A new method for operating a continuous-flow diffusion chamber to investigate immersion freezing: assessment and performance study

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Abstract. Glaciation in mixed-phase clouds predominantly occurs through the immersion-freezing mode where ice-nucleating particles (INPs) immersed within supercooled droplets induce the nucleation of ice. Model representations of this process currently are a large source of uncertainty in simulating cloud radiative properties, so to constrain these estimates, continuous-flow diffusion chamber (CFDC)-style INP devices are commonly used to assess the immersion-freezing efficiencies of INPs. This study explored a new approach to operating such an ice chamber that provides maximum activation of particles without droplet breakthrough and correction factor ambiguity to obtain high-quality INP measurements in a manner that previously had not been demonstrated to be possible. The conditioning section of the chamber was maintained at −20°C and water relative humidity (RHw) conditions of 113% to maximize the droplet activation, and the droplets were supercooled with an independently temperature-controlled nucleation section at a steady cooling rate (0.5°C min⁻¹) to induce the freezing of droplets and evaporation of unfrozen droplets. The measured ice nucleation efficiencies of model aerosols that had a surface active site density (nₛ) metric were higher but mostly agreed within 1 order of magnitude compared to results reported in the literature.

1 Introduction

Atmospheric ice nucleation plays an important role in initiating precipitation in clouds that consist of a mixture of supercooled liquid water droplets and ice crystals and in catalyzing the formation of ice particles within high-altitude cirrus clouds (Lohmann and Feichter, 2005; Boucher et al., 2013). This important step toward ice formation also affects the lifetime and radiative properties of these clouds; however, ice nucleation mechanisms are poorly understood and parameterized in cloud models (e.g., Hoose and Möhler, 2012; Murray et al., 2012; Kulkarni et al., 2012; Kanji et al., 2017; Knopf et al., 2018). Homogeneous ice nucleation is responsible for the formation of ice particles in dilute water and supercooled solution droplets at temperatures lower than ≈−38°C (Pruppacher and Klett, 1997). Ice nucleation can also proceed through heterogeneous ice nucleation triggered by ice-nucleating particles (INPs) (Vali et al., 2015). Multiple heterogeneous ice nucleation mechanisms have been
proposed, such as deposition nucleation (ice formation on INPs directly from the vapor phase), contact freezing (freezing initiated by INPs the moment they come into contact with a supercooled droplet), and condensation and immersion freezing (freezing initiated by immersed INPs within the supercooled water or solution droplets). Deposition nucleation has also been referred to as a pore condensation and freezing mechanism because it is similar to immersion freezing but occurs in pores (Marcolli 2014). Nevertheless, the immersion-freezing mode is thought to be the most important process for the formation of ice particles within mixed-phase clouds (e.g., Ansmann et al., 2009; Westbrook and Illingworth, 2013).

Immersion-freezing measurements are commonly made using continuous-flow diffusion chamber (CFDC) devices (e.g., Rogers, 1988; Chen et al., 1998; Stetzer et al., 2008; Kanji and Abbatt, 2009; DeMott et al., 2010; Friedman et al., 2011; Chou et al., 2011; Jones et al., 2011; Kanji et al., 2013; Booze et al., 2016; Garimella et al., 2016; Schiebel, 2017; Zenker et al., 2017). These chambers consist of two ice-coated parallel walls held at different temperatures, and different ice supersaturations are achieved by regulating the temperature gradient. Aerosols sampled into CFDCs are subjected to known discrete temperature and relative humidity conditions, and when the water relative humidity (RHw) is more than a few percent above 100 %, droplets will activate on the majority of particles within the growth section of the chambers. CFDCs also typically have an evaporation section at the bottom of the chamber where the wall temperatures are controlled in order to evaporate the droplets that do not freeze. Frozen droplets are counted using an optical particle counter (OPC) to determine the atmospheric INP concentrations (Garimella et al., 2017). However, the maximum RHw values achievable in this manner can limit the ability to determine the maximum immersion-freezing fraction (DeMott et al., 2015).

Here, we have expanded the capabilities of the CFDC-style device to achieve the maximum activation of particles to detect the immersion-freezing number concentrations of INPs at various supercooled temperatures. We present data to assess the performance from a newly developed CFDC in which all individual aerosol particles are activated to droplets, and these droplets are exposed to a sequence of supercooled temperatures. This is accomplished by modifying the existing design of the Pacific Northwest National Laboratory (PNNL) ice chamber (e.g., Friedman et al., 2011; Kulkarni et al., 2012). In the modified version, the growth section of the chamber was maintained at a higher RHw level under moderate supercooling conditions to activate all particles to supercooled droplets, whereas the evaporation section was always held at ice-saturated conditions and cooled over a range of temperatures at a known constant rate. The evaporation section serves two purposes in this case: it induces the freezing of droplets and evaporates the unfrozen droplets. Validation experiments using standard salt solutions are presented. Various INP proxies for mineral dust types that have previously shown ice nucleation ability over a wide span of supercooled temperatures were used to test the performance of the modified chamber.

2 Experimental design and performance validation

2.1 Description of the existing and modified chamber

The PNNL CFDC-style ice chamber operated in the traditional mode, referred to as the Compact Ice Chamber (CIC)-PNNL, has been described previously (e.g., Friedman et al., 2011; Kulkarni et al., 2012). The chamber consists of two sections: a growth section and an evaporation section joined together but thermally isolated from each other. Each section consists of two parallel vertical surfaces that are coated with a thin layer of ice, and these plates are independently temperature-controlled using external cooling baths (Lauda Brinkmann Inc.). Application of an ice layer (≈0.5 mm thick) on these surfaces involved three consecutive steps: cooling the plates of the chamber to −25 °C, filling the gap between the two parallel surfaces with deionized water (≈18 MΩ cm), and expelling the water after 20 s. To produce the desired water or ice supersaturation conditions, a horizontal linear temperature gradient between the plates was applied, and the corresponding temperature and RHw or relative humidity with respect to ice (RHice) were calculated using the Murphy and Koop (2005) vapor pressure formulations. The single sheath and sample flow rates were 5 and 1 liters per minute (L min$^{-1}$), respectively, resulting in a total particle residence time of ≈10 s in the chamber. The temperature gradient was applied such that supersaturation conditions RHw ≈106 % were achieved in order to investigate the immersion-freezing efficiencies of both atmospheric and laboratory-generated INPs. An OPC (CLIMET, model CI-3100) was used to classify the particles as ice crystals if they were greater than a certain size threshold (≈3 µm in diameter). The ice fraction ($F_{\text{ice}}$) was calculated by using the ratio of the ice crystal concentration classified by the OPC to the total condensation nuclei (CN) concentration that entered the chamber. The CN concentration was provided by a condensation particle counter (CPC; TSI 3775). Blank experiments using dry and filtered sample air were also performed at the beginning and end of each experiment for ≈10 min to calculate the background number of ice particles. Further, these ice particles were subtracted from the ice crystal concentration measured by the OPC, and the $F_{\text{ice}}$ was corrected.

Figure 1 shows the vertical cross-sectional geometry of the modified mode PNNL ice chamber, which is now referred to as the modified compact ice chamber (MCIC). This chamber design has a parallel-plate CFDC-style geometry, whose principle of generating a supersaturation between the two parallel surfaces and determining the $F_{\text{ice}}$ is similar to that of the existing CIC chamber but with modifications as
described here. The growth and evaporation sections of the MCIC chamber are now referred to as the conditioning and nucleation sections, respectively. The lengths of the two sections are identical (0.45 m), which limits the total particle residence time to \( \approx 10 \text{s} \). The droplet residence and nucleation times within the chamber are a maximum of 6.5 and 2 s, respectively. The chamber wall temperature values as a function of time during one typical ice nucleation experiment are shown in Fig. 2. During our study, the temperature controller of the cooling thermostat was programmed such that the warm and cold wall temperatures of the conditioning section were set to \(-9^\circ\text{C}\) and \(-27^\circ\text{C}\), respectively. The resulting water and ice saturation conditions are shown in Fig. 3. Here, the conditioning section temperature was chosen based on previous knowledge about the onset temperature of the INP test species being needed to induce the nucleation of ice at colder temperatures (\(\approx -20^\circ\text{C}\)) and the lower detection limit being needed to measure ice concentrations for temperatures warmer than \(-20^\circ\text{C}\). The shaded region shows the period (\(\approx 30 \text{min}\)) of one ice nucleation measurement; i.e., the OPC data from this period only are analyzed. The \(F_{\text{ice}}\) now indicates the cumulative fraction of droplets frozen as a function of the decreasing temperature in the nucleation section (see Text S1 in the Supplement). This metric of reporting ice nucleation results is commonly used to report frozen fraction vs. temperature (\(F_{\text{ice}} \text{ vs. } T\)) results (see e.g., DeMott et al., 2018; Kanji et al., 2017; Kohn et al., 2016). The isothermal conditions of the nucleation section are maintained at ice saturation and cooled at a steady rate (0.5 \(^\circ\text{Cmin}^{-1}\)) by a separate cooling bath in order to determine the immersion-freezing efficiency of INPs as a function of supercooled temperature. The choice of steady-state cooling rate is empirical at this moment, and the experiment is terminated when the nucleation section reaches \(\approx -44^\circ\text{C}\). This additional supercooling below the onset of homogeneous freezing temperature allowed for freezing data to be obtained for use in controlling the data quality and in accounting for the uncertainty within the temperature. The implications of higher cooling rates for INP measurements were also explored. The particle residence time (\(\approx 5 \text{s}\)) and ice-saturated conditions of the nucleation section allow a sufficiently sized differential between supercooled droplets and ice crystals and in fact prevent “droplet breakthrough” (Stetzer et al., 2008). While keeping the conditioning section conditions constant, the temperature of the nucleation section is raised to \(\approx -20^\circ\text{C}\) to induce freezing of droplets and evaporate unfrozen supercooling droplets. The residence time in each section of the chamber is \(\approx 5 \text{s}\), and the ice layer spans both sections of the chamber. A cyclone impactor upstream of the ice chamber is used to remove the larger particles (>1.5 \(\mu\text{m}\) in diameter) while sampling airborne arable dust particles. The heating tapes (red rectangular strip) are attached to the walls to precisely control the temperature of the walls. W: warm wall; C: cold wall; E: nucleation section wall. The length of both the conditioning and nucleation section is 0.45 m. The width of the chamber is 0.15 m. The gap between warm and cold walls is 0.01 m.

2.2 Numerical modeling

At the entrance of the nucleation section, the temperature and \(RH_w\) profiles can be unsteady. To better understand the flow patterns of these profiles within the transition zone and the zone’s impact on droplet behavior, numerical simulations using computational fluid dynamics (CFD) were performed. In
colder walls (Fig. 3). The results show that the operating con-
ditions of the chamber produce a skewed velocity profile and
that the aerosol lamina is displaced toward the colder wall.
The aerosol lamina is surrounded by filtered sheath flow,
and its width is determined by the ratio of sample to sheath
airflow. Because sample flow ideally occupies this fraction
of the total flow, the aerosol lamina experiences a range of
temperature and saturation conditions. The center tempera-
ture and RH\textsubscript{w} conditions, including uncertainty across the
aerosol lamina (assuming ideal confinement in the lamina),
are \(-19.7 \pm 0.7\, ^\circ\text{C} \) and \(113 \pm 0.5\, \%\), respectively. Additional
simulations were performed to understand the center temper-
ture and RH\textsubscript{w} conditions required to confine the particles
that are moved out of the aerosol lamina. We found that the
revised uncertainties for center temperature and RH\textsubscript{w} condi-
tions were \( \pm 0.9\, ^\circ\text{C} \) and \( \pm 0.7\, \%\), respectively.

CFD simulations were performed to achieve a complete
description of the velocity, ice saturation, and temperature
conditions within the chamber (Fig. 4a). A three-dimensional
mesh of the chamber geometry was generated and ex-
ported to the commercially available CFD software ANSYS
Inc (2016). The CFD software solver was the pressure-based
steady-state Navier–Stokes equation, which has implicit and
absolute velocity formulations. The viscous model – the stan-
dard renormalization group theory (RNG) \( \kappa-\varepsilon \) turbulence
model – was used. This model treats velocity fluctuations
better than other turbulence models for such a geometry. This
turbulence model was used in conjunction with species trans-
port modeling capability such that the effects of smaller ed-
dies of fluid motion were better captured. The pressure out-
let boundary condition was used, as were the CFD solution
method to couple the pressure–velocity and the default SIM-
PLE scheme. The Lagrangian discrete-phase model was used
to simulate the potential INP trajectories released from the
sample injection region to the outlet end of the chamber. The
simulations were performed using an “uncoupled approach”,
which means the motion of INP particles does not influence
the fluid-flow pattern. The temperature and RH\textsubscript{w} fields of the
INP trajectories were used to calculate the droplet growth and
evaporation trajectories using a water vapor diffusion growth
theory (Rogers and Yau, 1989) that neglects temperature cor-
corrections and kinetic and ventilation effects and assumes
perfect mass and thermal accommodation coefficients (Text S2).

The CFD-simulated airflow velocity and RH\textsubscript{ice} profiles
from the central region of the conditioning section are nearly
similar to the analytical solution (Fig. 4a). Both calculations
show the presence of maximum humidity values near the
middle of the chamber but slightly displaced values toward
the cold wall. The fluid-flow temperature characteristics from
the moment the aerosol lamina joins the sheath flow show
that the aerosol sample quickly (< 0.5 s) cools at the en-
trance of the conditioning section. To gain a better under-
standing of RH\textsubscript{w} and temperature conditions within the con-
ditioning and nucleation sections, the simulated data set of
a potential INP trajectory transiting within the chamber is
shown (Fig. 4b). The potential INPs experience nearly con-

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**Figure 2.** Measured temperature of warm, cold, and nucleation sec-
tion walls during a typical experiment. The shaded area indicates the
experimental conditions during one INP measurement. During this INP measurement, the temperature of both warm and
cold walls is kept constant, while the nucleation section is cooled at
a steady rate (0.5 °C min\(^{-1}\)).

**Figure 3.** Steady-state airflow velocity and relative humidity (RH)
conditions calculated using the mathematical model developed by
Rogers (1988) within the conditioning section of the ice chamber.
The chamber warm wall (left) and the cold wall (right) are at \(-9\) and
\(-27\, ^\circ\text{C} \), respectively. The shaded area between the two verti-
cal dash-dotted lines shows the boundaries of aerosol lamina un-
der these above temperatures and flow conditions (sheath flow:
5 L min\(^{-1}\); sample flow: 1 L min\(^{-1}\)). The profiles are asymmet-
ric because of the thermophoretic drift of the flow, caused by the
thermal gradient between the walls, towards the colder wall. The
conditioning section is always supersaturated with respect to ice
(RH\textsubscript{ice} > 100\, %), and except the near-wall positions, the section is
also supersaturated with respect to water (RH\textsubscript{w} > 100\, %).

In this study, the warm and cold walls of the conditioning sec-
tion were maintained at \(-9\) and \(-27\, ^\circ\text{C} \), respectively, and
the nucleation section was maintained at \(-20\, ^\circ\text{C} \). Analytical
steady-state calculations based on Rogers (1988) were
also used to understand the nature of the flow velocity profile
and the position of the aerosol lamina between the warm and
colder walls (Fig. 3). The results show that the operating con-

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Figure 4. (a) Contours of CFD-calculated airflow velocity, RH_{ice}, and temperature profiles within the ice chamber. Warm and cold walls of conditioning section are maintained at −9 and −27 °C, respectively. The nucleation section is maintained at −20 °C. The dashed line shows the trajectory of a single INP within the aerosol lamina transiting through the chamber. (b) CFD-calculated temperature and RH_{w} profiles of a potential INP released from the sample injection region to the outlet end of the chamber. Analytical calculations of droplet growth and evaporation of such a potential INP (0.3 µm in diameter) are also shown. The left and right sides of the vertical dotted line represent the conditioning and nucleation sections, respectively. See the text for more details. Simulation results at other nucleation section temperatures are shown in Figs. S1–S4.

The temperature conditions within a short time, ≈1 s, after entering the conditioning section. The potential INPs are assumed (i.e., sub-saturated particle growth is ignored) to activate to droplets because they are greater in size than cloud CN sizes (Seinfeld and Pandis, 2016) and grow as long as RH_{w} is increasing or remains constant. As the INPs enter the nucleation section, their RH_{w} and temperature values equilibrate with the nucleation section conditions. These calculations show that the droplets grow to ≈4 µm in diameter, and they shrink as the RH_{w} and temperature decrease (within the nucleation section). Note that droplet freezing within the nucleation section is not simulated in these simulations.

Additional simulations with the nucleation section temperature set to −30 and −37.5 °C were also performed (Figs. S2–S5). These simulations show that the RH_{w} field of a potential INP slightly decreases (≈0.5 %) and then increases within a very short period of time (<0.5 s) at the entrance region of the nucleation section. However, calculations show that such a perturbation does not affect the droplet evaporation behavior within the nucleation section because they all evaporate within ≈1 s after they enter the nucleation section and within the uncertainty limits of the set temperature of the nucleation section but not before they reach the set temperature (see Figs. S2–S4). The simulations are extended to understand the RH_{w} and temperature conditions of potential INPs released from different regions of the inlet section of the chamber. Simulations of five potential INPs are shown in Fig. S5. It is observed that INPs experience various temperature conditions (−17 to −19.5 °C) within the conditioning section, but after ≈0.5 s they all enter the nucleation section, and the temperature of each trajectory is identical.

Additional evaporative cooling calculations were performed to understand the suppression of droplet temperature while they enter the nucleation section. In the nucleation section the supercooled droplets experienced sub-saturation (RH_{w} > 0.8) and colder temperature conditions (>−37.5 °C). The Kulmala evaporative model (Su et al., 2018) was used to determine the surface temperature of these droplets using steady-state aerosol lamina airflow velocity (Fig. 3) and theoretically predicted RH_{w} fields (Figs. S2–S4). The calculations (Table S1) show the negligible effect of evaporative cooling on the droplet temperature such that additional supercooling is within the reported temperature uncertainty (±0.7 °C) across the aerosol lamina, and therefore evaporating-droplet cooling effects within the nucleation section are ignored.

2.3 Homogeneous freezing of ammonium sulfate particles

The temperature conditions within the nucleation section were validated using size-selected ammonium sulfate (AS) particles. These particles were generated by atomization of an aqueous solution made by dissolving AS (1 g) and Milli-Q water (18.2 MΩ cm; 100 g), resulting in a 1 wt % solution concentration using a constant output atomizer (TSI 3076). The atomized droplets were transported through a diffusion drier to obtain the dry particles, which were further trans-
ported to the differential mobility analyzer (DMA; TSI 3081) to obtain size-selected particles that had mobility diameters of 200 nm. The choice of this size allowed for the generation of the maximum number concentration of monodisperse particles. The concentration of these size-selected particles was measured using a CPC, and the particles were further transported to the ice chamber. As stated previously, the temperature and RH conditions within the conditioning section were −20 °C and ≈ 113 %, respectively, and these conditions were held constant, which led to droplet activation of size-selected AS particles. Next, the nucleation section was steadily cooled from −20 to −40 °C, and the ice particles exiting the chamber were classified as ice particles. The ice particle size distribution with supercooling is shown in Fig. 5a. The results show that the droplets began to freeze via a homogeneous freezing mode at ≈ −37.5 °C. The maximum number of ice particle concentrations was observed at ≈ −38.5 °C, when all the droplets froze. The nucleation section is always maintained at RH\textsubscript{ice} = 100 % (see Fig. 4a), and such ice saturation conditions do not grow or sublime the ice crystals. Therefore, ice particle size measured by the OPC can be representative of the size of the droplet while freezing. At slightly warmer temperatures (between −38.5 and −37.5 °C), we observe ice particles of ≈ 2.0 µm in diameter. The appearance of these smaller ice crystals could be because of the freezing of these smaller droplets (a consequence of evaporation within the entrance zone of the nucleation section) compared to ≈ 5.0 µm droplets at ≈ −38.5 °C. These homogeneous freezing threshold temperature values are in agreement with previous studies (e.g., Ignatius et al., 2016; Kohn et al., 2016). For example, Kohn et al. (2016) found 100 % freezing of supercooled dilute aqueous solution droplets at ≈ −38.2 °C. Theoretical calculations using a homogeneous nucleation rate (e.g., Earle et al., 2010; Atkinson et al., 2016) were performed to predict the homogeneous freezing curves of droplets that were 4 µm in diameter. Homogeneous freezing curves for various probable droplet residence times within the nucleation section are shown in Fig. S6. Note the good agreement between the experimental and predicted freezing temperatures. These results also show the complete evaporation of supercooled droplets within the nucleation section because no ice particles are observed above −37.5 °C, and therefore the freezing results (see Sect. 3) at warmer temperatures (−37.5 °C) can be ascribed to the heterogeneous freezing of the droplets or immersion freezing.

This experimental setup was further applied to understand the relationship between the F\textsubscript{ice} of AS particles relative to the RH\textsubscript{w} conditions within the conditioning section. The aim was to investigate the RH\textsubscript{w} value at which all the size-selected AS particles activate to droplets. Here, the nucleation section was held at −42 °C to induce homogeneous freezing of solution droplets, while the RH\textsubscript{w} within the conditioning section was steadily increased. It can be seen that RH\textsubscript{w} values close to 113 % are required before all the AS particles are activated to droplets and measured as ice crystals (Fig. 5b). Higher RH\textsubscript{w} values enable the encapsulation of all particles that are within and may spread outside of the width of aerosol lamina into droplets (Garimella et al., 2017). In addition, high-saturation conditions help grow the droplets to the larger size, so they survive long enough to induce the freezing of droplets within the nucleation section.

2.4 Sample preparation

The immersion-freezing efficiency of K-feldspar, illite-NX, Argentinian soil dust, and airborne soil dusts from arable region particles was measured to test the performance of the

Figure 5. Homogenous freezing of water droplets containing 1 wt % ammonium sulfate solution. (a) OPC-classified ice particle concentrations as a function of ice crystal diameter at different temperatures. Warm and cold walls of conditioning section are maintained at −9 and −27 °C, respectively. (b) The fraction of frozen solution droplets with RH\textsubscript{w}, where the temperature of the nucleation section is maintained constant at −42 °C to induce droplet freezing via the homogeneous freezing mode, and RH\textsubscript{w} within the conditioning section was steadily increased from 90 % to 120 %. Slightly colder temperature (−42 °C) than homogeneous freezing limit (≈ −38.5 °C; panel a) is used to account for the uncertainty within temperature and RH\textsubscript{w} conditions. The dashed line in panels (a) and (b) indicate the increase in freezing fraction of droplets trend (for illustration purposes) and the onset of saturation line, respectively. The uncertainty in RH\textsubscript{w} is shown as an error bar (see the text for more details). The uncertainty in F\textsubscript{ice} is 1 standard deviation (n = 3). For clarity, error bars are shown only for one data point.
MCIC. K-feldspar (BCS376) was purchased from the Bureau of Analysed Sampled Ltd, UK. Dry-dispersed (TSI 3433) K-feldspar particles that had a mobility diameter of 400 nm were size-selected by a DMA, and the nearly monodisperse particles were transported to the CPC and ice nucleation chamber. Based on theoretical calculations (Baron and Willeke, 2001), the distribution of these classified particles may also contain sub-populations of double- (≈ 700 nm) and triple-charged (≈ 985 nm) particles. Laboratory measurements showed that the contribution of double- and triple-charged particles was less than 7 % and 3 %, respectively, which also justified the choice of 400 nm size particles. Therefore, the multiply charged particle contribution is ignored, and the K-feldspar aerosol stream is assumed to consist only of particles whose mobility diameter equals 400 nm. However, the surface area of multiply charged particles could influence $F_{\text{ice}}$ because these large particles (> 400 nm) provide larger surface areas (Lüönd et al., 2010). Illite-NX and Argentinian soil dust were sampled at the AIDA (Aerosol Interaction and Dynamics in the Atmosphere) chamber facility during the Fifth International Ice Nucleation Workshop (FIN-02) campaign (DeMott et al., 2018). During the campaign, the two aerosol types were dry-dispersed in two different chambers: an 84 m$^3$ AIDA chamber and a 4 m$^3$ aerosol particle chamber (APC); but in this study, we sampled directly from the APC. The details of particle generation and aerosol properties are described by DeMott et al. (2018). The direct sampling of these two aerosol types corresponds to experiment numbers 8 and 10 conducted on 16 and 17 March 2015, respectively.

Airborne soil dust from an arable region or shorty airborne arable dust particles were sampled at the PNNL sampling site during a regional windblown dust event. The PNNL sampling site is located within the Columbia Plateau, WA, USA, which is confined by the Rocky Mountains to the east, the Blue Mountains to the south, and the Cascade Mountains to the west. The region was once covered with basalt lava but is now built up with loose topsoil–loess. This fine soil, which is erodible, and the agricultural dryland farming practices make this dry soil susceptible to wind erosion. The sampling was performed during one dust event on 11 May 2017, and the average temperature, humidity, and wind speed during that day were 18 °C, 60 %, and 6.26 m s$^{-1}$, respectively. The sampling port was ≈ 9 m above the ground on the rooftop of the Atmospheric Measurements Laboratory located on the PNNL campus in Richland, WA. The airborne dust particles were drawn into the laboratory through a cyclone impactor (URG-200-30EH), which was operated at 30 L min$^{-1}$ to obtain a cut point diameter equal to 1.5 μm. This size-selective sampling allowed for removal of the larger particles (> 1.5 μm) and therefore helped to classify unambiguously the ice crystals larger than 3 μm using an OPC. The CN concentration of airborne arable dust particles (> 0.1 μm) was measured using a laser aerosol spectrometer (LAS; TSI 3340). The $F_{\text{ice}}$ was calculated by determining the ratio of ice crystals provided by the OPC to the CN counts measured by the LAS. In parallel to INP measurements, the particles were collected on a carbon type-B film (Ted Pella Inc.; 01814-F) for scanning electron microscopy–energy dispersive X-ray spectroscopy (SEM–EDS) analysis to better understand the size distribution and composition of these airborne dust particles. The films were mounted on the C and D stages of an SKC Sioutas impactor that had 50 % cut points of 0.5 and 1.0 μm, respectively. The impactor was operated at 9 L min$^{-1}$, and a total of 1183 particles were analyzed. Figure S7 shows the exemplary SEM images. The images reveal that the particles are mostly composed of minerals, and the size distribution shows the mean area equivalent diameter of ≈ 0.53 μm. The input aerosol concentration of all four INP species varied from 100 to 800 cm$^{-3}$, and the sampling duration was ≈ 30 min.

3 Results and discussion

The MCIC was operated to measure the maximum immersion-freezing fraction of INPs. The modified design allowed for the faster (~ 30 min) accumulation of immersion-freezing data points to develop a continuous representation of the immersion-freezing behavior of INPs compared to the traditional CIC-PNNL design, in which immersion freezing was investigated at discrete temperatures. These expanded capabilities were demonstrated by measuring the immersion-freezing properties of four INP substances: K-feldspar, illite-NX, Argentinian soil dust, and airborne arable dust particles.

The measurements of the immersion-freezing properties of the four samples were investigated at temperatures between −20 and −38 °C. The averaged $F_{\text{ice}}$ data over ΔT = 0.25 °C temperature intervals were plotted against the midpoint temperature of each bin (Fig. 6). The vertical and horizontal error bars are equal to 1 standard deviation of the $F_{\text{ice}}$ measurements ($n = 3$) and temperature uncertainty (±0.4 °C) across the nucleation section, respectively. Freezing experiments with AS solution droplets show the homogeneous freezing threshold temperature conditions being below ≈ −38 °C, and therefore $F_{\text{ice}}$ data points above this temperature can be attributed to the immersion-freezing mode only. Figure 6 shows that four INP materials exhibit a distribution of immersion-freezing temperatures. The $F_{\text{ice}}$ of all INP species increased with decreasing temperature, consistent with many past studies (e.g., Kanji et al., 2017). The droplets containing immersed K-feldspar particles froze at the higher temperatures. The median freezing temperatures (i.e., the temperature at which 50 % of the droplets froze) of K-feldspar, illite-NX, Argentinian soil dust, and airborne arable dust particles were −25.4, −32.6, −31.4, and −31.8 °C, respectively, and the difference between freezing temperatures corresponding to $F_{\text{ice}}$ equal to 90 % and 10 %
post-processing INP instruments would be needed. Future studies that involve collocated direct and post-processing INP instruments and particle generation methods. The \( n_s \) indicates the cumulative number of ice active sites that are present per unit area of particle surface and that induce nucleation of ice upon cooling from 0 °C to experimental temperature \( T \). In this calculation, time dependence is neglected, and it is assumed that the different active sites present within the droplets are responsible for the nucleation of ice. The \( n_s \) calculation follows Hiranuma et al. (2015) and DeMott et al. (2018):

\[
\ln n_s(T) = -\frac{\ln(1 - F_{\text{ice}})}{A} \approx \frac{F_{\text{ice}}}{A},
\]

where \( A \) is the surface area per particle, and the approximation is valid for \( F_{\text{ice}} \ll 1.0 \) in Eq. (1). For K-feldspar and airborne arable dust particle analysis, the surface area is calculated assuming the particles are spherical, and this assumption may overestimate the \( n_s \); therefore, calculations should be viewed as the upper estimates of \( n_s \). The size distribution and CPC concentrations were used to calculate the \( A \) of individual airborne arable dust particles, as described by Niemand et al. (2012). For illite-NX and Argentinian soil dust particles, the \( A \) was obtained from the FIN-02 data archive (DeMott et al., 2018). The error in \( n_s \) (Eq. 1) was calculated using the error propagation method based on the uncertainties in the \( F_{\text{ice}} \) and \( A \).

Figure 7 shows \( n_s \) for the four INP materials tested in this work in comparison to parameterizations reported in previous studies. The \( n_s \) for K-feldspar is compared to the fit published by Atkinson et al. (2013). There is a good agreement with our measurements for temperatures warmer than −26 °C. Atkinson et al. (2013) used a droplet-freezing cold stage technique, in which a known amount of K-feldspar material was present in each droplet sized between 14 and 16 µm. These droplets were cooled at a rate of 1 °C min⁻¹, and droplet-freezing temperature data were used to construct the \( n_s \) parameterization. Note that the \( n_s \) fit from Atkinson et al. (2013) is valid up to −25 °C. In our work, we extrapolated the fit outside this limit to colder temperatures for comparison. However, such linear extrapolation to colder temperatures may not be correct because, as both Niedermeier et al. (2015) and DeMott et al. (2018) (the latter from the FIN-02 campaign) have shown, the \( n_s \) values level off at temperatures colder than −25 °C. The \( n_s \) for airborne arable dust was compared with the previous studies. Niemand et al. (2012) derived the \( n_s \) fit using combined immersion-freezing data from various natural dusts (Asian soil dust, Canary island dust, Saharan dust, and Israel dust). Recently, Ulrich et al. (2017) developed \( n_s \) parameterization using immersion-freezing \( n_s \) densities of various arable dusts (Saharan desert dust, Asian desert dust, Israeli desert dust, Canary Island
Figure 7. Ice nucleation active site density ($n_s$) as a function of temperature for four INP test species tested in this study. The panels (a) to (d) show $n_s$ densities for K-feldspar, airborne arable dust, illite-NX, and Argentinian soil dust, respectively. Solid and dash-dotted lines represent various parameterizations from the literature. See the text for details. Dashed lines in panels (a) and (d) indicate the extrapolated data calculated outside the temperature limits recommended in these $n_s$ parameterizations. The black color symbols represent $n_s$ values from various other instruments that participated in FIN-02 activity (DeMott et al., 2018). Filled color symbols show the data from the CIC-PNNL chamber but operated at steady-state temperature and RH$_w = 106\%$ conditions at FIN-02. For clarity, confidence intervals are shown only for one data point from each study.

dust) for the temperature range from $−14$ to $−30$ °C. Tobo et al. (2014) investigated the INP abilities of agriculture soils dusts collected from Wyoming, USA. Boose et al. (2016) investigated the INP efficiencies of airborne dust samples from four locations (Crete, Egypt, Peloponnese, and Tenerife) and generated the minimum to maximum bounds of $n_s$ from $−29$ to $−37$ °C. The comparison of our results with these previous results shows good agreement within 1 order of magnitude at colder temperatures, but the data diverge at warmer temperatures. This could be the consequence of a particularly active soil dust present in the local region. The $n_s$ for illite-NX was compared to that of Hiranuma et al. (2015), who combined immersion-freezing data from several direct-processing INP methods to develop an $n_s$ parameterization. Here, we used the Gumbel cumulative distribution linear fit parameters derived from dry dispersion measurements to generate the $n_s$ fit. Our data agree within 1 order of magnitude at warmer ($−28$ to $−30$ °C) and colder temperatures ($−34$ to $−38$ °C), but at other temperatures ($−30$ to $−34$ °C) the data diverge. Finally, we compared our data with $n_s$ parameterization from Steinke et al. (2016). Steinke et al. (2016) used immersion-freezing data from four soil dust samples (Mongolian soil, Karlsruhe soil, German soil, and Argentinian soil) to produce an $n_s$ fit that is valid over a temperature range between $−26$ and $−11$ °C. We extrapolated the $n_s$ fit toward colder temperatures, and the comparison shows higher $n_s$ values but overlaps within the order of magnitude with others.

Figure 7a, c, and d also show the $n_s$ results reported by five different direct-processing INP instruments used in the FIN-02 campaign (DeMott et al., 2018). Our data for K-feldspar nearly align with the others at warmer temperatures ($> −28$ °C). For the illite-NX sample, agreement with the PIMCA–PINC (Portable Immersion Mode Cooling chAmber–Portable Ice Nucleation Chamber) method

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is within 1 order of magnitude, but the agreement is observed within 2 orders of magnitude with others. The present data for Argentinian soil dust align with the PIMCA–PINC method and agree with the others within 1 order of magnitude. The discrepancy between our results and others’ could be attributed to the different capabilities employed by individual measurement methods to investigate the immersion-freezing properties. The experimental uncertainties (e.g., ice crystal detection limit, RH, and temperature error limits) from these methods could also influence the $n_s$ results. Previously, evaporative freezing by contact nucleation inside-out has been hypothesized to explain the higher freezing temperatures and rates of ice formation observed during droplet evaporation (Durant and Shaw, 2005). Durant and Shaw (2005) showed that water droplets containing individual insoluble INPs freeze at a higher temperature compared to the immersion-freezing mechanisms. We cannot rule out that the evaporative freezing mechanism may be occurring in our experiments, and it would be responsible for the higher $n_s$ values to be compared to those of other studies. The comparison with the CIC-PNNL chamber showed that our data agree within 1 order of magnitude. Note that CIC-PNNL (PNNL ice chamber but operated in a traditional mode; Friedman et al., 2011; Kulkarni et al., 2012) was operated at RH$_w = 106$ %, and its operation limited investigating immersion freezing on the entire particle population. It can be observed that for illite-NX and Argentinian soil dust samples a correction factor of 4 to 5 is needed to apply to the CIC-PNNL data to match with the data from the MCIC.

Our results can guide design considerations for future CFDC-style ice chambers. The length of the conditioning section can be increased so that higher RH$_w$ values would not be necessary to activate all the particles to sufficiently large droplet sizes ($\approx 2 \mu m$ in diameter). This design feature could help to increase the lifetime of the ice layer. Based on CFD results (Figs. S3–S5), the minimum evaporation or nucleation length required is 0.2 m. In addition, by implementing a separate refrigeration system to independently cool the nucleation section, the new operation mode presented here can be adapted.

4 Conclusions

An alternative method of operating a CFDC-style ice chamber – the MCIC – was explored to determine the immersion-freezing ability of INPs. This new mode of operation allowed us to obtain maximum immersion-freezing fractions of INPs without droplet breakthrough ambiguity. Here, instead of investigating immersion freezing at discrete temperatures, immersion freezing was investigated by activating particles to droplets at high RH$_w$ followed by steady cooling under imposed ice-saturated conditions. The chamber performance was evaluated by testing the ice nucleation ability of four INP materials: K-feldspar, illite-NX, Argentinian soil dust, and airborne arable dust particles. In addition, we performed CFD simulations to evaluate flow, humidity, and temperature performance. The results indicate that these three thermodynamic conditions are locally fully developed, which confirms constant mass and thermal flux and therefore steady operating conditions within the chamber. Tests using size-selected AS particles showed that homogeneous freezing of solution droplets occurs in agreement with theory and previous study results and that, to activate all the particles to droplets, high RH$_w$ values of $\approx 113$ % are needed. Analytical and CFD calculations indicate that such high values are needed to grow the droplets to larger sizes so that they can survive long enough to induce freezing and to allow the particles that may have escaped the aerosol lamina to activate into droplets. Tests using the four INP materials demonstrated the activation of all individual particles to generate immersion-freezing spectra in terms of $F_{\text{Ice}}$ and $n_s$. Experimental results indicate that K-feldspar minerals induced detectable ice formation at $\approx -22 ^{\circ} C$, and maximum $F_{\text{Ice}} (\approx 90 \%)$ was observed at $-28 ^{\circ} C$. The other three samples induced nucleation of ice at temperatures colder than $-26 ^{\circ} C$, and their maximum $F_{\text{Ice}} (\approx 90 \%)$ was observed to be $\approx -36 ^{\circ} C$. The $F_{\text{Ice}}$ was normalized using particle surface area to calculate the $n_s$, and these $n_s$ calculations show that our results are comparable to the parameterizations and data reported in the literature. We find that the majority of our $n_s$ results are higher within 1 order of magnitude than others. Analysis of such high-temporal-resolution immersion-freezing measurements could offer better insights into the freezing properties of INPs, thereby moving us toward improved representations of the immersion-freezing ability of INPs for cloud models.

Data availability. Data plotted in this paper are available upon request.

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Author contributions. GK analyzed the data and wrote the paper. NH, OM, KH, SC, DJC, and PJD contributed and commented on all results. SC provided airborne arable dust composition and morphology results.

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