https://doi.org/10.5194/acp-2021-537
Preprint. Discussion started: 12 July 2021

Atmospheric Chemistry and Physics Discussions

The effect of marine ice nucleating particles on mixed-phase clouds

Tomi Raatikainen[1], Marje Prank[1], Jaakko Ahola[1], Harri Kokkola[2], Juha Tonttila[2], and Sami Romakkaniemi[2]

[1]Finnish Meteorological Institute, Helsinki, Finland
[2]Finnish Meteorological Institute, Kuopio, Finland

Correspondence: Tomi Raatikainen (tomi.raatikainen@fmi.fi)

Abstract. Shallow marine mixed-phase clouds are important for the radiative balance, but modelling their formation and dynamics is challenging. These clouds depend on boundary layer turbulence and cloud top radiative cooling, which is related to the cloud phase. The fraction of frozen droplets depends on the availability of suitable ice nucleating particles (INPs), which initiate droplet freezing. While desert dust is the dominating INP type in most regions, remote boundary layer clouds are dependent on local marine INP emissions, which are often related to biogenic sources including phytoplankton. Here we use high resolution large eddy simulations to examine the potential effects of marine emissions on boundary layer INP concentrations and their effects on clouds. Surface emissions have a direct effect on INP concentration in a typical well-mixed boundary layer whereas a steep inversion can block the import of background INPs from the free troposphere. The importance of the marine source depends on the background INP concentration, so that marine emissions become dominant with low background concentrations. For the INP budget it is also important to account for INP recycling. Finally, with the high-resolution model we show how ice nucleation hotspots and high INPs concentrations are focused on updraught regions. Our results show that marine INP emissions contribute directly to the boundary layer INP budget and therefore have an influence on mixed-phase clouds.

1 Introduction

Stratocumulus clouds are shallow and thin clouds that cover large parts of the oceans and for this reason they have a significant effect on the radiative balance (Wood, 2012). Large uncertainties are related to mixed-phase clouds which contain both liquid cloud droplets and frozen particles (e.g., Korolev et al., 2017). Although this state is unstable as ice crystals tend to grow with the expense of liquid droplets, boundary layer mixed-phase clouds can persist for several hours and even days (e.g., Morrison et al., 2012). Ice crystal number concentration is important for the balance as too high concentration will lead to cloud glaciation (e.g., Murray et al., 2021).

Shallow mixed-phase clouds are typically in the temperature range from -20 to -10 °C, which means that a seed called ice nucleating particle (INP) is needed for the ice formation (e.g., Murray et al., 2012; Kanji et al., 2017). Heterogeneous ice formation (or nucleation) includes deposition of water vapour on a dry particle or the freezing can start at the surface of an insoluble particle immersed in a liquid droplet (e.g., Hoose and Möhler, 2012). Contact nucleation refers to a case where freezing happens right after a collision between liquid droplet and an INP (e.g., Ladino Moreno et al., 2013). Immersion

https://doi.org/10.5194/acp-2021-537
Preprint. Discussion started: 12 July 2021


Atmospheric Chemistry and Physics Discussions

freezing is the dominating ice nucleation mode for the temperature and humidity conditions in typical marine mixed-phase clouds (e.g., Murray et al., 2012).

Models based on the classical nucleation theory (e.g., Khvorostyanov and Curry, 2004; Chen et al., 2008; Hoose et al., 2010) can be adjusted to match with laboratory observations from typical freezing experiments (e.g., Murray et al., 2011; Hoose and Möhler, 2012). This adjustment relies mostly on INP specific parameters such as the contact angle (e.g., Chen et al., 2008; Ervens and Feingold, 2012, 2013; Ickes et al., 2017). Using these parametrizations in simulating ice formation is complicated by the fact that the ambient INP population is a complex mixture of different chemical species. Although field observations can provide the number of ice crystals as a function of temperature and humidity (e.g., Hartmann et al., 2020), there is limited information about the ambient INP distribution at least for the level of detail that is needed for ice nucleation modelling.

The most important INPs for shallow clouds include dust, soot and biogenic particles (e.g., Hoose et al., 2010). Desert dust is globally the most common INP type, and it includes several different mineral compositions mainly related to their source regions (e.g., Boose et al., 2016; Kok et al., 2021). Relatively high desert dust concentrations can be seen at continental outflow regions, but concentrations are lower especially in the Southern hemisphere (e.g., Prospero et al., 2002). In the absence of dust, local marine INP sources are important especially for the Southern Ocean (e.g., Burrows et al., 2013; Vergara-Temprado et al., 2017; Zhao et al., 2021; Huang et al., 2021).

At moderate wind speeds, sea spray aerosol is produced mainly by bubbles bursting at the sea surface (e.g., Mårtensson et al., 2003). Soluble sea salt aerosol is generally quite poor INP, but sea spray contains other material that may initiate droplet freezing. For example, dust may be re-emitted from the sea surface (Cornwell et al., 2020). Currently, most research is focused on biogenic or organic material that can act as an INP. There are experiments showing that artificially generated sea spray aerosol contains INPs (e.g., Wilson et al., 2015; DeMott et al., 2016; McCluskey et al., 2018b; Wolf et al., 2020; Gong et al., 2020; Ickes et al., 2020; Mitts et al., 2021) and studies on ambient INPs linked to marine origin (e.g., Hartmann et al., 2020; McCluskey et al., 2018a, c). Most of these studies link INPs to phytoplankton biological activity, which is typically related to chlorophyll concentrations. The actual INPs can be composed of molecules, intact cells, or microbe fragments (e.g., Burrows et al., 2013; McCluskey et al., 2018b; Knopf et al., 2018).

The details of the marine INPs related to their origin, emission rates and ice nucleation properties are still highly unclear. Nevertheless, there are a few large-scale studies exploring the potential importance of marine INPs (e.g., Burrows et al., 2013; Vergara-Temprado et al., 2017; Huang et al., 2018; Zhao et al., 2021). The results depend strongly on the assumed properties of marine INPs, but these studies support the view on important role of marine INPs in remote high latitude regions and at low altitudes. These regions are dominated by low-level mixed-phase clouds, which are known to be problematic for large scale models due their coarse resolution. Cloud resolving models are more suitable tools for exploring the effects of boundary layer dynamics on marine and dust INPs, and their interactions with clouds.

In this study we explore the potential effects of marine INPs on mixed-phase boundary layer clouds by using UCLALES-SALSA, which a cloud resolving large eddy simulator (LES) coupled with detailed aerosol-cloud-ice microphysics (Tonttila et al., 2017; Ahola et al., 2020). Specifically, we will examine how marine INP emissions impact boundary layer INP concen-

https://doi.org/10.5194/acp-2021-537
Preprint. Discussion started: 12 July 2021


Atmospheric Chemistry and Physics Discussions

trations and vertical distributions when compared with the effects of background dust aerosol and dust entrained from the free troposphere. We will also examine the impacts of INPs on mixed-phase cloud dynamics and stability.

2 Methods

2.1 The ISDAC case study

Our LES simulations are based on a case study, which is briefly presented here. Observations from the Semi-Direct Aerosol Campaign (ISDAC) focused on Arctic mixed-phase clouds (McFarquhar et al., 2011). Ovchinnikov et al. (2014) used these to derive setups for a LES model inter-comparison study. The setup is based on the observed single-layer mixed-phase stratiform cloud, which persisted for 15 hours. Most ice particles were pristine dendrite crystals and drizzle was absent, which justified the exclusion of ice aggregation and warm rain processes. The case study included several LES models with different ice microphysics. To remove the effect of different ice nucleation parametrizations, all models were configured to use an approach where the ice number concentration was set to a specific value ranging from zero up to 4 L^{-1}. In this diagnostic approach, in-cloud ice nucleation rate was calculated so that the target ice crystal number concentration was reached.

2.2 LES modelling

Current simulations are made with large eddy simulator UCLALES-SALSA. This model is based on the commonly used UCLALES (Stevens et al., 1999, 2005; Stevens and Seifert, 2008) where cloud microphysics is replaced by the SALSA aerosol module (Kokkola et al., 2008, 2018) extended for warm (Tonttila et al., 2017) and mixed-phase (Ahola et al., 2020) clouds. Because UCLALES-SALSA has been described in previous publications, only a brief description of SALSA is given here. Aerosol, cloud droplet and ice particle chemical composition (water, dust, sulphate, organics, etc.) and size distributions are described using sectional approach based on dry particle size bins. Water vapour partitioning is based on diffusion limited non-equilibrium growth (except for aerosol when RH<98 %) and this determines cloud activation, which takes place when aerosol wet size exceeds the critical droplet size. Common cloud microphysical processes like sedimentation and coagulation are implemented for each hydrometeor. Different ice nucleation modes were implemented, but here the focus is on immersion freezing. It is based on the stochastic approach presented by Khvorostyanov and Curry (2000), where freezing rates depend on ambient conditions (temperature and relative humidity) and properties of the INP (particle composition and ice nucleation parameters).

Ahola et al. (2020) used the ISDAC case study (Ovchinnikov et al., 2014) to validate the newly implemented ice microphysics, and then briefly compared simulations with modelled ice nucleation (cloud droplet freezing based on prognostic INPs) to those with diagnostic ice nucleation (cloud droplet freezing rate calculated based on the target ice number concentration of 4 L^{-1}). Due to the relatively high temperature (minimum temperature about 258 K or -15 °C just below the cloud top), it was assumed that droplet freezing occurred via the immersion freezing mechanism. Because the focus was on aerosol-cloud-ice interactions instead of the details of the freezing mechanism, suitable ice crystal number concentrations were obtained by

https://doi.org/10.5194/acp-2021-537
Preprint. Discussion started: 12 July 2021

Atmospheric Chemistry and Physics Discussions

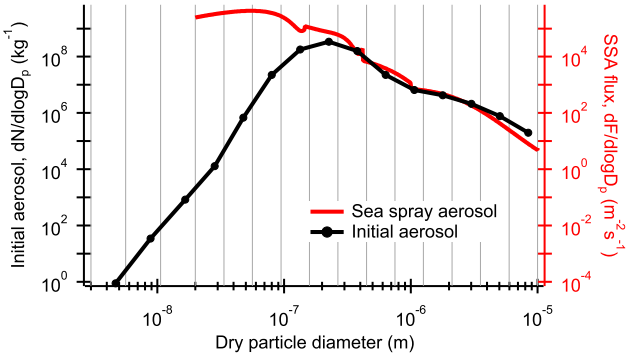


Figure 1. Initial aerosol size distribution for the ISDAC case (black line, left axis) and parametrized sea spray aerosol (SSA) emission flux for 6 $\mathrm{m\,s^{-1}}$ wind speed and 271.15 K temperature (red line, right axis). Vertical grey lines represent aerosol dry size bin limits for SALSA

adjusting the initial concentration of INPs. INPs were assumed to be composed of dust, which is the insoluble material that can initiate freezing, and soluble ammonium bisulphate with equal dry particle volume fractions. All other ice nucleation parameters had their default values, except cosine of the contact angle was increased from 0.50 to 0.57 to enhance freezing at these relatively high temperatures. The only adjustable parameter in these simulations was the fraction of externally mixed INPs in the initial aerosol size distribution (set to 0.00015). With this fraction, the initial ice crystal number concentration was about 4 $\mathrm{L^{-1}}$ as in the simulation with diagnostic cloud droplet freezing. Comparison between diagnostic and prognostic ice nucleation simulations showed the importance of modelled freezing rate and INP circulation on cloud stability (Ahola et al., 2020).

Here we examine how an additional or substitutive continuous marine INP source changes the situation. Most model settings are the same as in the original ISDAC case study (Ovchinnikov et al., 2014) and implemented as described in Ahola et al. (2020). These include bimodal initial ammonium bisulphate aerosol size distribution (see Fig. 1), simplified microphysics (disabled all collision processes, no rain, and assuming a spherical low-density ice particles), parametrized radiation scheme, large scale subsidence based on a constant divergence ($Q = 1.5 \cdot 10^{-6}$ $\mathrm{s^{-1}}$), zero surface sensible and latent heat fluxes, and a weak nudging of winds and free tropospheric humidity and temperature towards their initial values. The original case study had initial temperature and humidity profiles that represent de-coupled boundary layer, but eventually the boundary layer became coupled in the model simulations (Ovchinnikov et al., 2014). To allow vertical mixing from the start, we initialize simulations with well-mixed profiles (constant liquid water potential temperature and total water mixing ratio in the boundary layer). The effect of boundary layer de-coupling on marine INP emissions will be examined in Sect. 3.5.

Ice nucleation follows the approach used in Ahola et al. (2020). Background INPs are composed of dust and ammonium bisulphate, and these are an externally mixed fraction of the initial aerosol. Droplet freezing occurs via the immersion freezing mechanism and it is limited to cloud droplets. The effect of allowing aerosol freezing (interstitial INPs and those outside clouds) will be examined in Sect. 3.5. In the following simulations we examine the potential effects of marine INPs on mixed-phase clouds with different background INP concentrations. With this approach we will have a range of simulations representing cases from purely marine to purely background INPs. The downside of including background INPs in these simulations is that

https://doi.org/10.5194/acp-2021-537
Preprint. Discussion started: 12 July 2021


Atmospheric Chemistry and Physics Discussions

we can have externally mixed ammonium bisulphate aerosol and INP modes, which is critical for modelling ice nucleation, but marine and background INPs become internally mixed. This is a limit set by our model where the number of externally mixed particle modes is limited to two. Due to this limitation and the fact that properties of marine INPs are practically unknown, we assume that physically marine INPs are like those in the background aerosol (internally mixed "dust" and ammonium bisulphate) and that these particles are emitted as an externally mixed fraction of the sea spray aerosol (SSA) flux. In practise, we adjust the fractions of INPs in the initial background aerosol and SSA flux so that a wide range of marine INP contributions can be examined.

Here we use an upgraded version of UCLALES-SALSA where size-dependent SSA emissions are parametrized as a function of the domain mean wind speed at the height of 10 m and sea surface temperature (here constant 271.15 K). For the dry particle size range 0.020–1 μm the parametrization is from Mårtensson et al. (2003) and for the 1–10 μm size range it is from Monahan et al. (1986). For the latter size range, the temperature dependency term is from Jaeglé et al. (2011). Figure 1 shows the SSA flux for 6 $\mathrm{m\,s^{-1}}$ wind speed and 271.15 K sea surface temperature. If the sea spray aerosol is evenly mixed within the boundary layer (assuming top at 840 m and 1.3 $\mathrm{kg\,m^{-3}}$ air density), aerosol number concentration would increase by $1{\cdot}10^{6}$ $\mathrm{kg^{-1}}$ per hour. This is a small number compared with the initial total aerosol number concentration $165{\cdot}10^{6}$ $\mathrm{kg^{-1}}$.

Figure 1 shows the initial aerosol size distribution for the ISDAC simulations (Ovchinnikov et al., 2014). It is based on observations by Earle et al. (2011) covering a size range from 100 nm to about 10 μm, so the number concentration of sub-100 nm particles is most likely underestimated. This explains the difference between aerosol size distribution and SSA flux parametrization. These sub-100 nm particles have a minor role for clouds, because there are enough larger particles. For the current simulations, the first SALSA particle size range for the nucleation mode (bins 1–3) covers dry diameters from 3 to 20 nm while the default upper limit is 50 nm. In addition, the number of bins in the second size range was increased from the default of seven to twelve. This improves the size resolution for cloud droplets and ice particles, which are limited to the second size range. Externally mixed INPs are described by using another set of bins from the second size range for aerosol, cloud droplets and ice particles (so-called b-bins). In practice, the a-bins describe the distributions of soluble ammonium bisulfate aerosol and related cloud droplets (no ice in the a-bins in the absence of ice nuclei), and b-bins describe aerosol INPs and related INP-containing cloud droplets and ice crystals.

As mentioned above, reasonable ice crystal number concentrations are targeted primarily by adjusting the fractions of INPs in the initial background aerosol and in the SSA flux. Although ice crystal number concentrations as high as 4 $\mathrm{L^{-1}}$ were used in the ISDAC simulations (Ovchinnikov et al., 2014), observations made by Hiranuma et al. (2013) showed that ice number concentrations were about 0.4 $\mathrm{L^{-1}}$. High ice number concentrations are interesting, because these can lead to complete cloud glaciation. Therefore, we will focus on the range from the observations (0.4 $\mathrm{L^{-1}}$) up to 4 $\mathrm{L^{-1}}$.

https://doi.org/10.5194/acp-2021-537
Preprint. Discussion started: 12 July 2021


Atmospheric Chemistry and Physics Discussions

3 Results and discussion

3.1 Cloud response to INPs

As the first step we made eight simulations where marine INP emissions were either on or off and the initial background aerosol INP number concentration had four different values. Marine INP emissions are specified as a fraction of the SSA flux, and here emissions on and off mean fractions 0.005 and 0.0, respectively. Initial background INP number concentrations are specified as a fraction of the initial aerosol, and here the cases are called zero (fraction is 0.0), low (0.00001), medium (0.00005), and high (0.00010). Background INP fractions were selected so that the simulations without marine emissions cover the range from an ice-free case up to a cloud that is becoming mostly glaciated. When marine emissions are switched on, the fraction of INPs in the sea spray aerosol is high enough to have an impact on clouds. Simulation time was set to 24 h including a 1 h spin-up for SSA emissions and a 2 h spin-up for ice microphysics. Because most adjustments take place during the first 10 h and the trends are steady after 12 h, we will focus on the first 16 hours.

Results from our simulations are shown in Fig. 2. Cloud base and top heights represent the domain minimum and maximum values, respectively. Ice crystal number concentrations are averaged over grid cells where ice mass mixing ratio exceeds $1 \cdot 10^{-8}$ $\mathrm{kg\,kg^{-1}}$. Liquid (LWP) and ice (IWP) water paths are domain mean values. Simulations where marine INP emissions are switched on and off are shown with the solid and dashed lines, respectively. The effect of marine INP emissions is clearly seen in the initial ice crystal number concentration trends, which means that the aerosol is effectively transported from the sea surface up to the cloud layer (between 600 m and 850 m). IWP depends mostly on ice crystal number concentration because the mean ice crystal diameters are similar in all simulations (400–430 $\mu\mathrm{m}$ at 10 h). Condensible water is limited so an increase in IWP is seen as a decrease in LWP. For this reason, ice number concentration is the most important parameter for these mixed-phase clouds.

Marine INP emissions become more important with decreasing background INP concentration. In fact, marine INP emissions alone seem to have the same effect as having the medium INP background without marine INP emissions. There are two simulations, both without marine INP emissions, where the INP concentration is so low that the result is a thick almost purely liquid cloud. The other simulations end up to ice number concentration of about 2000 $\mathrm{kg^{-1}}$ (IWP about 10 $\mathrm{g\,m^{-2}}$), because precipitation increases with increasing INP concentration. This is the case even with the highest INP concentrations, but then there is also a reduction in LWP after the first 8 hours, which leads to a mostly glaciated cloud state.

The cloud starts to glaciate (LWP decreases and IWP increases) rapidly when ice crystal number concentration approaches 3000 $\mathrm{kg^{-1}}$, which was already confirmed in our previous study (Ahola et al., 2020), but in this case the limit can be exceeded due to an additional INP sea surface source. The drop in the ice number concentration in these simulations after 8 h is related to precipitation. In fact, the removal of INPs with precipitation saves the cloud from complete glaciation, but it also removes part of the condensible water. This has an impact on cloud stability as cloud top radiative cooling requires liquid water. The reduction in liquid water content explains why cloud top heights decrease in the three simulations with the lowest LWPs.

https://doi.org/10.5194/acp-2021-537
Preprint. Discussion started: 12 July 2021

Atmospheric Chemistry and Physics Discussions

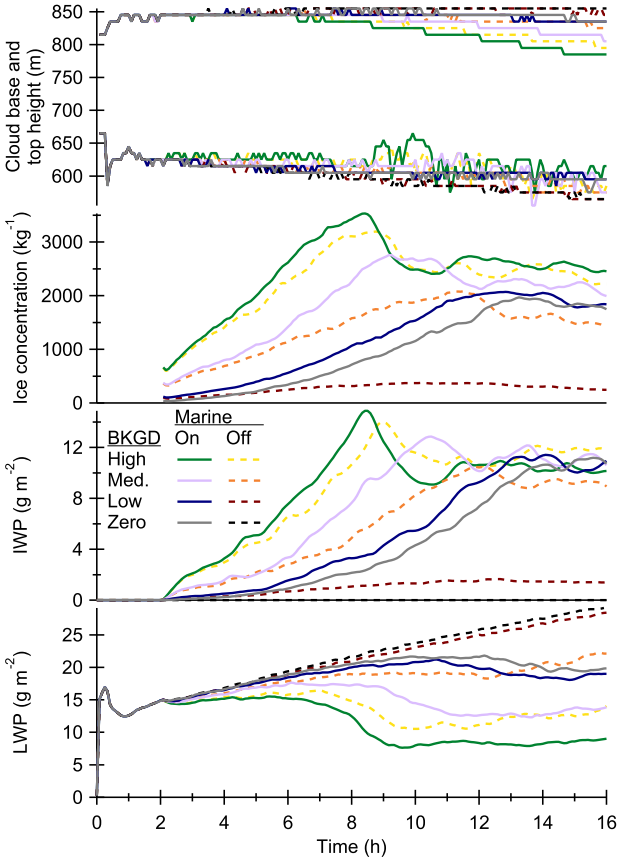


Figure 2. Time series of cloud base and top heights, ice crystal number concentration, and ice (IWP) and liquid (LWP) water paths from the eight model simulations with different background (BKGD) aerosol INP concentrations (zero, low, medium, and high) and marine INP emissions switched on (solid lines) or off (dashed lines).

3.2 INP budget

Cloud development in these simulations depend mainly on ice crystal number concentration, which is related to the availability INPs, so here we focus on the INP budget. Figure 3 shows the changes in column total INP mass (left) and number (right) concentrations due to the common removal and production mechanisms. Simulations where marine INP emissions are switched
180 off are excluded for clarity. The INP mass includes the total dust mass in aerosol, cloud droplets and ice crystals. Calculations cover the whole domain, but changes are negligible in the free troposphere. INP mass is related to large particles which are effective INPs, but number concentration depends mostly on small particles that are simply too small to be effective ice nuclei (size distribution shown in Fig. 1). This is not an issue for precipitation, because it includes only ice crystals. To have a more realistic estimate of the INP number budget, subsidence and surface fluxes include particles larger than 159 nm in dry diameter.

https://doi.org/10.5194/acp-2021-537
Preprint. Discussion started: 12 July 2021

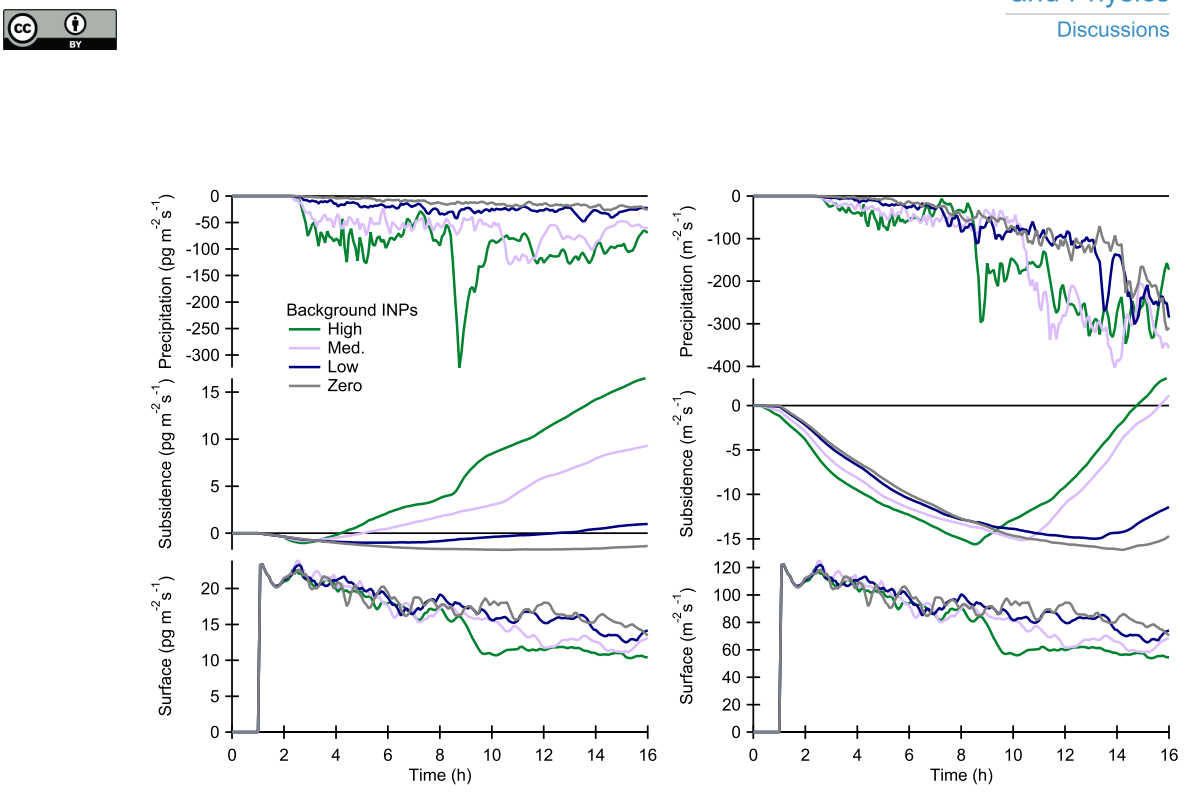


Figure 3. The three mechanisms (precipitation, subsidence and surface emissions) affecting on INP mass (left) and number (right) concentrations. Simulations are initialized with different background INP concentrations and marine INP emissions are switched on. The effects of surface emissions and subsidence on INP number concentration are calculated for particles larger than 159 nm in dry diameter.

This is the lower limit of the first size bin (Fig. 1) that has a significant fraction of ice in all our simulations. Because this limit is somewhat subjective and time and case dependent, we will focus more on the INP mass.

Precipitation is the main INP removal mechanism, and it can easily exceed production. In fact, the total INP mass is decreasing in all other simulations except the one without background INPs (Fig. 4). Surface INP emissions are a fraction (0.005) of the total SSA flux (Fig. 1), which depends on the 10 m wind speed (approx. 5–6 $\mathrm{m\,s^{-1}}$) and sea surface temperature (fixed to 271.15 K). Changes in the 10 m wind speed cause the slow decrease in surface fluxes. Subsidence is described by a downward vertical velocity related to altitude (z) and the fixed large scale divergence $Q = 1.5 \cdot 10^{-6}\ \mathrm{s^{-1}}$. Subsidence velocity ($= Qz$) is applied to all prognostic variables and it has an effect whenever there are vertical concentration gradients. Subsidence has a fairly small impact on INPs due to competing effects: while subsidence brings INP-rich aerosol from the free troposphere, it depletes boundary layer cloud and ice species at the same time (profiles discussed below). In addition, subsidence has a negative effect when surface concentration is increased due to marine INP emissions. This is the reason for the clear decrease in INP number and also for the initial decrease in INP mass. Subsidence becomes significant INP source when the initially high boundary layer concentration decreases due to precipitation while that at the free troposphere stays high. However, subsidence continues to have a small influence when the initial INP concentration is low enough to avoid the rapid removal of boundary layer INPs.

https://doi.org/10.5194/acp-2021-537
Preprint. Discussion started: 12 July 2021

Atmospheric Chemistry and Physics Discussions

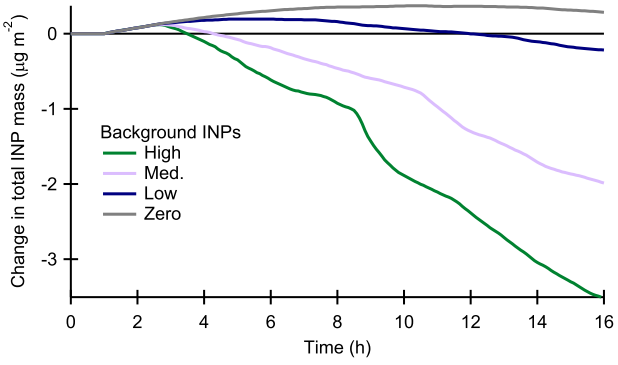


Figure 4. Changes in the column integrated INP mass from the initial values. The initial values for the four different background INP concentrations (high, medium, low and zero) are 9.6, 4.8, 1.0 and 0.0 μg m^{-2}.

Figure 5 shows horizontally averaged profiles of the main processes affecting on vertical INP mass distributions from simulations with (solid lines) and without (dashed lines) marine INP emissions (high background INP concentration). Corresponding INP mass mixing ratios in each phase and in total are shown in the right panel. The profiles are averages of the instantaneous model outputs (produced after every 300 s; original units used) over simulation time 7–8 h. This time interval was selected as an example, because the highest ice number concentrations are seen at that time just before precipitation rates increase significantly. The other simulations show similar behaviour, but magnitudes of these processes depend mostly on ice crystal number concentration.

Figure 5 emphasizes the important role of the vertical fluxes in recycling INPs compared with the relatively small contributions from sources and removal process (Fan et al., 2009; Solomon et al., 2015). Advection (aerosol, cloud, and ice) and precipitation (ice only) have the largest and almost opposite effects on the vertical INP distribution. Advection means mixing within the domain (i.e., no net effect on mass or number), so it practically reduces concentration differences caused by precipitation. Precipitation carries INPs from the cloud droplet freezing region to the near surface sublimation region where most INPs are released back to aerosol (more details in the next section). Advection and precipitation maintain steady profiles by recycling INPs while a fraction of particles is removed by surface precipitation and some particles are entering from the sea surface and free troposphere (subsidence).

Subsidence introduces aerosol particles from the free troposphere (positive values at the cloud top), but at the same time depletes cloud and ice species in the cloud (negative values below the cloud top). This is related to the steep gradient in total INP mass concentration (Fig. 5, right panel). When the total mass concentration is larger in the free troposphere than in the boundary layer (e.g., when precipitation removes the largest particles), the net effect of subsidence is positive (see Fig. 3 above). However, surface emissions change the concentration gradient so that subsidence has a negative effect near surface. In this case the difference between boundary layer and free troposphere dominates. In these simulations, subsidence and entrainment mixing are balanced so that the cloud top height is almost constant. Otherwise changes in the mixing layer depth would influence INP concentrations.

https://doi.org/10.5194/acp-2021-537
Preprint. Discussion started: 12 July 2021

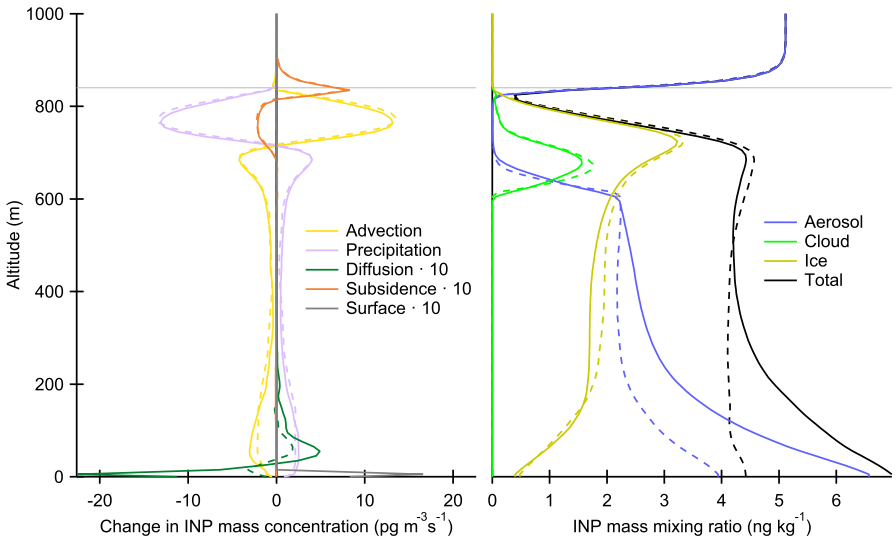


Figure 5. Horizontally averaged profiles of the main processes affecting on INP mass concentrations from simulations with (solid lines) and without (dashed lines) marine INP emissions (high background INP concentration). Diffusion, subsidence, and surface are multiplied by a factor of ten for clarity. Corresponding INP mass concentrations in aerosol, cloud, ice and in total are shown in the right panel. The profiles are averaged over simulation time 7–8 h. Cloud top height (840 m) is indicated by the horizontal lines.

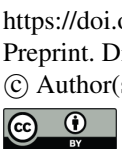
The only clear difference between simulations with and without marine INP emissions is seen in the near surface diffusion and surface emissions rates. Marine INP emissions influence only the first model layer, and sub-grid scale diffusion is the main mechanism transporting particles from the first model layer to the layers above where advection dominates. Diffusion reduces concentration differences within the domain just like advection, but diffusion is significantly weaker and limited to the lowest model layers due to the dependency on eddy diffusivity. Diffusion is not causing INP removal to the sea surface, because aerosol sedimentation (includes the effect of particle diffusivity) is disabled in these simulations. A test will be conducted in Sect. 3.5 where aerosol sedimentation is enabled.

The effect of marine INP emissions can be seen in the total INP mass concentration profiles as an increase near the sea surface. On the other hand, aerosol phase INP mass concentration profiles are similar. They both show decreasing trend above sea surface, which is typically related to a surface source. In this case, however, ice crystal sedimentation and sublimation near the surface is an additional reason. We will focus on this topic in the next section.

3.3 Ice budget

Because ice crystal mass and number concentrations are important for the time evolution of the cloud and the process are related to the INP budget, we will briefly examine the ice budget. Figure 6 shows the effects of the main production and removal mechanisms for ice mass mixing ratio (left) and ice crystal number concentration (right). Simulations where marine INP emissions are switched off are excluded for clarity. Nucleation (freezing of cloud droplets) is the only mechanism producing

https://doi.org/10.5194/acp-2021-537
Preprint. Discussion started: 12 July 2021



Atmospheric Chemistry and Physics Discussions

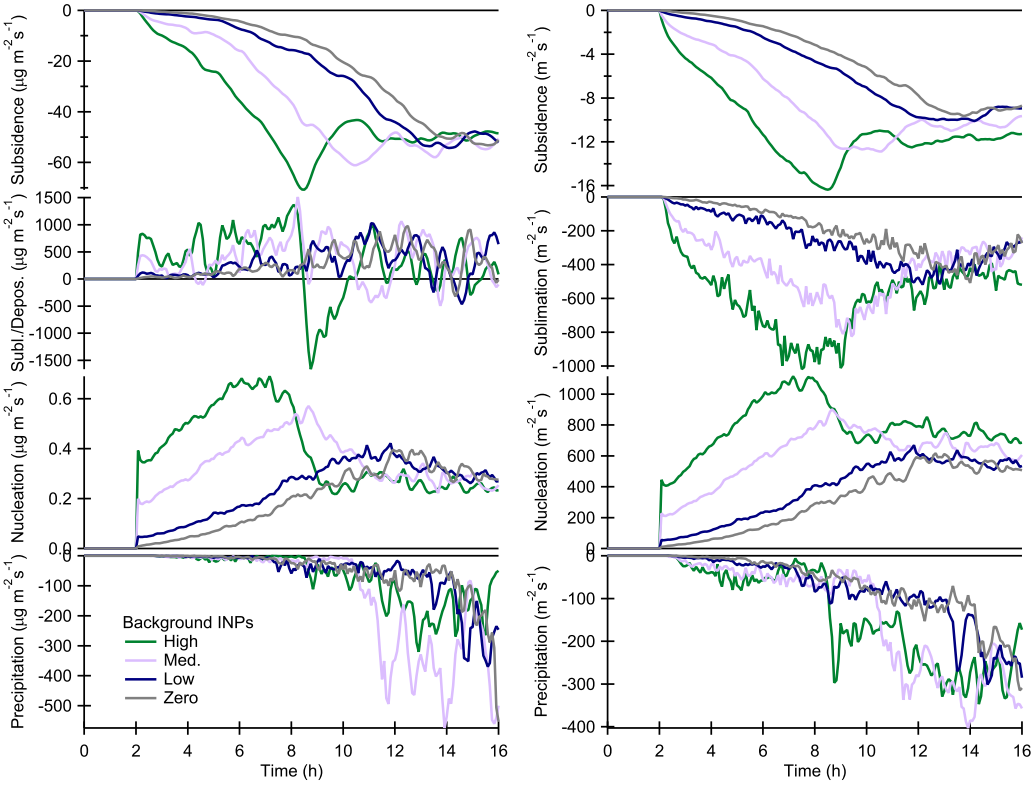


Figure 6. The four mechanisms (subsidence, water vapour sublimation and deposition, nucleation, and precipitation) affecting ice mass (left) and number (right) concentrations. Simulations are initialized with different background INP concentrations and marine INP emissions are switched on.

new ice particles, but nucleation has a negligible contribution to the ice mass. Ice mass depends mainly on water vapour sublimation and deposition rates, but the importance of precipitation increases with time. Only the largest ice crystals survive the fall through the sublimation layer, which means that they are permanently removed by precipitation. Other ice crystals are moved back to aerosol bins when essentially all ice has been sublimated. Here subsidence reduces ice concentrations at the top of the ice layer by bringing ice-free air from above.

Figure 7 shows the key processes affecting on vertical ice mass (left) and number (middle) concentration profiles for the simulation with high background INP concentration and marine INP emissions switched on. The profiles are averaged over simulation time 7–8 h. The right panel shows normalized (by the maximum value) profiles of cloud water and ice mass and ice crystal number concentrations. Advection, precipitation and partitioning of water vapour (sublimation/deposition) have the largest effects on vertical ice mass distribution. Ice crystals grow by deposition of water vapour both in-cloud and below cloud when RH with respect to ice exceeds 100 % (above 470 m) and sublimation takes place otherwise. Precipitation redistributes ice (and INPs) from the freezing and growth regions to the regions below. Advection reduces concentration differences caused

https://doi.org/10.5194/acp-2021-537
Preprint. Discussion started: 12 July 2021

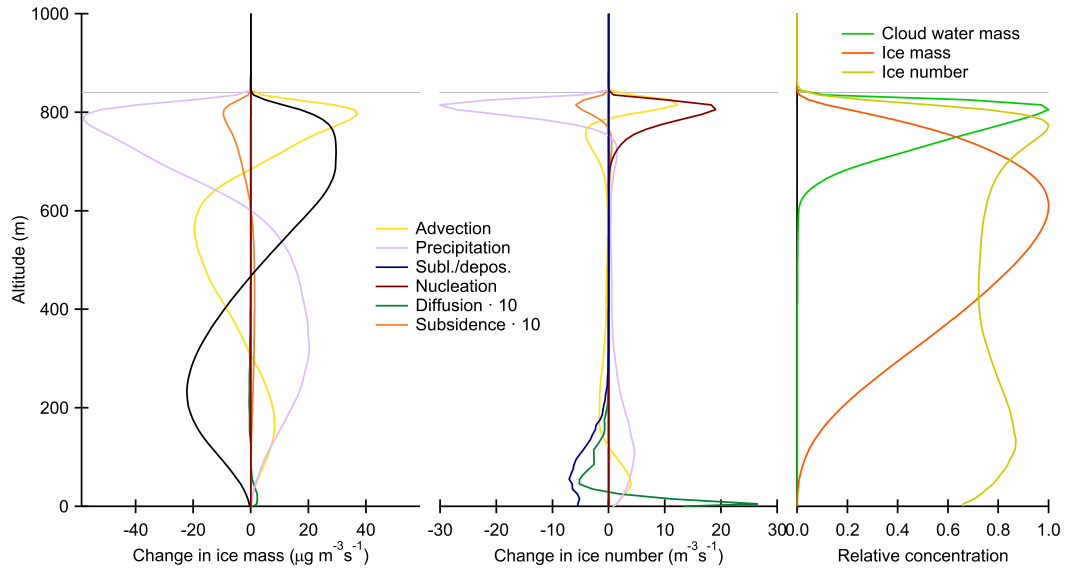


Figure 7. Horizontally averaged profiles of the main processes affecting on ice mass (left) and number (middle) concentrations. Diffusion and subsidence are multiplied by a factor of ten for clarity. Cloud water and ice mass mixing ratios and ice number concentration profiles normalized by their maximum values are shown in the right panel. The profiles are from the high background marine INP simulation averaged over 7–8 h. Cloud top height (840 m) is indicated by the horizontal lines.

by the other processes. Ice nucleation is important for the number concentration, and it takes place at the top of cloud where the lowest temperatures are seen. The smallest ice crystals may lose all ice in the sublimation region and in that case they are released back to aerosol (sublimation). This is an important INP source for the near-surface layer (mostly below 200 m), and most of the time sublimation rates exceed particle losses with precipitation (Fig. 6). Subsidence has the largest effect at the top of ice layer due to the steep gradient in mass and number concentrations. Diffusion has the largest effect on transporting ice from above to the lowest level where particle diffusivity increases precipitation removal rates.

Ice nucleation in these simulations is focused on the cloudy region and especially closer to the cloud top. However, ice nucleation is limited to cloud droplets, so aerosol freezing below and above the cloud is prohibited. The effect of aerosol freezing is tested in Sect. 3.5.

3.4 Details about cloud ice formation

In the above it was shown that advection transports INPs to the cloud top where most of the droplet freezing takes place. However, a closer look at 3D data shows that there is significant horizontal variability in the freezing rates and INP mass mixing ratios, and this variability can be best explained by vertical velocity. This is not a surprise knowing the importance of vertical velocity for cloud activation.

https://doi.org/10.5194/acp-2021-537
Preprint. Discussion started: 12 July 2021

Atmospheric Chemistry and Physics Discussions

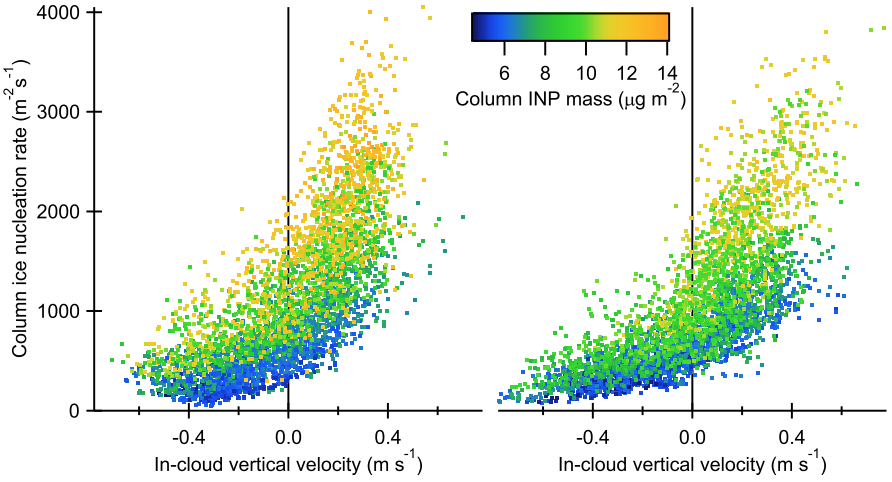


Figure 8. Vertically integrated ice nucleation rate as a function mean in-cloud vertical velocity for each column from a single time step at 8 h. Marker colour is based on vertically integrated INP mass mixing ratio. Simulations are with (left) and without (right) marine INPs and high background INP concentration.

Figure 8 shows vertically integrated ice nucleation rates as a function of mean in-cloud vertical velocity for each column of the domain from a single time step at 8 h (4096 columns in total). Marker colour is based on the corresponding vertically integrated column INP mass mixing ratio. Figures on the left and right show simulations with and without marine INP emissions, respectively. Once again, marine emissions do not have a clear direct effect on nucleation rate but contribute indirectly via INP number concentration. This is the main reason for the differences between these two figures.

Higher cloud droplet freezing rates are related to updraughts (positive vertical velocity) and marker colour shows that the updraughts have higher INP mass concentrations. INP concentrations range from less than 5 $\mu g\,m^{-2}$ to above 14 $\mu g\,m^{-2}$, so the variability is as high as ± 50 % compared to the 9.6 $\mu g\,m^{-2}$ background concentration. Nucleation rate and INP mass are also linked so that higher INP mass leads to higher nucleation rate at a constant vertical velocity. The brief explanation for these findings is related to the vertical transport of INPs. Unfrozen aerosol-phase INPs have high concentration near surface (see Fig. 5) mainly due to ice crystal sublimation. Updraughts bring these INPs to clouds where they become cloud droplets which continue rising until they reach temperatures low enough for freezing. Stronger updraughts can reach lower temperatures just below inversion layer, which means higher freezing rates. Freezing rates decrease when the most effective INPs have been frozen. Following ice crystal growth is not dependent on vertical velocity, but downdraughts increase sedimentation rates, which reduce INP concentrations. The strongest downdraughts also originate from the cloud top region which is depleted from INPs (Fig. 5) due to ice crystal sedimentation.

https://doi.org/10.5194/acp-2021-537
Preprint. Discussion started: 12 July 2021

Atmospheric Chemistry and Physics Discussions

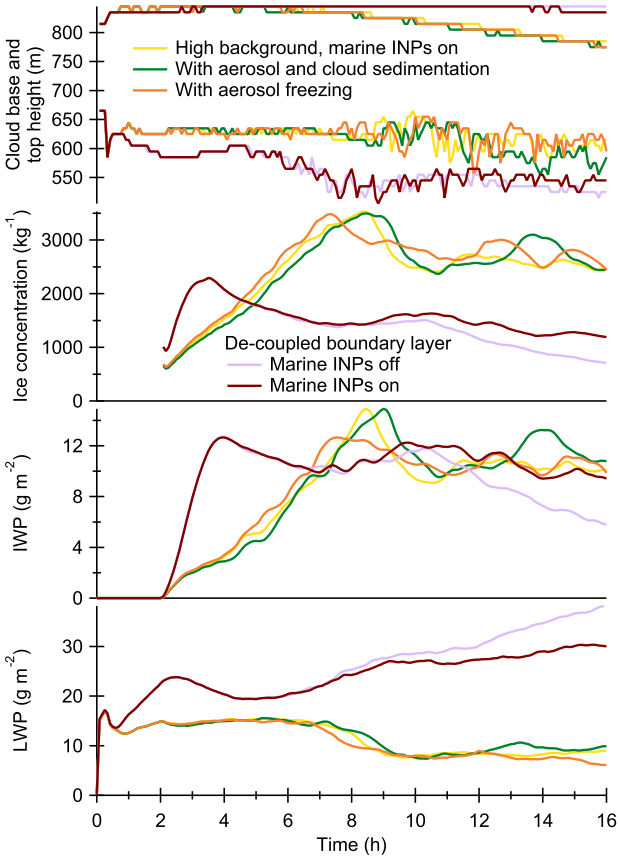


Figure 9. Sensitivity tests related to aerosol and cloud droplet sedimentation, aerosol freezing and marine INPs in the de-coupled boundary layer.

3.5 Sensitivity tests

Here we examine the effects of microphysical (aerosol and cloud droplet sedimentation, and aerosol freezing) and meteorological (de-coupled boundary layer) model considerations mentioned above. Figure 9 shows four test simulations and the high background marine INP simulation as the reference case for the sedimentation and aerosol freezing simulations. The effect of de-coupled boundary layer is tested by running simulations with and without marine INP emissions.

Allowing aerosol and cloud droplet sedimentation (disabled in the default simulations) has two potential effects on INPs. First, aerosol sedimentation could bring INPs from the free troposphere or remove those from the near surface layer by dry deposition. Second, sedimentation could have an impact on vertical distributions. Cloud droplet sedimentation redistributes cloud water, which influences clouds as explained in Ovchinnikov et al. (2014). Due to this side effect on clouds, it is not possible to fully isolate the effect of sedimentation on INPs. However, simulations made with and without aerosol and cloud

https://doi.org/10.5194/acp-2021-537
Preprint. Discussion started: 12 July 2021


Atmospheric Chemistry and Physics Discussions

droplet sedimentation show negligible differences. Advection and ice crystal sedimentation dominate vertical mixing, and the slow removal of INPs by dry deposition is almost fully compensated by a flux of INPs coming from the free troposphere.

Figure 9 shows that allowing aerosol freezing has a small impact on ice crystal number concentration. The main reason for this is the fact that freezing is practically limited to the cloudy regions (sub-saturated regions are too warm) where the aerosol can freeze before or after cloud activation. Aerosol freezing rate is about 10 % of the total freezing rate, so it is not insignificant. However, due to the above-mentioned reason, cloud droplet freezing rate is reduced by the same amount so that the total freezing rate is about the same as that in the simulation without aerosol freezing (Fig. 6). Most of the aerosol freezing takes place at the top of cloud. This is not related to downdraughts (or subsidence) bringing new INPs from the free troposphere. Instead, spatial correlation between cloud activation/de-activation and aerosol freezing rates at the cloud top indicates that temperature fluctuations, for example due to radiative cooling or mixing, first cause the release of INPs from evaporating cloud droplets and later initiate cloud activation and aerosol freezing. In the latter case, the instantaneous aerosol freezing can take place before the diffusion limited droplet growth leads to cloud activation.

Meteorological conditions are crucially important for clouds, but here we focus on the one that has direct relevance for the vertical transport of marine INP emissions, namely de-coupled boundary layer. The original ISDAC simulations were initialized with a de-coupled boundary layer, which reduces vertical mixing and partially isolates the near surface layer from the rest of the boundary layer (e.g., Wood, 2012). Figure 9 shows simulations with and without marine INP emissions (both with high concentrations of background INPs) when the boundary layer is de-coupled and humid as in the original ISDAC case study (Ovchinnikov et al., 2014). Temperature is increasing 0.004 $\mathrm{K\,m^{-1}}$ and total water mixing ratio is decreasing 0.00075 $\mathrm{g\,kg^{-1}\,m^{-1}}$ within the lowest 400 m. Due to the different heat and humidity contents, cloud states are different for coupled and de-coupled boundary layers. Comparing simulations made with and without marine INP emissions when boundary layer is de-coupled shows that marine INP emissions have negligible effect at least during the first 10 hours. This shows that de-coupling can indeed prevent marine INPs reaching clouds. After the first 10 hours, boundary layer becomes more coupled, so marine INP emissions start to influence ice crystal number concentrations.

4 Conclusions

In this study we examined the potential effects of marine ice nucleating particles (INPs) on shallow mixed-phase clouds by using a large eddy simulator UCLALES-SALSA (Tonttila et al., 2017; Ahola et al., 2020). Simulations were made by adjusting initial background INP concentrations and INP emissions with sea spray so that reasonable cloud ice crystal number concentrations were seen for a wide range of source strengths. Our simulations show that in the case of well mixed (coupled) boundary layer, updraughts are efficient in transporting marine INPs up to the clouds where droplet freezing can take place. When the background INP concentration is low, which means that free troposphere is not a significant INP source, marine emissions can maintain mixed-phase clouds. While the free troposphere is separated from the clouds by an inversion layer, which reduces vertical mixing, marine INPs are emitted directly to the boundary layer.

https://doi.org/10.5194/acp-2021-537
Preprint. Discussion started: 12 July 2021


Atmospheric Chemistry and Physics Discussions

Our simulations with UCLALES-SALSA, which has prognostic aerosol, cloud and ice phase INP size distributions, support the previous findings about the importance of INP recycling (Fan et al., 2009; Solomon et al., 2015). This means that the smallest precipitating ice crystals lose all ice in the near surface sublimation layer so that the original INPs are released. Updraughts transport these INPs, as well as those from the sea spray aerosol, back to the clouds where they may again initiate droplet freezing. Detailed examination of the 3D model outputs show that ice nucleation is focused on the updraught regions and these regions may have up to 50 % more INP mass compared with the background INP mass. On the other hand, downdraughts are depleted by up to 50 %, because they originate from the cloud top where ice formation and subsequent sedimentation reduce INP concentrations.

Prognostic ice microphysics including explicitly modelled ice nucleation is important for the simulations as this allows feedbacks between INPs and clouds (e.g., Paukert and Hoose, 2014; Savre and Ekman, 2015; Ahola et al., 2020). Precipitation removal is the most important feedback in our simulations. Increasing INP concentration increases precipitation removal rates, so most of our simulations ended up having similar cloud ice contents. Precipitation feedback also prevents complete glaciation, which happens in the case of fixed ice crystal number concentration (Ahola et al., 2020).

Efficient INP recycling, feedbacks between INP emissions and precipitation removal, and the fact that marine INPs are emitted directly to the mixed layer mean that modest marine INP emissions can maintain mixed-phase clouds at least in our simulations. Although significant uncertainties are still related to ambient INP emissions, our simulations support the current view (e.g., Vergara-Temprado et al., 2017; Huang et al., 2018; McCluskey et al., 2019; Zhao et al., 2021) that marine INPs can have a dominant role in remote regions far from continental dust sources.

Code and data availability. The source code of UCLALES-SALSA version used in this work is available from https://github.com/UCLALES-SALSA/UCLALES-SALSA/tree/isdac_poa (last access 3 March 2021). Brief description of the simulations and the data used in this publication are available at https://a3s.fi/moac_marine_inp_ms/readme.txt (Raatikainen, 2021, Last access 14.6.2021).

Author contributions. TR designed and performed the model simulations. All authors have contributed to developing the UCLALES-SALSA model. TR prepared the manuscript with contributions from all co-authors.

Competing interests. The authors declare that they have no conflict of interest.

Acknowledgements. This research has been supported by the Academy of Finland (grant nos. 322532 and 309127) and Horizon 2020 Research and Innovation Programme (grant no. 821205). The authors wish to acknowledge CSC – IT Center for Science, Finland, for computational resources.

https://doi.org/10.5194/acp-2021-537
Preprint. Discussion started: 12 July 2021

Atmospheric Chemistry and Physics Discussions

References

Ahola, J., Korhonen, H., Tonttila, J., Romakkaniemi, S., Kokkola, H., and Raatikainen, T.: Modelling mixed-phase clouds with the large-eddy model UCLALES–SALSA, Atmos. Chem. Phys., 20, 11 639–11 654, https://doi.org/10.5194/acp-20-11639-2020, 2020.

Boose, Y., Welti, A., Atkinson, J., Ramelli, F., Danielczok, A., Bingemer, H. G., Plötze, M., Sierau, B., Kanji, Z. A., and Lohmann, U.: Heterogeneous ice nucleation on dust particles sourced from nine deserts worldwide – Part 1: Immersion freezing, Atmos. Chem. Phys., 16, 15 075–15 095, https://doi.org/10.5194/acp-16-15075-2016, 2016.

Burrows, S. M., Hoose, C., Pöschl, U., and Lawrence, M. G.: Ice nuclei in marine air: biogenic particles or dust?, Atmos. Chem. Phys., 13, 245–267, https://doi.org/10.5194/acp-13-245-2013, 2013.

Chen, J.-P., Hazra, A., and Levin, Z.: Parameterizing ice nucleation rates using contact angle and activation energy derived from laboratory data, Atmos. Chem. Phys., 8, 7431–7449, https://doi.org/10.5194/acp-8-7431-2008, 2008.

Cornwell, G. C., Sultana, C. M., Prank, M., Cochran, R. E., Hill, T. C. J., Schill, G. P., DeMott, P. J., Mahowald, N., and Prather, K. A.: Ejection of Dust From the Ocean as a Potential Source of Marine Ice Nucleating Particles, J. Geophys. Res.-Atmos., 125, e2020JD033 073, https://doi.org/https://doi.org/10.1029/2020JD033073, 2020.

DeMott, P. J., Hill, T. C. J., McCluskey, C. S., Prather, K. A., Collins, D. B., Sullivan, R. C., Ruppel, M. J., Mason, R. H., Irish, V. E., Lee, T., Hwang, C. Y., Rhee, T. S., Snider, J. R., McMeeking, G. R., Dhaniyala, S., Lewis, E. R., Wentzell, J. J. B., Abbatt, J., Lee, C., Sultana, C. M., Ault, A. P., Axson, J. L., Diaz Martinez, M., Venero, I., Santos-Figueroa, G., Stokes, M. D., Deane, G. B., Mayol-Bracero, O. L., Grassian, V. H., Bertram, T. H., Bertram, A. K., Moffett, B. F., and Franc, G. D.: Sea spray aerosol as a unique source of ice nucleating particles, P. Natl. Acad. Sci. USA, 113, 5797–5803, https://doi.org/10.1073/pnas.1514034112, 2016.

Earle, M. E., Liu, P. S. K., Strapp, J. W., Zelenyuk, A., Imre, D., McFarquhar, G. M., Shantz, N. C., and Leaitch, W. R.: Factors influencing the microphysics and radiative properties of liquid-dominated Arctic clouds: Insight from observations of aerosol and clouds during ISDAC, J. Geophys. Res.-Atmos., 116, https://doi.org/https://doi.org/10.1029/2011JD015887, 2011.

Ervens, B. and Feingold, G.: On the representation of immersion and condensation freezing in cloud models using different nucleation schemes, Atmos. Chem. Phys., 12, 5807–5826, https://doi.org/10.5194/acp-12-5807-2012, 2012.

Ervens, B. and Feingold, G.: Sensitivities of immersion freezing: Reconciling classical nucleation theory and deterministic expressions, Geophys. Res. Lett., 40, 3320–3324, https://doi.org/https://doi.org/10.1002/grl.50580, 2013.

Fan, J., Ovtchinnikov, M., Comstock, J. M., McFarlane, S. A., and Khain, A.: Ice formation in Arctic mixed-phase clouds: Insights from a 3-D cloud-resolving model with size-resolved aerosol and cloud microphysics, J. Geophys. Res.-Atmos., 114, https://doi.org/https://doi.org/10.1029/2008JD010782, 2009.

Gong, X., Wex, H., van Pinxteren, M., Triesch, N., Fomba, K. W., Lubitz, J., Stolle, C., Robinson, T.-B., Müller, T., Herrmann, H., and Stratmann, F.: Characterization of aerosol particles at Cabo Verde close to sea level and at the cloud level – Part 2: Ice-nucleating particles in air, cloud and seawater, Atmos. Chem. Phys., 20, 1451–1468, https://doi.org/10.5194/acp-20-1451-2020, 2020.

Hartmann, M., Adachi, K., Eppers, O., Haas, C., Herber, A., Holzinger, R., Hünerbein, A., Jäkel, E., Jentzsch, C., van Pinxteren, M., Wex, H., Willmes, S., and Stratmann, F.: Wintertime Airborne Measurements of Ice Nucleating Particles in the High Arctic: A Hint to a Marine, Biogenic Source for Ice Nucleating Particles, Geophys. Res. Lett., 47, e2020GL087 770, https://doi.org/https://doi.org/10.1029/2020GL087770, 2020.

https://doi.org/10.5194/acp-2021-537
Preprint. Discussion started: 12 July 2021


Atmospheric Chemistry and Physics Discussions

Hiranuma, N., Brooks, S. D., Moffet, R. C., Glen, A., Laskin, A., Gilles, M. K., Liu, P., Macdonald, A. M., Strapp, J. W., and McFarquhar, G. M.: Chemical characterization of individual particles and residuals of cloud droplets and ice crystals collected on board research aircraft in the ISDAC 2008 study, J. Geophys. Res.-Atmos., 118, 6564–6579, https://doi.org/https://doi.org/10.1002/jgrd.50484, 2013.

Hoose, C. and Möhler, O.: Heterogeneous ice nucleation on atmospheric aerosols: a review of results from laboratory experiments, Atmos. Chem. Phys., 12, 9817–9854, https://doi.org/10.5194/acp-12-9817-2012, 2012.

Hoose, C., Kristjánsson, J. E., Chen, J.-P., and Hazra, A.: A Classical-Theory-Based Parameterization of Heterogeneous Ice Nucleation by Mineral Dust, Soot, and Biological Particles in a Global Climate Model, J. Atmos. Sci., 67, 2483–2503, https://doi.org/10.1175/2010JAS3425.1, 2010.

Huang, S., Hu, W., Chen, J., Wu, Z., Zhang, D., and Fu, P.: Overview of biological ice nucleating particles in the atmosphere, Environ. Int., 146, 106 197, https://doi.org/https://doi.org/10.1016/j.envint.2020.106197, 2021.

Huang, W. T. K., Ickes, L., Tegen, I., Rinaldi, M., Ceburnis, D., and Lohmann, U.: Global relevance of marine organic aerosol as ice nucleating particles, Atmos. Chem. Phys., 18, 11 423–11 445, https://doi.org/10.5194/acp-18-11423-2018, 2018.

Ickes, L., Welti, A., and Lohmann, U.: Classical nucleation theory of immersion freezing: sensitivity of contact angle schemes to thermodynamic and kinetic parameters, Atmos. Chem. Phys., 17, 1713–1739, https://doi.org/10.5194/acp-17-1713-2017, 2017.

Ickes, L., Porter, G. C. E., Wagner, R., Adams, M. P., Bierbauer, S., Bertram, A. K., Bilde, M., Christiansen, S., Ekman, A. M. L., Gorokhova, E., Höhler, K., Kiselev, A. A., Leck, C., Möhler, O., Murray, B. J., Schiebel, T., Ullrich, R., and Salter, M. E.: The ice-nucleating activity of Arctic sea surface microlayer samples and marine algal cultures, Atmos. Chem. Phys., 20, 11 089–11 117, https://doi.org/10.5194/acp-20-11089-2020, 2020.

Jaeglé, L., Quinn, P. K., Bates, T. S., Alexander, B., and Lin, J.-T.: Global distribution of sea salt aerosols: new constraints from in situ and remote sensing observations, Atmos. Chem. Phys., 11, 3137–3157, https://doi.org/10.5194/acp-11-3137-2011, 2011.

Kanji, Z. A., Ladino, L. A., Wex, H., Boose, Y., Burkert-Kohn, M., Cziczo, D. J., and Krämer, M.: Overview of Ice Nucleating Particles, Meteor. Mon., 58, 1.1–1.33, https://doi.org/10.1175/AMSMONOGRAPHS-D-16-0006.1, 2017.

Khvorostyanov, V. I. and Curry, J. A.: A new theory of heterogeneous ice nucleation for application in cloud and climate models, Geophys. Res. Lett., 27, 4081–4084, https://doi.org/https://doi.org/10.1029/1999GL011211, 2000.

Khvorostyanov, V. I. and Curry, J. A.: The Theory of Ice Nucleation by Heterogeneous Freezing of Deliquescent Mixed CCN. Part I: Critical Radius, Energy, and Nucleation Rate, J. Atmos. Sci., 61, 2676–2691, https://doi.org/10.1175/JAS3266.1, 2004.

Knopf, D. A., Alpert, P. A., and Wang, B.: The Role of Organic Aerosol in Atmospheric Ice Nucleation: A Review, ACS Earth Space Chem., 2, 168–202, https://doi.org/10.1021/acsearthspacechem.7b00120, 2018.

Kok, J. F., Adebiyi, A. A., Albani, S., Balkanski, Y., Checa-Garcia, R., Chin, M., Colarco, P. R., Hamilton, D. S., Huang, Y., Ito, A., Klose, M., Li, L., Mahowald, N. M., Miller, R. L., Obiso, V., Pérez García-Pando, C., Rocha-Lima, A., and Wan, J. S.: Contribution of the world's main dust source regions to the global cycle of desert dust, Atmos. Chem. Phys., 21, 8169–8193, https://doi.org/10.5194/acp-21-8169-2021, 2021.

Kokkola, H., Korhonen, H., Lehtinen, K. E. J., Makkonen, R., Asmi, A., Järvenoja, S., Anttila, T., Partanen, A.-I., Kulmala, M., Järvinen, H., Laaksonen, A., and Kerminen, V.-M.: SALSA – a Sectional Aerosol module for Large Scale Applications, Atmos. Chem. Phys., 8, 2469–2483, https://doi.org/10.5194/acp-8-2469-2008, 2008.

Kokkola, H., Kühn, T., Laakso, A., Bergman, T., Lehtinen, K. E. J., Mielonen, T., Arola, A., Stadtler, S., Korhonen, H., Ferrachat, S., Lohmann, U., Neubauer, D., Tegen, I., Siegenthaler-Le Drian, C., Schultz, M. G., Bey, I., Stier, P., Daskalakis, N., Heald, C. L., and

Romakkaniemi, S.: SALSA2.0: The sectional aerosol module of the aerosol–chemistry–climate model ECHAM6.3.0-HAM2.3-MOZ1.0, Geosci. Model Dev., 11, 3833–3863, https://doi.org/10.5194/gmd-11-3833-2018, 2018.

Korolev, A., McFarquhar, G., Field, P. R., Franklin, C., Lawson, P., Wang, Z., Williams, E., Abel, S. J., Axisa, D., Borrmann, S., Crosier, J., Fugal, J., Krämer, M., Lohmann, U., Schlenczek, O., Schnaiter, M., and Wendisch, M.: Mixed-Phase Clouds: Progress and Challenges, Meteor. Mon., 58, 5.1–5.50, https://doi.org/10.1175/AMSMONOGRAPHS-D-17-0001.1, 2017.

Ladino Moreno, L. A., Stetzer, O., and Lohmann, U.: Contact freezing: a review of experimental studies, Atmos. Chem. Phys., 13, 9745–9769, https://doi.org/10.5194/acp-13-9745-2013, 2013.

Mårtensson, E. M., Nilsson, E. D., de Leeuw, G., Cohen, L. H., and Hansson, H.-C.: Laboratory simulations and parameterization of the primary marine aerosol production, J. Geophys. Res.-Atmos., 108, https://doi.org/https://doi.org/10.1029/2002JD002263, 2003.

McCluskey, C. S., Hill, T. C. J., Humphries, R. S., Rauker, A. M., Moreau, S., Strutton, P. G., Chambers, S. D., Williams, A. G., McRobert, I., Ward, J., Keywood, M. D., Harnwell, J., Ponsonby, W., Loh, Z. M., Krummel, P. B., Protat, A., Kreidenweis, S. M., and DeMott, P. J.: Observations of Ice Nucleating Particles Over Southern Ocean Waters, Geophys. Res. Lett., 45, 11,989–11,997, https://doi.org/https://doi.org/10.1029/2018GL079981, 2018a.

McCluskey, C. S., Hill, T. C. J., Sultana, C. M., Laskina, O., Trueblood, J., Santander, M. V., Beall, C. M., Michaud, J. M., Kreidenweis, S. M., Prather, K. A., Grassian, V., and DeMott, P. J.: A Mesocosm Double Feature: Insights into the Chemical Makeup of Marine Ice Nucleating Particles, J. Atmos. Sci., 75, 2405–2423, https://doi.org/10.1175/JAS-D-17-0155.1, 2018b.

McCluskey, C. S., Ovadnevaite, J., Rinaldi, M., Atkinson, J., Belosi, F., Ceburnis, D., Marullo, S., Hill, T. C. J., Lohmann, U., Kanji, Z. A., O'Dowd, C., Kreidenweis, S. M., and DeMott, P. J.: Marine and Terrestrial Organic Ice-Nucleating Particles in Pristine Marine to Continentally Influenced Northeast Atlantic Air Masses, J. Geophys. Res.-Atmos., 123, 6196–6212, https://doi.org/https://doi.org/10.1029/2017JD028033, 2018c.

McCluskey, C. S., DeMott, P. J., Ma, P.-L., and Burrows, S. M.: Numerical Representations of Marine Ice-Nucleating Particles in Remote Marine Environments Evaluated Against Observations, Geophys. Res. Lett., 46, 7838–7847, https://doi.org/https://doi.org/10.1029/2018GL081861, 2019.

McFarquhar, G. M., Ghan, S., Verlinde, J., Korolev, A., Strapp, J. W., Schmid, B., Tomlinson, J. M., Wolde, M., Brooks, S. D., Cziczo, D., Dubey, M. K., Fan, J., Flynn, C., Gultepe, I., Hubbe, J., Gilles, M. K., Laskin, A., Lawson, P., Leaitch, W. R., Liu, P., Liu, X., Lubin, D., Mazzoleni, C., Macdonald, A.-M., Moffet, R. C., Morrison, H., Ovchinnikov, M., Shupe, M. D., Turner, D. D., Xie, S., Zelenyuk, A., Bae, K., Freer, M., and Glen, A.: Indirect and Semi-direct Aerosol Campaign: The Impact of Arctic Aerosols on Clouds, Bull. Amer. Meteor. Soc., 92, 183–201, https://doi.org/10.1175/2010BAMS2935.1, 2011.

Mitts, B. A., Wang, X., Lucero, D. D., Beall, C. M., Deane, G. B., DeMott, P. J., and Prather, K. A.: Importance of Supermicron Ice Nucleating Particles in Nascent Sea Spray, Geophys. Res. Lett., 48, e2020GL089 633, https://doi.org/https://doi.org/10.1029/2020GL089633, 2021.

Monahan, E., Spiel, D., and Davidson, K.: A Model of Marine Aerosol Generation Via Whitecaps and Wave Disruption, in: Oceanic Whitecaps, edited by Monahan, E. and Niocaill, G., vol. 2, pp. 167–174, Springer, Dordrecht, https://doi.org/10.1007/978-94-009-4668-2_16, 1986.

Morrison, H., de Boer, G., Feingold, G., Harrington, J., Shupe, M. D., and Sulia, K.: Resilience of persistent Arctic mixed-phase clouds, Nat. Geosci., 5, 11–17, https://doi.org/10.1038/ngeo1332, 2012.

Murray, B. J., Broadley, S. L., Wilson, T. W., Atkinson, J. D., and Wills, R. H.: Heterogeneous freezing of water droplets containing kaolinite particles, Atmos. Chem. Phys., 11, 4191–4207, https://doi.org/10.5194/acp-11-4191-2011, 2011.

https://doi.org/10.5194/acp-2021-537
Preprint. Discussion started: 12 July 2021

Atmospheric Chemistry and Physics Discussions

Murray, B. J., O'Sullivan, D., Atkinson, J. D., and Webb, M. E.: Ice nucleation by particles immersed in supercooled cloud droplets, Chem. Soc. Rev., 41, 6519–6554, https://doi.org/10.1039/C2CS35200A, 2012.

Murray, B. J., Carslaw, K. S., and Field, P. R.: Opinion: Cloud-phase climate feedback and the importance of ice-nucleating particles, Atmos. Chem. Phys., 21, 665–679, https://doi.org/10.5194/acp-21-665-2021, 2021.

Ovchinnikov, M., Ackerman, A. S., Avramov, A., Cheng, A., Fan, J., Fridlind, A. M., Ghan, S., Harrington, J., Hoose, C., Korolev, A., McFarquhar, G. M., Morrison, H., Paukert, M., Savre, J., Shipway, B. J., Shupe, M. D., Solomon, A., and Sulia, K.: Intercomparison of large-eddy simulations of Arctic mixed-phase clouds: Importance of ice size distribution assumptions, J. Adv. Model. Earth Sy., 6, 223–248, https://doi.org/https://doi.org/10.1002/2013MS000282, 2014.

Paukert, M. and Hoose, C.: Modeling immersion freezing with aerosol-dependent prognostic ice nuclei in Arctic mixed-phase clouds, J. Geophys. Res.-Atmos., 119, 9073–9092, https://doi.org/https://doi.org/10.1002/2014JD021917, 2014.

Prospero, J. M., Ginoux, P., Torres, O., Nicholson, S. E., and Gill, T. E.: Environmental characterization of global sources of atmospheric soil dust identified with the Nimbus 7 Total Ozone Mapping Spectrometer (TOMS) absorbing aerosol product, Rev. Geophys., 40, 2–1–2–31, https://doi.org/https://doi.org/10.1029/2000RG000095, 2002.

Raatikainen, T.: UCLALES-SALSA data set focusing on marine ice nucleating particles, https://a3s.fi/moac_marine_inp_ms/readme.txt, 2021.

Savre, J. and Ekman, A. M. L.: Large-eddy simulation of three mixed-phase cloud events during ISDAC: Conditions for persistent heterogeneous ice formation, J. Geophys. Res.-Atmos., 120, 7699–7725, https://doi.org/https://doi.org/10.1002/2014JD023006, 2015.

Solomon, A., Feingold, G., and Shupe, M. D.: The role of ice nuclei recycling in the maintenance of cloud ice in Arctic mixed-phase stratocumulus, Atmos. Chem. Phys., 15, 10 631–10 643, https://doi.org/10.5194/acp-15-10631-2015, 2015.

Stevens, B. and Seifert, A.: Understanding macrophysical outcomes of microphysical choices in simulations of shallow cumulus convection, J. Meteorol. Soc. Jpn. Ser. II, 86A, 143–162, https://doi.org/10.2151/jmsj.86A.143, 2008.

Stevens, B., Moeng, C.-H., and Sullivan, P. P.: Large-Eddy Simulations of Radiatively Driven Convection: Sensitivities to the Representation of Small Scales, J. Atmos. Sci., 56, 3963–3984, https://doi.org/10.1175/1520-0469(1999)056<3963:LESORD>2.0.CO;2, 1999.

Stevens, B., Moeng, C.-H., Ackerman, A. S., Bretherton, C. S., Chlond, A., de Roode, S., Edwards, J., Golaz, J.-C., Jiang, H., Khairoutdinov, M., Kirkpatrick, M. P., Lewellen, D. C., Lock, A., Müller, F., Stevens, D. E., Whelan, E., and Zhu, P.: Evaluation of Large-Eddy Simulations via Observations of Nocturnal Marine Stratocumulus, Mon. Weather Rev., 133, 1443–1462, https://doi.org/10.1175/MWR2930.1, 2005.

Tonttila, J., Maalick, Z., Raatikainen, T., Kokkola, H., Kühn, T., and Romakkaniemi, S.: UCLALES–SALSA v1.0: a large-eddy model with interactive sectional microphysics for aerosol, clouds and precipitation, Geosci. Model Dev., 10, 169–188, https://doi.org/10.5194/gmd-10-169-2017, 2017.

Vergara-Temprado, J., Murray, B. J., Wilson, T. W., O'Sullivan, D., Browse, J., Pringle, K. J., Ardon-Dryer, K., Bertram, A. K., Burrows, S. M., Ceburnis, D., DeMott, P. J., Mason, R. H., O'Dowd, C. D., Rinaldi, M., and Carslaw, K. S.: Contribution of feldspar and marine organic aerosols to global ice nucleating particle concentrations, Atmos. Chem. Phys., 17, 3637–3658, https://doi.org/10.5194/acp-17-3637-2017, 2017.

Wilson, T. W., Ladino, L. A., Alpert, P. A., Breckels, M. N., Brooks, I. M., Browse, J., Burrows, S. M., Carslaw, K. S., Huffman, J. A., Judd, C., Kilthau, W. P., Mason, R. H., McFiggans, G., Miller, L. A., Nájera, Juan J. and-Polishchuk, E., Rae, S., Schiller, C. L., Si, M., Temprado, J. V., Whale, T. F., Wong, J. P. S., Wurl, O., Yakobi-Hancock, J. D., Abbatt, J. P. D., Aller, J. Y., Bertram, A. K., Knopf, D. A., and Murray, B. J.: A marine biogenic source of atmospheric ice-nucleating particles, Nature, 525, 234–238, https://doi.org/10.1038/nature14986, 2015.

https://doi.org/10.5194/acp-2021-537
Preprint. Discussion started: 12 July 2021

Atmospheric Chemistry and Physics Discussions

Wolf, M. J., Goodell, M., Dong, E., Dove, L. A., Zhang, C., Franco, L. J., Shen, C., Rutkowski, E. G., Narducci, D. N., Mullen, S., Babbin, A. R., and Cziczo, D. J.: A link between the ice nucleation activity and the biogeochemistry of seawater, Atmos. Chem. Phys., 20, 15 341–15 356, https://doi.org/10.5194/acp-20-15341-2020, 2020.

Wood, R.: Stratocumulus Clouds, Mon. Weather Rev., 140, 2373–2423, https://doi.org/10.1175/MWR-D-11-00121.1, 2012.

Zhao, X., Liu, X., Burrows, S. M., and Shi, Y.: Effects of marine organic aerosols as sources of immersion-mode ice-nucleating particles on high-latitude mixed-phase clouds, Atmos. Chem. Phys., 21, 2305–2327, https://doi.org/10.5194/acp-21-2305-2021, 2021.