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

Ben Kravitz¹, Hailong Wang¹, Philip J. Rasch¹, Hugh Morrison² and Amy B. Solomon³

¹Atmospheric Sciences and Global Change Division, Pacific Northwest National Laboratory, PO Box 999, MSIN K9-24, Richland, WA 99352, USA
²Mesoscale and Microscale Meteorology Division, NCAR Earth System Laboratory, National Center for Atmospheric Research, Boulder, CO 80301, USA
³University of Colorado Cooperative Institute for Research in Environmental Sciences, NOAA Earth System Research Laboratory, National Oceanic and Atmospheric Administration, Boulder, CO 80309-0216, USA

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 owing 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 [1]. 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 [2]. 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 [3,4]. These aerosol effects are most dramatically seen in marine low clouds, which cover on average 34.0% of the ocean surface [5].

Evidence for the aerosol indirect effects can be seen in ship tracks (brighter clouds owing to injection of particles from ship plumes) [6,7] and in process modelling studies [8,9]. Although ship plumes do not always result in brighter clouds [10], this concept, in part, inspired Latham [11] 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. Owing to the ubiquity of marine low clouds, it has been estimated that a 4% increase in global cloud fraction [12] or a 6% increase in albedo of existing marine low clouds [13] could offset atmospheric warming owing to a doubling of the CO₂ concentration from preindustrial times. Numerous modelling studies have found that with sufficient, controlled aerosol injection, global warming could be offset, although MCB may not return other fields, such as temperature and Arctic sea ice, to their previous levels [14].

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. [15] showed that achievable CDNC correlates with updraft velocity, explaining in part why the resulting CDNC in simulations by Korhonen et al. [16] were substantially lower than in simulations by Partanen et al. [17], who used more realistic updraft velocities. Many of these microphysical processes operate on the subgrid 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. [18] 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. [19] 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-modelling studies, the injected aerosol particles were assumed to be uniformly distributed in the model grid box (a volume of the order of 10⁶ m³) within seconds. Stuart et al. [20] explicitly modelled an aerosol injection plume, showing that owing to in-plume aerosol coagulation, the number of aerosols that reach the cloud layer strongly depends upon meteorological conditions.

These past process-modelling 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
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>
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<tbody>
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<td>1.5 + 0.00075($z - 400$)</td>
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<td>0</td>
</tr>
<tr>
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</tr>
<tr>
<td>825–1500</td>
<td>266 + ($z - 825$)$^{0.3}$</td>
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</table>

warm marine clouds (see §2b). Interactions between ice particles and liquid drops add additional levels of complexity to the aerosol effects [21]. 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 owing 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 [21].

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.

(i) According to our model simulations, does CCN injection in the Arctic increase low-cloud albedo?
(ii) Are the albedo effects of aerosol–cloud interactions stronger for supercooled liquid or mixed-phase clouds?
(iii) Is there a difference in albedo effects between injection of CCN into a clean environment versus a polluted one?

2. Model and methods

(a) Model set-up

Our simulations are conducted using the Advanced Research Weather Research and Forecasting (WRF) model (v. 3.3) [22], 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 [23]. The fine-resolution WRF model has been used in process-modelling studies of warm clouds and mixed-phase clouds and compared with other models participating in intercomparison cases [23,24]. It has proven to be a useful tool for studying aerosol–cloud interactions.

We use a modified version of the set-up of the model intercomparison based on the Indirect and Semi-Direct Aerosol Campaign (ISDAC) [24,25]. 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) towards this initial profile with a time scale of 1 h, and winds at all levels are nudged with a time scale of 2 h. This is accomplished by adding an unphysical tendency term to all of the nudged fields with a height-dependent maximum magnitude of $\Delta t \Delta \phi/(1 \text{ h})$ or $\Delta t \Delta \phi/(2 \text{ h})$, respectively, where $\Delta t$ is the model timestep of 3 s, 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 set-up for model
intercomparisons of Arctic mixed-phase clouds [24,26,27], all of which were based on observed temperature and humidity profiles. Ovchinnikov et al. [24] specified non-zero wind shear in the initial meteorological profile, but the present 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 §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 [27]. 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 0 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 behaviour. These values can be compared with 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 [28]. The implications of these choices are discussed in §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) [29]. 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 [30].

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 approx. 30 m thick). This is the same domain size used by Wang et al. [18]. 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.

### (b) Microphysical parametrizations

A two-moment bulk microphysics scheme based on Morrison et al. [31,32] 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

<table>
<thead>
<tr>
<th>name</th>
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<th>ice processes included</th>
<th>CCN injection (geoengineering)</th>
</tr>
</thead>
<tbody>
<tr>
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</tr>
<tr>
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<tr>
<td>L200G</td>
<td>200</td>
<td>no</td>
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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 §2.
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. [23] for a different microphysical scheme. Droplet activation was parametrized following Abdul-Razzak & Ghan [33] as a function of the vertical velocity, temperature, pressure and aerosol size distribution parameters. Subgrid scale vertical velocity is not parametrized, 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 [34,35], 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. This study includes heterogeneous nucleation from cloud droplet freezing, which is one major difference from the SHEBA and ISDAC intercomparisons [24,36].

The microphysical scheme uses a lognormal size distribution with a fixed modal radius of 0.1 μm and a geometrical 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 the sake of 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 μm, is based on Morrison & Pinto [37]. Immersion freezing of cloud droplets and rain drops follows the drop-volume-dependent parametrization of Bigg [38]. 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 timestep.

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 §4.

(c) Simulation design

We perform a suite of eight simulations, each lasting 30 h, starting at 18.00 on 26 April and ending at 00.00 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 [39]. These values are also consistent with measurements taken in the Arctic: during the mixed phase Arctic cloud experiment (M-PACE) [40], background CCN was measured to be approximately 40 cm−3, and during SHEBA/FIRE-ACE, the background was approximately 200 cm−3 [41,42].

Wang et al. [18] 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 owing to coalescence of cloud drops, and subsequent wet removal that may result in a super-clean collapsed boundary layer [43,44]. In this 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 [43], 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. [18]. 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 $5 \text{ m s}^{-1}$, where the point source begins at the west side of the domain and travels eastwards; then, because the domain is doubly periodic, the point source reappears at the west side of the domain after it passes the east border and continues to emit particles, travelling eastwards. This emission rate of the total number of particles injected to the atmosphere is the same as suggested by Salter et al. [45] 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 [20], 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 §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 2h 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 [24].

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 §3, 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 §3a, 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 $0.01 \text{ g kg}^{-1}$. 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 MCB in §3a with a discussion of the injected particles, their transport and their activation into cloud droplets. Section 3b explores changes in the clouds as a result of injection, including cloud fraction, cloud albedo and cloud depth. Finally, §3c explores the susceptibility of the clouds to brightening.

(a) Particle injection and activation

Although our set-up was different from the specifications given by Ovchinnikov et al. [24] 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. [24] is that in lieu of the complex ice particle activation scheme employed in our simulations, Ovchinnikov et al. [24] prescribed the ice nucleation rate to be

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

otherwise,

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 $N_{i0} = 0, 1, \text{ or } 41^{-1}$. Within the first 8h of simulation (the duration of simulations described by
Figure 1. Domain-averaged timeseries 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 (3.4)); and (f) ice crystal volume mean radius (equation (3.7)). 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. (Online version in colour.)

Ovchinnikov et al. [24]), the liquid-only simulations in this study have similar liquid water paths to the $N_{li}=10^{-1}$ case (figure 1c). Inclusion of ice processes yields liquid and ice water paths similar to $N_{li}=10^{-1}$ (figure 1d). The liquid and ice water paths depicted in figure 1 of this study show greater spread after 12 h 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. [24]. 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 owing to the inclusion of diurnal variation or whether this divergence is simply owing 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 2a). In the absence of injection, the boundary layer collapses (i.e. cloud top decreases; figures 3b and 4b) and clouds dissipate, coincident with a rapidly declining CDNC to $1-2 \text{ cm}^{-3}$ 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 3 h.
The plots of CDNC show an oscillatory pattern for the injection simulations, but not the no-injection simulations. This oscillation is likely, in part, owing to the CCN point source traversing the periodic domain repeatedly, which has a period of approximately 7 h; 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 h. 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

\[
\text{TE} = \frac{N_P - B}{N_P^{\text{surf}} - B},
\]

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.
Figure 3. Domain-averaged timeseries of (a) scavenging efficiency (equation (3.3)); (b) cloud top; (c) liquid precipitation; (d) ice precipitation; (e) liquid cloud albedo (equation (3.6)); (f) ice cloud albedo (equations (3.5)–(3.7)); and (g) cloud fraction. Panels (e, 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 §3c. Experiments are listed in table 2. (Online version in colour.)

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 subcloud layer, to a state in which vertical transport from the surface to the cloud layer is stronger owing 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 ($\sigma_{w}^{2}$) for all simulations. A decoupled system is sometimes characterized by negative buoyancy flux near the cloud base and a cloud-base minimum in $\sigma_{w}^{2}$ [46]; both 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 $\sigma_{w}^{2} \approx 0$ at the surface to slightly positive values. Moreover, buoyancy fluxes and $\sigma_{w}^{2}$ values transition away from features indicating a decoupled 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 3a and 4a) is defined here as

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

where $N_T$ is the total number of cloud droplets (CDNC) plus the number of interstitial CCN, and $N_{\text{Pcloud}}$ is the number concentration of the passive tracer that is in cloudy grid cells. Because $N_T$ is affected by scavenging processes, but $N_{\text{P}}$ 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 owing to scavenging of existing particles via precipitation (figures 3c,d, 4c,d and 10) or collision/coalescence; both these processes reduce $N_T$. Figures 1e,f and 2e,f show that droplet size
Figure 5. Transport efficiency (equation (3.2)) for all injection simulations (table 2). All values shown are calculated only from passive tracers. Thick black line indicates the base of the cloud. (Online version in colour.)

Figure 6. Buoyancy flux source of TKE (shading; \text{cm}^2 \text{s}^{-3}) and vertical velocity variance (contours; \text{m}^2 \text{s}^{-2}) for each experiment as a function of time (x-axis) and height (y-axis). Buoyancy flux is calculated as \( (g/\bar{\theta}_v)\bar{w}'\theta'_v \), 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 \( w'w' \). (Online version in colour.)
increases before the collapse of the boundary layer (formal definitions of cloud droplet effective radius as calculated in this study are given in §3b); this is concurrent with increases in liquid water path and/or decreases in CDNC, indicating a combination of growth of existing particles and collision/coalescence. Figures 3a 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 2e 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 & Feingold [9] and Wang et al. [18] 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 neighbouring off-track clouds. The thickened clouds eventually produce stronger precipitation, counteracting the aerosol indirect effects. Likely owing to the dry conditions, low liquid water paths (figures 1c and 2c), 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 owing to precipitation. There are some dynamical circulation changes owing to precipitation, as is evident from the oscillation period discussed in §3a, 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 behaviour in the no-injection simulations. The background concentration of particles decays owing 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 I200N/L200N takes over 6 h longer to reach less than 10 cm$^{-3}$ than in I50N/L50N. CCN injection results in a steady accumulation of particles, reaching more than 2000 cm$^{-3}$ in some places directly along the centre of the injection plume. The increase in the in-cloud particle number generally remains more confined to the centre of the domain, spreading towards 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 with the results of Wang et al. [18] reveal that the spreading of CCN throughout the domain is slower in this 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 subsaturation 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 2d) and ice water content (figure 8) are much smaller than those of liquid clouds, and the majority of ice water is represented as snow.
Figure 7. Shading shows $N_I$ (CDNC plus interstitial CCN), and contours show $N_P$ minus the background CCN (cm$^{-3}$; see §3a). The $x$-axis indicates time, and $y$-axis indicates distance in the $y$-direction (km). $N_I$ 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 behaviour of no-injection simulations can be inferred from values away from the centre of the domain. Contours are placed at 50, 100, 200, 500, 1000, and 2000 cm$^{-3}$. (Online version in colour.)

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 [36]. 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.

(b) 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 owing 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 parametrization. The cloud top effective radius ($r_e$) can be approximated as

$$r_e \approx 1.08 r_v,$$

where $r_v$ is the cloud drop mean volume radius [47]. Liquid cloud optical depth ($\tau$) is calculated as

$$\tau = \frac{3}{2 \rho_w} \frac{\text{LWP}}{r_e},$$

where $\rho_w$ is the density of water.
Figure 8. Liquid and ice water content after 12 h of simulation (06.00 local time). Ice water content consists of ice crystals, snow and graupel. The x-axis indicates horizontal distance (km) in the y-direction, and y-axis indicates height (km). (Online version in colour.)

where $\rho_w$ is the density of liquid water, and LWP is the liquid water path [48]. From this, liquid cloud albedo ($\alpha$) can be calculated using the two-stream approximation [49]:

$$\alpha = \frac{(1 - g)r}{2 + (1 - g)r},$$  \hspace{1cm} (3.6)

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. Owing to significant heterogeneity of ice crystal shapes, there is no standardized concept for an ice crystal effective radius [50]. As a proxy, we use the volume mean radius of the ice crystals if taken as spherical particles:

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

(3.7)

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\(^{-3}\) [51]. Stephens et al. [52] define the ice cloud droplet effective radius in terms of equivalent volume spheres, yielding a similar value of optical depth to that of equation (3.5). As such, our gross simplification is potentially reasonable for bulk parametrizations. Regardless, as we discuss below, ice optical depth is far lower than liquid optical depth, so the simplification used in equation (3.7) 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 [53].

Taking the values calculated in equation (3.7) as the ice crystal volume mean radius \( (r_v) \), Ebert & Curry [54] provide bulk calculations for ice cloud optical thickness \( (\tau; \text{visible wavelengths}) \) and asymmetry parameter \( (g) \):

\[ \tau = \text{IWP} \left( 3.448 \times 10^{-3} + \frac{2.431}{r_v} \right), \]

(3.8)

and

\[ g = 0.7661 + 5.851 \times 10^{-4} \cdot r_v, \]

(3.9)

where IWP has units of g m\(^{-2}\) and \( r_v \) has units of \( \mu \text{m} \). Ice cloud albedo can then be calculated using the two-stream approximation, as in equation (3.6).

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 6 h in the polluted case when compared with the clean case. There are no large differences in liquid cloud droplet effective radius between mixed-phase and liquid-only phase simulations.

Cloud fraction shows substantial differences between liquid-only and mixed-phase simulations (figures 3g and 4g). In the liquid-only simulations, cloud fraction decreases substantially (greater than 40%) when the boundary layer collapses (also indicated by a substantial decrease in cloud top; figures 3b 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 3e and 4e). Although domain-averaged ice cloud albedo decreases throughout the simulations in the mixed-phase cases (figures 3f 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 the areas of less liquid cloud cover. Liquid clouds are the dominant source of reflectivity (figures 3, 4 and 9).

The substantial differences between the mixed-phase simulations and the liquid-only simulations are likely owing to the WBF process. Even slightly cooler temperatures in mixed-phase clouds (when compared with 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 owing to CCN injection; the maximum increase in domain albedo among all simulations is 0.23 (figure 3e). 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. [18], dynamical changes in circulation owing 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 centre 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).

**Figure 10.** Precipitation rate (rain and snow; mm day\(^{-1}\)) for all simulations. The x-axis indicates time, and y-axis indicates distance in the y-direction (km). All values are averaged in the x-direction. (Online version in colour.)
Cloud albedo susceptibility can provide a useful indication of cloud modification in response to aerosols. Susceptibility of the cloud to brightening is given by

\[ S = \frac{\text{d} \ln \alpha}{\text{d} \ln N_T}, \quad (3.10) \]

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 & Twomey [55] 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 is 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 this 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 applicable to all meteorological conditions in the Arctic (e.g. M-PACE), and thus the calculations of susceptibility presented here may differ for different situations [26].

4. Discussion and conclusion

Our results show that injection of aerosols into the Arctic marine boundary layer, either deliberately (geoengineering) or owing 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 converted 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. Precipitation may indeed contribute to 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. [18] 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
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 §3a) 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 (3.10)) and can be inferred from the joint histogram. Histograms show results for all times after the first 3 h of simulation to allow the cloud layer and ice processes to fully develop. (Online version in colour.)

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 [26]. 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 parametrization). Our results show this holds for both liquid-only and mixed-phase cold clouds. As stated in §3b, the results presented here are likely dependent upon the microphysical scheme used and may not be broadly generalizable [36]. 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 set-up. 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 [46]. Our chosen vertical resolution of approximately 30 m is too coarse to accurately capture these features. We performed additional simulations with a vertical resolution of approximately 10 m but did not find any substantive changes in our results (not pictured). The results of Stevens et al. [46] 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 under-representation 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 [18].

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–90°N) as calculated from monthly mean sea ice extent data for 2012 [56,57]. Taking the maximum domain albedo increases from our results of 0.23, the additional radiative forcing from Arctic geoengineering would be an average of $-0.45 \text{ W m}^{-2}$ globally, or $-10.94 \text{ W m}^{-2}$ 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 [58]. 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 [21,59,60]. For low liquid water paths (less than 30–50 g m$^{-2}$), there is also a droplet size effect on longwave cloud forcing [59,60].

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. [17] and Alterskjær et al. [61] showed the aerosol direct effect of sea salt aerosols could be a substantial portion of the total radiative impact of MCB. Jenkins & Forster [62] explicitly modelled 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 owing 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 when compared with 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. [24] found that ice water path, and hence calculations of cloud albedo (equation (3.6)), 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. [61] and Wang et al. [44]. 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 [63]; 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 §2c. Although having no aerosol source in the model domain is unrealistic for a 30h 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 owing to the small liquid water content when compared with warm clouds. In the current set-up, when cloud particles evaporate and resuspend the aerosols, the aerosols return to their original size, whereas they should grow in size owing 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 & Morrison [64], 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 and 3, 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 simulated only 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 represent only 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-modelling studies like ours can be useful in determining some of the behaviours and underlying physical mechanisms behind natural and anthropogenic emissions of CCN into Arctic marine low clouds.

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