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Possible roles of ice nucleation mode and ice nuclei depletion in the extended lifetime of Arctic mixed-phase clouds

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[1] The sensitivity of Arctic mixed phase clouds to the mode of ice particle nucleation is examined using a 1-D cloud model. It is shown that the lifetime of a simulated low-level Arctic mixed-phase stratus is highly sensitive to the number concentration of deposition/condensation-freezing nuclei, and much less sensitive to the number of contact nuclei. Simulations with prognostic ice nuclei concentration exhibit rapid depletion of deposition/condensation-freezing nuclei due to nucleation scavenging which significantly extends the mixed-phase cloud lifetime. In contrast, scavenging has little impact on the number of contact nuclei. Thus, contact mode nucleation generally dominates in the cloud layer when both modes are simultaneously considered. The dominance of contact nucleation in Arctic mixed-phase clouds is consistent with a number of in situ observations, remote retrievals, and laboratory experiments. A conceptual model of long-lived Arctic mixed-phase clouds is developed that explains their persistence through the rapid depletion of deposition/condensation-freezing ice nuclei and a self-regulating drop-contact freezing feedback. **Citation:** Morrison, H., M. D. Shupe, J. O. Pinto, and J. A. Curry (2005), Possible roles of ice nucleation mode and ice nuclei depletion in the extended lifetime of Arctic mixed-phase clouds, *Geophys. Res. Lett.*, *32*, L18801, doi:10.1029/2005GL023614.

1. Introduction

[2] Recent field experiments in the Arctic have highlighted the common occurrence of mixed-phase stratiform clouds throughout the year [e.g., *Shupe et al.*, 2005a]. These clouds have a strong impact on the surface radiative fluxes [e.g., *Zuidema et al.*, 2005, hereinafter referred to as Z05] and hence the sea ice mass balance [e.g., *Curry and Ebert*, 1992]. Despite recent advances in the characterization of Arctic mixed-phase clouds (AMPC) by in situ and remote measurements, there remain fundamental uncertainties in our understanding of these clouds. In particular, it is uncertain how these clouds are able to persist for extended time periods (several days) while maintaining substantial amounts of both supercooled water and ice. It has been suggested that AMPC may be maintained by cloud-top radiative cooling and/or large-scale moisture convergence [*Pinto*, 1998; *Harrington et al.*, 1999; *Jiang et al.*, 2000]. Depletion of IN within the cloudy boundary layer may also play a role in their persistence, since cloud models show strong sensitivity of AMPC lifetime to the ice nuclei (IN) or ice crystal concentration [e.g., *Harrington et al.*, 1999; *Jiang et al.*, 2000].

[3] Heterogeneous nucleation is responsible for ice formation in AMPC. Heterogeneous nucleation may occur through a number of distinct modes. These modes include deposition (nucleation on aerosol directly from vapor), condensation-freezing (defined here as deliquescence or wetting of aerosol in water subsaturated or supersaturated conditions followed by freezing), immersion-freezing (freezing caused by resident aerosol within drops), and contact-freezing (collision of aerosol with drops followed by rapid freezing). Note that the same particle can potentially nucleate ice through more than one mode. In practice, it is difficult to distinguish between deposition and condensation-freezing, particularly in water supersaturated conditions [*Meyers et al.*, 1992]. Hereafter we will refer to their combined action as 'deposition/condensation-freezing' nucleation.

[4] The differing roles of contact and deposition/condensation-freezing nucleation are explored using a 1-D cloud model. The consistency of these results with in situ observations, remote retrievals, and laboratory experiments is described. A conceptual model is then developed that explains the persistence of supercooled water in AMPC through the depletion of deposition/condensation-freezing nuclei and a self-regulating drop-contact freezing feedback.

2. Modeling Results

[5] To examine how ice nucleation mode and IN depletion may impact AMPC, a case observed during the Surface Heat Budget of the Arctic Ocean (SHEBA) [*Uttal et al.*, 2002] and First ISCCP Regional Experiment – Arctic Clouds Experiment (FIRE-ACE) [*Curry et al.*, 2000] in May, 1998, is simulated using a 1-D cloud model with a dual moment bulk microphysics scheme [*Morrison et al.*, 2005]. This cloud system consisted of a horizontally-extensive low-level deck (Figure 1) that persisted over the ice-covered western Arctic Ocean for several weeks during late April and May. The period of May 7–10 is chosen here since there were few upper-level clouds; seeding of the AMPS from above was therefore negligible. The FIRE-ACE flight on May 7 measured maximum liquid water contents of about 0.06 g m^{-3} [Z05] and cloud-top temperatures of ∼−25°C.

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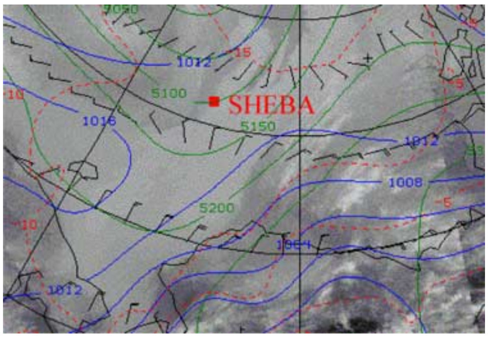

Figure 1. Advanced Very High Resolution Radiometer satellite infrared image overlayed with National Centers for Environmental Prediction (Medium Range Forecast) analysis of sea level pressure (blue, mb), surface temperature (red, °C), near-surface winds (full barb = 5 m s^{-1}), and 500 mb height (green, m) at 0000 UTC May 7.

[6] The model predicts profiles of temperature and water vapor mixing ratio along with the mixing ratios and number concentrations of cloud droplets, ice, rain, and snow. Since local updrafts are unresolved, a sub-grid model of vertical velocity is coupled to the cloud microphysics scheme that allows for realistic prediction of droplet number concentration and size [*Morrison et al.*, 2005]. The vertical resolution in the cloud layer is ~60 m with a time step of 20 s. Large-scale temperature and water vapor advection and vertical velocity are derived from the European Centre for Medium Range Weather Forecasts (ECMWF), and have been adjusted vertically to reduce biases in the cloud top height. Other physical parameterizations and aspects of the model configuration are described by *Morrison et al.* [2005].

[7] To isolate the impact of nucleation mode, two parallel sets of simulations without prognostic IN number concentration are run with 1) ice nucleation by contact freezing of drops and no deposition/condensation-freezing nucleation ('CON' simulations), and 2) ice nucleation by deposition/condensation-freezing and no contact freezing ('DCF' simulations). Two additional sets of simulations (both CON and DCF) are run with prognostic IN number concentration, allowing depletion by nucleation scavenging and entrainment of IN into the cloudy boundary layer from above (assuming that the supply of IN is constant in the free atmosphere). In situ sources of IN are neglected since these sources appear to be limited over the ice-covered Arctic Ocean [e.g., *Bigg*, 1996]. Recycling of IN is neglected since crystal sublimation is fairly insignificant here. The bulk entrainment velocity, w_e, is assumed to be 0.3 cm/s based on the mean ECMWF synoptic-scale subsidence rate at the top of the boundary layer and the assumptions that the mass flux of air into the free atmosphere and horizontal advection of mixed layer height are negligible, and the height of the boundary layer is constant. While this value of w_e is uncertain, the model exhibits little sensitivity to values of w_e < 1 cm/s typical of stratiform cloud-topped boundary layers [e.g., *Ackerman et al.*, 2004].

[8] The N_{IN} associated with contact freezing, $N_{IN,C}$, and deposition/condensation-freezing nucleation, $N_{IN,D}$, are given by *Meyers et al.* [1992] as a function of temperature (assuming a monodisperse size with radius of 0.1 μm) and ice supersaturation, respectively. Note that these formulations are based upon mid-latitude measurements and have a high degree of uncertainty; N_{IN} may vary several orders of magnitude in time and space [*Meyers et al.*, 1992]. Although the actual $N_{IN,C}$ and $N_{IN,D}$ may have varied significantly from the Meyers curve during this case, we use this formulation to illustrate the sensitivity of AMPS lifetime to $N_{IN,C}$ and $N_{IN,D}$. The collection rate of contact IN by droplets follows *Young* [1974]. Since local vertical motion and hence supersaturation fluctuations are unknown and not resolved, we assume that collection occurs through convective Brownian motion and that phoretic forces are approximately balanced over the domain. The impact of phoretic forces on the aggregate-scale collection rate is uncertain, particularly for IN larger than 0.1 μm.

[9] The mode of nucleation has a strong impact on the model results in simulations without prognostic IN. In the baseline CON simulation, the model produces a low-level AMPC that is fairly similar to observations (although somewhat less persistent), with supercooled water near cloud top and ice falling from the cloud layer to the surface (Figure 2). While the baseline DCF run produces a low-level cloud, it consists mostly of ice with little supercooled water (not shown). This difference in cloud phase is primarily attributed to large differences in ice crystal concentration (N_i) between the simulations (maximum of 0.3 vs. 10.4 L^{-1} in the CON and DCF runs, respectively) despite the fact that the baseline $N_{IN,C}$ and $N_{IN,D}$ (at water saturation) near cloud top are similar (~10–20 L^{-1}). This large difference in N_i occurs because an active contact nucleus must collide with a droplet to initiate an ice crystal in the CON run, resulting in $N_i \ll N_{IN,C}$. In contrast, an active deposition/condensation-freezing nucleus initiates an ice crystal by definition in the DCF run, so that in the absence of ice multiplication, $N_i \sim N_{IN,D}$. Because of these differences in N_i, the rate of water vapor uptake (deposition) by ice particles (i.e., Bergeron-Findeisen process) is about one order of magnitude smaller for a given bulk ice mass and ice supersaturation in the CON run compared to the DCF run. In the CON run, the condensate supply rate (produced mostly by cloud-top radiative cooling and large-scale moisture convergence) is able to balance the rate of water vapor uptake by ice particles due to the Bergeron-Findeisen

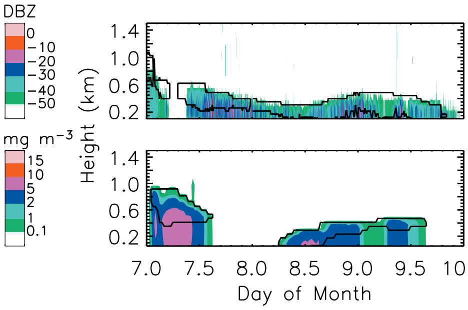

Figure 2. (top) Reflectivity (contoured) and liquid water boundaries (line) determined from SHEBA millimeter cloud radar and depolarization lidar from May 7–10, 1998, (bottom) modeled ice water content (contoured) and liquid water boundaries (line) over the same period for the baseline CON run (with no prognostic IN).

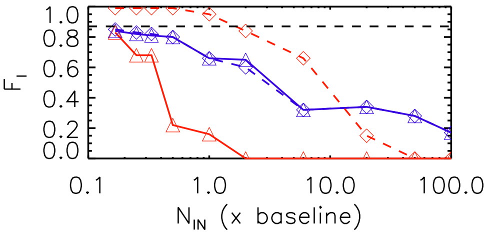

Figure 3. Sensitivity of the fraction of time with liquid water (F_l) to IN number concentration (N_{IN}) for the simulations assuming contact (blue) or deposition/condensation-freezing (red) nucleation. The solid line indicates simulations without prognostic IN number concentration, while the dashed lines indicate simulations with prognostic IN. The black dashed line indicates the observed F_l.

process and thus is able to maintain the liquid layer. This result is consistent with the explanation for the presence of supercooled water in mid-latitude cold clouds described by *Rauber and Tokay* [1991]. In contrast, water vapor uptake by ice particles due to the Bergeron-Findeisen process rapidly depletes liquid in the DCF run (depletion of liquid by riming is negligible). Note that the bulk deposition rate of water vapor onto ice particles is also influenced by the crystal habit and fallspeed [e.g., *Harrington et al.*, 1999], but the impact of these parameters is not investigated in detail here.

[10] The lifetime of the AMPC, indicated by the fraction of time liquid is present in the simulations, is much more sensitive to $N_{IN,D}$ than $N_{IN,C}$ in simulations without prognostic IN (Figure 3). As $N_{IN,C}$ is increased in the CON runs, the amount of supercooled water available for subsequent freezing is decreased. This self-regulating drop-freezing feedback limits the increase in N_i relative to the increase in $N_{IN,C}$, and hence reduces the sensitivity of the liquid fraction to $N_{IN,C}$. In contrast, an increase in $N_{IN,D}$ produces a corresponding increase in N_i (for a given temperature at water saturation), since $N_i \sim N_{IN,D}$ as described above. Thus, liquid fraction is much more sensitive to $N_{IN,D}$.

[11] In simulations with prognostic IN, depletion of contact nuclei is limited since few active contact nuclei actually initiate ice particles as described above. In contrast, all *active* deposition/condensation-freezing nuclei initiate ice particles that precipitate and are eventually removed from the system. The mean fraction of in-cloud IN that are depleted is 0.13 in CON, compared to 0.98 in DCF, assuming w_e = 0.3 cm/s and baseline values of $N_{IN,D}$ and $N_{IN,C}$. Thus, depletion has little impact on liquid fraction in the CON runs, while it substantially decreases N_i in the DCF runs, leading to a reduced Bergeron-Findeisen process and substantially increased liquid fraction for nearly all values of $N_{IN,D}$ tested (see Figure 3). Although our estimate of w_e is uncertain, the entrainment of IN is significant in the baseline DCF run only when w_e > 10 cm/s. Since deposition/condensation-freezing nuclei are depleted much more rapidly than contact nuclei, contact nucleation dominates in the cloud layer when both modes are simultaneously considered, except when w_e > 10 cm/s.

[12] We have thus far not considered the role of immersion freezing. Including immersion freezing (following *Bigg* [1953]) along with deposition/condensation-freezing and contact nucleation has little impact on the model results, except when $N_{IN,C}$ is reduced by a factor of ~2–3 or more. Note that numerous laboratory studies suggest the superior nucleating activity of particles acting in contact rather than immersion mode (see section 3). We leave further investigation of this subject for future work.

3. Observations

[13] Several observational analyses are consistent with the dominant role of drop freezing through contact nucleation in AMPC suggested by these simulations. Large numbers of spheroid- and irregularly-shaped ice crystals were observed in situ in mixed-phase clouds during FIRE-ACE [*Lawson et al.*, 2001; *Rangno and Hobbs*, 2001]. *Rangno and Hobbs* [2001] observed that maximum ice particle concentrations were dependent upon the largest droplets generated in AMPC (even in clouds occurring outside of the rime-splintering zone). Numerous laboratory experiments have demonstrated the superior nucleating ability of contact mode compared with deposition/condensation-freezing or immersion modes, particularly at temperatures above ~−20°C (summarized by *Young* [1974] and *Pruppacher and Klett* [1997]). Thus, deposition/condensation-freezing nuclei may be depleted in AMPC, but many other particles may remain active as contact nuclei. The number concentrations of deposition/condensation-freezing nuclei measured by a Continuous Flow Diffusion Chamber (CFDC) (which does not specifically measure contact nuclei) during FIRE-ACE were fairly limited, with ~50% of the measurements at zero [*Rogers et al.*, 2001]. Note that in-cloud measurements are uncertain due to potential contamination by cloud particles shattering, evaporating, and entering the CFDC [*Rogers et al.*, 2001]. Typical values of N_i in AMPC are uncertain. Measurements range from ~0.01–1 L^{-1} [e.g., *Pinto*, 1998; *Rogers et al.*, 2001] to values 2–3 orders of magnitude higher [e.g., *Lawson et al.*, 2001]. These differences are likely the result of different instrumentation and observed size ranges, and natural variability including possible ice multiplication.

[14] Ground-based cloud phase retrievals during SHEBA using cloud radar, depolarization lidar, and microwave radiometer [*Shupe et al.*, 2005b] further sug-

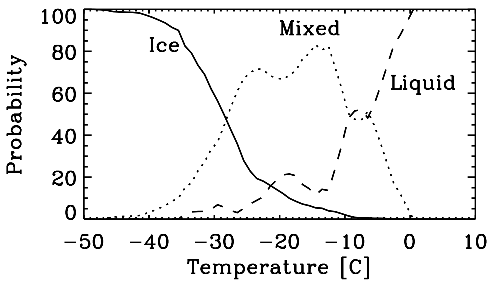

Figure 4. Percent of clouds retrieved at SHEBA as liquid, ice, or mixed-phase as a function of the minimum in-cloud temperature. Cloud systems consisting of ice precipitating into liquid or mixed-phase regions are excluded.

gest the importance of cloud drops in initiating ice formation. The fraction of clouds occurring as ice, liquid, or mixed-phase have been binned according to minimum in-cloud temperature, T_{min} (Figure 4). The presence of ice is strongly correlated with occurrence of liquid at T_{min} greater than about −20°C. Only 12% of clouds at $T_{min} = -20°C$ were all-ice; this ratio decreases further at warmer temperatures. A lack of ice clouds at temperatures above −20°C is consistent with limited deposition/condensation-freezing nucleation at these temperatures, at least in water subsaturated conditions.

4. A Conceptual Model of AMPS

[15] We now describe a conceptual model of long-lived, low-level AMPC that is consistent with the observations and modeling results described above.

[16] i) In regions that are favored for cloud development, droplets tend to form in-situ at $T > \sim -20°C$, while either droplets or all-ice clouds (at water subsaturation through deposition/condensation-freezing nucleation) form at about $-30 < T < -20°C$.

[17] ii) Ice particles nucleate by contact freezing of the largest drops and by deposition/condensation-freezing if active nuclei are initially present. Secondary ice production may occur under favorable conditions. Clouds with $T > -20$ to $-15°C$ and high droplet concentration (and thus small droplets and limited contact freezing) may remain composed primarily of supercooled drops with little or no ice.

[18] iii) Deposition/condensation-freezing nuclei are rapidly depleted by nucleation scavenging, while scavenging has much less impact on the concentration of contact nuclei. Thus, contact nucleation generally dominates in the cloud layer.

[19] Since contact freezing dominates in this conceptual model, the primary formation of ice exhibits a self-regulating feedback that helps to explain the persistence of liquid water in AMPC; as described previously, an increase in the contact nucleation rate reduces the amount of liquid water available for subsequent freezing. On the other hand, limited depletion of contact nuclei helps to maintain ice.

5. Conclusions

[20] A series of simulations using a 1-D cloud model have suggested that the ice nucleation mode has a strong impact on the lifetime of Arctic mixed-phase stratus. In simulations without prognostic IN, assuming that deposition/condensation-freezing nucleation was dominant led to strong sensitivity of mixed-phase cloud lifetime to IN concentration consistent with previous studies [e.g., *Pinto*, 1998; *Harrington et al.*, 1999; *Jiang et al.*, 2000]. In simulations with prognostic IN, rapid depletion of deposition/condensation-freezing nuclei occurred through nucleation scavenging and precipitation, leading to substantially increased mixed-phase cloud lifetime. In contrast, depletion had little impact on contact nucleation, since few active contact nuclei actually collided with droplets and initiated ice. Thus, contact nucleation dominated in the cloud layer when both modes were simultaneously considered except under very large entrainment velocities (>10 cm/s). The dominant role of contact nucleation is consistent with in situ observations, ground-based retrievals, and laboratory experiments. The potentially dominant role of contact nucleation in mid-latitude mixed-phase clouds has also been suggested [e.g., *Young*, 1974; *Hobbs and Rangno*, 1985]. Based upon the observations and modeling results highlighted in this paper, a conceptual model of long-lived Arctic mixed-phase clouds was developed that explains their persistence in terms of the depletion of deposition/condensation-freezing nuclei and a self-regulating feedback involving drop freezing by contact nucleation. Detailed, high-resolution 3-D simulations with a binned representation of the cloud microphysics are needed to better understand and quantify this conceptual model. The potentially dominant role of contact nucleation in Arctic mixed-phase clouds, and some mid-latitude mixed-phase clouds, suggests the need for its treatment in climate and weather prediction models and the development of field-deployable instruments to measure the size and number concentration of active contact nuclei in the atmosphere.

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