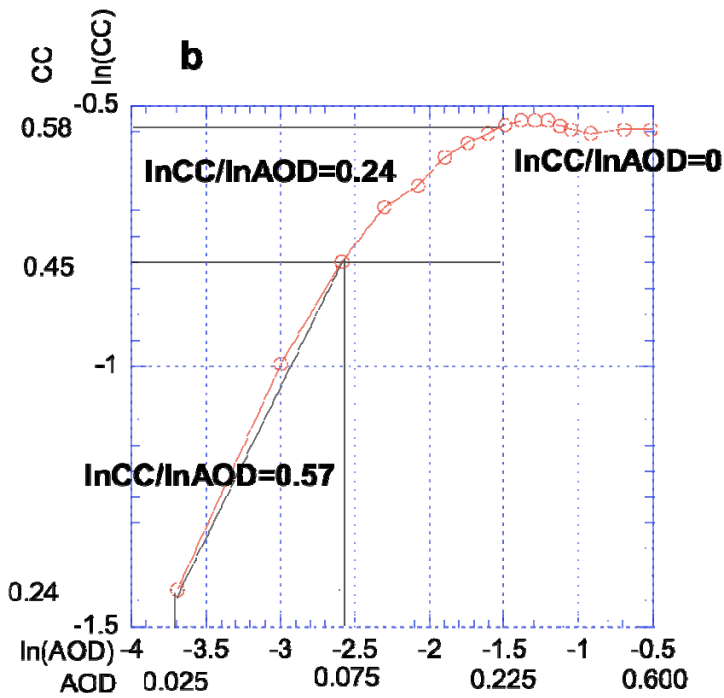
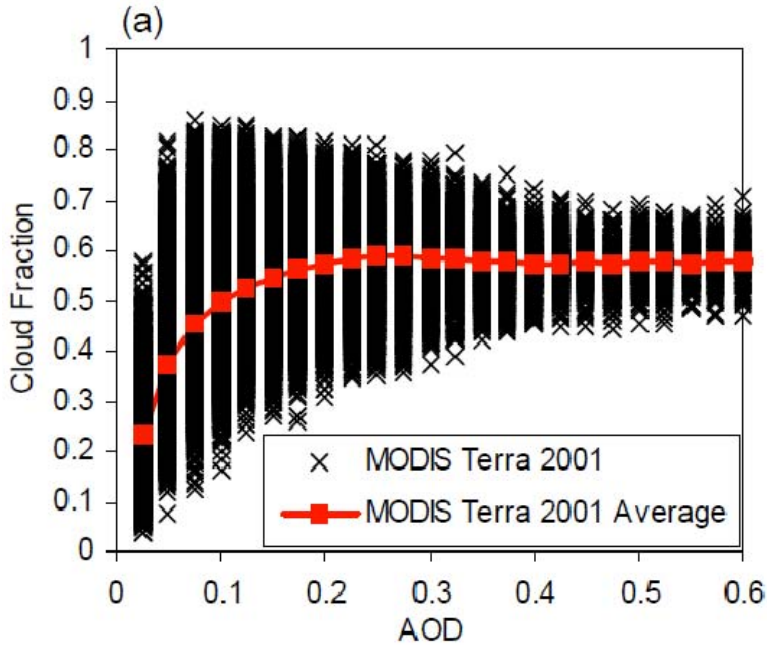


Figure 7: Annual global MODIS retrieved cloud cover as a function of AOD. (a) as presented by Myhre et al (2007); (b), presented on a logarithmic scale, with $\ln(CC)/\ln(AOD)$ calculated for three AOD intervals.

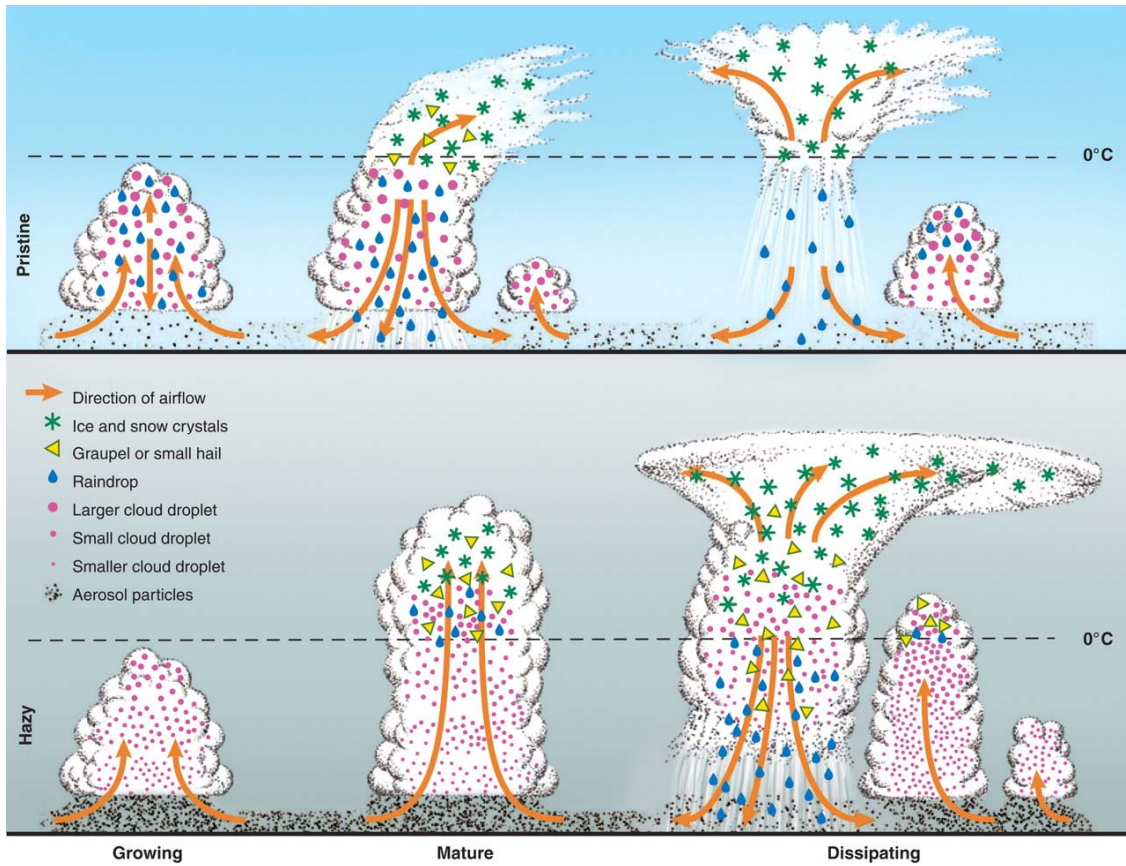


Figure 8: Illustration of the aerosol cloud invigoration hypothesis. Top: Clouds in pristine air rain-out their water before reaching the freezing level. Bottom: The aerosols delay the rain until the cloud reaches the freezing level, where the water freezes into ice hydrometeors and releases more intensely the latent heat of freezing, which invigorates the cloud. The cloud tops grow to greater heights and expand to larger anvils. From Rosenfeld et al., 2008.

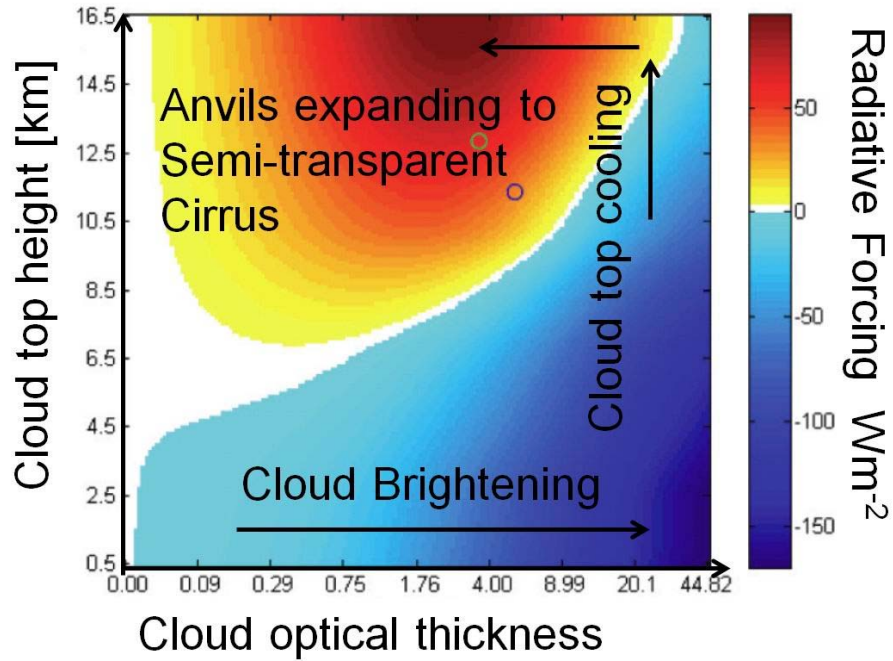


Figure 9: The net TOA radiative forcing of a cloud in a tropical atmosphere, as a function of its cloud top height and optical thickness. After Koren et al., 2010.

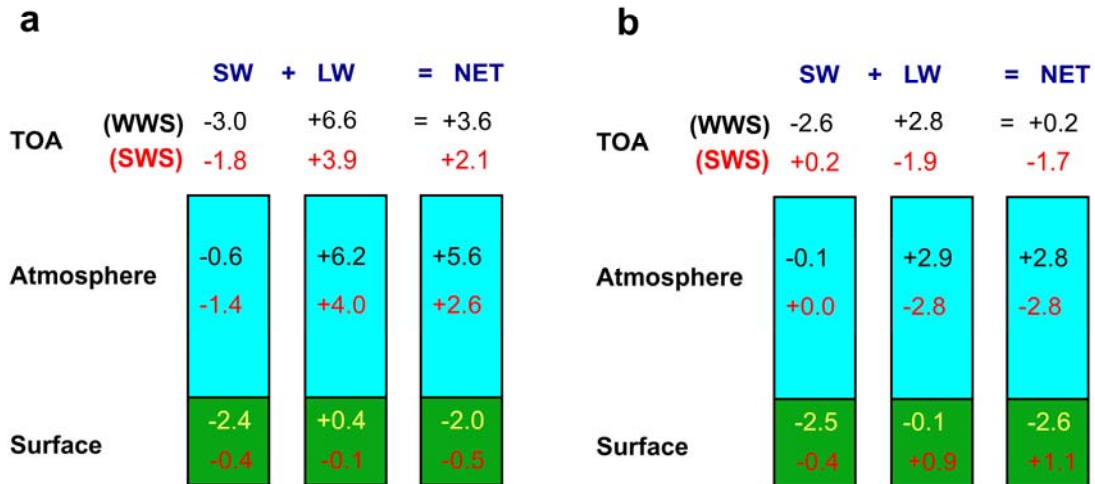


Figure 10. Short wave (SW), long wave (LW), and net radiative forcing of aerosol cloud mediated effect at the top of atmosphere (TOA), atmosphere, and surface (SFC) for the China tropical (a) and SGP temperate (b) cases of deep convective cloud system, with weak wind shear (WWS) and strong wind shear (SWS). Values in red are for the stronger wind shear condition. Values are averaged over the last day of simulations (From Fan et al., 2012).

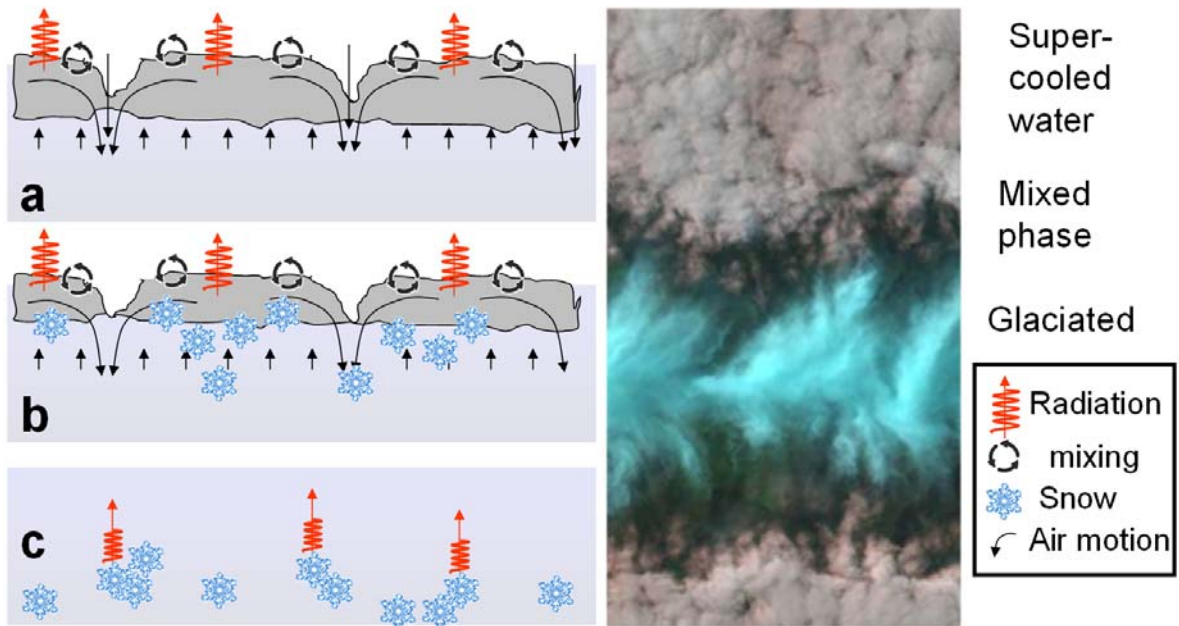


Figure 11: A schematic illustration of the mechanism for transition from persistent supercooled layer clouds to a stable situation of glaciated or no clouds. The mechanism that maintains the supercooled layer clouds is the radiative cooling and mixing at their tops, the same as for closed marine stratocumulus. The inverted convection replenishes the cloud water that is lost to ice precipitation (a). An increased loss of cloud water to ice precipitation, due to increased concentrations of IN and/or decrease in CCN, makes the cloud thinner and broken with less water (b). When the cloud water is fully consumed by the ice crystals that precipitate, there is nothing that will keep the radiative cooling that regenerated and maintained the cloud at the first place, and the cloud dissipates and leaves some falling ice crystals or no cloud at all (c). The image was taken by LANDSAT over the eastern USA on 11 December 2009. The glaciation in this case occurred probably due to addition of IN by aircraft exhaust to clouds at temperatures of -30 to -35°C . Source: NASA Visible Earth.