Aerosol- and Droplet-Dependent Contact Freezing: Parameterization Development and Case Study

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ABSTRACT

A parameterization for contact freezing is presented that combines theoretical expressions for determining the collision efficiency with experimentally determined freezing efficiency results. The parameterization has dependencies on aerosol and cloud droplet physical properties, including electric charges, as well as ambient temperature and humidity. The highest freezing rate is obtained at large aerosol and large cloud droplet sizes, and at cold temperatures and low relative humidities, with typical dust aerosol and droplet properties. The number concentration of ice nucleating particles (INPs) in the contact freezing mode are generally lower than those in the immersion freezing or deposition nucleation mode; however, under certain conditions contact INP concentrations can exceed those of the other modes. The new parameterization is used in a high-resolution, semi-idealized simulation of a deep convective cloud, and a number of sensitivity studies are performed. Results indicate the greatest sensitivity is to the best-fit function to laboratory data. The simulations show that droplet properties and ambient relative humidity contribute significantly to contact freezing.

1. Introduction

Ice particles in mixed-phase clouds have a large impact on cloud lifetime, precipitation amount, and cloud radiative properties (Boucher et al. 2013; Lau and Wu 2003; Lohmann and Feichter 2005; Morrison et al. 2005), so correctly modeling ice formation processes is important for simulations performed on all spatial and temporal scales. Ice forms on aerosol particles, known as ice nucleating particles (INP), through four different processes: immersion freezing, condensation freezing, deposition nucleation, and contact freezing (Cantrell and Heymsfield 2005; Hoose and Möhler 2012; Murray et al. 2012; Vali et al. 2015). Immersion, condensation, and contact freezing require the presence of liquid water and are the primary ice formation mechanisms in mixed-phase clouds. There is only a subtle difference between immersion and condensation freezing, and as such they are often considered together. Deposition nucleation is active only above ice saturation and at colder temperatures compared to the other freezing modes.

The different ice nucleation processes depend primarily on environmental moisture and temperature, but particle properties (size and surface characteristics) and, in the case of contact freezing, droplet size and electrical charges, play a role. Hoose and Möhler (2012) state that the importance of particle size for contact freezing is not clear. In terms of particle type, recent studies suggest it has a significant influence on its ability to nucleate ice, with mineral dust being identified as a major contributor over a broad range of temperatures (Hoose and Möhler 2012; Murray et al. 2012; Atkinson et al. 2013). To account for the differences in nucleation ability of different aerosol species, parameterizations have been developed for immersion freezing and deposition nucleation (Niemand et al. 2012; Tobo et al. 2013; DeMott et al. 2010, 2014; Hiranuma et al. 2014; Steinke et al. 2015). However, a similar aerosol-dependent parameterization including dependencies on all the variables affecting nucleation has not yet been developed for contact freezing.

Developing such a parameterization remains a challenge because of a lack of quantitative experimental data systematically probing each of the dependencies for various aerosol species. Ladino et al. (2013)
provide a review of recent contact freezing studies and highlight the need for more atmospherically relevant laboratory experiments. Parameterizations based on observations lack a sufficient amount of detail, often neglecting a complete description of aerosol and droplet physical properties (Young 1974; Meyers et al. 1992). For example, Phillips et al. (2008) includes dependencies on aerosol chemical properties but assumes the only difference between immersion and contact freezing is a constant shift in the freezing temperature. Diehl and Mitra (2015) include a detailed description of contact freezing, with a dependency on aerosol and droplet size; however, only between two and five aerosol sizes are considered for aerosols between 0.1 and 1 μm, depending on particle type.

Classical nucleation theory has been employed to describe contact freezing (Cooper 1974; Hoose et al. 2010; Wang et al. 2014); however, these parameterizations are not constrained by measurements. Electrical effects have not been included in earlier parameterizations, despite both experiments and theory indicating that they play a role (Wang et al. 1978; Ladino et al. 2011). These parameterizations are still commonly used in weather and climate models, or in some cases contact freezing is completely neglected.

Here we present a new parameterization for contact freezing, which combines a classical theoretical framework for calculating the collision efficiency with recent experimentally determined freezing efficiency measurements. In this way, the new parameterization includes dependencies on aerosol and cloud droplet properties, as well as ambient temperature and humidity, thus providing a new level of completeness. To perform sensitivity studies, the parameterization is applied to a semi-idealized simulation of a convective cloud, and the results are also compared against the commonly used Meyers et al. (1992) parameterization for contact freezing. Finally, the new parameterization is used to quantify the relative importance of the different variables affecting contact freezing.

2. Parameterization development

a. Theoretical determination of collision efficiency

Contact freezing is a two-step process: first, a collision between a supercooled droplet and an aerosol particle must take place, and, second, freezing of the droplet must result from the collision. The collision efficiency, which describes the efficiency with which interstitial aerosol particles collide with supercooled cloud droplets, is governed by many forces. Brownian motion dominates at small particle sizes, whereas inertial impaction and interception dominate at large particle sizes. The phoretic forces, thermophoresis and diffusiophoresis, occur when cloud droplets are either evaporating or growing by condensation. These all combine to determine the total collision efficiency.

The flux of aerosols to a collector drop falling at terminal velocity was determined by Wang et al. (1978). This model includes the effect of Brownian motion and the phoretic forces on the collision efficiency, which is given by

\[ E_{\text{Ph,Br}}(a, r, RH) = \frac{K_{\text{Ph,Br}}}{\pi(a + r)^2|V_r - V_a|}. \] (1)

where

\[ K_{\text{Ph,Br}} = \frac{4\pi B_prC_{\text{Ph}}}{\exp(B_pC_{\text{Ph}}D_{\text{Diff}}a) - 1} \] (2)

is the collision kernel (m3 s−1) due to phoretic forces and Brownian motion;

\[ C_{\text{Ph}} = C_{\text{Th}} + C_{\text{Diff}} \] (3)

is the sum of phoretic forces (kg m3 s−2); and

\[ C_{\text{Th}} = -\frac{12\pi\mu_r r(k_A + 2.5k_p N_{\text{Kn}})k_A(\rho_{\text{air}} - \rho_{\text{v}})\rho_{\text{v}}}{5(1 + 3N_{\text{Kn}})(k_A + 2k_A + 5k_p N_{\text{Kn}})p} \] (4)

and

\[ C_{\text{Diff}} = -\frac{6\pi\mu_r r \times 0.74D_{\text{Diff}} M_{\text{air}} a(\rho_{\text{v,air}} - \rho_{\text{v}})\rho_{\text{v}}}{(1 + aN_{\text{Kn}})M_{\text{w}}\rho_{\text{air}}} \] (5)

are the thermophoretic force (kg m s−2) and diffusiophoretic force (km s−2), respectively.

The governing equations presented above are also described in numerous other references (Martin et al. 1980; Pruppacher et al. 1998; Ladino et al. 2011). In such a model, hydrodynamic effects are treated very simply compared to the trajectory model (Grover et al. 1977), where the actual vapor density, temperature, and velocity fields are evaluated and used to determine the precise trajectory of an aerosol particle around a droplet. However, these simplifications have a negligible effect on the particle capture efficiency (Wang et al. 1978).

This model was extended to include the effects of inertial impaction, which dominates at larger aerosol
sizes. According to Park et al. (2005), the collision efficiency due to inertial impaction is

$$E_{\text{Imp}}(a, r) = \left( \frac{\text{Stk}}{\text{Stk} + 0.35} \right)^2. \quad (6)$$

As a further extension, the effects of electrical charges on droplets and aerosols were parameterized through Wang et al. (2010). The collision efficiency due to electric charges is

$$E_E(a, r) = \frac{2KCq_ar^2}{3\pi\mu_a V_{fr}r^2a}, \quad (7)$$

where $K = 9 \times 10^9 \text{N m}^{-2} \text{C}^{-2}$,

$$q_r = 3.32 \times 10^{-6} \beta r^2, \quad \text{and} \quad (8)$$

$$q_a = 3.32 \times 10^{-6} \beta a^2 \quad (9)$$

are the mean charges (C) on the droplet and aerosol, respectively. The empirical parameter $\beta$ (C m$^{-2}$) is a coefficient that describes the amount of charge on the droplets and aerosols per unit surface area and can vary from between 0 (neutral) and 7 (highly electrified clouds). All the necessary equations for the undefined parameters above are presented in appendix A, and all symbols are defined in appendix B.

Assuming all particle–droplet collisions result in collection of the particle by the droplet, the collection efficiency (CE) is simply equal to the total collision efficiency:

$$CE(a, r, RH) = E_{\text{Ph,Br}}(a, r, RH) + E_{\text{Imp}}(a, r) + E_E(a, r). \quad (10)$$

Figure 1 shows the collection efficiency for varying aerosol radii and ambient RH values. A prominent feature of Fig. 1 is the presence of the “Greenfield gap,” which is the transition region between the Brownian-motion-dominated small-aerosol size range and the inertial-impaction-dominated region at larger aerosol sizes. This region is where the collection efficiencies are lowest, and it is where the phoretic forces become important. The Greenfield gap has been experimentally confirmed, recently by Ladino et al. (2011) under laboratory conditions.

Nagare et al. (2015) show that the expressions for thermophoresis provided by Wang et al. (1978) result in a collision efficiency that is almost independent of particle size, a different response compared to expressions provided by Andronache et al. (2006). Therefore, the collisions efficiencies shown in Fig. 1 could be an underestimation for submicron particles. Furthermore, Nagare et al. (2015) show a high sensitivity of the Wang et al. (1978) formulation of thermophoresis to temperature and note that this was formulated for warm rain processes, and its application to subzero temperatures may be problematic.

Other interesting features of Fig. 1 are the inverse relationships between the collection efficiency and the droplet radius and also the ambient RH. From Fig. 1, the increasing collection efficiency with decreasing droplet size is attributed to the increased contribution of Brownian motion to particle capture. Considering the trend with RH, the effect of diffusiophoresis is to create a force such that the aerosol particles move in the same direction as the vapor flux. For low-ambient-RH conditions, which corresponds to an evaporating droplet, this means a force away from the droplet and hence a decrease in the collection efficiency. However, thermophoresis is simultaneously present as a result of differences in droplet and air temperature.
which create a force in the same direction as the temperature gradient and the opposite direction as diffusiophoresis. There is conflicting evidence as to whether thermophoresis or diffusiophoresis dominates (Santachiara et al. 2012); however, the above theoretical treatment implies enhanced collision efficiency in low-ambient-RH conditions. In the atmosphere, this would correspond to regions of dry-air entrainment into cloudy air. Note that the ambient RH has the strongest influence on the collection efficiency in the Greenfield gap; that is, aerosol particles between about $10^{-2}$ and 1 $\mu m$.

Figure 1 also indicates that electrically charged droplets and aerosols act to increase the collection efficiency, particularly within the Greenfield gap. The influence of the relative humidity on the collection efficiency is reduced when charged droplets and aerosols are present. An electric charge obtained with $\beta = \pm 7$ corresponds to a highly electrified cloud associated with thunderstorms (Wang et al. 2010); thus, this represents a realistic upper bound. Yair et al. (2010) provide a parameterization for the lightning potential index (LPI), based on the vertical velocity, and cloud liquid and ice water content. Although this cannot be directly incorporated into the parameterization presented here, it could be used to inform the choice of $\beta$ in Eqs. (8) and (9). This could be done by assuming a linear scaling between LPI and $\beta$ and, hence, provide a more realistic time-varying estimate of the collection efficiency as a result of electrical charges.

The collection efficiency for interactions between small droplets and small aerosols is dominated by Brownian motion, and this can give efficiencies greater than 1, as seen in Fig. 1 for $r < 10 \mu m$ and $a < 10^{-2} \mu m$. It is therefore important to limit the maximum value of the collection efficiency to 1. Phoretic effects still influence the collection efficiency for droplet radii down to 6 $\mu m$, and below this droplet size the collection efficiency will always be unity for all aerosol sizes.

Diehl et al. (2006) show that interactions between small droplets and small aerosols contribute nearly 20 orders of magnitude less to the collection kernel, when compared to interactions between large droplets and large aerosols. This means the freezing rate for small droplet–small aerosol interactions would be vastly smaller than for large droplet–large aerosol interactions. The number concentration of smaller droplets is typically larger than the number concentration of large droplets, which would act to increase the freezing rate in this regime, but this is still insufficient to compensate for the difference in the collection kernel. Therefore, the impact of these interactions between small droplets and small aerosols on the contact freezing rate is small. This will be demonstrated in section 2c.

b. Laboratory results of freezing efficiency

Once an aerosol particle and cloud droplet collide, there is a certain probability that freezing of the droplet occurs. This second step in the process of contact freezing is quantified through the freezing efficiency, which depends on the ambient temperature, humidity, and the aerosol and droplet sizes. Recent experiments have attempted to quantify this in a systematic way, as summarized by Ladino et al. (2013).

Figure 2 shows a selection of results obtained from different laboratory instruments (Pitter and Pruppacher 1973; Svensson et al. 2009; Ladino et al. 2011; Hoffmann et al. 2013a,b; Niehaus et al. 2014; W. Cantrell 2015, personal communication), for dust particles with different mineralogies. Taken together, these data cover the complete temperature range over which contact freezing is thought to occur. There is considerable spread in the data, particularly at warmer temperatures. Nevertheless, the particle surface-area-normalized freezing efficiency (m$^{-2}$) can be described by the temperature-dependent function:

$$nFE(T_{air}) = A \exp[B(T_{air} - 273.15)],$$

(11)

where $A = 1.223 \times 10^7$, and $B = -2.686 \times 10^{-1}$, and were determined by means of a nonlinear least squares method, where the data are first approximated by a model, and the model parameters are refined through successive iterations that minimize the errors between the data and the model. The reported freezing efficiency was normalized by the particle surface area assuming active sites at the particle surface are responsible for initiating contact freezing, similar to the ice nucleation active surface site (INAS) density approach for immersion freezing (Hoose and Möhler 2012). This approach follows Hirahama et al. (2015).

Pitter and Pruppacher (1973) did not report the freezing efficiencies of their wind tunnel experiments; however, by assuming a certain number of collisions, as done by Ladino et al. (2011), the freezing efficiency can be estimated. These estimates were not used to define this function. At these warmer temperatures, the best-fit line is quite insensitive to the data used to define the temperature dependent function. Removing the Svensson et al. (2009) data results in an almost negligible change in the best-fit line, since the magnitudes of the errors between the data and the model are smaller than those at colder temperatures. A manually derived alternative fit is proposed in order to test the sensitivity of the parameterization to the
freezing efficiency data. This alternative fit is shown as the dashed line in Fig. 2 and will be employed in the sensitivity studies of section 3b. To prevent unphysical high values, the freezing efficiency must have an upper limit of unity: that is, $n_{FE} \leq 1$.

The freezing efficiency may have an additional dependency on droplet size, which is not considered in Eq. (11). Looking at the Ladino et al. (2011) data in Fig. 2, the effect of increasing the droplet size from 6.5 to 13 $\mu$m is to increase the freezing efficiency significantly. Since there are only two sizes considered in these experiments, extracting a relationship between the freezing efficiency and droplet size is prone to a significant amount of uncertainty. Therefore, it is of little use to attempt to parameterize this effect in the freezing efficiency until more data are available.

c. The contact freezing rate

The final step in defining the contact freezing rate is to combine the theoretically determined collection efficiency with the experimentally determined freezing efficiency results. The collection kernel ($m^2 s^{-1}$) first needs to be calculated from the collection efficiency, as shown below:

$$K_{coll} = \pi (r + a)^2 CE |V_r - V_a|,$$  \hspace{1cm} (12)

where $CE$ is given by Eq. (10), and $V_r$ and $V_a$ are given by Eqs. (A16) and (A17), respectively. Finally, the contact freezing rate ($m^{-3} s^{-1}$) is given by

$$\frac{dN_{inp}}{dt} = \int_{a_1}^{a_2} \int_{r_1}^{r_2} K_{coll} N_r \max(1, n_{FE} A_a) N_a \, dr \, da,$$  \hspace{1cm} (13)

where $N_r$ is the number concentration of droplets ($m^{-3}$), $n_{FE}$ is given by Eq. (11) ($m^{-2}$), $A_a$ is the surface area of aerosol particles with radius $a$ ($m^2$), and $N_a$ is the number concentration of aerosol particles ($m^{-3}$). The integration limits are given by the range of droplet and aerosol sizes over which the parameterization is valid: that is, $r = 1–535 \mu$m and $a = 10^{-3}–10 \mu$m. The dust size distribution must be defined by the user. The droplet size distribution can be either calculated from model-diagnosed quantities, or, in the case where a bin microphysics scheme is used, the same bins can be employed in the integration over droplet sizes. Also, the total number concentration of INPs must not exceed whichever is smaller: the number concentration of aerosols or the number concentration of droplets.

Figure 3 shows 2D histograms of the contact freezing rate as a function of aerosol and droplet radii (top) and ambient temperature and RH (bottom). The highest
freezing rates are obtained through this parameterization when large aerosol particles (≥0.3 μm) interact with large cloud droplets (≥30 μm). Indeed, at the very largest sizes, the frozen fraction is one. The reason for the high freezing rates at large aerosol and droplet sizes is twofold. First, at large aerosol and droplet sizes, the freezing efficiency approaches 1 (Fig. 1). Second, the collection kernel of Eq. (12) depends on the square of aerosol plus droplet radius.

The interactions between small aerosols (≤0.01 μm) and small droplets (≤10 μm) make a negligible contribution to the collection kernel, as also shown in Diehl et al. (2006). There is a small area of anomalously low freezing rates when aerosols and droplets of approximately the same size interact. This occurs when the terminal velocity of the droplets is approximately equal to the terminal velocity of the aerosol particles. It is offset to larger droplet sizes as a result of the difference in density between droplets and aerosols and results in a lower collision efficiency.

The bottom panel shows high freezing rates at cold temperatures and low-ambient-RH conditions, as a result of the increase in freezing efficiency with decreasing temperature and the increase in the collection efficiency with decreasing RH. At high values of RH and smaller droplet sizes, which are more relevant for in-cloud conditions, the phoretic forces have the strongest influence. At 255 K, a decrease in the RH of about 10% can increase the freezing rate by about an order of magnitude for 30-μm droplets. For large droplets, the contact freezing rate is mainly determined by temperature, meaning that large freezing rates can also occur in high-RH conditions. Figure 3 also indicates that, under the prescribed conditions, aerosol and droplet size has a larger influence on the freezing rate than ambient temperature and RH. The effects of electric charges were investigated in a similar manner, and found to be largely independent of ambient temperature and relative humidity.

To understand how the number concentration of ice nucleation events determined with the above parameterization compare to the other freezing modes and other contact parameterizations, Fig. 4 shows a comparison with the immersion freezing parameterization from Niemand et al. (2012) and deposition nucleation parameterization from Steinke et al. (2015).
Meyers et al. (1992) parameterization for contact freezing is also shown. The shaded areas show possible values based on the range of RH_{ice} values for deposition nucleation, and RH_{water}, r, N_r, and b for contact freezing. Here, a time step of 1 s is chosen in order to calculate contact number concentrations from the freezing rate given by Eq. (13). This is a typical microphysics time step in large-eddy simulations; however, numerical weather prediction models use a time step at least an order of magnitude larger, meaning an order-of-magnitude more contact INP concentrations than those shown in Fig. 4.

For the most part, number concentrations of INPs in the contact mode are several orders of magnitude below those in the immersion or deposition mode. However, in conditions that correspond to low ambient RH, large droplet and aerosol sizes, and large number concentrations, INP concentrations produced in the contact mode can become greater than those produced in the other freezing modes, since the maximum values in the contact mode are greater than those in the other modes. When the prescribed aerosol radius is increased, contact freezing can become significantly larger than the other modes. There is also a very large spread in possible values of contact freezing INPs, covering up to 15 orders of magnitude. This highlights the significant influence that droplet properties, ambient RH, and electrical effects have on determining the number concentration of INPs in the contact mode. These number concentrations are mostly lower than those suggested by the Meyers et al. (1992) parameterization, except when the aerosol size becomes large.

3. Application in a simulation of a deep convective cloud

a. Model description

The nonhydrostatic regional-weather-forecasting Consortium for Small-Scale Modeling (COSMO) Model, version 5.01 (Schättler et al. 2008), was run at high resolutions capable of resolving energy-containing turbulence (Barthlott and Hoose 2015). To evaluate the above parameterization and perform sensitivity experiments, COSMO simulations were performed with high grid resolution of 100 m for a semi-idealized case of a convective cloud. In conjunction with the contact freezing parameterization presented above, the Niemand et al. (2012) parameterization for immersion freezing on desert dust particles, and the Steinke et al. (2015) parameterization

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<td>Wet Dust</td>
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for deposition ice nucleation on Arizona test dust were used. The two-moment cloud microphysics scheme of Seifert and Beheng (2006) was also used, which uses the supersaturation to define a power law, in order to calculate CCN concentrations that are representative of continental conditions. The parameterization is only applied to cloud droplets, and contact freezing of rain droplets is not considered. Since rain drops collect many particles through collision–coalescence, they are efficient in the immersion mode (Paukert et al. 2017).

A vertically constant, two-mode lognormal dust aerosol size distribution was used, covering particle sizes from 0.1 to 100 μm, which is based on observations from Jungfraujoch research station (M. Niemand 2015, personal communication) (mode 1: $N = 0.015 \times 10^6 \text{m}^{-3}$, $\mu = 1.355 \times 10^{-6} \text{m}$, $\sigma = 1.443$; mode 2: $N = 0.00001 \times 10^6 \text{m}^{-3}$, $\mu = 8.518 \times 10^{-6} \text{m}$, $\sigma = 1.358$). The dust aerosols are not removed by precipitation in the model. Immersion freezing acts only on the immersed dust aerosols, and contact and deposition ice nucleation act on the interstitial dust. The droplet size distribution was calculated from the model-diagnosed cloud liquid water content and droplet number concentration in every grid box, assuming a modified gamma distribution, with parameters defined in Seifert and Beheng (2006) for droplets in the size range 1–535 μm.

The aerosol and droplet distributions were divided into 10 bins, over which the integration for the parameterizations was performed. The dust size distribution and the temporal and spatial mean of cloudy grid points for the droplet size distribution from one of the simulations, the Wet Dust run (Table 1), are shown in Fig. 5.

The simulations were initialized from a sounding, with CAPE = 2801 J kg$^{-1}$ indicated by the shaded region, and realistic topography was specified at each grid point, as shown in Fig. 6. The topography was chosen to represent the region near Jülich, in western Germany, with the mountains in the southwest of the domain. In this region, the measurement campaign High Definition Clouds and Precipitation for Advancing Climate Prediction (HD(CP)$^2$) Observational Prototype Experiment (HOPE; http://hdcp2.zmaw.de) was conducted in spring 2013, for which realistic COSMO runs were performed by Barthlott and Hoose (2015) with up to 250-m horizontal grid spacing. Random temperature and wind...
fluctuations with amplitude 0.02 K and 0.02 m s\(^{-1}\), respectively, were added in the lowest 100 hPa of the atmosphere. A horizontal grid spacing of 100 m was used, with 100 vertical levels, and 600 \times 600 grid cells horizontally. A time step of 2 s was used for the duration of the 9-h simulation.

A number of sensitivity experiments were performed to quantify the effect of various parameters on the contact INP number concentrations. These are detailed in Table 1. The parameters of interest in the sensitivity study are the ratio of immersed to interstitial dust aerosol \(\varepsilon\), the temperature-dependent freezing efficiency fit, where “high” denotes the solid line and “low” denotes the dashed line in Fig. 2 and the effect of charges on aerosol particles and cloud droplets. An additional simulation is performed with the Meyers et al. (1992) contact freezing parameterization. The Meyers run represents the contact freezing parameterization from these authors, used along with the Niemand et al. (2012) and Steinke et al. (2015) parameterizations. Note that the Meyers simulation is independent of aerosol properties, so stating \(\varepsilon = 90\) only implies that there is 90% of the aerosol available for immersion freezing. Given the low hygroscopicity of uncoated dust (Ghan et al. 2001), the Dry Dust run would be more representative of atmospheric conditions with fresh dust particles that have not undergone aging. The Charge run represents an expected maximum in contact INP concentrations, whereas the Low nFE simulation represents the expected minimum.

b. Sensitivity studies

The evolution of the deep convective cloud is shown in Fig. 7. Here, the spatial mean vertically integrated time series of the cloud liquid content (QC), ice water content (QI), cloud droplet number concentration (CDNC), and ice number concentration (INC) from the Wet Dust run are shown. QC and CDNC show a peak at 4 h, and QI and INC increase to a maximum shortly after, between 4 and 5 h into the simulation, after which the cloud decays. In the last hour of the simulation, the cloud starts to deepen once again.

Figure 8 shows the mean horizontal cross section of the contact INP number concentrations from the five runs using the contact freezing parameterization presented here, at 4 h into the simulation, when the number concentrations are among the highest. These plots, as well as the subsequent domain-mean cross sections, are averaged over latitudes. By multiplying the contact freezing rate, given by Eq. (13), by the model time step (here 2 s), the number concentration of contact INP can be calculated.

The largest sensitivity is shown in the Low nFE simulation, where INP concentrations are three to four orders of magnitude lower than the other simulations. This implies that defining an appropriate best-fit function to the laboratory freezing efficiency data is critical. Another interesting difference is that the maximum INP concentrations in Low nFE are located at altitudes around 8 K colder than Dry Dust, as a result of the horizontal difference between the two fit functions in Fig. 2. The Wet Dust simulation shows lower maximum concentrations compared to the Dry Dust and Charge simulations; however, the differences between these three simulations are relatively small. The Meyers simulation contains contact INP number concentrations, which are significantly lower than the Wet Dust simulation. This should be expected since the input dust size
is mostly greater than 1 μm, meaning there is an abundance of relatively larger particles that favor INP production via the parameterization presented here. Note that the Meyers et al. (1992) parameterization has no cold temperature limit; therefore, large INP concentrations are produced at very low temperatures.

The ice crystal number concentrations are shown in Fig. 9. Here, the number concentrations are lower than that of the total INP concentrations as a result of conversion of ice into other hydrometeors. The number concentration of INPs acts as the starting value for the ice crystal concentrations. As the simulation progresses, losses due to collision, riming, and evaporation, as well as gains due to freezing of rain and cloud droplets have the net effect of reducing ice crystal concentrations while producing snow, graupel, and hail. Ladino et al. (2017) present observations suggesting the opposite can occur: that is, that ice crystal concentrations are several orders of magnitude larger than INP number concentrations. This points to a missing ice multiplication process in the model. The largest sensitivities are in the simulations that have lower number concentrations of contact INP. There is a clear discontinuity in the number of ice crystals at 261 K for the Low nFE and Meyers simulations, where the immersion parameterization becomes active, and the concentrations at temperatures warmer than this are at least three orders of magnitude smaller.

c. **Dominant contributors to the contact freezing rate**

A unique feature of the new contact freezing parameterization is the dependence on many new variables which affect the freezing rate, specifically the aerosol and droplet size and number concentrations, the relative humidity, and electrical charges. An analysis of the contribution of the droplet properties and the relative humidity to the contact INP concentrations is presented here. During the simulations, the dust aerosol properties...
and the electrical charges were constant and are therefore not investigated here. Figures 10 and 11 show 2D histograms of the droplet properties, relative humidity, and contact freezing INP concentrations, with the colors indicating the density (bin count/sample count/bin area). The droplet sizes were calculated from the cloud water content and cloud droplet number concentration in 10 logarithmic-spaced bins between $10^{-6}$ and $10^{-4}$ m. These histograms represent in-cloud conditions, defined as cloud water plus cloud ice mixing ratio greater than zero.

Figure 10 indicates that droplets are present in the cloud with concentrations fairly uniformly distributed from about $10^4$ to $10^8$ m$^{-3}$, at temperatures warmer than 255 K. There is a preference for smaller droplets from between 1 and 10 $\mu$m, so much so that the largest droplet bins greater than about 20 $\mu$m are unoccupied. As expected, there are high values of relative humidity at warmer temperatures, and at temperatures relevant for contact freezing the in-cloud relative humidity can range from 20% to 100%. The sounding in Fig. 6 is quite dry in the midtroposphere. As such, there is a high occurrence of relative humidity values of 40%, which would correspond to cloud edge where one could see an enhancement in contact freezing in the lower-relative-humidity environment. The bottom-right panel indicates contact INPs are mostly present at low concentrations within the temperature range at which they are produced. However, contact INPs can sediment out or be transported in the updraft to colder temperatures.

Figure 11 shows that contact INPs are mostly present at lower concentrations. Higher contact INP number concentrations occur preferentially at larger droplet radii and over a larger range of droplet concentrations. The contact freezing events involving large droplets are very rare. Perhaps most interestingly, the relative humidity histogram indicates that contact INPs are present at lower relative humidities of about 40% almost as
often as at higher relative humidities over 90%. This does not necessarily mean INPs are produced in the lower-relative-humidity areas almost as often as in the higher-relative-humidity areas, only that they are present here. Advection acts on these particles, possibly transporting them into an area of different environmental conditions. This does indicate that droplet properties and the ambient humidity play a significant role in contact freezing, and these variables should be included in parameterizations aiming for a sophisticated representation of contact freezing.

4. Summary and conclusions

By combining theoretical expressions for determining the collection efficiency with experimentally determined freezing efficiency results, a parameterization for ice nucleation in the contact mode has been developed. The parameterization includes dependencies on more variables than other parameterizations, most notably the surface area of aerosols and cloud droplets, making it particularly useful for detailed process and sensitivity studies. A decrease in the ambient RH of 50% results in an increase in the collision efficiency of about an order of magnitude. Highly charged droplets and aerosol have a similar effect on the collision efficiency, and the effect of RH is significantly diminished in the presence of charged droplets and aerosols. The size of droplets and aerosols has the largest influence on the collision efficiency. A dependence on temperature enters the parameterization through a best-fit function to laboratory observations of the freezing efficiency from numerous experiments.

Putting all of this together, the final parameterization produces the highest freezing rates when large aerosols interact with large droplets. Here, the freezing rate is more than 14 orders of magnitude larger than interactions between small aerosols and small droplets. Higher freezing rates are produced at lower-RH conditions, implying that, in areas of dry-air entrainment, the contact freezing rate would be enhanced. A comparison of this new parameterization to other freezing modes, as well as other contact parameterizations, suggests the number concentration of INPs produced from the parameterization presented here should be mostly lower than in other modes and parameterizations. However, under certain conditions, which correspond to large droplet and aerosol interactions in a low-RH environment with large number concentrations, the number of INPs could exceed those of the other freezing modes.

![Two-dimensional histograms showing density (bin count/sample count/bin area) for a range of values of temperature and droplet number concentrations, droplet radii, relative humidity, and contact INP number concentrations.](image)
Semi-idealized, very-high-resolution simulations of a convective cloud were used to perform sensitivity studies and to evaluate the parameterization against another commonly used parameterization. The results indicate the largest sensitivity is to the best-fit function to laboratory data of the freezing efficiency. This underscores the importance of laboratory data to constrain ice nucleation parameterizations. Increasing the amount of interstitial aerosols resulted in a change in the contact INP number concentrations of a factor of 2. Using a realistic upper limit for the effect of charged droplets and aerosols resulted in only a slight increase in the contact INP concentrations.

The case study shows that in-cloud conditions are dominated by smaller droplets over a wide range of concentrations, and the relative humidity is often as low as 40%, presumably near the cloud edge. Contact freezing INPs are present most often in areas of high droplet concentrations and small droplet radii. The temperature and relative humidity do not have a large influence on where contact INPs are present in the cloud. This implies that contact freezing can be enhanced in low-relative-humidity environments near cloud edges, in contrast to the other freezing modes, which are not influenced by the relative humidity and are primarily dependent on temperature.

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**APPENDIX A**

**Theoretical Expressions**

The particle mobility (s kg$^{-1}$) is given by Pruppacher et al. (1998) [Eq. (11.20)] as follows:

$$B_p = \frac{1 + \alpha N_{Kn}}{6\pi\mu_d a},$$  \hspace{1cm} (A1)

where

$$\alpha = 1.257 + 0.4 \exp(-1.1/N_{Kn})$$  \hspace{1cm} (A2)

and

$$N_{Kn} = \frac{\lambda_d}{a}$$  \hspace{1cm} (A3)
is the Knudsen number (unitless):
\[
\mu = \frac{1.496 \times 10^{-6} T_{air}^{1.5}}{T_{air} + 120} \quad \text{(A4)}
\]

is the dynamic viscosity of air (kg m^{-1} s^{-1}) (Ladino et al. 2011). The mean free path (m) is given by Ladino et al. (2011) as follows:
\[
\lambda = \frac{21.55 \mu T_{air}^{0.5}}{p} \quad \text{(A5)}
\]

The particle diffusivity (m^2 s^{-1}) was taken from Pruppacher et al. (1998) [Eq. (11.21)]:
\[
D_{\text{Diff}} = B_p k_b T_{air} \quad \text{(A6)}
\]

The mean ventilation coefficients for aerosol particles (unitless), and for mass (unitless) were taken from Ladino et al. (2011):
\[
f_p = \begin{cases} 
1 + 0.108(\text{Sc}^{1/3} \text{Re}^{1/2}) & \text{if } r \leq 60 \mu m \\
0.78 + 0.308(\text{Sc}^{1/3} \text{Re}^{1/2}) & \text{if } 60 < r \leq 1500 \mu m 
\end{cases} \quad \text{(A8)}
\]

and
\[
f_v = \begin{cases} 
1 + 0.108(\text{Sc}^{1/3} \text{Re}^{1/2}) & \text{if } r \leq 60 \mu m \\
0.78 + 0.308(\text{Sc}^{1/3} \text{Re}^{1/2}) & \text{if } 60 < r \leq 1500 \mu m 
\end{cases} \quad \text{(A9)}
\]

It is standard to assume that \(f_h = f_v\), where \(f_h\) is the mean ventilation coefficient for heat transfer. Also
\[
V_r = \begin{cases} 
\mu \text{Re} \\
2\rho_{\text{air}} \lambda
\end{cases} \quad \text{if } r < 10 \mu m
\]

\[
\mu \text{Re} \quad \text{if } 10 \leq r \leq 535 \mu m \quad \text{(A16)}
\]

The terminal velocity (m s^{-1}) of aerosol particles is determined by applying Eq. (11.22) from Pruppacher et al. (1998):
\[
V_a = \frac{2(1 + aN_{Kn}a^2g)(\rho_p - \rho_{\text{air}})}{9\mu_a} \quad \text{(A17)}
\]

are the Schmidt numbers for collected particles (unitless) and for water vapor in air (unitless), respectively. To determine the terminal velocity of droplets in air, the Reynolds number must be computed:
\[
\text{Re} = \exp(Y) \quad \text{(A12)}
\]

with
\[
Y = -0.318657 \times 10^4 + 0.992696X - 0.153193 \times 10^{-2}X^2 \\
- 0.987059 \times 10^{-3}X^3 - 0.578878 \times 10^{-3}X^4 \\
+ 0.855176 \times 10^{-4}X^5 - 0.327815 \times 10^{-5}X^6 \quad \text{(A13)}
\]

and with the unitless number
\[
X = \ln(C_d \text{Re}^2), \quad \text{(A14)}
\]

where ln is the natural logarithm, and \(C_d\) is the drag coefficient, and, according to Eq. (10.142) of Pruppacher et al. (1998),
\[
C_d \text{Re}^2 = \frac{32r^3(\rho_p - \rho_{\text{air}}) \rho_{\text{air}} g}{3\mu_a^2}, \quad \text{(A15)}
\]

which is known as the Davies or Best number (unitless). Now the terminal velocity (m s^{-1}) of droplets can be calculated through Eqs. (10.139) and (10.146) of Pruppacher et al. (1998):
\[
V_r = \frac{\mu_{\text{air}}}{4\pi r^3 \rho_a kl} \frac{dm}{dr} + T_{\text{air}} \quad \text{(A18)}
\]
where

\[
\frac{dm}{dt} = \frac{4\pi r M_a D_w \chi(T_{\text{air}})}{1 + D_w \chi(T_{\text{air}})} \left( \frac{RH}{100} - 1 \right)
\]

is the mass growth rate of the droplet, and

\[
\chi(T_{\text{air}}) = \frac{p_{\text{sat}}(T_{\text{air}})}{R_v T_{\text{air}}}.
\]

Here we have assumed the saturation ratio is unity, which is a valid approximation for the size of droplets.

### Table B1. List of symbols.

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Name</th>
<th>Value/units</th>
</tr>
</thead>
<tbody>
<tr>
<td>(a)</td>
<td>Aerosol radius</td>
<td>m</td>
</tr>
<tr>
<td>(A_a)</td>
<td>Aerosol particle surface area</td>
<td>m²</td>
</tr>
<tr>
<td>(B_p)</td>
<td>Particle mobility</td>
<td>s kg⁻¹</td>
</tr>
<tr>
<td>(C)</td>
<td>Cunningham slip correction factor</td>
<td>—</td>
</tr>
<tr>
<td>(C_{\text{DF}})</td>
<td>Diffusiophoretic force</td>
<td>kg m⁻² s⁻²</td>
</tr>
<tr>
<td>(C_{\text{CE}})</td>
<td>Collection efficiency</td>
<td>—</td>
</tr>
<tr>
<td>(C_{\text{Ph}})</td>
<td>Phoretic force</td>
<td>kg m⁻² s⁻²</td>
</tr>
<tr>
<td>(C_{\text{Th}})</td>
<td>Thermophoretic force</td>
<td>kg m⁻² s⁻²</td>
</tr>
<tr>
<td>(D_{\text{Diff}})</td>
<td>Particle diffusivity</td>
<td>m² s⁻¹</td>
</tr>
<tr>
<td>(D_v)</td>
<td>Water vapor diffusivity</td>
<td>(0.225 \times 10^{-4} \text{ m}^2 \text{ s}^{-1})</td>
</tr>
<tr>
<td>(E_{\text{E}})</td>
<td>Collision efficiency due to electric charges</td>
<td>—</td>
</tr>
<tr>
<td>(E_{\text{Imp}})</td>
<td>Collision efficiency due to inertial impaction</td>
<td>—</td>
</tr>
<tr>
<td>(E_{\text{Ph,Br}})</td>
<td>Collision efficiency due to phoretic forces and Brownian motion</td>
<td>—</td>
</tr>
<tr>
<td>(f_p/f_s/f_h)</td>
<td>Mean ventilation coefficient for particle flux, mass, and heat transfer</td>
<td>—</td>
</tr>
<tr>
<td>(g)</td>
<td>Gravitational acceleration</td>
<td>(9.807 \text{ m} \text{ s}^{-2})</td>
</tr>
<tr>
<td>(k_a)</td>
<td>Thermal conductivity of moist air</td>
<td>J m⁻¹ s⁻¹ °C⁻¹</td>
</tr>
<tr>
<td>(k_b)</td>
<td>Boltzmann constant</td>
<td>(1.381 \times 10^{-23} \text{ kg m}^2 \text{ s}^{-2} \text{ K}^{-1})</td>
</tr>
<tr>
<td>(K_{\text{coll}})</td>
<td>Collection kernel</td>
<td>m⁻¹</td>
</tr>
<tr>
<td>(k_{\text{Ph,Br}})</td>
<td>Collision kernel due to phoretic forces and Brownian motion</td>
<td>m⁻¹</td>
</tr>
<tr>
<td>(l_v)</td>
<td>Latent heat of vaporization</td>
<td>(2274 \times 10^3 \text{ J kg}^{-1})</td>
</tr>
<tr>
<td>(M_{\text{air}})</td>
<td>Molecular weight of air</td>
<td>(28.96 \times 10^{-3} \text{ kg mol}^{-1})</td>
</tr>
<tr>
<td>(M_w)</td>
<td>Molecular weight of water</td>
<td>(18.02 \times 10^{-3} \text{ kg mol}^{-1})</td>
</tr>
<tr>
<td>(N_a)</td>
<td>Number concentration of aerosol particles</td>
<td>m⁻³</td>
</tr>
<tr>
<td>(nFE)</td>
<td>Aerosol particle surface area normalized freezing efficiency</td>
<td>m²⁻²</td>
</tr>
<tr>
<td>(N_{Kn})</td>
<td>Knudsen number</td>
<td>—</td>
</tr>
<tr>
<td>(N_r)</td>
<td>Number concentration of droplets</td>
<td>m⁻³</td>
</tr>
<tr>
<td>(p)</td>
<td>Ambient air pressure</td>
<td>Pa</td>
</tr>
<tr>
<td>(p_0)</td>
<td>Actual vapor pressure</td>
<td>Pa</td>
</tr>
<tr>
<td>(p_{\text{sat}})</td>
<td>Saturated vapor pressure</td>
<td>Pa</td>
</tr>
<tr>
<td>(q_e)</td>
<td>Aerosol charge</td>
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</tr>
<tr>
<td>(q_r)</td>
<td>Droplet charge</td>
<td>C</td>
</tr>
<tr>
<td>(r)</td>
<td>Droplet radius</td>
<td>m</td>
</tr>
<tr>
<td>(Re)</td>
<td>Reynolds number</td>
<td>—</td>
</tr>
<tr>
<td>(RH)</td>
<td>Relative humidity</td>
<td>—</td>
</tr>
<tr>
<td>(R_g)</td>
<td>Gas constant for water vapor</td>
<td>(461.5 \text{ J kg}^{-1} \text{ K}^{-1})</td>
</tr>
<tr>
<td>(S_c, S_{cw})</td>
<td>Schmidt number for collected particles and water vapor in air</td>
<td>—</td>
</tr>
<tr>
<td>(Stk)</td>
<td>Stokes number</td>
<td>—</td>
</tr>
<tr>
<td>(T_{\text{air}})</td>
<td>Ambient air temperature</td>
<td>K</td>
</tr>
<tr>
<td>(T_r)</td>
<td>Droplet temperature</td>
<td>K</td>
</tr>
<tr>
<td>(V_a)</td>
<td>Terminal velocity of aerosol</td>
<td>m s⁻¹</td>
</tr>
<tr>
<td>(V_r)</td>
<td>Terminal velocity of droplet</td>
<td>m s⁻¹</td>
</tr>
<tr>
<td>(\beta)</td>
<td>Coefficient of droplet and aerosol electric charge per unit surface area</td>
<td>C m⁻²</td>
</tr>
<tr>
<td>(\gamma)</td>
<td>Surface tension of water</td>
<td>(0.0727 \text{ N m}^{-1})</td>
</tr>
<tr>
<td>(\lambda_a)</td>
<td>Mean free path</td>
<td>m</td>
</tr>
<tr>
<td>(\mu_a)</td>
<td>Dynamic viscosity of air</td>
<td>kg m⁻¹ s⁻¹</td>
</tr>
<tr>
<td>(\rho_{\text{air}})</td>
<td>Air density</td>
<td>(1.1 \text{ kg m}^{-3})</td>
</tr>
<tr>
<td>(\rho_p)</td>
<td>Aerosol particle density</td>
<td>(1400 \text{ kg m}^{-3})</td>
</tr>
<tr>
<td>(\rho_w)</td>
<td>Water vapor density</td>
<td>(1000 \text{ kg m}^{-3})</td>
</tr>
<tr>
<td>(\rho_w)</td>
<td>Water density</td>
<td>—</td>
</tr>
</tbody>
</table>
considered in this parameterization. Using the droplet and air temperatures, Murphy and Koop (2005) can be used to calculate the saturated vapor pressure of the droplet \( p_{\text{sat}}(T_d) \) and air \( p_{\text{sat}}(T_{\text{air}}) \). Then, using the relative humidity, the actual vapor pressure of the droplet and air can be calculated as follows:

\[
\begin{align*}
  p_0(T_d) &= \frac{\text{RH}}{100} p_{\text{sat}}(T_d) \quad \text{and} \quad (A21) \\
  p_0(T_{\text{air}}) &= \frac{\text{RH}}{100} p_{\text{sat}}(T_{\text{air}}). \quad (A22)
\end{align*}
\]

Following this, the water vapor density at the droplet surface and the water vapor density of air can be computed (Ladino et al. 2011):

\[
\begin{align*}
  \rho_{v,f} &= \frac{p_0(T)}{T R_v} \quad \text{and} \quad (A23) \\
  \rho_{v,\text{air}} &= \frac{p_0(T_{\text{air}})}{T_{\text{air}} R_v}. \quad (A24)
\end{align*}
\]

The dimensionless Stokes number describes the inertial impact properties of a particle:

\[
\text{Stk} = \frac{\rho_p a^2 V_r}{9 \mu_a R}. \quad (A25)
\]

The Cunningham slip correction factor can be computed by

\[
C_e = 1 + \frac{a}{\lambda} \left[ 1.257 + 0.4 \exp\left(-1.1 \frac{a}{\lambda_p}\right) \right]. \quad (A26)
\]

APPENDIX B

List of Symbols

Table B1 shows the symbols used in this study, along with their definitions and units.

REFERENCES


