The Effect of Ice Nuclei Efficiency on Arctic Mixed-Phase Clouds from Large-Eddy Simulations

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ABSTRACT

The effects of ice nuclei (IN) efficiency on the persistent ice formation in Arctic mixed-phase clouds (AMCs) are investigated using a large-eddy simulation model, coupled to a bin microphysics scheme with a prognostic IN formulation. In the three cases where the IN efficiency is high, ice formation and IN depletion are fast. When the IN concentration is 1 and 10 g$^{-1}$, IN are completely depleted and the cloud becomes purely liquid phase before the end of the 24-h simulation. When the IN concentration is 100 g$^{-1}$, the IN supply is sufficient but the liquid water is completely consumed so that the cloud dissipates quickly. In the three cases when the IN efficiency is low, ice formation is negligible in the first several hours but becomes significant as the temperature is decreased through longwave cooling. Before the end of the simulation, the cloud is in mixed phase when the IN concentration is 1 and 10 g$^{-1}$ but dissipates when the IN concentration is 100 g$^{-1}$. In the case where two types of IN are considered, ice formation persists throughout the simulation. Analysis shows that as the more efficient IN are continuously removed through ice formation, the less efficient IN gradually nucleate more ice crystals because the longwave cooling decreases the cloud temperature. This mechanism is further illustrated with a simple model. These results indicate that a spectrum of IN efficiency is necessary to maintain the persistent ice formation in AMCs.

1. Introduction

Arctic mixed-phase cloud (AMC) consists of a thin mixed-phase layer and an underlying ice-phase layer (Morrison et al. 2012). It exerts a positive radiative forcing to the surface because it can absorb the upwelling longwave radiation and emit longwave radiation to the surface (Shupe and Intrieri 2004; Garrett and Zhao 2006; Dong et al. 2010). The representation of AMC and its climatic effect in large-scale models is still a big challenge (Xie et al. 2013; English et al. 2014). The difficulty mainly arises from the fact that the maintenance of AMC depends on the complex interactions between the large-scale environment, cloud microphysics, cloud dynamics, and radiation (Harrington et al. 1999; Harrington and Olsson 2001; Korolev and Isaac 2003; Ervens et al. 2011; Stramler et al. 2011; Yang et al. 2015), and these interactions have not been well understood (Morrison et al. 2012).

One of the unresolved problems is that the persistent ice formation in AMC cannot be reproduced in models with prognostic ice nuclei (IN) formulations. Cases from the Mixed-Phase Arctic Cloud Experiment (M-PACE; Verlinde et al. 2007) and the Beaufort Arctic Storms Experiment (BASE; Curry et al. 1997) were simulated with a large-eddy simulation (LES) model, which was incorporated with a detailed microphysics scheme (Fridlind et al. 2007). It was found that IN were quickly consumed and the clouds became completely liquid phase in a short time. This result is not consistent with the observed persistence of ice formation. A similar result was also found for a case from the Surface Heat Budget of the Arctic Ocean campaign (SHEBA; McFarquhar et al. 2011; Fridlind et al. 2012). This discrepancy between the simulation results and the observational facts indicates that the treatment of the ice-related processes in models needs further improvement.

Several mechanisms have been proposed that can possibly prolong the ice formation in models. The key point is that a prolonged IN supply is needed in models. For example, it is known that contact nuclei could not be efficiently removed through nucleation scavenging. When contact nuclei were treated separately with the other ice...
nuclei in the model, they could remain in the atmosphere, dominate ice formation, and contribute to the persistent ice formation (Morrison et al. 2005). However, the dominant role of contact nuclei in AMC was not supported by some observational studies (de Boer et al. 2010, 2011). A second mechanism is that IN could be entrained into the cloud from the layer below (Avramov et al. 2011). These IN could compensate for the loss of the in-cloud IN and the ice formation could be significantly prolonged. IN recycling was also found to contribute to the continuous ice formation. This is because the IN released by the complete sublimation of ice crystals could catalyze ice formation again (Solomon et al. 2015).

Ice nucleation was parameterized with the deterministic theory in most of the previous modeling studies (Morrison et al. 2005; Avramov et al. 2011; Solomon et al. 2015). In the deterministic theory, each IN instantaneously nucleates an ice crystal once its critical temperature is reached (Pruppacher and Klett 1997; Lamb and Verlinde 2011; Murray et al. 2012). For a given temperature, the concentration of IN can be obtained from an empirical formula (e.g., DeMott et al. 2010). However, using the deterministic theory and the measured cloud-top temperature, Westbrook and Illingworth (2013) found that the calculated number of IN was much smaller than the observed number of ice crystals in a quasi-stationary midlevel stratiform cloud case. They therefore suggested that the deterministic theory might not be applicable in stratiform mixed-phase clouds. Instead, they suggested that the classical nucleation theory might be able to explain the persistent ice formation in this type of cloud.

In the classical nucleation theory, ice nucleation is considered to be stochastic (Pruppacher and Klett 1997; Lamb and Verlinde 2011; Murray et al. 2012). For a given temperature, each IN has a probability of nucleating an ice crystal, and the IN with higher efficiency have larger probability. Westbrook and Illingworth (2013) suggested that an ensemble of inefficient IN is capable of nucleating ice crystals for a long time. Each IN in the ensemble only has a small probability of nucleating an ice crystal so that the depletion of IN is very slow and the IN can be available for a long time.

In the classical nucleation theory, the radius of the IN $r_N$ and the contact angle $\theta$ between the ice and the surface of the IN are important parameters determining the nucleation rate. Previous studies found that the nucleation rate was much more sensitive to $\theta$ than to $r_N$ (Ervens and Feingold 2012). Therefore, $r_N$ is usually set to be a fixed value, and $\theta$ is the only variable describing the property of the IN in models. On an IN with a smaller $\theta$, the formation of an ice embryo is associated with a lower energy barrier and the ice nucleation is easier. This means that a smaller $\theta$ corresponds to a higher IN efficiency.

In previous modeling studies, the same $\theta$ was usually assigned for all IN (e.g., Hoose et al. 2010; Li et al. 2013; Fan 2013; Ervens and Feingold 2012). This treatment implicitly assumed that the surface properties were homogeneous for all IN. However, some studies found that, if assuming the same $\theta$ for all IN, the predicted temporal variation of droplet frozen fraction deviated significantly from that of the experimental result, while if assuming more than one $\theta$ for the IN, the prediction matched the experimental result very well (Marcolli et al. 2007; Luond et al. 2010; Welti et al. 2012; Ervens and Feingold 2012; Kulkarni et al. 2012). This indicates that the surface properties are actually inhomogeneous for the IN population.

The so-called $\theta$-PDF scheme was implemented into a two-moment microphysics scheme to treat the inhomogeneity of the IN efficiency in an LES study of AMC (Savre and Ekman 2015). In the $\theta$-PDF scheme, different IN have different $\theta$ and hence different efficiencies. In Savre and Ekman (2015), a normal distribution of $\theta$ was assumed. Their results showed that ice formation persisted throughout the simulations. The IN with high efficiency were quickly depleted in the earlier time of the 6-h simulations, while the IN with low efficiency remained in the AMC and catalyzed the ice formation in the later time. Although the assumption of the normal distribution of $\theta$ has not been proved by observations, and the simulation time is relatively short, Savre and Ekman (2015) suggested that the inhomogeneity of the IN efficiency might be important for the maintenance of the ice phase in AMC.

In this study, we use an LES coupled with a detailed bin microphysics scheme that has a prognostic IN formulation for mixed-phase clouds to examine the effect of IN efficiency on the persistent ice formation in AMC. Section 2 describes the method. Section 3 presents the simulations with only the high-efficiency IN, and the simulations with only the low-efficiency IN as well, in various IN concentrations. This section also presents the results of a simulation with two types of IN. Section 4 describes the results of a simple model which shows the effect of both the variability of the IN efficiency and the longwave cooling in maintaining the ice formation. A summary is given in section 5.

2. Method

a. The microphysics scheme for mixed-phase clouds

A bin microphysics scheme for mixed-phase clouds is used in this study. For the liquid phase, we consider droplet activation, condensational growth/evaporation, collision–coalescence, sedimentation, and the conversion of liquid droplets into ice crystals through immersion freezing. The initial aerosols that can act as CCN are assumed to be ammonium sulfate following a lognormal
size distribution with a mean diameter of 0.2 μm and a geometric standard deviation of 1.5 μm. The activation of aerosols is based on the model-predicted supersaturation. The aerosols are activated if their dry sizes are larger than the critical dry aerosol size for that supersaturation. The droplet spectrum is represented by 25 mass-doubling bins. Details of the liquid-phase microphysics can be seen in Xue and Feingold (2006), Feingold et al. (1996), and Tzivion et al. (1987).

For the ice phase, we consider ice nucleation, depositional growth/evaporation, and sedimentation. Details of the initial setups of the IN size, efficiency, and number concentration will be given in section 2b. The number concentration of IN is predicted. IN are transported by the airflow and depleted by the nucleation of ice crystals. Upon the complete sublimation of ice crystals, IN are recycled. The recycled IN preserve their original size and efficiency and can be used for further ice nucleation. Because previous studies have pointed out that immersion freezing was the major heterogeneous nucleation mode through which IN catalyzed ice formation in AMC (e.g., de Boer et al. 2010, 2011), only immersion freezing is considered in this study. The spectrum of ice crystals are represented by 25 mass-doubling bins, similar to the treatment of the liquid droplet spectrum. Rimming (ice–liquid collection) and aggregation (ice–ice collection) processes are not considered in the model in this study. The microphysics scheme now can simulate the effects of IN on the mixed-phase clouds and can also simulate the interactions between the liquid phase and ice phase, such as the Wegener–Bergeron–Findeisen process.

Ice nucleation through immersion freezing is calculated based on the classical nucleation theory. The details of the theory can be found in textbooks (Pruppacher and Klett 1997; Lamb and Verlinden 2011) and previous literatures (e.g., Chen et al. 2008; Hoose et al. 2010; Li et al. 2013). We briefly present the theory here. The number of ice crystals formed in a time step Δt is \( n_{IN} \) \([1 - \exp(-J_{he} \Delta t)]\), where \( n_{IN} \) is the IN concentration and \( J_{he} \) the heterogeneous nucleation rate. As in Khvorostyanov and Curry (2000), \( J_{he} \) is expressed as

\[
J_{he} = \frac{k_B T}{h} 4\pi T_i c_{irs} \exp \left( -\frac{\Delta g^\phi}{RT} - \frac{\Delta F_{g,s}}{k_B T} \right),
\]

where \( k_B \) is the Boltzmann constant, \( T \) the temperature (K), \( h \) the Planck constant, \( c_{irs} \) the number of water molecules in contact with unit area of IN surface, and \( R \) the universal gas constant. The activation energy for water molecules to diffuse across the water–ice interface \( \Delta g^\phi \) (kcal mol\(^{-1}\)) is parameterized as (Pruppacher and Klett 1997)

\[
\Delta g^\phi = 5.55 \exp(-8.423 \times 10^{-3} T + 6.384 \times 10^{-4} T^2 + 7.891 \times 10^{-6} T^3),
\]

where \( T \) is in degrees Celsius. In Eq. (1) \( \Delta F_{g,s} \) is the energy barrier that has to be overcome in the ice nucleation event and satisfies \( \Delta F_{g,s} = (4/3) \pi r_{in}^2 f(m, x) \), where \( r_{in} \) is the surface tension of the water–ice interface and \( f(m, x) \) is the geometrical factor. The contact coefficient \( m = \cos \theta \) and \( x \) is the ratio of \( r_g \) to \( r_N \). The critical radius \( r_g \) is expressed as \( r_g = 2 \sigma_{in}/[L_{\text{ef}} \rho_i \ln(T_0/T)] \), where \( \rho_i \) is the density of ice. \( T_0 \) is 273.15 K, and \( L_{\text{ef}} \) is the effective melting heat introduced in Khvorostyanov and Sassen (1998). The model also records the ice-forming rate, which is used to study where and how fast the ice crystals form. Here, the ice-forming rate refers to the number of newly formed ice crystals in unit mass of air and unit time (g\(^{-1}\) h\(^{-1}\)). It is affected by the nucleation rate of every individual IN as well as the concentration of the IN.

The depositional growth of ice crystals follows the capacitance theory in Ovchinnikov et al. (2014), where the ice crystals were treated to be spherical and have a low effective density. The effects of ventilation and radiation on the growth of ice crystals are both neglected. The growth rate of an individual ice crystal is

\[
\frac{dm}{dt} = BC \Delta S_i,
\]

where \( m_i \) is the mass of the ice crystal. The capacitance \( C \) is related to \( m_i \) by \( C = a_i m_i^{b_i} \) with \( a_i = 0.09 \) m kg\(^{-b_i}\) and \( b_i = 0.5 \). The supersaturation over ice is \( \Delta S_i = q_i/q_{is} - 1 \), where \( q_i \) is the specific humidity and \( q_{is} \) is the saturation specific humidity over ice. The factor \( B \) satisfies

\[
B = \frac{4\pi}{q_{is} \left( \frac{1}{R_{\text{VT}}} - 1 \right) \frac{l_i}{kT} + \frac{R_i T}{D_{\text{es}}}},
\]

where \( l_i \) is the latent heat of sublimation, \( R_i \) the gas constant of vapor, \( k \) the heat conductivity of air, \( D \) the diffusivity of vapor in air, and \( e_{is} \) the saturation vapor pressure over ice.

The terminal velocity of an ice crystal \( V_i \) satisfies

\[
V_i = a_v D^{b_v},
\]

where \( a_v = 12 \) m\(^{-1}h^{-1}\) and \( b_v = 0.5 \). The maximum dimension of the ice crystal \( D \) satisfies \( D = \pi C \).

When only liquid-phase processes are considered in the microphysics scheme, the model predicts the liquid water potential temperature and the total water mixing ratio, which is the sum of the mixing ratios of water vapor and liquid water. When ice-phase processes are
added to the microphysics scheme, the effect of ice water on thermodynamics is also considered by extending the liquid water potential temperature to the liquid–ice water potential temperature (Tripoli and Cotton 1981). The total water mixing ratio is now the sum of the mixing ratios of water vapor, liquid water, and ice water.

b. Large-eddy simulation model and case setup

LESs are capable of performing realistic simulations of the three-dimensional (3D) atmospheric flow and have been widely used to study the mixed-phase clouds (e.g., Fridlind et al. 2007, 2012; Solomon et al. 2011, 2015). In this study, UCLA-LES is used (Stevens et al. 1999). This model solves the 3D anelastic momentum equation and the thermodynamics equation. Liquid–ice water potential temperature and total water mixing ratio (including water vapor, liquid water, and ice water) are the prognostic variables (Yamada and Mellor 1979; Tripoli and Cotton 1981). The longwave radiation is parameterized with a two-stream radiative transfer scheme (Larson and Kotenberg 2007). Previous studies found that shortwave radiation also played an important role in the development of AMCs (Solomon et al. 2015; Wang et al. 2015). However, in order to avoid the complexity induced by shortwave radiation and focus on the effect of IN efficiency, shortwave radiation is not considered in this study. The model has $64 \times 64 \times 131$ grid points with a resolution of $35\,\text{m} \times 35\,\text{m} \times 10\,\text{m}$. In addition, the ice-phase microphysics is turned on at 2 h in order to let the turbulence develop.

The case selected for this study is a semi-idealized case adopted from an intercomparison project of AMC (Ovchinnikov et al. 2014). It is a typical long-lived single-layer decoupled mixed-phase cloud. The initial profile is a composite of the idealization of a sounding at Barrow, Alaska, (now known as Utqiaġvik) and the aircraft observation in the Indirect and Semi-Direct Aerosol Campaign (ISDAC; McFarquhar et al. 2011). All the case-dependent settings are as in Ovchinnikov et al. (2014) except that no nudging is performed for wind components, temperature, or moisture. The initial profiles are shown in Fig. 1. A remarkable feature is that the layer below 0.4 km is stable and has a high total water mixing ratio. In addition, during the selected case, the majority of ice crystals were pristine dendrites. Both riming and aggregation events were negligible (Ovchinnikov et al. 2014). As an approximation, the ice crystals are treated to be dendrites and are represented by the spherical ice crystals with low density, as presented in section 2a.

We use different contact coefficients to represent different types of IN in this study. We first perform simulations where each case has only one type of IN, that is, one contact coefficient. These simulations are performed to unravel the complicated feedbacks in the AMC without involving the complex interaction between different types of IN. As will be shown, the results of these simulations can greatly help us to understand the simulation which has two types of IN. The value of contact coefficient cannot be directly measured and is usually inversely derived from experimental data (Chen et al., 2014).
The derived value is usually associated with significant uncertainty (Chen et al. 2008; Hoose et al. 2010; Murray et al. 2012), so that previous studies used significantly different values for contact coefficients, such as 0.5 (Khourystyanov and Curry 2005; Li et al. 2013) and 0.86 (Hoose et al. 2010; Fan 2013). In this study, we use a contact coefficient of 0.63 for the high-efficiency IN and 0.54 for the low-efficiency IN. The corresponding nucleation rates are shown in Fig. 2. At the same temperature, the IN with a contact coefficient of 0.63 has a higher nucleation rate than the IN with a contact coefficient of 0.54. The onset of nucleation is defined as the point when the nucleation rate $J_{th}$ becomes greater than $10^{-5}\text{s}^{-1}$ and the corresponding temperature is defined as the onset temperature (Hoose et al. 2010). Based on this definition, the onset temperature is 261.0 K for the contact coefficient of 0.63 and 258.5 K for the contact coefficient of 0.54. As will be shown, the contact coefficient of 0.63 is selected for the high-efficiency IN because its onset temperature is higher than the initial cloud-top temperature, so that the IN with a contact coefficient of 0.63 have a high nucleation rate from the very beginning of the simulation. The contact coefficient of 0.54 is selected for the low-efficiency IN because its onset temperature is lower than the initial cloud-top temperature and can be reached as the longwave cooling cools the cloud. The IN with a contact coefficient of 0.54 can therefore have a very low nucleation rate at the early stage and a relatively high nucleation rate as the cloud-top temperature decreases.

In the simulations with only one type of IN, the IN concentration is set to be 1, 10, and 100 g$^{-1}$. These concentrations are within the observed concentration of dust aerosol in the Arctic. Previous studies found that dust aerosol could be efficiently transported into the Arctic (Quinn et al. 2007; Fan 2013; Atkinson et al. 2013) and catalyze ice formation (Rogers et al. 2001; Prenni et al. 2009). The concentration of dust aerosol in the Arctic varies from nearly 0 to as high as 1500 $\mu$g m$^{-3}$ (Fan 2013). This corresponds to about 1400 g$^{-1}$ when assuming the density of dust aerosol to be 2000 kg m$^{-3}$ and $r_N$ to be 500 nm. The estimated concentration of dust aerosol is consistent with that in Atkinson et al. (2013).

A simulation that has two types of IN is then performed. This simulation is a simple implementation of the $\theta$-PDF scheme. Ideally, far more than two types of IN should be used in the $\theta$-PDF scheme. For example, a continuous normal distribution was used to represent the $\theta$-PDF in Savre and Ekman (2015). However, using many types of IN in the bin microphysics scheme is computationally expensive. We therefore use only two types of IN to demonstrate the importance of the variability of the IN efficiency in maintaining the ice formation in AMC. DeMott et al. (2010) found that the IN concentration increases exponentially as temperature decreases. This indicates that the lower-efficiency IN, which are activated at lower temperature, have a higher concentration than the higher-efficiency IN. Thus, we set the high-efficiency IN (contact coefficient of 0.63) to have a concentration of 1 g$^{-1}$ and the low-efficiency IN (contact coefficient of 0.54) to have a concentration of 10 g$^{-1}$. In the simulation with two types of IN, ice crystals nucleated on each type of IN are treated separately. There are 25 bins for the spectrum of ice crystals from each type of IN. This treatment allows the recycled IN to preserve their original efficiencies.

3. Results

a. Simulations with high-efficiency IN

Figure 3 shows the evolution of the cloud in the case with a contact coefficient of 0.63 and an IN concentration of 1 g$^{-1}$. We first use this case as an example to explain the feedbacks involving microphysics, radiation, and dynamics in AMC. Liquid water forms between 0.65 and 0.8 km immediately after the simulation begins (Fig. 3a). The longwave cooling, which is sustained by the liquid water, decreases the temperature near the cloud top (Fig. 3b). The decreased temperature then enhances the growth of liquid droplets. More importantly, this negative buoyancy near the cloud top induces turbulence in the boundary layer. Water vapor near the surface is then transported upward (Fig. 3c) and contributes to the rapid thickening of the liquid layer within the first 6 h (Fig. 3a). Therefore, the base of the liquid layer is lowered to 0.45 km, and the maximum liquid water mixing ratio increases to about 0.2 g kg$^{-1}$.

Because the temperature of the liquid layer is lower than 261.0K, which is the onset temperature of ice
formation for the IN with a contact coefficient of 0.63, ice crystals readily form between 0.6 and 0.8 km right after the ice-phase processes are turned on at 2 h (Figs. 3c and 3d). The ice-forming rate generally decreases with time (Fig. 3d). This is because the ice formation continuously consumes IN, as evident from the decreasing IN concentration (Fig. 3d). The slight increase of the ice-forming rate from 5 to 6 h is caused by the IN that are transported into the cloud from the surface. After formation, the ice crystals grow and some of the ice crystals fall out as precipitation. The combined effect of the decreasing ice-forming rate and the precipitation removal of ice crystals results in the decrease of the ice water mixing ratio (Fig. 3c). The ice water mixing ratio is on the order of 0.001 g kg\(^{-1}\) (Fig. 3c). This is much smaller than the liquid water mixing ratio. As a result, the depositional growth of ice crystals does not substantially affect the liquid droplets.

After 6 h, the water vapor is well mixed in the boundary layer (Fig. 3c). The longwave cooling continues decreasing the temperature and maintains the growth of liquid droplets. The liquid layer therefore keeps thickening. In the meanwhile, the ice-forming rate decreases because of the depletion of IN. In addition, the ice crystals keep falling out. Consequently, the ice water mixing ratio decreases and becomes negligible after 10 h.

The time series of the averaged cloud properties for the three cases where the IN efficiency is high are shown in Fig. 4. When the IN concentration is 1 g\(^{-1}\), the LWP rapidly increases before 6 h and then slowly increases until the end of the simulation (Fig. 4a). This is consistent with the characteristics of the liquid layer development in Fig. 3a. The evolution of the LWP is also quantitatively consistent with that in the intercomparison project (Ovchinnikov et al. 2014). Both the IWP and the ice concentration peak at 3 and 6 h and then become negligible after 10 h. The IN are quickly depleted and only 1% of the initial IN are left in the domain at 10 h. The increases of the IWP and the ice concentration before 3 h result from the fast formation of ice crystals, while the decreases after 3 h are caused by the IN depletion and also the precipitation removal of ice particles. The peaks at 6 h result from the vertical IN transport from the layer near the surface, as shown in Fig. 3d. In this case, the ice formation is strongly limited by the low initial IN concentration and the fast depletion of IN. The rapid consumption of IN is also seen in previous studies where IN concentration is treated as a prognostic variable (Harrington and Olsson 2001; Fridlind et al. 2007, 2012).

Compared to the case with an IN concentration of 1 g\(^{-1}\), the case with an IN concentration of 10 g\(^{-1}\) has a
smaller LWP, a larger IWP, and a higher ice concentration (Fig. 4). Both the IWP and the ice concentration peak at 3 and 8 h, and become negligible after 20 h, indicating that the ice phase persists for several hours longer than that in the case with an IN concentration of 1 g$^{-1}$. The higher IN concentration in this case results in the formation of more ice crystals (Fig. 4c), which can significantly affect the liquid phase through the competition for vapor. The depositional growth of these ice crystals leads to the rapid increase of the IWP (Fig. 4b) and the fast decrease of the LWP from 2 to 3 h (Fig. 4a). Because the ice formation through immersion freezing needs supplies of liquid water, the reduction of liquid water then limits the following ice formation. The IWP and the ice concentration then quickly decrease from 3 h (Figs. 4b and 4c) as ice formation is slowed down and precipitation removal of ice crystals continues. When the LWP is sufficiently small (i.e., <30 g m$^{-2}$; Garrett et al. 2002), a thinner liquid layer produces a weaker longwave cooling and hence a weaker turbulence, causing the vapor below 0.4 km to be transported into the cloud at a slower rate but for a longer time. Therefore, both the LWP and the IWP increase until 8 h, which is later than that in the case with an IN concentration of 1 g$^{-1}$. In addition, the entrainment also adds IN into the cloud and contributes to the increase of the ice concentration from 7 to 8h. After 8 h, the IN are significantly depleted so that both the IWP and the ice concentration decrease, and the LWP increases as a result of the longwave cooling. In this case, the liquid water amount is the limiting factor for ice formation before 8 h but the IN concentration becomes the limiting factor after 8 h.

In the case with an IN concentration of 100 g$^{-1}$, the very high IN concentration results in an ice concentration of 30 g$^{-1}$ (Fig. 4c) and an IWP of 60 g m$^{-2}$ (Fig. 4b). The depositional growth of such a large amount of ice crystals strongly suppresses the growth of liquid droplets so that the LWP becomes negligible in a very short time (Fig. 4a). Consequently, the longwave cooling is shut down, and the ice layer dissipates in less than 1 h. In this case, the IN are depleted by less than 20% in the short lifetime of the cloud. It is the amount of the liquid water instead of the IN that limits the formation of ice crystals.

**b. Simulations with low-efficiency IN**

Figure 5 shows the evolutions of the averaged cloud properties in the three cases where IN efficiency is low (i.e., contact coefficient of 0.54). Before 6h, the LWPs in the three cases are almost identical and increase rapidly, whereas the IWPs are almost negligible. In these cases, the initial temperature of the cloud is higher than 258.5 K, which is the onset temperature of ice formation for the IN with a contact coefficient of 0.54. The initial ice formation is therefore very slow. However, as the longwave cooling decreases the temperature, the ice formation becomes faster and the IWP starts to increase. After 6 h, in the case with an IN concentration of 1 g$^{-1}$, the evolution of the LWP is almost the same as that in the case where the contact coefficient is 0.63 and the IN
The IWP slowly increases as a result of the longwave cooling but is always less than 1 g m$^{-2}$ (Fig. 5b). The IN concentration decreases with time (Fig. 5c). The ice concentration increases with time before 20 h but is still very low owing to the very low IN concentration. After 20 h, the ice concentration starts to decrease. This is because the ice formation is reduced as a result of the severe IN depletion and also because precipitation removes ice crystals.

Compared to the case with an IN concentration of 1 g$^{-1}$, the case with an IN concentration of 10 g$^{-1}$ has a smaller LWP, a larger IWP, and a higher ice concentration (Fig. 5c). In this case, the IN concentration is larger so that more ice crystals are nucleated (Fig. 5c) and the IWP is larger. This leads to a smaller LWP because of the competition for water vapor between the ice crystals and the liquid droplets. After 20 h, the formation of ice crystals is also reduced (Fig. 5c), similar to the case with an IN concentration of 1 g$^{-1}$.

In the case with an IN concentration of 100 g$^{-1}$, the ice concentration is even larger because of the very large IN concentration (Fig. 5c). The growth of these ice crystals rapidly increases the IWP (Fig. 5b). It also strongly suppresses the growth of the liquid droplets, resulting in the fast decrease of the LWP (Fig. 5a). When the LWP is too small, the longwave cooling becomes too weak to sustain the turbulence so that the cloud dissipates. In this case, the depletion of IN is also less than 20%.

For the three cases where the IN efficiency is low, ice crystals do not form in the first several hours but form in the later hours, because it takes a long time for the longwave cooling to cool the clouds before the low-efficiency IN can take effect. This is different from the cases where the IN efficiency is high. In those cases, ice crystals form in the first several hours but do not form in the later hours. In addition, no matter what the IN efficiency is, the clouds dissipate when the IN concentration is 100 g$^{-1}$. It is therefore clear that both the efficiency and the concentration of the IN have important effects on the evolutions of the AMCs.

c. Simulations with two types of IN

Results from the simulation with two types of IN are shown in Fig. 6. A striking feature is that the formation of ice crystals can persist throughout the time period from 2 to 24 h (Figs. 6b and 6c). In the first half of the simulation, the formation of ice crystals is dominated by the high-efficiency IN, as evident from the fact that the high-efficiency IN is significantly depleted while the low-efficiency IN remains unchanged (Fig. 6c). The depletion of the high-efficiency IN leads to the decrease of the ice concentration (Fig. 6c) as well as the IWP (Fig. 6b), similar to those in the case where the contact coefficient is 0.63 and the IN concentration is 1 g$^{-1}$. In this period, the high-efficiency IN have been significantly depleted, while the ice formation due to the low-efficiency IN has become sufficiently fast because of the decreased temperature driven by the longwave cooling. Consequently, the ice concentration (Fig. 6c) and the IWP (Fig. 6b) both increase, as in the case where the contact coefficient is 0.54 and the IN concentration is 10 g$^{-1}$.
The height at which the IN nucleate ice crystals is closely related to their efficiency. For the high-efficiency IN, they nucleate ice crystals at all heights in the cloud during the short time right after the ice process is turned on (near 2 h in Fig. 6d). However, they are quickly depleted because of the rapid ice formation. More high-efficiency IN are transported into the cloud from the subcloud layer and are quickly consumed near the cloud base. Consequently, the effect of the high-efficiency IN is confined near the cloud base. For the low-efficiency IN, the effects are mainly confined near the cloud top because they require a low temperature to take effect. There are no ice crystals forming in the region between the cloud-base area and the cloud-top area (Fig. 6d). This is because the two types of IN have a very big difference in efficiency. When a more continuous spectrum of IN efficiency is used, ice crystals could form contiguously in the cloud.

4. A simple model with multiple types of IN

A simple model with multiple types of IN is developed to further investigate the role of the variability of IN efficiency in ice formation. In terms of the contact coefficient, or equivalently the IN efficiency, the IN are divided into $k$ bins. The initial IN number distribution is $n(m) = dn/dm = 5(k - i + 1)^2$, where $i$ is the bin index from 1 to $k$, and $m$ is the contact coefficient of IN. Here, we again assume that the concentration of the IN decreases with its efficiency, similar to the case with two types of IN in section 3c. We have $k = 9$ bins ranging from $m = 0.54$ to 0.63 with a bin width of 0.01. The total IN concentration is hence $14.25 \, \text{g}^{-1}$. Note that the spectrum of IN efficiency used in this study is quite different from the spectrum used in Savre and Ekman (2015), where a normal distribution of IN efficiency is used. The fact that low-efficiency IN has a higher number concentration than the high-efficiency IN (as discussed in section 2b) might be very important for the persistent ice formation in AMCs. The number of ice crystals formed in a time step $\Delta t$ is $n(m)[1 - \exp(-J_{bc}\Delta t)]dn$, which is the same as that in the LES model. Every time ice crystals form, the same amount of IN are depleted from the IN bin. IN recycling is not considered in this simple model.

The calculation of $J_{bc}$ requires the temperature. The initial temperature in the simple model is 259 K, which is the initial cloud-top temperature in the LES. The cooling rate in the simple model is prescribed to be 1, 2, and 3 K day$^{-1}$. The cooling rate of 2 K day$^{-1}$ is similar to that of the case where the contact coefficient is 0.63 and the IN concentration is 1 g$^{-1}$ (Fig. 4b). In a real cloud, the cooling rate depends on the strength of the cloud-top longwave cooling, which further depends.

FIG. 6. The evolutions of (a) LWP, (b) IWP, (c) ice concentration (solid line), and IN concentration with contact coefficient of 0.63 (thick dashed line) and 0.54 (thin dashed line), and (d) ice-forming rate, for the cases with two types of IN (one with a contact coefficient of 0.63 and an IN concentration of 1 g$^{-1}$, and the other with a contact coefficient of 0.54 and an IN concentration of 10 g$^{-1}$). Both the LWP and the IWP are averaged over the whole domain. The ice concentration and the IN concentration are averaged horizontally over the whole domain and vertically from the surface to 0.8 km. The ice-forming rate is horizontally averaged over the whole domain.
on the LWP. When the LWP is sufficiently large (i.e., $>30 \text{ g m}^{-2}$), the cloud is like a blackbody in the longwave band and has a nearly constant longwave emissivity of 1, so that the longwave cooling is not sensitive to the LWP (Garrett et al. 2002). However, when the LWP is very small, the cloud emissivity decreases with decreasing the LWP. As a result, different clouds can have different cooling rates.

Figures 7a–c show the distributions of the ice-forming rate. For a given cooling rate, the peak of the ice-forming rate is initially at regions with $m > 0.57$, which corresponds to the very efficient IN. These IN readily nucleate ice crystals and result in a large total ice-forming rate before 1 h (Fig. 7d). As time goes on, the peak of the ice-forming rate shifts from the larger $m$ to the smaller $m$. This means that the more efficient IN are gradually depleted, while the less efficient IN gradually nucleate more ice crystals as the longwave cooling decreases the cloud temperature. Consequently, the ice formation persists for a long time (Fig. 7d). For a given IN ensemble, increasing the cooling rate increases the total ice-forming rate but decreases the persistence of ice formation (Fig. 7d). Therefore, under the considered scenario, an appropriate cooling rate must be provided by the longwave cooling so that both the rate and the duration of ice formation can be maintained. Although using a different distribution for IN efficiency, Savre and Ekman (2015) also found that longwave cooling was important in maintaining the persistent ice formation in AMCs.

In this simple model, it is implicitly assumed that the parcel is always at the cloud top. Because the cloud top has the lowest temperature, the depletion of the IN is the fastest. In a real cloud, the parcel can stay at the cloud top for only a short time before it is transported to a warmer place by the turbulence. The depletion of IN is hence slower than that at the cloud top. As a result, the duration of ice formation in a real cloud is much longer than that in the simple model. Besides, the fixed cooling rate means that the simple model cannot represent the feedbacks involving liquid water, ice crystal, longwave cooling, and turbulence. Apart from these limitations, the results from this simple model show the importance of both the variability of the IN efficiency and the longwave cooling in maintaining the ice formation.

5. Summary

The impact of the IN efficiency on the maintenance of ice formation in AMC is investigated with an LES model, coupled with a prognostic IN formulation and a bin microphysics scheme for mixed-phase clouds.

Three simulations with only the high-efficiency IN are first investigated. In these simulations, ice crystals form
right after the ice process is turned on. When the IN concentration increases from 1 to 10 g\(^{-1}\), the IWP becomes larger while the LWP becomes smaller. This is because increasing IN concentration results in more ice crystals. The growth of these ice crystals consumes a lot of vapor and hence reduces the growth rate of liquid droplets. In addition, the fast depletion of the IN and the precipitation removal of ice crystals in the cases with IN concentrations of 1 and 10 g\(^{-1}\) leads to purely liquid-phase clouds before the end of the 24-h simulation. When the IN concentration is 100 g\(^{-1}\), the ice concentration is very high. The growth of these ice crystals substantially suppresses the growth of liquid droplets. The longwave cooling is then shut down and the cloud dissipates quickly.

In the simulations with only the low-efficiency IN, ice crystals do not form until 6 h, when the longwave cooling has decreased the temperature to be sufficiently low for these low-efficiency IN to take effect. In the cases with IN concentrations of 1 and 10 g\(^{-1}\), the clouds remain mixed phase until the end of the simulations. In the case with an IN concentration of 100 g\(^{-1}\), the cloud also dissipates before the end of the simulation because the liquid water is completely depleted.

For the case that has two types of IN (the high-efficiency IN at a concentration of 1 g\(^{-1}\) and the low-efficiency IN at a concentration of 10 g\(^{-1}\)), ice formation can persist throughout the simulation. In the first half of the simulation, the ice formation is dominated by the high-efficiency IN because only the high-efficiency IN can nucleate ice crystals at the relatively high temperature. In the second half of the simulation, the ice formation is dominated by the low-efficiency IN. This is because the high-efficiency IN have been severely depleted as the cloud develops but the longwave cooling has decreased the temperature so that the low-efficiency IN can readily nucleate ice crystals.

A simple model is developed to illustrate the importance of both the variability of the IN efficiency and the longwave cooling in maintaining the ice formation. It is found that as the more efficient IN are continuously depleted, the decreasing temperature can make the less efficient IN start nucleating ice crystals. As a result, the cloud has a continuous supply of IN and the ice formation persists for a long time. This simple model also shows that the cooling rate of the cloud must be appropriate to maintain the ice formation. If the cooling rate is too small, the ice-forming rate will be too low and cannot produce enough ice crystals. If the cooling rate is too large, the depletion of IN will be too fast so that the ice formation cannot persist for a sufficiently long time.

This study indicates that it is important to characterize the variability of the IN efficiency in the atmosphere and represent this variability in the models in order to better understand the continuous ice formation in AMCs.

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REFERENCES


