Process-model simulations of cloud albedo enhancement by aerosols in the Arctic

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Abstract

A cloud-resolving model is used to simulate the effectiveness of Arctic marine cloud brightening via injection of cloud condensation nuclei (CCN), either through geoengineering or other increased sources of Arctic aerosols. An updated cloud microphysical scheme is employed, with prognostic CCN and cloud particle numbers in both liquid and mixed-phase marine low clouds. Injection of CCN into the marine boundary layer can delay the collapse of the boundary layer and increase low-cloud albedo. Albedo increases are stronger for pure liquid clouds than mixed-phase clouds. Liquid precipitation can be suppressed by CCN injection, whereas ice precipitation (snow) is affected less; thus the effectiveness of brightening mixed-phase clouds is lower than for liquid-only clouds. CCN injection into a clean regime results in a greater albedo increase than injection into a polluted regime, consistent with current knowledge about aerosol-cloud interactions. Unlike previous studies investigating warm clouds, dynamical changes in circulation due to precipitation changes are small. According to these results, which are dependent upon the representation of ice nucleation processes in the employed microphysical scheme, Arctic geoengineering is unlikely to be effective as the sole means of altering the global radiation budget but could have substantial local radiative effects.
1 Introduction

Aerosol-cloud interactions, including the so-called aerosol indirect effects, are responsible for some of the largest sources of uncertainty in computing the global radiation budget (Boucher et al. 2013). The first aerosol indirect effect, also called the cloud albedo effect, refers to the consequences of adding aerosols that act as cloud condensation nuclei (CCN) to clouds under an assumption of fixed liquid water path: CCN may increase the liquid cloud droplet number concentration (CDNC) and thus reduce droplet size, resulting in an increased albedo (Twomey, 1977). The second aerosol indirect effect, also called the cloud lifetime effect, describes how the additional CCN in liquid clouds might change liquid water path causing increases in cloud lifetime, cloud opacity, and areal extent (Albrecht, 1989; Wood, 2012). These aerosol effects are most dramatically seen in marine low clouds, which cover on average 34.0% of the ocean surface (Warren et al., 1988).

Evidence for the aerosol indirect effects can be seen in ship tracks (brighter clouds due to injection of particles from ship plumes; e.g., Radke et al., 1989; Coakley et al., 2000) and in process modeling studies (e.g., Ackerman et al. 2003; Wang and Feingold, 2009). Although ship plumes do not always result in brighter clouds (e.g., Chen et al., 2012), this concept in part inspired Latham (1990) to suggest the possibility of deliberately injecting aerosols into the marine boundary layer to increase planetary albedo and cool the planet, counteracting some of the warming effects of anthropogenic greenhouse gas emissions. This proposal is typically known as Marine Cloud Brightening (MCB) and is part of a broader set of strategies called solar geoengineering. Due to the ubiquity of marine low clouds, it has been estimated that a 4% increase in global cloud fraction (Randall et al., 1984) or a 6% increase in albedo of existing marine low clouds (Latham et al. 2008) could offset atmospheric warming due to a doubling of the CO$_2$ concentration from preindustrial times. Numerous
modeling studies have found that with sufficient, controlled aerosol injection, global warming could be offset, although MCB may not return other fields, like temperature and Arctic sea ice, to their previous levels (e.g., Rasch et al., 2009).

Several previous studies have highlighted key points in which microphysical uncertainties have strong influences on the overall uncertainty in the effectiveness of MCB. For example, Pringle et al. (2012) showed that achievable CDNC correlates with updraft velocity, explaining in part why the resulting CDNC in simulations by Korhonen et al. (2010) were substantially lower than in simulations by Partanen et al. (2012), who used more realistic updraft velocities. Many of these microphysical processes operate on the sub-grid scale of global-scale models; process models (such as cloud resolving models) can explicitly resolve the small-scale turbulent updrafts and better represent these microphysical mechanisms that cause some of the uncertainties in global models, providing a useful complement. Wang et al. (2011) investigated the effects of MCB in warm marine clouds, revealing the dynamical feedbacks associated with aerosol-induced changes in precipitation and the dependence of MCB effectiveness on meteorological and background aerosol conditions. Jenkins et al. (2013) examined the effects of the diurnal cycle on MCB, showing the time of day of injection has a profound impact on aerosol indirect effects; they also found that the aerosol direct effect of scattering solar irradiance can, to some degree, complement the effects of MCB in cloud-free areas. In these two previous process-modeling studies, the injected aerosol particles were assumed to be uniformly distributed in the model grid box (a volume on the order of $10^6$ m$^3$) within seconds. Stuart et al. (2013) explicitly modeled an aerosol injection plume, showing that due to in-plume aerosol coagulation, the number of aerosols that reach the cloud layer strongly depends upon meteorological conditions.

These past process-modeling studies have focused on liquid clouds in a warm marine
boundary layer. Doing so spans the vast majority of marine low clouds, but in low temperatures, such as in the Arctic, many marine low clouds exist in the mixed phase (i.e., supercooled liquid droplets and ice particles coexist). Cloud microphysical processes in clouds that contain ice, and hence the mechanisms that drive aerosol-cloud interactions, are different from the processes in warm marine clouds (see Section 2.2 below). Interactions between ice particles and liquid drops add additional levels of complexity to the aerosol effects (e.g., Morrison et al., 2012). As such, a set of questions could be asked about the effectiveness of MCB in ice-containing clouds in cold environments; such questions clearly have relevance for future climate. For example, increasing the shortwave reflectivity of the Arctic could offset some of the effects of reduced Arctic albedo due to receding sea ice. Moreover, if sufficient portions of the Arctic ocean become ice-free, Arctic shipping could increase, and hence the presence of ship tracks could be more prevalent. These effects could be counterbalanced by the longwave impacts of mixed-phase clouds, which are known to dominate cloud radiative effects in the Arctic (Morrison et al. 2012).

In this study, we use a cloud resolving model to assess some of the effects of introducing CCN into marine low clouds in the Arctic from a single point source representing a ship. This is one of the proposed methods in MCB of introducing CCN into marine low clouds. In particular, we address the following questions throughout the course of this paper:

1. According to our model simulations, does CCN injection in the Arctic increase low-cloud albedo?

2. Are the albedo effects of aerosol-cloud interactions stronger for supercooled liquid or mixed-phase clouds?

3. Is there a difference in albedo effects between injection of CCN into a clean environment
versus a polluted one?

2 Model and Methods

2.1 Model Setup

Our simulations are conducted using the Advanced Research Weather Research and Forecasting (WRF) model (Version 3.3; Skamarock et al. 2008), used as a cloud-resolving model. Third-order Runge-Kutta time stepping is applied to the dynamics. Fifth and third order advection is employed in the horizontal and vertical directions, respectively, with a monotonic limiter applied to the time integration scheme; this advection scheme is particularly important for the transport of tracers (Wang et al. 2009). The fine-resolution WRF model has been used in process-modeling studies of warm clouds and mixed-phase clouds and compared with other models participating in intercomparison cases (e.g., Wang et al. 2009; Ovchinnikov et al. 2014). It has proven to be a useful tool for studying aerosol-cloud interactions.

We use a modified version of the setup of the model intercomparison based on the Indirect and Semi-Direct Aerosol Campaign (ISDAC; McFarquhar et al. 2011; Ovchinnikov et al. 2014). The initial profile for our simulations is given in Table 1; the temperature and specific humidity in the free troposphere (altitudes above 1200 m) are nudged (Newtonian relaxation) toward this initial profile with a time scale of one hour, and winds at all levels are nudged with a time scale of two hours. This is accomplished by adding an unphysical tendency term to all of the nudged fields with a height-dependent maximum magnitude of $\Delta t \Delta \phi / (1 \text{ hour})$ or $\Delta t \Delta \phi / (2 \text{ hours})$, respectively, where $\Delta t$ is the model timestep of three seconds, and $\Delta \phi$ is the departure of the nudged field from the initial profile. The initial profile is characterized by an inversion at 825 m altitude and supersaturated conditions so
a cloud forms underneath the inversion level immediately after the simulation starts. This is a common setup for model intercomparisons of Arctic mixed phase clouds (Klein et al., 2009; Morrison et al., 2011; Ovchinnikov et al., 2014), all of which were based on observed temperature and humidity profiles. Ovchinnikov et al. 2014 specified nonzero wind shear in the initial meteorological profile, but this study specifies no initial wind shear so as not to further complicate detection of signals in our output; turbulence-induced wind shear is allowed to develop over the course of the simulation. The implications of this modification are discussed in Section 4. The surface pressure is specified to be 1020 mb, and the surface skin temperature is 267 K. The surface roughness length is 0.004 m (Morrison et al. 2011). Large-scale air subsidence is calculated by vertical integration of a specified horizontal wind divergence \((5 \times 10^{-6} \text{ s}^{-1})\) from the surface to the inversion, with zero divergence above the inversion, and is applied to temperature and humidity. Sensible and latent heat fluxes at the surface are set to zero for the entire simulation. The choices of no heat and moisture surface fluxes were to reduce the number of sources of externally-driven variability in model behavior. These values can be compared to observed values of sensible and latent heat fluxes of -2.07 and -5.09 W m\(^{-2}\), respectively, taken during August 2001 over a region of drifting pack ice (Birch et al. 2009). The implications of these choices are discussed in Section 4. A sensitivity study to explore different values of surface heat fluxes will be undertaken in the future. As is typical in process-model simulations, the domain is doubly periodic in the horizontal directions, without advective forcing for heat and moisture in the domain, although the nudging provides a source term for temperature and humidity to prevent significant drift in the environmental conditions. We include both shortwave and longwave radiation schemes (CAMRT; Collins et al. 2006). The boundary-layer turbulence is initiated via a small random perturbation to the temperature field. The 1.5 order turbulent
kinetic energy (TKE) closure is used to calculate subgrid-scale diffusion (Deardorff 1972).

The model domain is 120 km in the $x$ direction (400 grid cells, each 300 m in size), 60 km in the $y$ direction (200 grid cells, each 300 m in size), and 1.5 km in the $z$ direction (50 layers, each approximately 30 m thick). This is the same domain size used by Wang et al. (2011). The location of the domain is 71.32°N, 156.61°W, which is relevant only for shortwave radiative flux calculations. At this latitude, our model domain covers an area that is approximately equivalent to a single grid box corresponding to the current resolution of many global models.

### 2.2 Microphysical Parameterizations

A two-moment bulk microphysics scheme based on Morrison et al. (2005, 2009) is used in all model simulations. Some simulations have the ice processes switched off to focus on pure supercooled liquid clouds (Table 2). This scheme is one of the microphysical options in the WRF model; however, we have modified the default scheme for this study, mostly to accommodate the injection of aerosols from a moving point source. A prognostic variable for interstitial accumulation-mode aerosol number concentration has been added, as was done by Wang et al. (2009) for a different microphysical scheme. Droplet activation was parameterized following Abdul-Razzak and Ghan (2000) as a function of the vertical velocity, temperature, pressure, and aerosol size distribution parameters. Sub-grid scale vertical velocity is not parameterized, and all vertical velocities are calculated at the grid scale, which could potentially lead to underprediction of the number of cloud droplets that are activated. Aerosol particles are incorporated into liquid drops upon activation (i.e., a sink of particle number) and returned to the interstitial state after drop evaporation (i.e., a source of particle number) in any given grid box. It is assumed that each evaporated liquid drop releases one
single aerosol particle (Feingold et al. 1996; Mitra et al. 1992), indicating that drop coalescence acts as a removal process for aerosol particle number in addition to the sedimentation and ultimate loss of larger drops to the surface. Scavenging processes considered in the model simulations include nucleation scavenging, collision/coalescence, resuspension, and wet deposition; collision/coalescence and wet deposition directly reduce total CCN number concentration in the boundary layer. The present study includes heterogeneous nucleation from cloud droplet freezing, which is one major difference from the SHEBA and ISDAC intercomparisons (Morrison et al. 2011; Ovchinnikov et al. 2014).

The microphysical scheme uses a lognormal size distribution with a fixed modal radius of 0.1 $\mu$m and a geometric standard deviation of 1.5 to represent accumulation-mode aerosols. Therefore, the injected and resuspended aerosols do not modify the particle sizes and spectrum width. For simplicity, the coarse-mode aerosol is also switched off for all simulations. Ice nucleation through contact and immersion freezing of cloud droplets and immersion freezing of rain drops is included in the model. The effective diffusivity of contact ice nuclei, assuming a size of 0.1 $\mu$m, is based on Morrison and Pinto (2005). Immersion freezing of cloud droplets and rain drops follows the drop-volume dependent parameterization of Bigg (1953). In conditions of temperature less than -8°C and water saturation or ice supersaturation greater than 8%, deposition and condensation freezing nucleation processes are represented by a relaxation of the cloud ice number concentration to 0.16 L$^{-1}$ if the existing total ice (cloud ice, snow, and graupel) concentration falls below this value in one model time step.

For the purposes of activation into cloud droplets, aerosols are assumed to have the same properties as ammonium sulfate aerosols. Because these are the only aerosols considered in this study, and because ammonium sulfate aerosols are efficient CCN, we use the
terms aerosol and CCN interchangeably. We discuss the implications of this assumption in Section 4.

2.3 Simulation Design

We perform a suite of eight simulations, each lasting 30 hours, starting at 6 p.m. on 26 April and ending at 12 a.m. on 28 April (local time). In these simulations, we specify the background concentration of CCN, whether the ice processes were included or excluded, and whether CCN injection was included or excluded. Table 2 details the eight different simulations, as well as the naming conventions for each experiment. The “clean” case is considered to have an initial background CCN of 50 cm$^{-3}$, and the “polluted” case to have an initial background CCN of 200 cm$^{-3}$, consistent with global mean values (Latham, 2012). These values are also consistent with measurements taken in the Arctic. During the Mixed Phase Arctic Cloud Experiment (M-PACE; Verlinde et al. 2009), background CCN was measured to be approximately 40 cm$^{-3}$, and during SHEBA/FIRE-ACE, the background was approximately 200 cm$^{-3}$ (Curry et al. 2000; Fridlind et al. 2012).

Wang et al. (2011) included a uniform background source of CCN of 2 mg$^{-1}$ h$^{-1}$ in each grid box within the boundary layer to account for natural sea-salt emissions. This source term was also used to balance the loss of CCN due to coalescence of cloud drops and subsequent wet removal that may result in a super-clean collapsed boundary layer (Ackerman et al. 1993; Wang et al. 2010). In the current study, we did not include such a background source, as we were interested in diagnosing the ability of CCN injection to prevent the boundary layer from collapsing. Low CCN conditions that are incapable of sustaining a boundary layer structure have been observed in nature (Ackerman et al. 1993), so although such conditions are not necessarily ubiquitous, our simulation design is relevant to potential
real-world conditions.

To simulate particle injection, we follow the method of Wang et al. (2011). From the beginning of the simulation, CCN are emitted into the lowest atmospheric layer from a single moving point source (one grid box in size), representing injection from a ship. This design is equally applicable for both deliberate CCN injection (MCB) or emissions from cargo shipping. The CCN emission rate is $1.45 \times 10^6 \text{ m}^{-2} \text{s}^{-1}$ from a point source moving at 5 m s$^{-1}$, where the point source begins at the West side of the domain and travels Eastward; then because the domain is doubly periodic, the point source re-appears at the West side of the domain after it passes the East border and continues to emit particles, traveling Eastward. This emission rate of the total number of particles injected to the atmosphere is the same as suggested by Salter et al. (2008) for geoengineering purposes, except that the injected CCN take the same lognormal size distribution as the background aerosols rather than a uniform size. We did not consider the potential for in-plume aerosol coagulation (Stuart et al. 2013), instead implicitly assuming a sufficient amount of particles are injected to achieve the mass loading used in this study. The aerosol particles have composition properties corresponding to ammonium sulfate. Further investigations could explore the effects of particle composition on our results; we discuss some of the implications of this assumption in Section 4. CCN number is reported as two separate prognostic variables: the “active” CCN number is calculated based on full interactivity with cloud scavenging processes, and the “passive” CCN number does not include scavenging within clouds.

In all simulations of mixed-phase clouds, the ice processes were not included until two hours after the beginning of the simulations. This is consistent with the ISDAC model intercomparison; this specification was included to allow the boundary-layer turbulence to develop before the ice processes kick in (Ovchinnikov et al. 2014).
All reported values of liquid water path were calculated using cloud water only. Inclusion of rain in these calculations has negligible impacts on values of liquid water path. Conversely, as is shown in the following section, most of the ice in these simulations is in the form of snow, so all calculations of ice water path include ice crystals, snow, and graupel.

In Section 3.2, there is a discussion of cloud fraction. For the purpose of this calculation, a grid box is said to have liquid cloud if the cloud liquid water mixing ratio is at least 0.01 g kg$^{-1}$. A grid box has ice cloud if the ice water mixing ratio is at least $10^{-5}$ g kg$^{-1}$. Column cloud fraction is defined as the fraction of all model columns in which cloud optical thickness is at least 2.

### 3 Results

We begin our investigation of marine cloud brightening in Section 3.1 with a discussion of the injected particles, their transport, and their activation into cloud droplets. Section 3.2 explores changes in the clouds as a result of injection, including cloud fraction, cloud albedo, and cloud depth. Finally, Section 3.3 explores the susceptibility of the clouds to brightening.

#### 3.1 Particle Injection and Activation

Although our setup was different from the specifications given by Ovchinnikov et al. (2014) for the ISDAC intercomparison, we nevertheless find it useful to contextualize results from our non-injection simulations through a comparison with the results from that study. One major difference between our simulations and those of Ovchinnikov et al. (2014) is that in lieu of the complex ice particle activation scheme employed in our simulations, Ovchinnikov
et al. (2014) prescribed the ice nucleation rate to be

\[
\frac{dN_i}{dt} = \max \left( 0, \frac{N_{i0} - N_i}{\Delta t} \right) \quad S_i \geq 0.05 \text{ or } q_i \geq 0.001 \text{ g kg}^{-1}
\]

(1)

where \( N_i \) is ice crystal number, \( S_i \) is the supersaturation fraction, and \( q_i \) is the liquid water mixing ratio. \( N_{i0} \) is a prescribed target ice particle concentration; simulations were conducted for which \( N_{i0} = 0, 1, \) or \( 4 \text{ L}^{-1} \). Within the first eight hours of simulation (the duration of simulations described by Ovchinnikov et al., 2014), the liquid-only simulations in this study have similar liquid water paths to the \( N_{i0} = 0 \text{ L}^{-1} \) case (Figure 1c). Inclusion of ice processes yields liquid and ice water paths similar to \( N_{i0} = 1 \text{ L}^{-1} \) (Figure 1d). The liquid and ice water paths depicted in Figure 1 of the present study show greater spread after 12 hours of simulation, approximately incident with sunrise, so we are unable to definitively state that the results presented here exactly replicate a particular simulation described by Ovchinnikov et al. (2014). Moreover, the simulations in the ISDAC intercomparison were only performed for night times, so we do not know whether differences between simulations in the calculated liquid and ice water paths begin to grow larger due to the inclusion of diurnal variation or whether this divergence is simply due to a sufficiently long simulation time.

The idea of MCB hinges on the injected CCN activating into cloud droplets. CCN injection clearly results in an increase in CDNC (Figures 1a and 3a). In the absence of injection, the boundary layer collapses (i.e., cloud top decreases; Figures 2b and 4b) and clouds dissipate, coincident with a rapidly declining CDNC to 1-2 cm\(^{-3}\) by the end of the simulations. The decreasing trend of CDNC shows little dependence on whether ice processes are included, nor whether the background is clean or polluted. Despite having no sensible or latent heat fluxes at the surface, CDNC increases in the injection simulations, showing
that the boundary-layer turbulence, driven primarily by cloud radiative cooling, is sufficient to loft the injected particles from the surface into the cloud within about three hours.

The plots of CDNC show an oscillatory pattern for the injection simulations but not the no-injection simulations. This oscillation is likely in part due to the CCN point-source traversing the periodic domain repeatedly, which has a period of approximately 7 hours; CDNC values reach a dynamic balance between additional injection of CCN and scavenging of CCN via activation, collision/coalescence, and resuspension. There are additional oscillations with a period of approximately 3 hours. These could be due to local precipitation-dynamical interactions, or they could be related to the eddy turnover time in the decoupled boundary layer, but we are as of yet unable to attribute these oscillations to a particular mechanism.

Figure 5 shows transport efficiency, which we define as

$$\text{TE} = \frac{N_P - B}{N_{P,\text{surf}} - B}$$  \hspace{1cm} (2)

where $N_P$ is the number concentration of the passive tracer at each level, $N_{P,\text{surf}}$ is the number concentration of the passive tracer at the level of injection (the lowest model layer), and $B$ is the background concentration of particles (either 50 or 200 cm$^{-3}$). This quantity indicates how efficiently the injected particles are transported from the surface layer to other model layers in the absence of cloud scavenging processes. All simulations show very efficient transport at or below the cloud layer, in some cases exceeding 95%. In the second half of the simulation, some of the aerosol is transported into the free troposphere above the boundary layer top.

The sudden jump in TE within the first few hours of simulation (Figure 5) could be due to a transition from the initial part of the simulation, in which the cloud layer is decoupled
from the sub-cloud layer, to a state in which vertical transport from the surface to the cloud layer is stronger due to increased coupling. To ascertain the mechanisms behind this transition, Figure 6 shows the buoyancy flux of turbulence and the variances of the vertical velocity ($\sigma^2_w$) for all simulations. A decoupled system is sometimes characterized by negative buoyancy flux near the cloud base and a cloud-base minimum in $\sigma^2_w$ (Stevens et al. 2005); both of these features are seen early in the simulations. However, all simulations undergo a transition from negative buoyancy flux at the surface to near-neutral buoyancy flux and from $\sigma^2_w \approx 0$ at the surface to slightly positive values. Moreover, buoyancy fluxes and $\sigma^2_w$ values transition away from features indicating a de-coupled state to a more strongly coupled state. This suggests that there is sufficient vertical motion to transport the aerosols up into the cloud layer despite the lack of surface heat and moisture fluxes.

The scavenging efficiency (Figures 2a and 4a) is defined here as

$$SE = 1 - \frac{N_T}{N_{P\text{cloud}}}$$

where $N_T$ is the total number of cloud droplets (CDNC) plus the number of interstitial CCN, and $N_{P\text{cloud}}$ is the number concentration of the passive tracer that is in cloudy grid cells. Because $N_T$ is affected by scavenging processes, but $N_P$ is not, the quantity SE gives an indication of how efficiently the scavenging processes (collision/coalescence and precipitation) are operating to reduce the number of CCN. SE values increase over the course of the simulations due to scavenging of existing particles via precipitation (Figures 2c-d, 4c-d, and 10) or collision/coalescence; both of these processes reduce $N_T$. Figures 1e-f and 3e-f show that droplet size increases before the collapse of the boundary layer (formal definitions of cloud droplet effective radius as calculated in this study are given in Section 3.2); this is concurrent with increases in LWP and/or decreases in CDNC, indicating a combination of
growth of existing particles and collision/coalescence. Figures 2a and 4a show that SE is
lower for all injection simulations, indicating two main effects. The dominant effect of CCN
injection is to provide new particles for activation into cloud droplets, thus reducing SE. A
secondary effect is to increase the number of interstitial aerosols, thus increasing $N_T$.

SE values show almost no dependence upon whether ice processes are included in the
simulations, which is perhaps not surprising, as only a small fraction of droplets freeze and
form ice. In the no-injection cases, the background CCN number has little effect on SE.
However, injection into a relatively clean environment results in a lower SE than injection
into a polluted environment. Figures 1e and 3e show that injection into a clean regime
results in a lower cloud droplet size; smaller droplets have lower collection efficiencies, which
would contribute to a lower SE than in the no-injection simulations.

Wang and Feingold (2009) and Wang et al. (2011) showed that reduced precipitation
along the ship plume can induce dynamical feedbacks that lead to moist convergent flow
into the ship track, thickening clouds along the track but thinning the neighboring off-track
clouds. The thickened clouds eventually produce stronger precipitation, counteracting the
aerosol indirect effects. Likely due to the dry conditions, low liquid water paths (Figures
1c and 3c), and a more stable boundary layer in the Arctic than in the subtropical marine
boundary layer, the dominant modifications to cloud properties here can be explained by
the conventional aerosol indirect effects, without the additional complications of dynamical
effects due to precipitation. There are some dynamical circulation changes due to precipita-
tion, as is evident from the oscillation period discussed in Section 3.1, but the net effects on
clouds are small.

Figure 7 gives the spatio-temporal distribution of the injected particle concentration. The
off-track parts of the domain give an indication of behavior in the no-injection simulations.
The background concentration of particles decays due to cloud scavenging. The decay rate is dependent upon drop sizes and number concentration, and thus upon the background concentration of aerosols. The total particle number concentration ($N_T$) in $I_{200N}/L_{200N}$ takes over 6 hours longer to reach below $10 \, \text{cm}^{-3}$ than in $I_{50N}/L_{50N}$. CCN injection results in a steady accumulation of particles, reaching over $2000 \, \text{cm}^{-3}$ in some places directly along the center of the injection plume. The increase in the in-cloud particle number generally remains more confined to the center of the domain, spreading toward the edges more slowly.

The passive tracer is not scavenged by in-cloud processes, so it represents the maximum possible area affected by CCN injection. Comparisons to the results of Wang et al. (2011) reveal that the spreading of CCN throughout the domain is slower in the present study.

This model configuration is not equipped to provide calculations of supersaturation over liquid water for accurate calculation of liquid water condensation/evaporation. As such, the Wegener-Bergeron-Findelsen (WBF) process, whereby ice crystals grow at the expense of liquid droplets in conditions where the air parcel is supersaturated with respect to ice but subsaturated with respect to liquid water, is captured by the model, but the small liquid sub-saturation is not explicitly represented. Although the impact of CCN injection on droplet sizes does not directly affect droplet evaporation in the model, there is still substantial impact on liquid water through interactions with ice nucleation and growth. In the microphysical scheme used here, the initiation of ice nucleation by freezing droplets depends on the availability of droplet number, and the freezing rate increases with drop size. Both liquid water path and ice water path increase early in the simulations, with greater increases for the more polluted background and the injection cases. In the injection cases, liquid droplet number and ice crystal number also increase early in the simulations. This likely indicates that existing ice particles are increasing in size, and new particles are being
nucleated. Injection causes a substantial decrease in liquid water particle size and a slight increase in ice particle size. The total ice water path (Figures 1d and 3d) and ice water content (Figure 8) are much smaller than those of liquid clouds, and the majority of ice water is represented as snow.

The results presented here are consistent with frequently observed Arctic cloud regimes that are CCN-limited. With weak aerosol sources, effective wet deposition of CCN, and aerosol-cloud-precipitation interactions, Arctic clouds can become depleted (Mauritsen et al. 2011). These situations resemble our no-injection simulations, which are characterized by low CCN and low amounts of cloud cover. This suggests that the regions of boundary layer collapse shown in multiple figures are not necessarily uncommon in the Arctic.

3.2 Cloud Extent and Albedo Changes

The most salient question for MCB is whether our results indeed show cloud brightening, i.e., whether there are increases in cloud extent and cloud albedo due to particle injection. Because total liquid water is, to a large extent, controlled by ice nucleation processes, which differ between models, caution should be used when generalizing these results.

The model does not explicitly include calculations of cloud albedo, so we represent this quantity with a parameterization. The cloud top effective radius ($r_e$) can be approximated as

$$r_e \approx 1.08 r_v$$  \hspace{1cm} (4)

where $r_v$ is the cloud drop mean volume radius (Rosenfeld et al. 2012). Liquid cloud optical depth ($\tau$) is calculated as

$$\tau = \frac{3}{2 \rho_w} \frac{LWP}{r_e}$$  \hspace{1cm} (5)
where $\rho_w$ is the density of liquid water, and LWP is the liquid water path (Stephens 1978).

From this, liquid cloud albedo ($\alpha$) can be calculated using the two-stream approximation (Bohren 1987):

$$\alpha = \frac{(1 - g)\tau}{2 + (1 - g)\tau}$$  \hspace{1cm} (6)

where $g$ is the (dimensionless) asymmetry parameter of the cloud droplets, assumed here to be 0.85.

Ice processes require a different formulation for calculating cloud albedo. Due to significant heterogeneity of ice crystal shapes, there is no standardized concept for an ice crystal effective radius (McFarquhar and Heymsfield, 1998). As a proxy, we use the volume mean radius of the ice crystals if taken as spherical particles:

$$r_v = \left[ \frac{\text{IWP}}{N_i} \frac{3}{4\pi\rho_i} \right]^{1/3}$$  \hspace{1cm} (7)

where IWP denotes the ice water path (ice crystals only), $N_i$ is the number of ice crystal particles, and $\rho_i$ is the density of ice, taken here to be 0.9 g cm$^{-3}$ (Morrison and Grabowski, 2008). Stephens et al. (1990) define the ice cloud droplet effective radius in terms of equivalent volume spheres, yielding a similar value to that of Equation 5. As such, our gross simplification is potentially reasonable for bulk parameterizations. Regardless, as we discuss below, ice optical depth is far lower than liquid optical depth, so the simplification used in Equation 7 should have negligible effects on our results. Given the vast uncertainty inherent in calculations of ice cloud albedo, we are unlikely to find a substantially better estimate of ice cloud albedo without detailed computations of the scattering phase functions of the implicitly assumed particle shapes (Mishchenko et al. 1996).

Taking the values calculated in Equation 7 as the ice crystal volume mean radius ($r_v$),
Ebert and Curry (1992) provide bulk calculations for ice cloud optical thickness ($\tau$; visible wavelengths) and asymmetry parameter ($g$):

$$\tau = \text{IWP} \cdot (3.448 \times 10^{-3} + 2.431/r_v); \quad (8)$$

$$g = 0.7661 + 5.851 \times 10^{-4} \cdot r_v \quad (9)$$

where IWP has units of g m$^{-2}$ and $r_v$ has units of $\mu$m. Ice cloud albedo can then be calculated using the two stream approximation, as in Equation 6.

Domain-averaged liquid cloud drop effective radius increases throughout the simulations until the timing of the collapse of the boundary layer, at which point it sharply decreases (Figure 1e). Similarly, ice crystal volume mean radius remains relatively constant until collapse (Figure 1f). CCN injection results in a vastly reduced liquid cloud drop effective radius, consistent with the first aerosol indirect effect, although liquid water content increases along the injection track over the initial part of the simulation (Figures 1e and 8). As multiple fields in Figures 1-4 show, the boundary layer collapse occurs in all no-injection simulations, indicating that CCN injection as simulated here is sufficient to prevent this collapse. Collapsed regions are characterized by small-scale convection that shows no evidence of organization (not shown); collapsed regions also arise in the injection simulations away from the injection track. Because these simulations have no surface fluxes and no wind shear in the initial meteorological profile, small-scale convection is likely driven primarily by radiation, possibly in combination with latent heat exchange below the cloud base. This collapse results in substantial decreases in liquid and ice water path, and the collapse is delayed by approximately six hours in the polluted case as compared to the clean case. There are no large differences in liquid cloud droplet effective radius between mixed-phase and liquid-only
Cloud fraction shows substantial differences between liquid-only and mixed-phase simulations (Figures 2g and 4g). In the liquid-only simulations, cloud fraction decreases substantially (> 40%) when the boundary layer collapses (also indicated by a substantial decrease in cloud top; Figures 2b and 4b), and cloud fraction remains lower throughout the remainder of the simulation. The timing of the decrease in cloud fraction is consistent with diurnal variation. Conversely, in the mixed-phase simulations, the cloud fraction decreases when the boundary layer collapses, but it then returns to cover nearly the entire domain. Liquid cloud albedo also decreases in all simulations, coincident with the boundary layer collapse (Figures 2e and 4e). Although domain-averaged ice cloud albedo decreases throughout the simulations in the mixed-phase cases (Figures 2f and 4f), it increases in areas where the liquid cloud albedo decreases (Figure 9). This suggests that inclusion of ice processes results in a layer of optically thin ice clouds that has greater thickness in areas of less liquid cloud cover. Liquid clouds are the dominant source of reflectivity (Figures 2, 4, and 9).

The substantial differences between the mixed phase simulations and the liquid-only simulations are likely due to the WBF process. Even slightly cooler temperatures in mixed-phase clouds (as compared to liquid-only clouds) can sufficiently lower the minimum supersaturation over ice required to form ice crystals, allowing the WBF process to occur. The consequent creation of ice cloud lowers the liquid water path, reducing shortwave absorption. As such, we would expect the changes in cloud fraction to be consistent with diurnal variations in shortwave radiation.

All simulations have initial increases in albedo due to CCN injection; the maximum increase in domain albedo among all simulations is 0.23 (Figure 2e). Figure 9 shows an initial increase in ice albedo along the CCN injection track, providing additional evidence...
that CCN injection accelerates the WBF process.

CCN injection suppresses liquid precipitation (Figures 1c and 10), consistent with the second aerosol indirect effect. However, unlike the results of Wang et al. (2011), dynamical changes in circulation due to modification of precipitation are small, possibly in part because of the stability of the boundary layer. As such, we do not find regions of reduced albedo on the edges of the center track. The clouds along the injection track tend to persist and spread out over time, also consistent with the second indirect effect. In the mixed-phase simulations, although liquid precipitation is suppressed along the injection track, snow precipitation predominantly occurs along the track (Figure 10).

3.3 Cloud Susceptibility

Cloud albedo susceptibility can provide a useful indication of cloud modification in response to aerosols. Susceptibility of the cloud to brightening is given by

$$S = \frac{\mathrm{d} \ln \alpha}{\mathrm{d} \ln N_T}$$

(10)

where $\alpha$ is the cloud albedo, and $N_T$ is the total number of potential CCN, taken here to be the sum of CDNC and interstitial CCN. Platnick and Twomey (1994) define susceptibility with the denominator only including CDNC and not interstitial CCN. We have opted for a modified definition in our study to include albedo increases for all potential CCN, not just active cloud droplets. One could analogously define the susceptibility of liquid water path to particle injection. We do not include such calculations here, as the low liquid water paths in our simulations make calculations of susceptibility very sensitive to variability.

Figure 11 shows a joint histogram of $N_T$ and $\alpha$, from which susceptibility can be inferred.
Comparison of the injection and no-injection simulations reveals two different regimes in each
simulation. The bow-shaped pattern that appears in all panels of Figure 11 are due to cloud
particles that are off the main injection track; the shape of this pattern is likely due to
the diurnal variation in liquid and ice water path (although conclusive attribution to this
mechanism is beyond our capabilities in the present study). The values corresponding to the
injection track are shown as a curve with lower frequency of occurrence that spans a larger
range of values of \( N_T \). If only considering the values corresponding to the injection track, \( \alpha \)
shows a positive increase with \( N_T \), consistent with the first indirect effect that introducing
additional CCN will increase albedo. The concavity of these curves in Figure 11 is due to a
saturation effect: as additional CCN become CDNC, the relative amount of available water
for condensational growth of existing particles decreases, so changes in the size of the cloud
droplets are smaller.

As shown in the previous sections, the cloud layer in our simulations is, to some extent,
decoupled from the surface and is not heavily precipitating. This situation may not be appli-
cable to all meteorological conditions in the Arctic (e.g., M-PACE), and thus the calculations
of susceptibility presented here may differ for different situations (Klein et al. 2009).

4 Discussion and Conclusions

Our results show that injection of aerosols into the Arctic marine boundary layer, either
deliberately (geoengineering) or due to other mechanisms that would increase CCN in the
Arctic region, has the potential to brighten low clouds. Most of the albedo effects occur
in the liquid phase, the features of which are consistent with current knowledge of aerosol
indirect effects. In the simulations that include ice processes, some of the liquid water is con-
verted to ice, resulting in low in-cloud ice water content. Injection of CCN into mixed-phase
clouds results in smaller liquid water increases than in the liquid-only simulations through interactions with ice nucleation and growth. The boundary layer collapse is coincident with substantial precipitation of water (Figure 10), both in liquid and ice form. The precipitation may indeed contribute toward the boundary layer collapse through reductions in CCN and liquid water path. Injection of CCN into a relatively unpolluted environment results in greater albedo increases than injection into polluted environments, consistent with current knowledge about aerosol-cloud interactions.

The mechanisms governing the albedo and lifetime increases of the clouds in our simulations are straightforward. Evidence for the first and second aerosol indirect effects is clearly visible, with few complicating factors, unlike the results of Wang et al. (2011) for warm clouds, in which precipitation induced strong dynamical circulation changes in cloud cover. The lack of strong circulation changes in our study is likely due to a combination of low temperatures and low precipitation, resulting in small latent heating and cooling, and thus limiting mesoscale dynamical changes. The effects of including surface sensible and latent heat fluxes on dynamical circulation changes will be addressed in future work; these choices could partially explain both the initial decoupling of the cloud layer from the subcloud layer and the substantially lower liquid and ice water paths than in M-PACE, which was a more well-mixed case (Klein et al. 2009). Altering the surface fluxes could in turn change the susceptibility of the clouds to changes in aerosol concentration. Insofar as the microphysical schemes used in our simulations accurately represent the processes involved in MCB, the microphysical effects of Arctic CCN injection appear to be more predictable than those of MCB in warm clouds, although some of this predictability is likely due to our simplified experimental design (no initial wind shear, no surface fluxes, and a particular ice nucleation parameterization). Our results show this holds for both liquid-only and mixed-
phase cold clouds. As stated in Section 3.2, the results presented here are likely dependent upon the microphysical scheme used and may not be broadly generalizable (e.g., Morrison et al., 2011). As of yet, the effects of the initial meteorological profile on our results are unclear, particularly the effects of the strong inversion present in our setup. Potential future work could include initial profiles that correspond to measurements taken during other field campaigns, such as M-PACE or SHEBA. Moreover, a horizontal grid spacing of 300 m may not fully resolve large eddies in a boundary layer that is 1.5 km deep; further investigations could explore the effects of horizontal resolution on our results.

Entrainment processes at the top of marine low clouds occur in very thin layers (Stevens et al., 2005). Our chosen vertical resolution of $\sim 30$ m is too coarse to accurately capture these features. We performed additional simulations with a vertical resolution of $\sim 10$ m but did not find any substantive changes in our results (not pictured). The results of Stevens et al. (2005) suggest that to properly resolve entrainment issues would require vertical resolution that is an order of magnitude finer than in our simulations, accompanied by commensurately fine horizontal resolution to properly resolve eddy structure. The required computational power to conduct the present simulations with such fine resolution is beyond our means. The coarse vertical resolution of our simulations could result in underrepresentation of many different processes, each of which either enhances or diminishes entrainment. As such, we are unable to make conclusions about the effects of vertical resolution on our results. Regardless, we believe the effect of entrainment on mixing was reasonably captured in our simulations, based on previous studies of subtropical marine boundary layer clouds with a similar model configuration (Wang et al., 2011).

To put our results into context, we can do a simple back-of-the-envelope calculation of the effects of Arctic MCB on the global radiation budget, assuming that the results in our
domain can be extrapolated to all open ocean regions of the Arctic. We focus only on open ocean areas, as sea ice and snow-covered land already have higher albedos than marine low clouds, so brighter clouds over these regions will have minimal impacts on the radiation budget. As an illustration, we determined open ocean fraction in the Arctic (66.56°N to 90°N) as calculated from monthly mean sea ice extent data for 2012 (Meier et al., 2013; Peng et al., 2013). Taking the maximum domain albedo increase from our results of 0.23, the additional radiative forcing from Arctic geoengineering would be an average of -0.45 W m\(^{-2}\) globally, or -10.94 W m\(^{-2}\) over the Arctic. At most, the radiative impacts would have a small (although potentially non-negligible) effect on the global radiation budget, suggesting Arctic MCB could not serve as the sole means of offsetting the net radiative forcing from greenhouse gas emissions. However, the local effects on the Arctic radiation budget could be quite substantial, even if the actual effect is an order of magnitude smaller than the maximum effect as calculated from our results. These results may also be diminished if cooling causes sea ice growth, reducing the area of open ocean. Moreover, longwave forcing from Arctic clouds has a positive correlation with liquid water path, which is enhanced by CCN injection (Shupe and Intrieri, 2004). This increase in longwave forcing could offset some of the shortwave forcing from brightening, although the increase in longwave surface cloud forcing saturates at liquid water path values of approximately 60 g m\(^{-2}\), so determining the net impact of this longwave effect is not straightforward. The increase in downwelling longwave radiation could also increase latent heating and hence moisture flux from the surface into the clouds, forming a feedback loop (Garrett and Zhao, 2006; Lubin and Vogelmann, 2006; Morrison et al. 2012). For low liquid water paths (<30-50 g m\(^{-2}\)), there is also a droplet size effect on longwave cloud forcing (Garrett and Zhao, 2006; Lubin and Vogelmann, 2006).

The simulations presented here involve injection of CCN with aerosol properties, including
hygroscopicity and thermodynamics related to cloud particle activation, corresponding to ammonium sulfate. Explicitly representing a different type of aerosol (the most commonly studied aerosol for MCB is sea salt) could modify the effectiveness of MCB. Partanen et al. (2012) and Alterskjær et al. (2013) showed the aerosol direct effect of sea salt aerosols could be a substantial portion of the total radiative impact of MCB. Jenkins and Forster (2013) explicitly modeled the effects of creating CCN via evaporating sea water droplets; including these mechanisms can lessen albedo increases, in some cases nearly negating the aerosol indirect effects. Moreover, the size of the sea salt aerosols can affect the results due to a competition effect between the injected sea salt and other particles. Further studies could incorporate all of these effects by explicitly simulating microphysics specific to sea salt aerosols and how they differ as compared to sulfate aerosols. Alternatively, simulating different aerosols that serve as effective ice nuclei could result in more water being retained as ice, enhancing ice cloud albedo.

Cloud ice crystal size is assumed to have a fixed size distribution width, although the modal radius is allowed to vary. Ovchinnikov et al. (2014) found that ice water path, and hence calculations of cloud albedo (Equation 6), is strongly dependent upon accurate representations of the ice crystal size distribution. According to the results of Ovchinnikov et al., our calculations of ice water path may have been underestimated, implying our results for ice albedo could be more dramatic. Replications of our study with different microphysical schemes could be useful in verifying our results.

The clean cases have a collapsed boundary layer away from the injection track, consistent with features described by Ackerman et al. (1993) and Wang et al. (2010). No simulation shows indications of organized convection or cellular structure. This is possibly in part due to holding heat and moisture surface fluxes at zero (Kazil et al. 2014); a future study is
planned that will investigate the effects of these fluxes on the results presented here. The boundary layer collapse could also be due to the choice of not including a background aerosol source, as discussed in Section 2.3. Although having no aerosol source in the model domain is unrealistic for a 30 hour period, our simulations include the process of gradual aerosol removal. As such, our simulations show the impact of aerosol injection into a wide variety of different aerosol and cloud background conditions. Another potential exploration for the lack of cellular structure is reduced evaporative cooling below the cloud base due to the small liquid water content as compared to warm clouds. In the current setup, when cloud particles evaporate and resuspend the aerosols, the aerosols return to their original size, whereas they should grow in size due to collision/coalescence. Thus the resuspended aerosols are too small, making them less effective CCN than they would otherwise be if aerosol mass were conserved. Conserving aerosol mass, such as in the scheme of Lebo and Morrison (2013) could delay the collapse of the boundary layer, although further simulations would be needed to test this. A further reason for the boundary layer collapse could be the choice of no initial wind shear. In the presence of stronger shear throughout the simulation, the injected CCN would be distributed more evenly across the domain, which could prevent boundary layer collapse in regions away from the injection track and help to maintain the cloud through increased turbulence. Inclusion of initial shear may also reduce the oscillatory patterns in Figures 1-2, as the redistribution of the CCN by wind would mean that additional injection would not be into such CCN-rich areas.

Some of the results show a dependence upon the diurnal cycle, although we are unable to make firm conclusions about the effects of diurnal variation from our study. We only simulated one full diurnal cycle, so it is unclear whether the features we show have some component of diurnal variation that is masked by the transient nature of the simulations.
Moreover, we used a particular shortwave radiation scheme; different radiation schemes may have different impacts on the results. Isolating the effects of the boundary layer collapse, as well as inclusion of surface sensible and latent heat fluxes, could give a better indication of the timing of shortwave impacts on our results.

Our results only represent process-level studies. Determining the actual effects of CCN injection into the Arctic marine boundary layer, either inadvertently or advertently, would require a great deal of further work. Moreover, there are many concerns with geoengineering that are not represented here, all of which would be assessed by appropriate governance structures before a decision to deploy geoengineering is made. Nevertheless, process-modeling studies like ours can be useful in determining some of the behaviors and underlying physical mechanisms behind natural and anthropogenic emissions of CCN into Arctic marine low clouds.

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Table 1: The initial meteorological profile used in all simulations. $\theta$ denotes potential temperature (K), $q$ is the total water mixing ratio (g kg$^{-1}$), $u$ and $v$ are horizontal wind speeds in the $x$ and $y$ directions, respectively (m s$^{-1}$), and $z$ denotes altitude (m).

<table>
<thead>
<tr>
<th>Altitude (m)</th>
<th>$\theta$ (K)</th>
<th>$q$ (g kg$^{-1}$)</th>
<th>$u$ (m s$^{-1}$)</th>
<th>$v$ (m s$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0-400</td>
<td>$265 + 0.004(z - 400)$</td>
<td>$1.5 - 0.00075(z - 400)$</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>400-825</td>
<td>265</td>
<td>1.5</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>825-1500</td>
<td>$266 + (z - 825)^{0.3}$</td>
<td>1.2</td>
<td>0</td>
<td>0</td>
</tr>
</tbody>
</table>
Table 2: Naming conventions and descriptions of the eight simulations used in this study. “I” indicates inclusion of ice processes, and “L” indicates liquid only. “N” indicates no CCN injection, and “G” indicates CCN injection (geoengineering). Further descriptions of the sensitivity studies performed here are given in Section 2.

<table>
<thead>
<tr>
<th>Name</th>
<th>Background CCN (cm$^{-3}$)</th>
<th>Ice processes included</th>
<th>CCN Injection (geoengineering)</th>
</tr>
</thead>
<tbody>
<tr>
<td>I50N</td>
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</tr>
<tr>
<td>I50G</td>
<td>50</td>
<td>yes</td>
<td>yes</td>
</tr>
<tr>
<td>I200N</td>
<td>200</td>
<td>yes</td>
<td>no</td>
</tr>
<tr>
<td>I200G</td>
<td>200</td>
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<td>yes</td>
</tr>
<tr>
<td>L50N</td>
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</tr>
<tr>
<td>L50G</td>
<td>50</td>
<td>no</td>
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</tr>
<tr>
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<td>200</td>
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<td>no</td>
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<tr>
<td>L200G</td>
<td>200</td>
<td>no</td>
<td>yes</td>
</tr>
</tbody>
</table>
Figure 1: Domain-averaged time series of a) liquid droplet number; b) ice crystal number; c) liquid water path; d) ice water path (includes ice crystal, snow, and graupel); e) liquid cloud droplet effective radius (Equation 4); and f) ice crystal volume mean radius (Equation 7). Values in panels a, c, and e are averaged over all grid boxes containing liquid cloud. Panels b, d, and f are averaged over grid boxes containing ice. Experiments are listed in Table 2.
Figure 2: Domain-averaged time series of a) scavenging efficiency (Equation 3); b) cloud top; c) liquid precipitation; d) ice precipitation; e) liquid cloud albedo (Equation 6); f) ice cloud albedo (Equations 5, 6, 7); and g) cloud fraction. Panels e and f are averaged over the entire domain; grid boxes with no cloud are given an albedo of 0. Panel g is calculated by dividing the total number of columns with cloud by the total number of columns in the domain. Criteria for determining cloudy grid cells are given at the end of Section 2.3. Experiments are listed in Table 2.
Figure 3: Same as Figure 1 but showing differences between injection vs no injection (dashed lines) and inclusion of ice processes vs exclusion of ice processes (solid lines).
Figure 4: Same as Figure 2 but showing differences between injection vs no injection (dashed lines) and inclusion of ice processes vs exclusion of ice processes (solid lines).
Figure 5: Transport efficiency (Equation 2) for all injection simulations (Table 2). All values shown are calculated only from passive tracers. Thick black line indicates the base of the cloud.
Figure 6: Buoyancy flux source of TKE (shading; cm$^2$ s$^{-3}$) and vertical velocity variance (contours; m$^2$ s$^{-2}$) for each experiment as a function of time ($x$-axis) and height ($y$-axis). Buoyancy flux is calculated as $(g/\theta_v)\overline{w'\theta'}$, where $g$ is acceleration due to gravity, $\theta_v$ is virtual potential temperature, $w$ is vertical velocity, a bar indicates the domain mean, and a prime indicates the perturbation from the mean. Vertical velocity variance is calculated as $\overline{w'w'}$. 
Figure 7: Shading shows $N_T$ (CDNC plus interstitial CCN), and contours show $N_P$ minus the background CCN (cm$^{-3}$; See Section 3.1). $x$-axis indicates time, and $y$-axis indicates distance in the $y$ direction (km). $N_T$ values are averages in the $x$ and $z$ directions over all grid boxes containing cloud, and $N_P$ values are averages in the $x$ and $z$ directions over all grid boxes in the domain. Only injection simulations are shown, as behavior of no-injection simulations can be inferred from values away from the center of the domain. Contours are placed at 50, 100, 200, 500, 1000, and 2000 cm$^{-3}$. 
Figure 8: Liquid and ice water content after 12 hours of simulation (6 a.m. local time). Ice water content consists of ice crystals, snow, and graupel. $x$-axis indicates horizontal distance (km) in the $y$-direction, and $y$-axis indicates height (km).
Figure 9: Cloud albedo (liquid or ice) for each injection simulation as a function of time (x-axis) and y-dimension (y-axis). All values are averaged in the x direction and show values for cloud only, not the entire domain. y-axis indicates horizontal distance and has units of km. Only injection simulations are shown, as behavior of no-injection simulations can be inferred from values away from the center of the domain. Ice cloud albedo has no values for the first two hours, as ice processes were not included during this time period (Section 2.3).
Figure 10: Precipitation rate (rain and snow; mm day$^{-1}$) for all simulations. x-axis indicates time, and y-axis indicates distance in the y-direction (km). All values are averaged in the x direction.
Figure 11: Joint histogram of CDNC plus interstitial CCN ($N_T$) and cloud albedo ($\alpha$) for each simulation. Values show frequency of occurrence of each binned combination of $N_T$ and $\alpha$. Each $N_T$ value (See Section 3.1) indicates an average in the $x$ and $z$ directions over all grid boxes containing clouds. Each $\alpha$ value indicates an average in the $x$ direction over all columns containing clouds (liquid or ice, reported separately). Susceptibility is defined as $d \ln \alpha / d \ln N_T$ (Equation 10) and can be inferred from the joint histogram. Histograms show results for all times after the first three hours of simulation to allow the cloud layer and ice processes to fully develop.