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

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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. Added aerosols to low clouds generally incur 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 some times in harmony and most 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. It 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, causing changes of cloud regimes in some cases. This leads to very complicated and uneven AIE. Present observations assume logarithmic relation between aerosol amount and cloud response. This hides the physics of much more complicated behavior, which its present understanding is described in this paper. Process models at high resolution (LES) have reached in the last few years the development stage that they can capture much of this complicated behavior of shallow clouds The implementation into a GCM 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

Added aerosols to deep clouds generally incurs 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 the deep clouds are much more complicated, because 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. Respectively, the parameterization of these processes for GCMs is further away than for the low clouds.

It is important to emphasize here that we 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.
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 et al. 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 effect to be larger.

Second, aerosols serve as the nuclei (CCN) for cloud droplets and can alter the albedo of clouds. 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 significantly exceed the results from satellite observations, which range from about -0.3 to -0.5 Wm^{-2} (Quass et al., 2009). Quass 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.

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. Values stronger than -1.5 Wm^{-2} would negate the impact of CO_2 since 1850, When considering the lag of oceans even -1.0 Wm^{-2} implies 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.

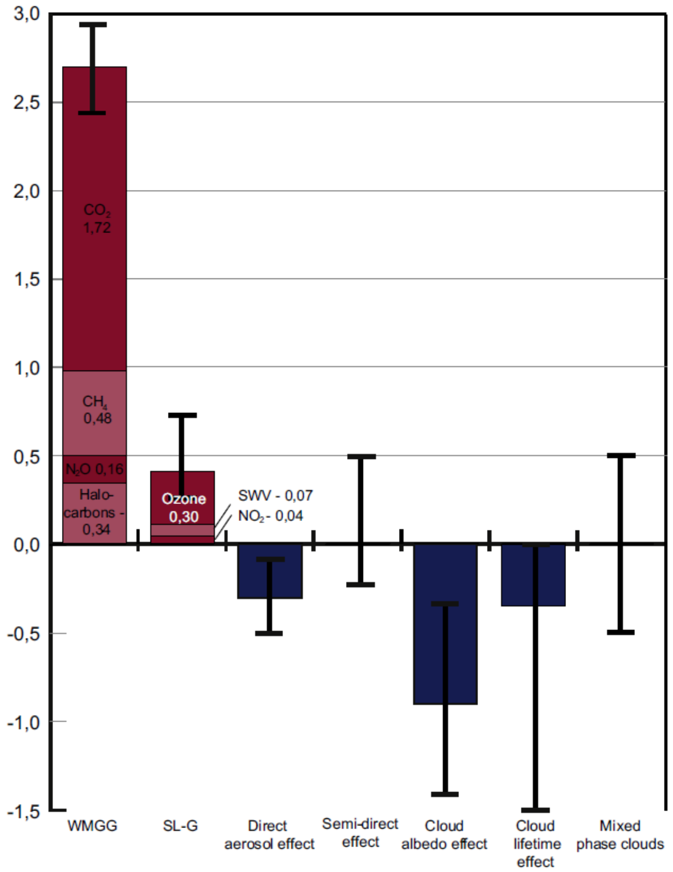


Figure 1. Radiative forcing estimates of atmospheric compounds from the pre-industrial period 1750 to 2007. From Isaksen et al., 2009.

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 -1.5 W m^{-2} was reduced to -1.2 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

2. Aerosol induced radiative forcing of 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 (Brenguier 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. We can write the aerosol indirect effect as the sensitivity of the albedo α 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. The Twomey term is called the albedo susceptibility (Platnick and Twomey 1994), and is well-approximated (e.g. Twomey, 1991) by

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

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.

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 forcing (Ackerman et al., 1995; Rosenfeld et al., 2006; 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.

Rain intensity in stratocumulus depends on N_d and cloud thickness h (Fig. 2). Van Zanten et al. (2005) showed that aircraft-measured cloud base rain rate $R \alpha h^3/N_d$. Since effective radius $r_e^3 \alpha$ LWC/$N_d \alpha h/N_d$, then $R \alpha h^2 r_e^3$. This was also reproduced by the simulations of Wang et al. (2009), but only for clouds with N_d <100 mg^{-1}, and h of about 600 m. For clouds with similar h but $N_d \approx 150$ mg^{-1} the surface rain rate was zero. This implies cloud top r_e of about 15 μm. Wang et al. (2009b and 2011) found similar results of complete suppression of surface precipitation at high N_d and respectively small r_e. The relation of $R \alpha h^3/N_d$ 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 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 (2011) 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; Goren and Rosenfeld, 2012). Mean r_e of areas of closed cells occasionally reaches 18 μm before breaking into open cells (Goren and Rosenfeld, 2012). The average N_d in the brightest 5% of the clouds is estimated to be approximately 50 and 15 cm^{-3} in the closed and open cells regimes, respectively. In comparison, Wood et al. (2011) aircraft-measured an average N_d of 21 cm^{-3} near cloud base of the open cells.

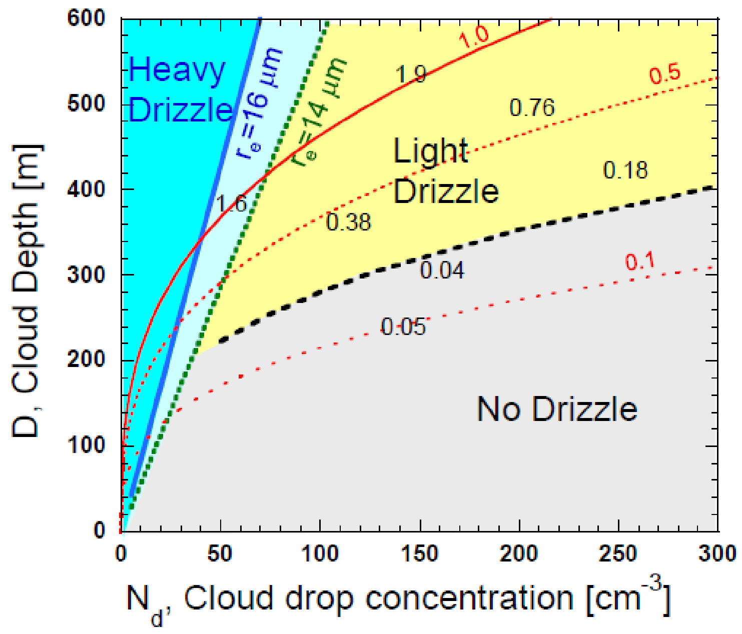


Fig. 2. The dependence of drizzling regimes in marine stratocumulus clouds on drop number concentration and cloud depth. Heavy drizzle is defined where most water resides in the drizzle drops. Light drizzle is defined where most water resides in the cloud drops. The cloud drop effective radius of r_e=16 µm 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 mm day^{-1} is based on DYCOMS-II observations. The red lines show the approximation of R α D^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). The After Rosenfeld et al. (ACP 2006).

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. 2012). 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, Christensen and Stephens).

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 D 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 Nd 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, leading 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 cells regimes was proposed by Rosenfeld et al. (2006). Based on this mechanism, it is 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 Figure 3). The onset of heavy precipitation that occurs when the cloud top r_e exceeds 16 μm, 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 Figure 3). This picture is complicated by the evaporative cooling of the rain shafts, which form mini gust fronts at the surface that regenerates 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 (PNAS 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, and so that cycles of clouds building and raining out occurs. This corresponds to the regime of oscillating and raining open cells.
3. The heavily precipitating clouds, where all incipient cloud water effectively precipitated 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, Nature 2009), but not between the regimes. 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 areas c and d of Figure 3, where most of the albedo changes due to N_d changes (Twomey effect) of -19 Wm^{-2} is offset by those due to LWP of +15 Wm^{-2}, leaving a net effect of only -4 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 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.

2.4 The components of cloud radiative effect

A method for partitioning the aerosol indirect effect into three components, namely the cloud cover, LWP and droplet radius effects was developed and tested by Goren and Rosenfeld (2012). It is based on the assumption that changes in MSC cloud regimes that occur at short distance in homogeneous meteorological conditions are associated with respective changes in the concentration of CCN, as approximated by the retrieved N_d. The method was applied to 50 cases of well defined transitions from closed to open cells (see examples in areas **a** and **b** of Figure 3). It was found that the negative CRE over the closed cells is on average higher by 110±18 Wm^{-2} than the adjacent open cells. This large negative CRE is composed of the cloud cover (42±8%), LWP (31±8%) and radius (26±6%) effects. This shows that the radius effect, which is caused by the change in N_d for a given

LWP, contributes on average only a quarter of the forcing, whereas the rest is contributed by added cloud water to the closed cells both in the horizontal (cloud cover effect) and in the vertical (LWP effect). However, the brightest and thickest clouds in the open cells were shown to have often more LWP than the thickest clouds in the closed cells, as observed from both satellite (Goren and Rosenfeld, 2012) and aircraft (Wood et al., 2011) observations.

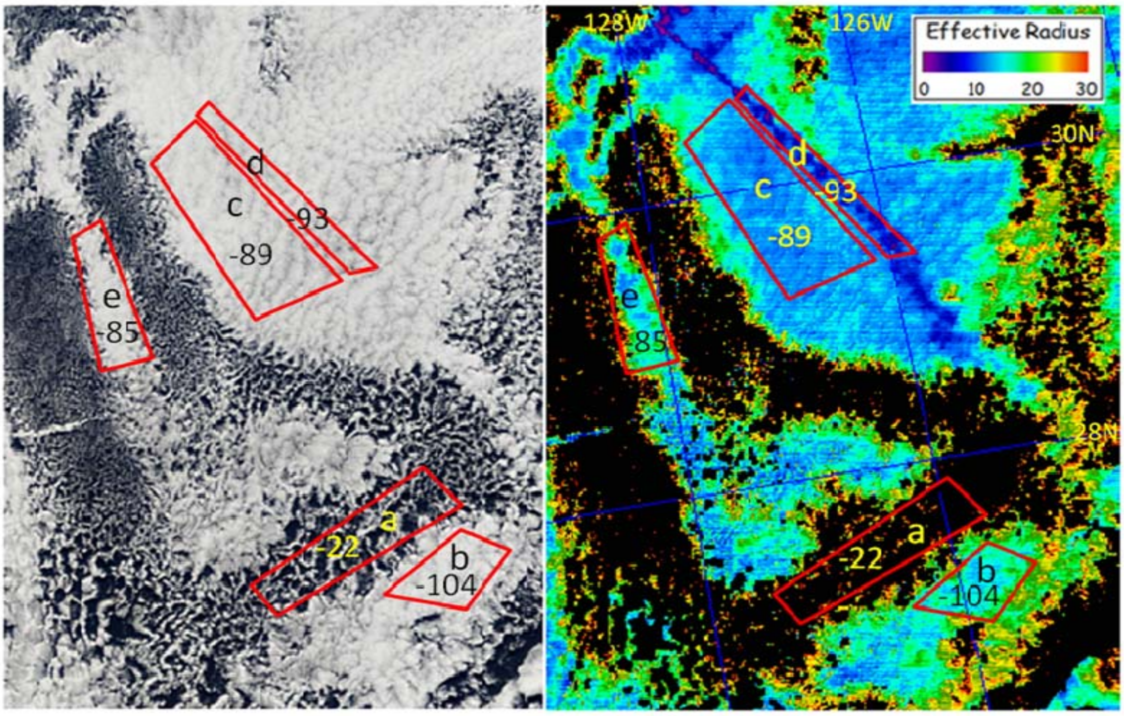


Figure 3. MODIS satellite image of open and closed cells in marine stratocumulus with ship tracks in an area of about 500x550 km off the west of the coast of California on 7 January 2009 19:05 UTC. The ship tracks appear as a marked decrease in cloud drop effective radius (r_e in μm) on the right panel. The ship tracks are barely discernible in the true color image on the left panel, except for areas where r_e>16 μm, 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 areas b and a is -82 Wm^{-2}. The 24 hour average differential CRE would have been -130 Wm^{-2} with a diurnally averaged normalized solar position to 30N on the equinox. From Goren and Rosenfeld (2012).

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. These results should be taken 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 the similarity between the relative contributions of CRE found in these two studies (George and Wood, 2010; Goren and Rosenfeld, 2012) mean that most of the variability in the cloud RF in the southeastern Pacific is 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 equilibrium state of weakly precipitating closed cells is large inversion height with well mixed boundary layer and strong entrainment at the top of the inversion. For very pristine drizzling clouds or a thin layer of very low clouds a stable situation is a very low inversion height, also defined as a "collapsed" boundary layer (Brethreton et al., 2010). 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.

2.5 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 4 presents a map of the occurrence of mesoscale cellular convection over the eastern

Pacific Ocean, 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. The first two 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 (Fig. 5) and increasing cloud thickness (see e.g. George and Wood 2010), the combination of which increases precipitation dramatically (Fig. 5, see also Bretherton et al. 2010). 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.

Globally, almost all of the increase in cloud cover f_c with AOD occurs at AOD<0.2 (see Figure 6). For AOD≤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 to a large part of the dependence of f_c on AOD.

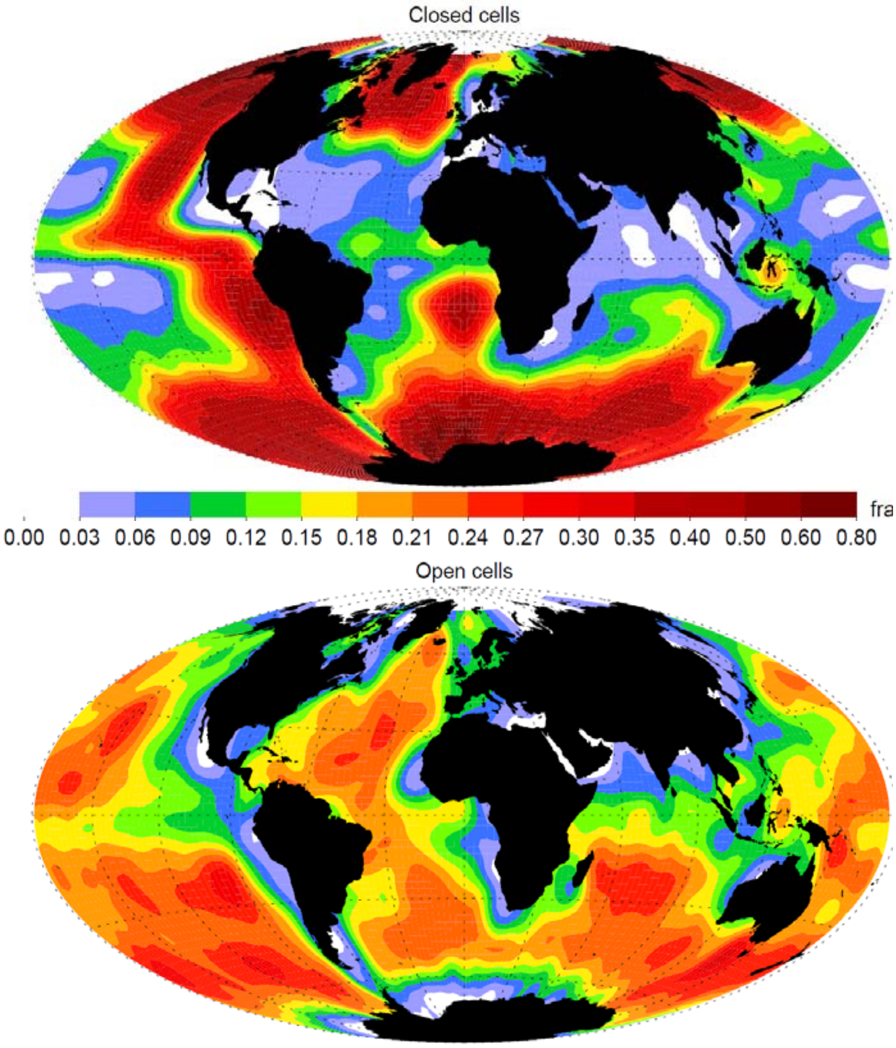
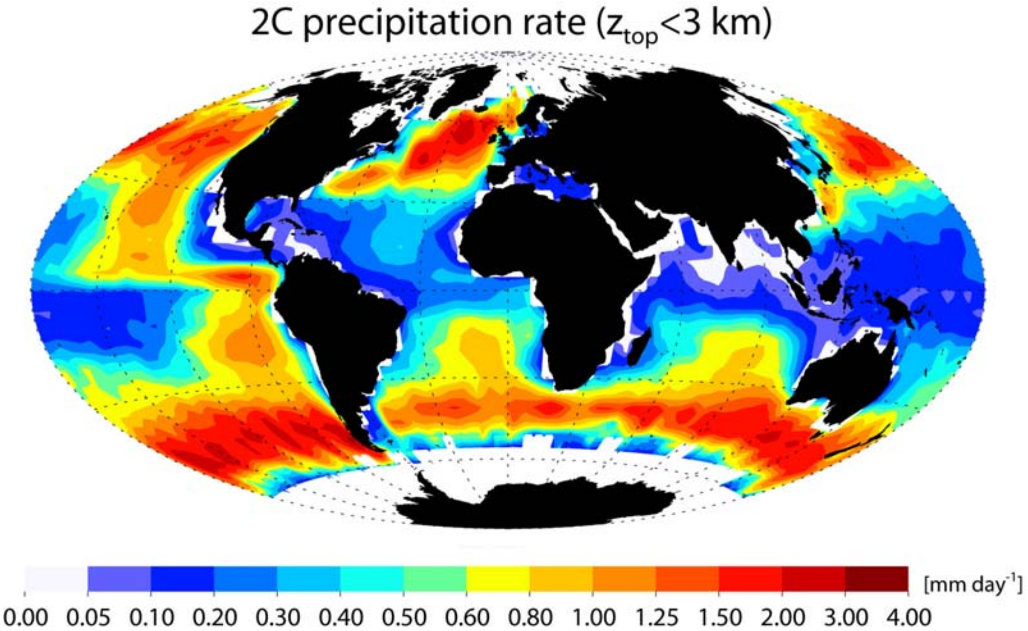


Figure 4. Frequency of occurrence of closed (top) and open (bottom) mesoscale cellular convection (MCC), based on all available MODIS data from 2008, using method of Wood and Hartmann (2006).



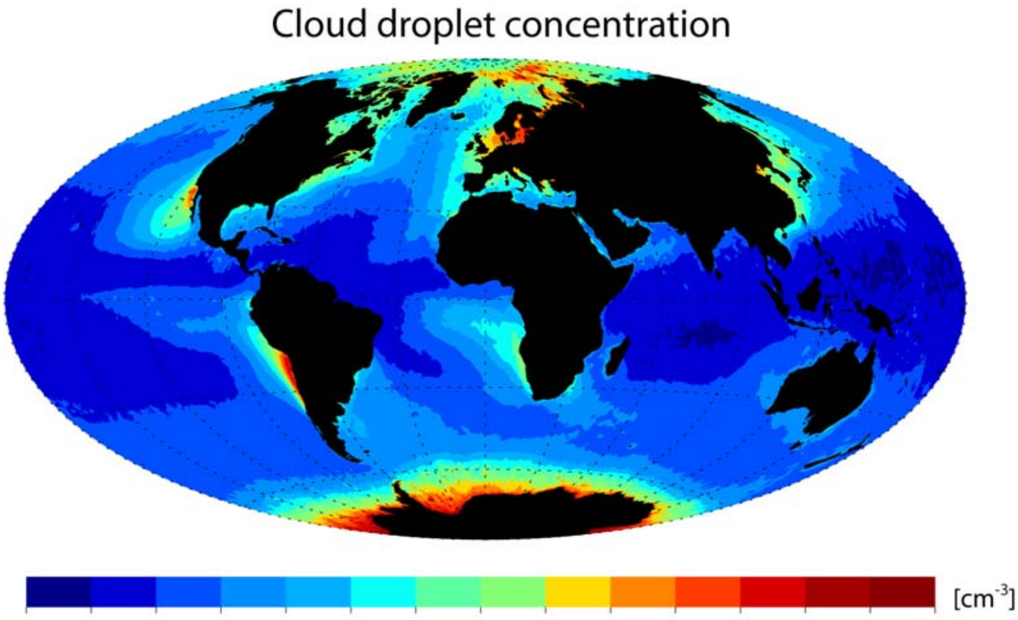


Figure 5. Top: Annual mean map of column maximum precipitation rate from clouds with tops below 3 km altitude, from the CloudSat Precipitation Radar (Lebsock et al. 2011). Bottom: Annual mean cloud droplet number concentration for horizontally extensive (instantaneous cloud cover exceeding 0.8 for 1x1 degree boxes) liquid clouds N_d, using data from MODIS, following the method of Bennartz et al. (2007).

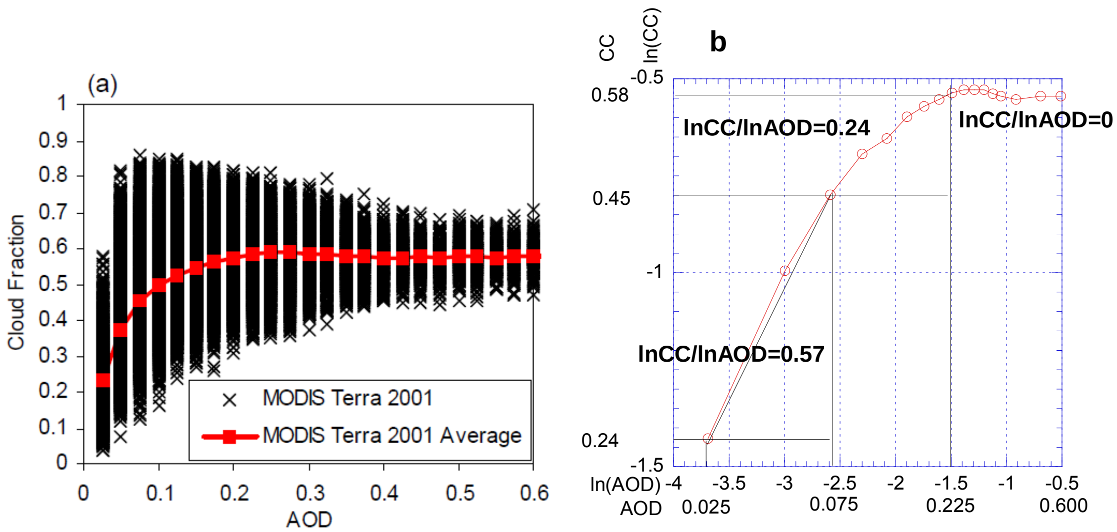


Figure 6: Annual global MODIS retrieved cloud cover as a function of AOD. (a) as presented by Myhre et al (2007); (b), presented on a logarithmic scale, with ln(CC)/ln(AOD) calculated for three AOD intervals.

2.6 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 (Fig 4). 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 Figure 3, where the large areas of closed cells appear 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 continent is 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 exceeds 100 Wm^{-2} (Goren and Rosenfeld, 2012).

2.7 The possible underestimate of the radiative forcing of low clouds

As we have discussed in Section 2.4, it is possible that the Twomey effect is 1/4 or less than the overall AIE from low clouds (Sekiguchi et al., 2003; Kaufman et al., 2005; Lebsock et al., 2008). Similar contribution of the Twomey effect was shown to occur due to regime changes in MSC (Goren et al., 2012), giving some insights to the causes of this partition between the components of the AIE. Yet, the IPCC AR4 found a radius effect of -0.7 Wm^{-2} with the large uncertainty range of -0.3 to -1.8 Wm^{-2}. If we multiply the AR4 range by a factor of four to account for the other, non-accounted for effects, we obtain a range of -7.2 to -1.2 Wm^{-2}. 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 one has to assume one of the two 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 equally strong positive RF that is also induced 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 the strong negative RF is hiding equally 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 imply that there should be a similarly large positive radiative forcing that balances this cooling effect. It is hypothesized here that this missing positive forcing is induced by the aerosol effect on deep and/or high clouds, through several possible mechanisms that are presented in this section.

3.1 Aerosol invigoration of deep tropical clouds

Most of the condensed cloud water in deep tropical convective clouds in pristine air mass is precipitated as warm rain (i.e., without the involvement of the ice phase) before reaching the freezing level. Adding CCN to the clouds causes Nd 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, 2011). As for the MSC in Figure 2 and for the same fundamental physical considerations, the number of activated cloud drops near cloud base scale linearly to the cloud depth for arriving at threshold r_e of ~14 μm for rain initiation in deep convective clouds (Freud et al., 2011; Freud and Rosenfeld, 2011). Increasing the number of activated aerosols into cloud drops by 100 cm^{-3} increases D for onset of rain by ~280 m. Therefore, in deep tropical cloud with freezing level of 3-4 km above cloud base, an adiabatic concentration of nearly 1000 drops cm^{-3} is required to nucleate near cloud base for delaying completely the onset of precipitation to above the freezing level.

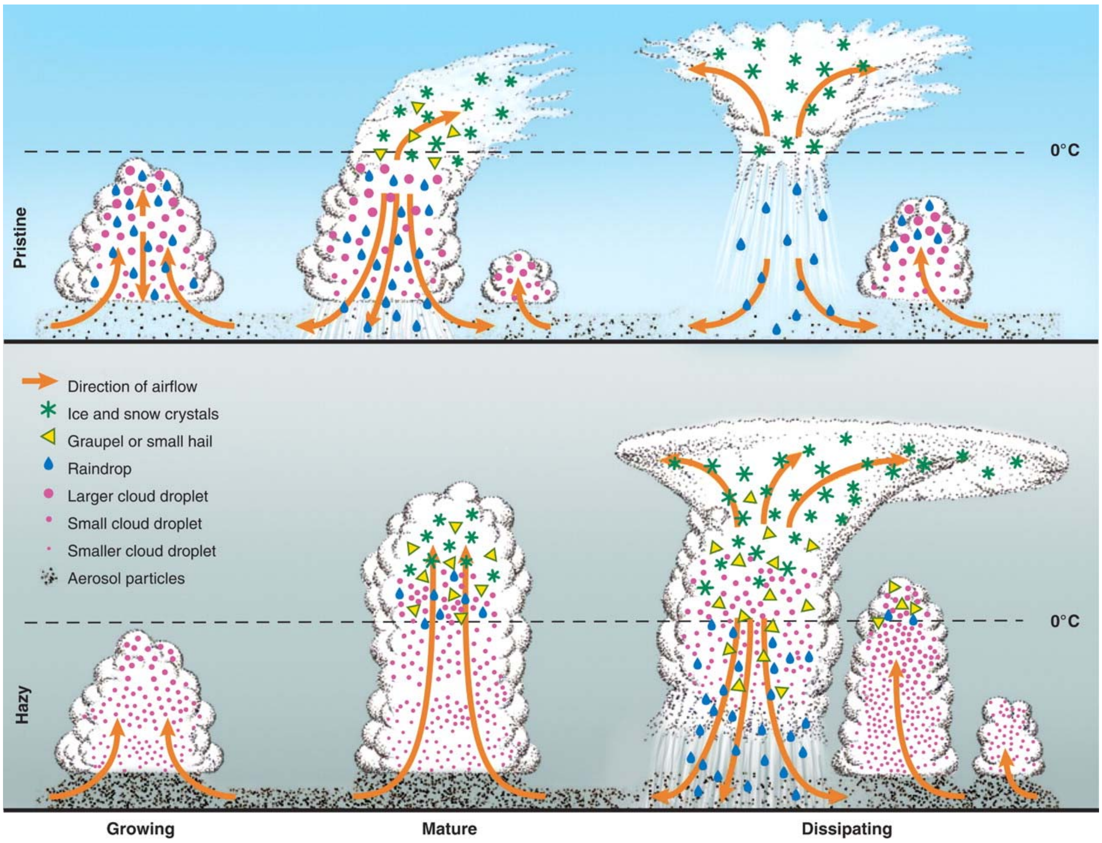


Figure 7: Illustration of the aerosol cloud invigoration hypothesis. Top: Clouds in pristine air rain-out their water before reaching the freezing level. Bottom: The aerosols delay the rain until the cloud reaches the freezing level, where the water freeze into more intense ice hydrometeors and release 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.

It was hypothesized (Rosenfeld et al., 2008) that delaying the precipitation to above the freezing level cause the cloud water to freeze first onto ice hydrometeors and so release the latent heat of freezing, which would have not been realized if the rain was not suppressed by the aerosols from occurring at lower levels (see illustration at Figure 7). 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 the lower levels. At the final account, more low level cooling and high level warming occurs 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).

Cloud simulation studies have generally confirmed the invigoration hypothesis for deep warm base clouds with weak wind shear in moist environment. 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 2008; Wang, 2005; Seifert and Beheng, 2006; van den Heever et al., 2006; Fan et al., 2007 and 2009). According to 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).

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 physical meaning of such associations is questionable due to possible problems of the retrieved aerosols by cloud contamination and other artifacts that are caused by the proximity to the clouds. Koren et al. (2010a) showed that this was not the cause for the findings, 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 about 3 km, which is well below the height of the anvils. 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 against the aerosol invigoration effect, at least on land. Therefore the aerosol effect is not monotonic, such that the invigoration effect reaches a maximum at AOD of ~0.3. This was shown theoretically by Rosenfeld et al. (2008) and observationally over the Amazon by Koren et al. (2008). Satellite measurements of AOD and rainfall showed that also the rainfall is enhanced over the Amazon with increased AOD, up to the optimal AOD of about 0.3, and showed smaller response or even some decrease when AOD is increased further (Lin et al., 2006).

Anvil clouds associated with deep convection exert a substantial longwave cloud forcing. 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., GRL 2011; Langenberg, Nature 2011). This lends credibility to the suggestion of Zhang et al. (2011) 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 tack 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., 2011).

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).

All these findings, and especially the long term measurements of Li et al. (2011), show that the aerosol invigoration effect is an important process in the climate system. The aerosol effect on deep convective clouds can influence radiative forcing in the several ways, as illustrated in Figure 8.

- Brightening of the clouds at a fixed cloud top leads to increasing of their albedo and greater negative RF. However, for already thick convective cloud, where the albedo effect is nearly saturated, the negative effect is expected to be rather small.
- The invigoration effect can cause the cloud tops to reach greater heights while keeping their albedo fixed at the nearly saturated value for thick clouds. The colder cloud tops emit less thermal radiation to space and hence induce positive RF.
- The anvils expand over larger areas and produce 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.
- The evaporation of the detrained aerosols enrich the upper troposphere and lower stratosphere (UTLS) with water vapor, which act there as potent green house gas with additional strong positive RF.

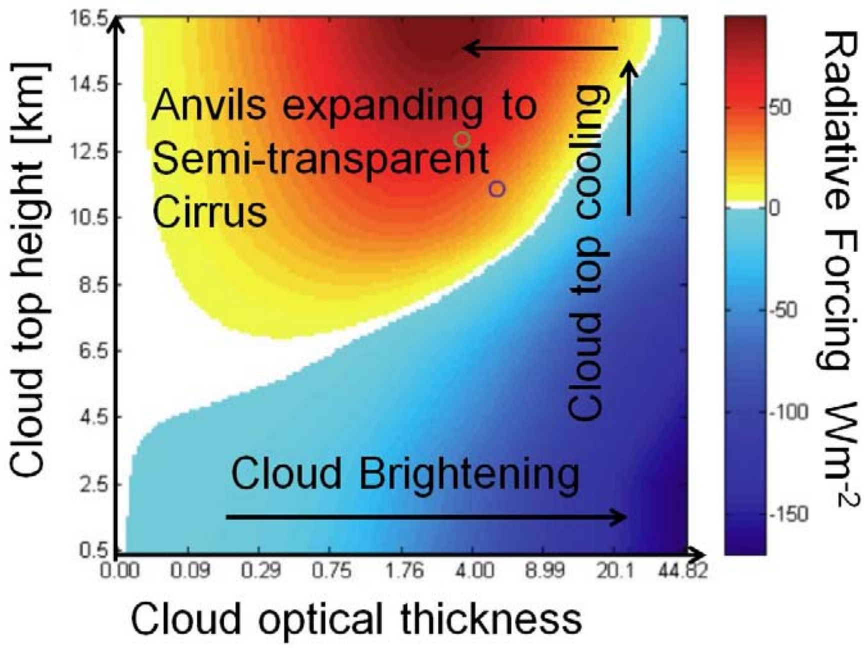


Figure 8: The net TOA radiative forcing of a cloud in a tropical atmosphere, as a function of its cloud top height and optical thickness. After Koren et al., 2010.

3.2 Aerosols enhancing detrainment of ice and vapor in the UTLS

Aerosols can 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. Sherwood (2002a) and J. H. Jiang et al. (2009b) found, based on satellite data, 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. This could be due to CCN nucleating small cloud drops that freeze homogeneously into respectively small ice particles, or to invigoration of the storms activating more aerosols aloft. 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 that must exist in the dense smoke keep the cloud drops extremely small up to the homogeneous ice nucleation level, where they become similarly small (r_e=~10 μm) ice particles, whereas ice in the ambient clouds formed mostly by mixed phase processes and formed large particles in the anvils, with r_e>30 μm (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).

Aircraft measurements and model simulations showed convincingly that aerosols indeed nucleated small cloud drops aloft that froze homogeneously into small ice crystals in the anvils of clouds over southern Florida when ingesting aerosols that came from Africa across the Atlantic Ocean (Fridland 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. The smaller ice crystals evaporate at the tropical tropopause layer (TTL) and moisten it, whereas without the added CCN the ice crystals would be larger and precipitate to lower levels. These microphysical effects occur regardless of the invigoration effects of the aerosols, whether the convection is enhanced or suppressed.

A simulation of the aerosol effect at a regional scale of 450X600 km with horizontal resolution of 2 km, done with a bin microphysics cloud model for two cloud regimes: (a) deep tropical convective clouds during summer over eastern China, and (b) mid-latitude cool base convective clouds over the central USA, centered over the SGP ARM site (Fan et al., 2012). Both cases were run with weak and strong wind shear, and small and high CCN concentrations. Large cloud invigoration occurred 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 9a. In the temperate case the net RF were weaker and of opposite signs for the different wind shears (Figure 9b).

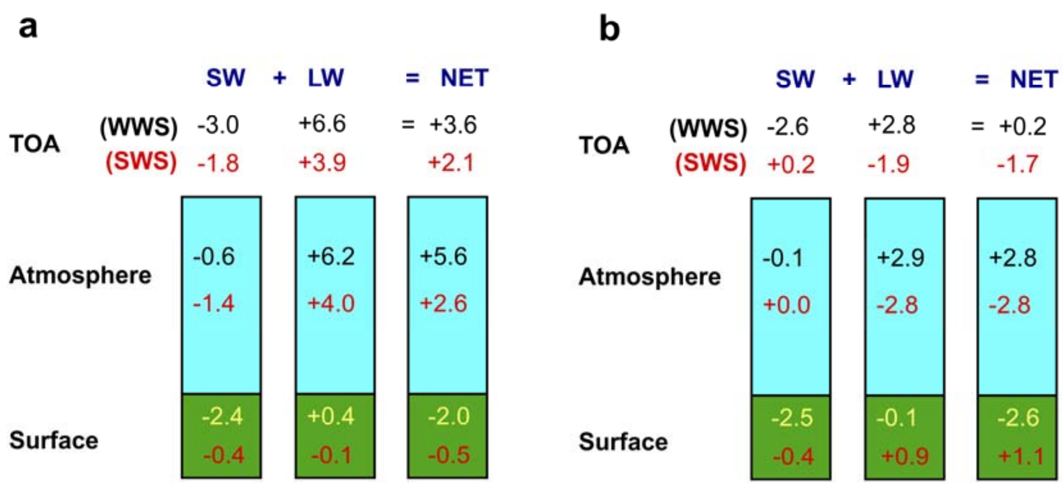


Figure 9. Short wave (SW), long wave (LW), and net radiative forcing of aerosol cloud mediated effect at the top of atmosphere (TOA), atmosphere, and surface (SFC) for the China tropical (a) and SGP temperate (b) cases of deep convective cloud system, with weak wind shear (WWS) and strong wind shear (SWS). Values in red are for the stronger wind shear condition. Values are averaged over the last day of simulations.

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, Myhre et al. 2007). While the decadal humidity variations are likely due to those of tropopause temperature, radiosonde data do not show a longer-term warming and the source of the moistening 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 it 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 (Sherwood 2002b, Chen and Yin 2011, P. K. Wang et al. 2011, Nielsen et al. 2011); back-of-the-envelope calculations suggest this mechanism 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 there also has been little trend in humidity since then. A similar microphysical effect from ice nuclei could also occur for cirrus clouds formed near the tropopause (Notholt et al 2005). Finally, there is evidence from observations (Su et al., 2011; Wu et al., 2011) and models (Liu et al. 2009) that pollution particles lofted in deep convection elevate cirrus cloud temperatures and water vapor mixing ratios, this 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 (Sherwood et al 2005, Seidel et al 2011). In summary, aerosols probably exert a second indirect warming effect through stratospheric water vapor, and this could be of nontrivial magnitude.

4. Possible interactions between the two effects

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

5. Implications to GCMs

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 and Coauthors (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 and Co-Authors (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 and Coauthors 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 and Coauthors (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 and Coauthors (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, including empirical methods relating aerosol concentrations to drop number (e.g., Lin and Leaitch, 1997) and 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 and Coauthors (2011)) in some models but calculated from prognostic aerosol modal equations (e.g., Neale and Co-Authors (2011)) in others.

The chief limitation in GCM representations of aerosol-cloud interactions arises from simplifications in their cloud macrophysics, which provide 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 are 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 and Coauthors 2011). The corresponding increase for GFDL CM2.1, which does not include cloud-aerosol interactions, is 0.66°C (Knutson and Co-Authors 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 and Coauthors (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 and Coauthors (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)) and the use of double-moment microphysics in deep convection in experimental versions of GFDL AM3 (Salzmann et al. 2010) and the NCAR Community Atmosphere Model (Song and Zhang 2011).

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-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 these 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. 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, which will allow retrieving Na from the vertical evolution of Nd 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 combine with Na yields the supersaturation and the CCN concentrations. Multiangular 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 aerosols, water and energy budgets, at a scale of a box of several hundred km on the side. This needs to 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 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. 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.

Added aerosols 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. It 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. This leads to very complicated and uneven AIE. Present observations assume 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. 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

Added aerosols to deep clouds generally incurs 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. Respectively, the parameterization of these processes for GCMs is further away than for the low clouds.

It is important to emphasize here that we 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 and hydrological cycle. Therefore, we have to quantify the AIE correctly not only for understanding the climate, but also for improving the 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, it is recommended here to plan coordinate 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.

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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