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1. INTRODUCTION

The so-called first indirect event first identified by Twomey (1974) suggests that increasing pollution results in greater CCN concentrations and greater numbers of cloud droplets, which, in turn, increase the reflectance of clouds. Subsequently, Albrecht (1989) hypothesized that the higher droplet concentrations in clouds would reduce the rate of formation of drizzle drops by collision and coalescence. The reduced rate of drizzle formation, in turn, would result in higher liquid water contents and lead to longer-lived clouds which by increasing cloud cover would lead to further enhancement of the albedo of those clouds. This is often referred to as the second indirect effect. General circulation model(GCM) simulations of the cloud albedo effect since pre-industrial times is estimated to be between -0.5 and -1.9 W/m² from different climate models and the cloud lifetime effect to be between -0.3 and -1.4 W/m² (Lohmann and Feichter, 2004). The parameterizations in all the GCMs assume that the second indirect effect always increases cloud lifetime, cloud liquid water contents, and cloud albedo. GCMs do not even agree on the relative importance of the albedo and lifetime effects. Differences are likely related to the range of treatments of droplet activation, assumptions of what constitutes the background aerosol, as well as autoconversion parameterizations.

In this paper we present evidence that cloud responses to increased CCN concentrations do not always yield a response that is in accordance to the Albrecht hypothesis. We do this first for boundary layer clouds and then for deep convective clouds.

2. AEROSOL INDIRECT EFFECTS IN BOUNDARY LAYER CLOUDS

Modeling studies suggest that cloud responses to increased CCN can vary depending on the intensity of drizzle in clean clouds, to the moisture content of air overlying the boundary layer, and lateral entrainment rates in cumuli.

For heavily drizzling stratocumulus clouds large eddy simulations (LES) by Stevens et al. (1998) showed that drizzle cools and stabilizes the subcloud layer. Thus if CCN reduce the drizzle rates the boundary layer becomes slightly more unstable,

eddies are more intense and transport more water into the cloud layer, clouds exhibit higher liquid water contents and liquid water paths, thereby yielding higher cloud albedo. This is a positive feedback to the Twomey hypothesis and is consistent with Albrecht's hypothesis. However, in Jiang et al's. (2002) LES of lightly precipitating marine stratocumulus, higher CCN concentrations suppressed drizzle which resulted in weaker penetrating cumulus and an overall reduction in the water content of the clouds. Thus cloud albedo was very little influenced by the increase in CCN concentrations. These results seem at first contradictory. But they are consistent with the inferences by Paluch and Lenschow (1991) from observations of boundary layer clouds. Their study and our modeling studies suggest that drizzle falling only partway through the sub-cloud layer can destabilize the boundary layer leading to cumulus under stratus; Whereas, drizzle falling through the entire sub-cloud layer can cool and stabilize the entire boundary layer. This leads to decoupling of the stratus layer from the surface. Thus enhanced CCN concentrations in a lightly drizzling boundary layer can stabilize the cloudy boundary layer leading to a response contrary to the Albrecht hypothesis.

In another modeling study Ackerman et al. (2004) showed that increases in CCN do not necessarily result in increases in LWP in stratocumulus clouds. A primary factor affecting the LWP response to aerosol changes is the profile of humidity above the inversion. Only when the humidity above the inversion was high did increases in aerosol result in an increase in LWP. When dry air overlies the inversion, increases in aerosol tend to decrease LWP because of enhanced entrainment drying. Similar results were obtained by Lu and Seinfeld (2005).

A very recent example of departures from the Albrecht hypothesis is Xue and Feingold's simulations of aerosol influences on tradewind cumulus. They found that increasing concentrations of CCN and droplets produced smaller droplets and suppressed drizzle and led to enhanced evaporation of droplets by entrainment. This is because, for a given LWC, smaller droplets evaporate more readily than larger droplets. Thus entrainment of dry air into a cloud composed of numerous small droplets(when CCN concentrations are high) experience a reduction in cloud fraction, cloud size, and cloud depth, thereby reducing cloud albedo.

These simulations highlight the nonlinearity of cloud systems when drizzle is present and suggests that increased concentrations of CCN may not always increase cloud water contents, cloud lifetimes, and

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cloud albedo. How often these exceptions occur is still unknown.

3. ENHANCED AEROSOL CONCENTRATIONS IN DEEP CONVECTIVE CLOUDS

A direct extension of the concept that enhanced CCN concentrations will lead to a reduction in precipitation to deep convective clouds would suggest that enhanced CCN in those clouds will result in suppressed precipitation. Indeed, simulations by Reisin et al. (1996a,b) with an axisymmetrical cloud model with a comprehensive bin microphysical representation of water and ice hydrometeors found just that. They showed that a simulation with relatively low CCN (100 cm^{-3} at 1% supersaturation) produced precipitation efficiently through the freezing of large droplets interacting with ice crystals. With increasing CCN concentrations and decreasing drop sizes, graupel growth was suppressed and the precipitation efficiency decreased (for $\text{CCN}=900 \text{ cm}^{-3}$, precipitation was reduced by 85%).

But in Khain et al.'s (2005) simulations of the effects of smoke-enhanced CCN concentrations, they found that smaller cloud droplets reduce the production of drizzle drops. When these droplets froze, the associated latent heat release resulted in more vigorous convection. In a clean cloud, on the other hand, drizzle depleted the cloud liquid water so that less latent heat was released when the cloud glaciated, resulting in less vigorous convection. Thus, they found that a squall line did not form under clean conditions, whereas a squall line developed under continental aerosol conditions and produced more precipitation after two hours. Zhang et al. (2005) came to similar conclusions in their model simulations for different three-week periods over the ARM site in Oklahoma.

Seifert and Beheng (2005), however, showed that the effect of changes in CCN on mixed phase convective clouds is quite dependent on cloud type. They found that for small convective storms, an increase in CCN decreases precipitation and the maximum updraft velocities. For multicellular storms, the increase in CCN has the opposite effect – namely, promoting secondary convection, and increasing maximum updrafts and total precipitation. Supercell storms were the least sensitive to CCN. Their study also showed that the most important pathway for feedbacks from microphysics to dynamics is via the release of latent heat of freezing.

All these simulations are mostly single-cloud or idealized multicell storms with either detailed bin microphysics or bulk microphysics.

To further confound the problem of how aerosol variability effects precipitation, I turn to our own simulations of dust influences on Florida convection

(van den Heever et al, 2006) and urban pollution effects (van den Heever and Cotton, 2006). These simulations are more properly called mesoscale simulations. The finest grid spacings are either 0.5km or 1.5km, respectively, and cover domains of roughly 150km. The period of the simulations is about 12h in one case and about 24h in the other. Storms are initiated via Florida sea-breeze forcing and land-surface heating including urban land-use effects, respectively. The simulations are cloud-resolving and the microphysics is a bulk microphysics model that includes explicit activation of CCN, giant CCN (GCCN) and ice-forming nuclei (IFN), with each aerosol species being prognostic variables. In addition precipitation physics emulates a full-bin microphysics model. The simulations are case study simulations of actual observed convective events.

In the Florida simulations a layer of dust is imposed that has properties of enhanced CCN, GCCN, and IFN in which simulations are performed for the collective effects of those aerosols as well as responses to enhanced CCN alone, GCCN alone, and IFN alone. Table 1 shows the range of

Table 1: Aerosol initialization profiles for the sensitivity tests.

Experiment	Name	IFN	CCN	GCCN
Exp1	CLN	Clean	Clean	Clean
Exp2	GCCN	Clean	Clean	Observed
Exp3	CCN	Clean	Observed	Clean
Exp4	IFN	Observed	Clean	Clean
Exp5	C+G	Clean	Observed	Observed
Exp6	I+G	Observed	Clean	Observed
Exp7	I+C	Observed	Observed	Clean
Exp8	OBS	Observed	Observed	Observed

experiments considered. Table 2 shows the results of accumulated surface precipitation (acre-feet) for the 8 sensitivity tests for those simulations first at 1800UTC and then 6 hours later at 00UTC. It can be seen that as the convection first becomes deep organized systems (1800UTC), dust serving as IFN enhances precipitation the most followed by GCCN. The clean control simulation is fourth in the ordering whereas dust serving to enhance solely CCN is lower still. Dust serving to enhance CCN, GCCN, and IFN (OBS) has the lowest amount of precipitation at that time. While dust enhances storm updrafts appreciably it does not increase precipitation. Six hours later the ordering of the experimental results on precipitation has changed dramatically with the highest simulated precipitation being for the clean control case, and with dust serving to enhance CCN in the middle of the pack, followed by dust serving as CCN, GCCN, and IFN (OBS). There are several reasons for these changes. First the cumulonimbi vents the dust so strongly that by

Table 2: Accumulated surface precipitation (acre-feet) for the eight sensitivity tests.

1800 UTC ACCUMULATED PRECIPITATION		0000 UTC ACCUMULATED PRECIPITATION	
Exp Name	Magnitude (a-f)	Exp Name	Magnitude (a-f)
IFN	66608	CLN	442168
GCCN	65874	GCCN	368053
I+G	63487	I+G	352112
CLN	63289	IFN	349373
CCN	61741	OBS	346309
C+G	58275	CCN	344338
I+C	57700	I+C	330610
OBS	57008	C+G	327560

00Z the lower atmosphere is largely cleaned of dust. Second, the early convection that responded quite vigorously to enhanced IFN and GCCN has put down cold pools which differ appreciably from the control simulations. As a result the impacts of dust on precipitation are now linked to the nonlinear responses of convection to varying cold-pool strengths. We explore this more fully in the urban simulations.

In the simulations of the St. Louis, MO urban area influence on ordinary thunderstorms, van den Heever and Cotton (2006) found that urban land-use determines whether or not storms actually develop in the downwind region. Air pollution which can serve to enhance CCN and GCCN concentrations does have an impact upon surface precipitation, however. Figure 1 illustrates the time series of accumulated surface precipitation for the region downwind of the St. Louis urban region. Early on we see the urban aerosol (enhanced CCN and GCCN) produce the largest amount of rainfall. However by 00UTC, the enhanced CCN produced the largest rainfall while the urban aerosol exhibited the least amount. This is further illustrated in Table 3 where the accumulated rainfall is presented for the entire fine grid. Here again, the final longer term response to aerosols is quite different than what occurs during the first few hours. Contrary to the Albrecht hypothesis, increasing CCN produced the largest amount of precipitation. The nonlinear behavior of the convective system once cold pool dynamics become dominant in controlling

storm dynamics is largely responsible for this behavior.

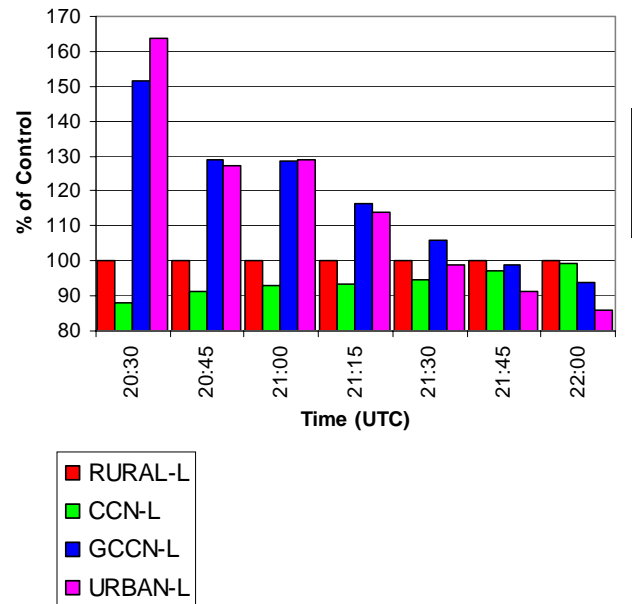


Figure 1: Time series of the accumulated volumetric precipitation in the downwind region expressed as a percentage of the RURAL-L simulation.

Table 3: Accumulated volumetric precipitation (acre-feet) for entire Grid 3.

TIME	RURAL	CCN	GCCN	URBAN
20:00	0	0	0	0
21:00	13956	13748	14411	14490
22:00	36338	35900	35173	35299
23:00	63409	63964	61451	58227
00:00	74370	75499	72511	69914

4. CONCLUDING REMARKS

We have seen that even in very light drizzling cloud systems that once the precipitation process is altered by varying aerosol concentrations the response of the clouds can be quite nonlinear. In the case of deep convective clouds where gravity wave and cold pool dynamics play a central role in the longer term response of clouds to varying aerosol concentrations, the ultimate consequences of varying aerosol amounts to precipitation is so nonlinear that predicting those responses is nearly impossible.

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