

1 **The SPectrometer for Ice Nuclei (SPIN): An instrument to**  
2 **investigate ice nucleation**

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1 **Abstract**

2 The SPectrometer for Ice Nuclei (SPIN) is a commercially available ice nucleating particle  
3 (INP) counter manufactured by Droplet Measurement Technologies in Boulder, CO. The SPIN is  
4 a continuous flow diffusion chamber with parallel plate geometry based on the Zurich Ice  
5 Nucleation Chamber and the Portable Ice Nucleation Chamber. This study presents a standard  
6 description for using the SPIN instrument and also highlights methods to analyze measurements  
7 in more advanced ways. It characterizes and describes the behavior of the SPIN chamber, reports  
8 data from laboratory measurements, and quantifies uncertainties associated with the  
9 measurements. Experiments with ammonium sulfate are used to investigate homogeneous  
10 freezing of deliquesced haze droplets and droplet breakthrough. Experiments with kaolinite, NX  
11 illite, and silver iodide are used to investigate heterogeneous ice nucleation. SPIN nucleation  
12 results are compared to those from the literature. Also presented (as an advanced use), is a  
13 machine learning approach for analyzing depolarization data from the SPIN Optical Particle  
14 Counter. Overall, we report that the SPIN is able to reproduce previous JNP counter  
15 measurements.

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## 1. Introduction

Aerosol particles facilitate the nucleation of cloud droplets and ice crystals in Earth's atmosphere (Pruppacher and Klett, 1997). Ice nucleating particles (INP) enable the formation of ice crystals via several possible mechanisms, including deposition nucleation, immersion freezing, and contact freezing (Rogers, 1989; Pruppacher and Klett 1997). Droplets freeze homogeneously below temperatures of  $\sim -38$  °C, including deliquesced haze droplets which do so below water saturation at such cold temperatures (Koop et al., 2000). Because of the complexity of the ice nucleation process, understanding INP interactions with water has been difficult (Hoose and Möhler, 2012; Boucher et al., 2013; Stocker et al., 2013). Despite this difficulty, the significant influence that mixed-phase, and ice clouds have on the Earth's radiative budget and hydrologic cycle makes understanding the microphysics of cloud formation an important step in quantifying their influence on climate (e.g. Storelvmo et al., 2011; Hoose and Möhler, 2012; Tao et al., 2012; Gettelman et al., 2013).

Laboratory measurements allow for the investigation of ice nucleation at specific conditions with controlled aerosol properties and provide insight into ice formation as it occurs in the atmosphere. Several types of instruments have been developed to measure the efficiency of heterogeneous nucleation of cloud droplets and ice crystals. Many of these have applicability for measurements in the laboratory, as well as intended application for field observations. Among these instruments, the Continuous Flow Diffusion Chamber (CFDC) (Rogers, 1988) has proven a useful tool to measure the conditions required to nucleate ice crystals on various INPs. Studies have been conducted on different nucleation and freezing mechanisms using many types of aerosol particles under a wide range of temperatures and relative humidities (RHs) (Rogers, 1988; Salam et al., 2006; Stetzer et al., 2008). Improved versions of the original cylindrical

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1 chamber described by Rogers (1988) have been successfully deployed in ground and aircraft  
2 based field campaigns (Chen et al., 1998; DeMott et al., 2003a; DeMott et al., 2003b). One  
3 contemporary (parallel plate) design is the Zurich Ice Nucleation Chamber (ZINC) (Stetzer et al.,  
4 2008), which has been used for several laboratory studies (e.g., Welti et al., 2009; Welti et al.,  
5 2014). The Portable Ice Nucleation Chamber (PINC), designed as a field-deployable version of  
6 the ZINC, has since been used to conduct several laboratory and field studies (Chou et al., 2011;  
7 Chou et al., 2013; Kanji et al., 2013). In addition, other research groups have also developed  
8 similar chambers (Kanji et al., 2009; Kulkarni et al., 2009; Friedman et al., 2011; Jones et al.  
9 2011; Saito et al., 2011). Adapting the parallel plate design and other features from the ZINC and  
10 PINC chambers, the SPectrometer for Ice Nuclei (SPIN) is a commercially available ice nuclei  
11 counter manufactured by Droplet Measurement Technologies (DMT) in Boulder, CO. This study  
12 characterizes the behavior of the SPIN chamber and reports data that characterize the general  
13 instrument design and performance.

## 2. Instrument theory and design

### 2.1 Operating principles

14  
15  
16  
17 CFDCs, such as the SPIN, are used for ice nucleation measurements by exposing aerosol  
18 particles to controlled temperature and RH conditions. The chamber walls (which are parallel  
19 plates in the SPIN chamber) are coated with a thin layer of ice (~1 mm thickness in the SPIN  
20 chamber, as inferred from the volume depleted from the water reservoir after icing). The water  
21 vapor partial pressure directly adjacent to the ice wall is the saturation vapor pressure over ice at  
22 the given ice wall temperature. A laminar air stream flows between the plates, and if the plate  
23 temperatures are different, water vapor and heat diffuse from the warmer to the colder wall. This

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1 idealized setup leads to linear profiles of water vapor partial pressure and temperature between  
2 the two walls. The exponential dependence of saturation vapor pressure on temperature,  
3 according to the Clausius-Clapeyron relation, leads to supersaturated conditions with respect to  
4 ice, between the two walls, with a maximum close to the position of the aerosol lamina (Rogers,  
5 1988; Stetzer et al., 2008). Aerosol particles are constrained within this lamina and surrounded  
6 by two sheath flows passed along each wall. This restricts the aerosol to a narrow range of  
7 temperature and supersaturation at which ice nucleation can take place. An example of the  
8 chamber flow and thermodynamic profile is shown in Figure 1.

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9 A sufficient temperature gradient between the walls results in the water vapor partial pressure  
10 in the aerosol lamina exceeding the saturation vapor pressure over liquid water. In this case  
11 droplets, in addition to ice, can nucleate on the aerosol particles. Though droplets can be  
12 identified using a depolarization optical particle counter (OPC) (such as the SPIN detector  
13 described in Section 2.2), increasing the size difference between droplets and ice helps in  
14 distinguishing the two phases. To accomplish this, CFDC chambers employ an evaporation  
15 section after the main chamber (Fig. 2) to shrink or eliminate droplets while retaining ice  
16 crystals. The ice walls in the evaporation section of the chamber are isothermal, so the water  
17 vapor partial pressure is equal to the saturation vapor pressure over ice. Droplets are therefore  
18 unstable and shrink in a manner akin to the Bergeron-Wegner-Findeisen process (Rogers, 1988;  
19 Pruppacher and Klett 1997). Depending on their residence time in the evaporation section,  
20 droplets over a critical size will not evaporate completely, and are large enough to be detected by  
21 the OPC. The main chamber conditions that generate droplets over this critical size are termed  
22 droplet breakthrough. These conditions are quantified for the SPIN chamber in experiments

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1 | described in Section 4 and represent an upper RH limit for ice nucleation experiments if droplets  
2 | and ice crystals are indistinguishable.

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### 3 | **2.2 SPIN chamber design**

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4 | Figure 3 shows a diagram of the SPIN system, illustrating the refrigeration, air flow control,  
5 | and water flow control components. The temperatures of the two chamber walls and the  
6 | evaporation section are controlled using compressor-driven refrigeration systems and heater  
7 | strips affixed to the walls. The warm wall and evaporation section are cooled using a single-stage  
8 | (with R404A refrigerant) refrigeration loop, while the cold wall is cooled using a two-stage (with  
9 | R404A first stage refrigerant and R508B second stage refrigerant) refrigeration loop. Ten  
10 | solenoid valves (four for the warm wall, four for the cold wall, and two for the evaporation  
11 | section) with proportional–integral–derivative (PID) control are used to regulate refrigeration.

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12 | Thirty 30 W heater strips (twelve on the warm wall, twelve on the cold wall, and six on the  
13 | evaporation section) are used to minimize deviations of temperature from the set point by  
14 | applying heating via twenty-six independent PID controllers (twelve for each of the warm and

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15 | cold walls and two for the evaporation section). T-type thermocouples that are inserted into the  
16 | walls and affixed with thermal epoxy are positioned at sixteen locations on each chamber wall  
17 | and two locations on the evaporation section to map variability in temperature (Fig. 2). The

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18 | chamber itself is machined from aluminum components, with the inner chamber walls  
19 | sandblasted for wettability, and junctions are sealed with rubber gaskets. The plate-to-plate  
20 | distance in the un-iced chamber is 1 cm, which is reduced on average to ~8 mm with the ice  
21 | layer. Qualitative inspection with an endoscope camera indicates that the ice layer is thicker  
22 | towards the bottom of the chamber and thinner towards the top. The ratio of the main chamber  
23 | length to the evaporation section length is 4:1.

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1 | A hepa-filtered and dried (using molecular sieve desiccant) sheath flow along each wall is  
2 | circulated through the chamber using a pump and mass flow controller (MFC). Sample air is  
3 | drawn into the system by an additional pump. The incoming sample air is drawn into the sheath  
4 | flow using a knife-edge inlet similar to the one used in the ZINC (Stetzer et al., 2008), which  
5 | splits the sheath into two flows that move along each wall. The knife-edge also focuses the  
6 | particle flow to the center of the chamber, which in laminar flow conditions, limit the  
7 | temperature and supersaturation range experienced by the particles. Figure 2 shows the  
8 | dimensions of the main chamber and evaporation section.

9 | After passing through the main chamber and evaporation section, the air stream flows  
10 | through a linear depolarization OPC that uses four optical detectors for counting, sizing, and  
11 | differentiating unactivated aerosol particles, droplets, and ice crystals in the 0.4 - 15 μm size  
12 | range. Figure 4 shows the optical diagram of the OPC. The side scatter detector is used for  
13 | particle sizing by total scattering intensity, and the backscatter detectors are used to measure P  
14 | (parallel to the incident laser light) and S (perpendicular to the incident laser light) polarization  
15 | for phase discrimination: ice crystals depolarize more light than water droplets because of  
16 | anisotropy of ice compared to liquid water (e.g. Wettlaufer et al., 1999; Thomson et al., 2009),  
17 | and this change in depolarization signal is used to differentiate the two phases (Liou and Lahore,  
18 | 1974; Nicolet et al., 2010; Clauss et al., 2013; Nichman et al., 2016). The OPC laser (Osela ILS-  
19 | 640-250-FTH-1.5MM-100uM) is a continuous wave 500 mW 670 nm laser with a top-hat beam  
20 | profile. One of two sets of backscatter optics has a polarizing beam splitter and measures  
21 | backscattered light in both P and S polarizations (P1 and S1, respectively). The second set of  
22 | backscatter optics measures only the P polarization (P2). The detection angle of both sets of  
23 | backscatter optics is centered at 135° and has a half angle of 20°.

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1 | LabVIEW software is used for instrument control and data acquisition. The SPIN software  
2 | program consists of several different loops and sub-programs and allows for significant  
3 | automation during operation. User control of the various SPIN components, including the  
4 | compressors, valves, and detector is also performed and automated through the LabVIEW  
5 | interface. Individual actions, such as toggling valves, as well as sequences, such as icing the  
6 | chamber walls, are controllable through software. The software also includes functionality to  
7 | create custom sequences, allowing for the majority of operations (including system and  
8 | compressor startup, cooling the chamber, icing the walls, and running the activation experiments  
9 | described in Section 3.1) to be automated for increased experimental reproducibility. High data  
10 | load corresponding to large OPC concentrations will cause a drop counting efficiency: for 1 Lpm  
11 | sample flow, this corresponds to particle counts higher than ~3900 per cm<sup>3</sup>, above which  
12 | additional particles are not recorded. In addition to the foreground sequences initiated by the  
13 | user, background sequences can also be run to monitor instrument performance. With remote  
14 | access enabled through virtual network computing (VNC) software (separate from the LabVIEW  
15 | software), much of the chamber operation can be performed remotely. In particular, starting the  
16 | compressors, cooling the chamber, icing the chamber, switching the chamber inlet and outlet  
17 | valves, and controlling chamber conditions can be preformed remotely through VNC, but  
18 | refilling the water reservoir, installing/removing the OPC, and switching other valves must be  
19 | performed on site.

### 21 | 3. Methodology

#### 22 | 3.1 Experimental methods

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1 Before beginning experiments, the chamber is dried, cooled, and the walls are coated with  
2 ice. This is accomplished by first flowing dry nitrogen through the chamber via the sample and  
3 sheath flow inlets to remove residual moisture; the flow exiting the chamber outlet is routed  
4 through a dew point sensor, (Vaisala DMT152 dewpoint transmitter), so the moisture content of  
5 the chamber can be directly measured to ensure the dew point is below  $-40^{\circ}\text{C}$ . The compressor  
6 system is then activated to cool the chamber (both walls of the main chamber and evaporation  
7 section) to the icing temperature of  $-25^{\circ}\text{C}$ . Before icing, the double distilled deionized 18.2 M $\Omega$   
8 Millipore (DDI) water in the reservoir is cooled to  $\sim 2^{\circ}\text{C}$  to reduce strain on the refrigeration  
9 system during icing and to ensure that the wall temperatures do not exceed  $0^{\circ}\text{C}$  over the course  
10 of the icing process. With the water reservoir attached to the two-way water pump, the icing  
11 sequence is activated in the software. This sequence controls the filling and emptying of the  
12 chamber with DDI water to form the ice layers. The ice dwell counter in the software specifies  
13 the amount of time the chamber is filled with water and is typically set to 5 s. During and after  
14 the icing sequence it is critical to prevent moist room air from entering the chamber, which can  
15 cause non-uniform ice on the chamber walls via the formation of frost. This is accomplished by  
16 flowing dry air or nitrogen through the sample and sheath flow inlets while allowing the excess  
17 flow pressure to be released into the room upstream of the chamber inlets. The entire filling  
18 sequence typically lasts  $\sim 5$  min. The difference in the volume of water in the reservoir before  
19 and after the icing process is used to infer the amount of ice formed, and this difference is  
20 typically  $\sim 1.25$  L. After the ice layer has been formed, the dry nitrogen flow through the  
21 chamber is continued to ensure that no frost accumulates in the chamber. Subsequent installation  
22 of the detector and activation of the sheath pump allows for assessment of background frost

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1 counts that may bias the reported INP concentrations. This background concentration (typically  
2 between a few counts to several 10s of counts L<sup>-1</sup>) influences the lower detection limit of INP.

3 Once the chamber is iced and has a sufficiently low background, it is ready to perform INP  
4 activation experiments. This is accomplished in one of two main ways: (1) ramping the wall  
5 temperatures to determine the temperature and/or supersaturation dependent ice crystal  
6 concentration or (2) keeping the walls at different but constant temperatures to measure the

7 temporal variability of ice crystal concentration at desired temperature and supersaturation  
8 conditions. For the former, increasing the temperature gradient between the walls increases the  
9 chamber supersaturation, and decreasing the gradient decreases chamber supersaturation.  
10 Ramping both wall temperatures allows for temperature scans at the same supersaturation. For  
11 experiments in this study, the evaporation section temperature is set to be the same as the  
12 average aerosol lamina temperature. Also, in all cases, the OPC reports side scatter (sizing) and  
13 backscatter (depolarization) spectra to infer size, concentration, and phase of counted particles.

14 Frost backgrounds are typically higher in the SPIN chamber at higher supersaturations, and  
15 the presence of frost in the chamber can lead to data artifacts. Therefore, periodically measuring  
16 the background frost counts with no particles in the chamber (by setting the inlet valve to the  
17 filter position for 3-5 minutes) is an important procedure during activation experiments  
18 (described below). For converging and diverging wall temperature ramps, (with typical ramp  
19 rates of  $dRH/dt$  at 2 % min<sup>-1</sup>), this check is performed at the beginning and end of each ramp.  
20 For constant supersaturation experiments, this check is performed at fixed time intervals,  
21 typically twice per hour. Experiments are automatable using sequences in the SPIN software.  
22 These sequences automate the periodic background checks as well as controlling the wall  
23 temperature set points. The background concentration increases over time as vapor is transferred

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1 from the warm wall to the cold wall, leading to irregularities in the ice layers: as a result, the  
2 experiment must be ended once it no longer meets the background levels required for the  
3 particular experiment. The exact time this occurs depends on the particular operating conditions  
4 for an experiment but is typically after 2-5 hours of operation. For example, if a laboratory  
5 experiment with 10 INP cm<sup>-3</sup> were to report activated fractions at the 1 % level, it would require  
6 a background of no more than 100 counts L<sup>-1</sup>.

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7 If the temperature gradient between the warm and cold walls is large (e.g. larger than ~ 10-15  
8 °C, depending on the actual temperatures) the buoyancy of the air adjacent to the warm wall is  
9 expected to overcome the mean flow and causes (upward) flow reversal along the warm wall  
10 (Rogers, 1988). The dashed line in the top panel of Figure 5 shows the ice saturation ratio (S<sub>ice</sub>)  
11 above which flow reversal is possible according to the calculations from Rogers (1988), and the  
12 bottom panels show two examples of normal and reversed flow profiles. If flow reversal  
13 interferes with the aerosol lamina, the chamber behavior may deviate from ideality (Rogers,  
14 1988; Stetzer et al., 2008). Increasing the sheath flow combats flow reversal, but it decreases the  
15 residence time of particles in the chamber, which reduces nucleation and crystal growth  
16 efficiencies (Rogers, 1988).

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## 17 **3.2 Data processing methods**

### 18 **3.2.1 Standard use**

19 The temperature and supersaturation conditions in the chamber are calculated assuming a  
20 linear temperature and water vapor partial pressure gradient between the walls. The width and  
21 location of the aerosol lamina (and therefore the range of temperatures and supersaturations  
22 experienced by the aerosols) is calculated extending the method from Rogers (1988) using the  
23 reported values for wall temperatures, (at all thermocouple locations), sheath flow rate, and

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1 sample flow rate. The thickness of the ice layer where particles nucleate is assumed to be  
2 negligible in the calculations, since the nucleation region (at the top of the chamber) has a much  
3 thinner ice layer than the chamber average. This approach provides a basis for calculating  
4 chamber conditions at each pair of thermocouples (on the warm and cold wall, respectively, at a  
5 given location). The flow rates and temperature conditions are used to calculate the velocity  
6 profile, and the ratio of the sample flow rate to the total flow rate is used to determine the width  
7 of the aerosol lamina (Rogers, 1988; Kulkarni and Kok, 2012). Since a velocity profile  
8 calculation is part of this procedure, the fraction of reversed flow is known.

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9 The above calculations provide temperatures and supersaturations as a time series along with  
10 the size distributions and depolarization signals measured by the OPC, which provides four  
11 values (one from each of the detectors) on a single particle basis. The number of aerosol particles  
12 that have activated into ice crystals or droplets in the standard use case is based on a size  
13 threshold, typically 3-5  $\mu\text{m}$ , above which only particles that have nucleated into droplets or  
14 crystals exist. Droplet breakthrough conditions represent an upper RH limit for the standard use  
15 case because nucleated droplets and ice crystals may be indistinguishable based on size alone.  
16 Experimentally determined droplet breakthrough thresholds are presented in Section 4. A  
17 condensation particle counter (CPC) is typically used in parallel to SPIN to measure the  
18 concentration of particles entering the chamber, so the estimate of the number of ice crystals  
19 from above is used to infer activated fractions by dividing the ice crystal concentration by the  
20 total particle concentration entering the chamber.

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### 21 **3.2.2 Advanced use**

22 In the advanced use case, the number of aerosol particles that have activated into ice crystals  
23 or droplets is inferred in post-processing from classification of the particle-by-particle (PbP) data

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1 | in the four-dimensional OPC parameter space via supervised machine learning, (ML) (Mohri et  
2 | al., 2012). This process is described below and illustrated in Figure 6 for an example freezing  
3 | experiment. *Please note that the ML procedure described in this section is not an out-of-the-box*  
4 | *functionality of the SPIN software but rather a method developed by the authors to utilize the*  
5 | *instrument in more advanced ways.*  
6 | CFDC OPC data has historically been analyzed using post-evaporation section particle size  
7 | as the sole determiner of activation into ice or droplets, since ice crystals grow to be much larger  
8 | than the seed particles (e.g. Rogers et al., 2001; DeMott et al., 2010; Chou et al., 2011). If  
9 | particles larger than a certain size are assumed to be ice crystals, then it must either be assumed  
10 | or imposed that all aerosol particles are smaller than the ice size and that droplets above that size  
11 | do not survive the evaporation section. In order to meet this constraint, particle impactors are  
12 | often used to prevent the largest (and potentially most ice active) particles from entering the  
13 | CFDC (e.g. Rogers et al., 2001; DeMott et al., 2010; DeMott et al., 2015). Also, if the main  
14 | chamber is supersaturated with respect to water, the aerosol particles may activate into droplets.  
15 | The evaporation section is designed to avoid counting these droplets in the OPC, but it will cease  
16 | to evaporate droplets completely above a threshold (temperature-dependent) supersaturation  
17 | level. This droplet breakthrough threshold typically provides an upper limit for measurable  
18 | supersaturation, above which droplets must be differentiated from ice crystals, and it marks the  
19 | upper RH limit for traditional CFDC operation and data analysis. However, the addition of  
20 | depolarization data and analysis using supervised ML algorithms allow for this size assumption  
21 | to be relaxed, since all detected particles are classified by phase and the uncertainty associated  
22 | with this classification is quantified (see below). In general, this ML approach provides accurate,  
23 | reproducible, and uncertainty-quantifying analysis of the OPC data using preexisting MATLAB

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libraries (<http://www.mathworks.com/help/stats/classification.html>;  
[http://www.mathworks.com/matlabcentral/fileexchange/41187-fast-kernel-density-estimator--  
multivariate-](http://www.mathworks.com/matlabcentral/fileexchange/41187-fast-kernel-density-estimator--multivariate-)). It also requires fewer assumptions to be made about particle classification and  
allows more flexibility in experimental design.

Supervised ML algorithms for classification are used to assign new data to predetermined  
classes based on the similarity of the new data to each of these classes (Mohri et al., 2012). The  
different output classes in the SPIN OPC data are aerosol particles, water droplets, and ice  
crystals. These classes must have training data that correspond to a known class (Mohri et al.,  
2012). However, the SPIN OPC data from an activation experiment typically contains mixtures  
of these classes rather than the pure end members, so choosing the data that correspond to a class  
is performed statistically. In particular, training data is sampled from a subset of the data that  
represents the different classes.

In order to inform this sampling, Kernel Density Estimation (KDE) (Rosenblatt, 1956;  
Parzen, 1962) is used to create probability density functions (PDFs) of the data in various time  
intervals. Using the KDE approach to sample training data factors in the relative likelihood that a  
given class of particle (aerosol, droplet, or ice) will appear in an area of the parameter space.  
This approach takes into account the structure of the underlying PDFs of the training data to  
incorporate training data uncertainty into estimates of classification uncertainties.

The SPIN OPC reports four intensity count values (size, S1, P1, and P2) in the PbP data, and  
KDE with automatic bandwidth estimation is used to create a four dimensional Gaussian mixture  
model (GMM) (McLachlan and Peel, 2000) of the PDFs (one dimension corresponding to each  
of the intensity count values from the SPIN OPC) in this particle data, following the method of  
Kristan et al., (2011). Figure 6 shows data from an illite NX freezing experiment at -40 °C with

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1 ramping supersaturation. Specifically, Figure 6a shows PbP data from the beginning of the  
2 experiment during an *aerosol only* time interval (time 1, when only aerosol particles are present  
3 in the chamber). It also shows the corresponding KDE; for ease of visualization the 2D GMM-  
4 KDE in S1/P1 vs. size parameter space shown in lieu of the full 4D GMM-KDE, used in the  
5 analysis. Figure 6b similarly shows data and the GMM-KDE during an *aerosol + ice* time  
6 interval (time 2, at a supersaturation higher than that required for the onset of ice formation,  
7 where both particles and ice are present). The PDF in Figure 6a is deconvolved from the one in  
8 Figure 6b and is used to create a residual PDF, which is then used to inform the weighted  
9 sampling of data from time 2 to create training data for the *ice only* output class (corresponding  
10 to the region of the parameter space only ice crystals occupy, Fig. 6c). The data corresponding to  
11 the *aerosol only* (Fig. 6a) and *ice only* (Fig. 6c) classes are then used to train a supervised ML  
12 algorithm. In this example, a Gaussian kernel support vector machine (SVM) (Mohri et al., 2012)  
13 is used, and classification uncertainties are quantified via 5-fold cross validation of the training  
14 data (Fig. 6d). Once trained, the SVM classifies the OPC data from an experiment with a known  
15 missed classification percentage (Fig. 6e).

16 As in the standard use case, a CPC is used in parallel to SPIN to measure the concentration of  
17 particles entering the chamber, so the estimate of the number of ice crystals from above is used  
18 to infer activated fractions by again dividing the ice crystal concentration by the total particle  
19 concentration entering the chamber.

20 Figure 7 shows an extension of this approach to discriminate aerosol particles, water droplets  
21 and ice crystals using an application of 3-class supervised ML (Mohri et al., 2012). Bootstrap  
22 aggregated decision trees (Breiman, 1996) are used for the classification instead of SVM in this  
23 case, because this classification algorithm outperforms SVM in terms of classification error in

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1 the 3-class case: both algorithms are operationally interchangeable, so the better performing one  
2 with respect to classification error was chosen. This example shows a (dry-generated,  
3 polydisperse) silver iodide (AgI) activation experiment at -17\_°C where the chamber RH is  
4 increased. Though a 4D parameter space is again used for this classification, Figure 7a shows  
5 only three of the PBP data dimensions along with 2D projections at the axes limits to illustrate  
6 the portions of the parameter space occupied by each class. With this approach, the  
7 concentrations (Fig. 7b) and fractions (Fig. 7c) of ice crystals and water droplets along with  
8 classification uncertainty is reported as a function of chamber conditions, (note that evaporation  
9 section reduces droplet fractions, even above the droplet breakthrough point). The time series  
10 data are corrected for background frost by quantifying the frost counts that are classified as ice  
11 crystals by the algorithm. At the subsaturated RH conditions in Figure 7c, data with lower error  
12 bars that are not greater than zero show activated fractions that are statistically indistinguishable  
13 from zero. Overall, with the large datasets (up to thousands of data points per second) generated  
14 by the SPIN OPC, particle classification is performed in a reproducible manner with  
15 classification accuracies of 95.0-99.9%.

16 Once similarly large datasets are generated for field measurements in future studies, ML  
17 classifiers can be used for distinguishing frost from real ice in the field. Also, the general ML  
18 approach can be used for other instruments with size-only data: for example, an SVM that uses  
19 size only would find the optimal size (by maximizing the margin between ice and aerosol  
20 training data) to distinguish the two classes and quantify the uncertainty associated with  
21 choosing this size via cross-validation.

#### 23 4. Results and comparisons to literature

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1 To evaluate the performance of the SPIN OPC, sizing and detection experiments are  
2 performed with different sizes of monodisperse spheres. Glass beads, polystyrene latex spheres,  
3 and melamine resin spheres are used for the characterizations. The results from these calibrations  
4 are shown in Figure 8, and illustrate how the SPIN OPC sizes particles (Fig. 8a). The sizing  
5 behavior follows a power law fit for particle sizing between 0.5 and 11 μm. Smaller particles  
6 scatter the laser light less efficiently, and rapidly decreasing detection efficiency with decreasing  
7 size is observed for sub-micrometer particles (Fig. 8b). In the super-micrometer size range  
8 (where ice crystals are expected to be present) the counting efficiency is very close to 1.

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9 To evaluate the behavior of the SPIN chamber, several types of freezing experiments are  
10 reported. Specifically, 200 nm (nebulized, dried, and mobility selected) ammonium sulfate (AS)  
11 aerosol is used to determine the droplet breakthrough line and to compare the deliquesced haze  
12 droplet homogeneous freezing points reported by SPIN to literature values. Also, polydisperse,  
13 dry-generated NX illite and AgI (Sigma Aldrich >99 %, with a narrow mobility size distribution  
14 between 200-300 nm) particles are used to investigate the heterogeneous ice activation for  
15 comparison to literature, (note that the purity levels of AgI in previous studies is unknown). The  
16 homogeneous freezing results shown in Figure 9 suggest that the temperatures and  
17 supersaturations reported by SPIN are indicative of the real conditions in the chamber. Similarly,  
18 the results from the heterogeneous freezing experiments are shown in Figure 10 and indicate  
19 freezing occurs in the expected regions of phase space for the different seed particles.

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20 AS data is used to characterize SPIN's behavior in humid (near or above water saturation)  
21 conditions across a wide range of temperatures. AS freezing experiments at colder temperatures  
22 compare the chamber performance to the well-characterized homogeneous freezing behavior of  
23 deliquesced haze droplets. Though solid AS has been observed to nucleate in the deposition

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1 | mode (Abbat et al., 2006), this effect is negligible for the temperatures and activated fractions  
2 | considered in this study. The data in Figure 9 show that this behavior is captured in both  
3 | temperature and RH ramps and occurs in the expected region of phase space. They demonstrate  
4 | that droplets in SPIN begin to freeze when the chamber temperature falls below  $\sim 38$  °C and that  
5 | homogeneously frozen ice crystals begin to disappear above this temperature. Similarly, the RH  
6 | ramp data show that supercooled aqueous AS haze droplets freeze at RH levels similar to those  
7 | predicted by the Koop et al., (2000) line for a homogeneous nucleation rate coefficient  
8 |  $J = 10^{11} \text{ cm}^{-3} \text{ s}^{-1}$ . The effects of multiply charged particles is also negligible because the  
9 | uncertainty contributed by these particles on expected homogenous freezing supersaturation ( $\pm 1$   
10 | % RH using the relevant values from Koop et al., (2000)) is smaller than the other measurement  
11 | uncertainties of these experiments ( $\pm 5$  % RH).

12 | At warmer temperatures, homogeneous freezing does not occur, so AS is used to measure  
13 | SPIN's droplet breakthrough threshold. Though post processing of the PbP detector data can  
14 | distinguish droplets from ice (Section 3.2), droplet breakthrough is quantified using AS (which  
15 | is much more hygroscopic than many INPs) and provides a conservative (lower RH) estimate for  
16 | where this occurs. The measured droplet breakthrough line indicates that SPIN's evaporation  
17 | section extends (a few percent above water saturation) the region of phase space where  
18 | experiments can be performed without needing to differentiate droplets from ice crystals.

19 | AgI, despite not being found in the atmosphere, nucleates very efficiently in the deposition  
20 | mode across a range of temperatures and provides a benchmark to assess chamber performance  
21 | (e.g. Detwiler and Vonnegut, 1981; Stetzer et al., 2008). The SPIN results shown in Figure 10  
22 | match literature data across a wide temperature range and demonstrate that SPIN accurately  
23 | captures ice nucleation at warmer temperatures where there are many atmospherically relevant

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1 INP that activate into ice crystals. In addition to AgI results, heterogeneous freezing results for  
2 NX illite and kaolinite are also included, because they are commonly used surrogates for  
3 atmospheric dust, which is important for ice nucleation at colder temperatures (DeMott et al.,  
4 2003b; Welti et al., 2009; Cziczo et al., 2013). Unlike AgI, NX illite and kaolinite show a strong  
5 temperature dependence in freezing behavior and nucleate ice much less efficiently at  
6 temperatures warmer than  $\sim -35$  °C. Previous investigation of these materials has shown this  
7 transition (e.g., Welti et al., 2009), and the SPIN data in this study also capture this temperature  
8 dependence. Additionally, the coldest illite data points demonstrate that SPIN can access  
9 temperatures in the cirrus cloud regime.

10 Figure 11 shows SPIN performance during an exemplary ambient measurement taken in  
11 Cambridge, MA on April 14<sup>th</sup>, 2016 with an aerosol temperature of  $\sim -30$  °C and water saturation  
12 ratio ( $S_{liq}$ ) of 1.05 (reported lamina temperature was  $-30.7$  °C  $\pm$  0.2 °C and lamina  $S_{liq}$  was  $1.05 \pm$   
13 0.01 over the measurement period). Polydisperse ambient aerosol was sampled through an inline  
14 molecular sieve dryer with no upstream impaction. The SPIN sheath flow was provided from a  
15 dry nitrogen cylinder (instead of recirculating flow through a dryer) to minimize frost. Assuming  
16 a 5  $\mu$ m threshold for ice yields 13-40 INP  $L^{-1}$  in the measurement periods and 3-9 frost counts  $L^{-1}$   
17 during the filter periods: the INP concentration adjusted for frost was then 9-36 INP  $L^{-1}$  over the  
18 entire measurement period.

## 20 **5. Measurement uncertainty**

21 With a CFDC instrument, there are various sources of uncertainty that influence  
22 experimental results (e.g. Rogers, 1988; Stetezer et al., 2008). Uncertainties in wall temperatures,  
23 flows, numbers of counted ice crystals, flow reversal, and other deviations from ideality must be

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1 considered when interpreting results from a CFDC chamber. In SPIN, wall temperatures are  
2 recorded at sixteen locations on each wall. The aerosol lamina temperature and supersaturation  
3 conditions are calculated based on the measurements at these locations, providing a way to infer  
4 the variations in thermodynamic conditions experienced by the aerosol. Since the three  
5 bottommost thermocouples are strongly coupled to the evaporation section and occupy the  
6 transition region between the moist main chamber and dry evaporation section, only the top  
7 thirteen thermocouples are used for reporting average chamber conditions. Therefore, uncertainty  
8 in chamber conditions is reported as the standard deviation of the lamina temperature and RH at  
9 these thirteen locations. Variability in the wall temperatures depends on operating conditions  
10 (but typically has a standard deviation of 0.5-2 °C), and these variations must be included in  
11 uncertainty estimates. Along with these temperature gradients, variations in temperature and  
12 supersaturation across the width of the aerosol lamina are sources of uncertainty in SPIN  
13 measurements. The width of the aerosol lamina itself can be a source of uncertainty (Garimella et  
14 al., 2015), and merits investigation in future work. For the purposes of this study,  
15 misclassification error from the ML approach is used for reporting uncertainties in ice crystal  
16 concentrations and activated fractions.

17 Computational fluid dynamics (CFD) calculations in this section are used to explore how  
18 closely simulated chamber conditions match ideality. Figure 12 shows results from ANSYS  
19 Fluent CFD modeling (Fluent, 2015) of the SPIN chamber. Several simulations were performed  
20 and results from two of these are shown. Specifically, the left column in Figure 12 shows results  
21 for nominal lamina temperature of -40 °C and lamina  $S_{ice}$  at 1.3, and the right column shows  
22 results for nominal lamina temperature of -30 °C and lamina  $S_{ice}$  at 1.1. Overall, simulated  
23 chamber temperature, RH, and flow velocity match the ideal case predicted by Rogers (1988) for

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1 these and other Fluent experiments. Flow reversal along the warm wall (Fig. 5) is also observed  
2 in high RH simulations, but as with the physical experiments, this effect does not appear to have  
3 an appreciable influence on the aerosol lamina conditions beyond that which is predicted by  
4 Rogers (1988) model. Since the analytical and CFD models show qualitatively similar flow  
5 reversal, this effect is accounted for when reporting chamber conditions with the analytical  
6 model. Furthermore, agreement between the homogeneous freezing data and expected  
7 homogeneous freezing conditions provides evidence, similar to that shown by Richardson  
8 (2009), that this effect does not bias results, since these experiments are performed where the  
9 largest degree of flow reversal is expected.

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## 11 6. Conclusions

12 This study outlines the operating principles, chamber design, and experimental results from  
13 the SPIN chamber, a commercially available CFDC chamber manufactured by DMT. The results  
14 from this study evaluate the SPIN chamber performance using a variety of experimental tests and  
15 CFD modeling.

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16 The introduction of SPIN as a commercially available ice chamber is similar to the  
17 introduction of the DMT Cloud Condensation Nuclei Counter (Roberts and Nenes, 2005; Lance  
18 et al., 2006), potentially allowing these measurements to be made with higher temporal and  
19 spatial coverage. In addition, the methodologies highlighted in this study facilitate performing  
20 INP measurements, with increased experimental flexibility and reproducibility and also with  
21 fewer assumptions in the analysis. In particular, the addition of a depolarization detector coupled  
22 with supervised ML algorithms for data analysis allows for robust determination of particle  
23 phase with uncertainty quantification. These results motivate future work to extend the ML

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1 approach to field data analysis and to intercompare the SPIN chamber performance with other  
2 INP counters in the laboratory and in the field.

3 The experiments presented in this study illustrate SPIN's measurements of freezing behavior  
4 of both the heterogeneous and homogeneous regimes and demonstrate that the SPIN chamber  
5 reproduces freezing data measured in previous studies. The AgI measurements span a wide range  
6 of temperatures, extending to regions where the more efficient atmospheric INP would activate  
7 into crystals. The NX illite and kaolinite measurements demonstrate that SPIN measures into the  
8 heterogeneous freezing portion of the cirrus cloud regime and also captures the temperature  
9 dependence of mineral dust ice activity. The AS experiments provide an estimate of the droplet  
10 breakthrough level of the SPIN chamber at warmer temperatures, and at colder temperatures  
11 shows that homogeneous freezing of deliquesced haze droplets occurs in agreement with  
12 previous experiments and theory. By using an uncertainty quantifying OPC analysis technique,  
13 recording high spatial resolution temperature measurements along the chamber walls, and  
14 investigating the chamber conditions using CFD modeling, the main sources of uncertainty in  
15 SPIN measurements have also been studied.

16 Overall, the SPIN chamber reproduces laboratory data measured by previous CFDC  
17 chambers, and the uncertainties in the measurements have been investigated. The commercial  
18 availability of such an instrument may allow for increased coverage of INP measurements that  
19 will help constrain the influence of ice nucleation on the atmospheric radiation budget and the  
20 initiation of precipitation, thereby leading to a better understanding of the impact of ice  
21 formation on the Earth's climate and water budget.

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1 **Acknowledgements**

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3 Office of Science, STTR program under Award Number DE-SC-0004258. S. Garimella and D. J.  
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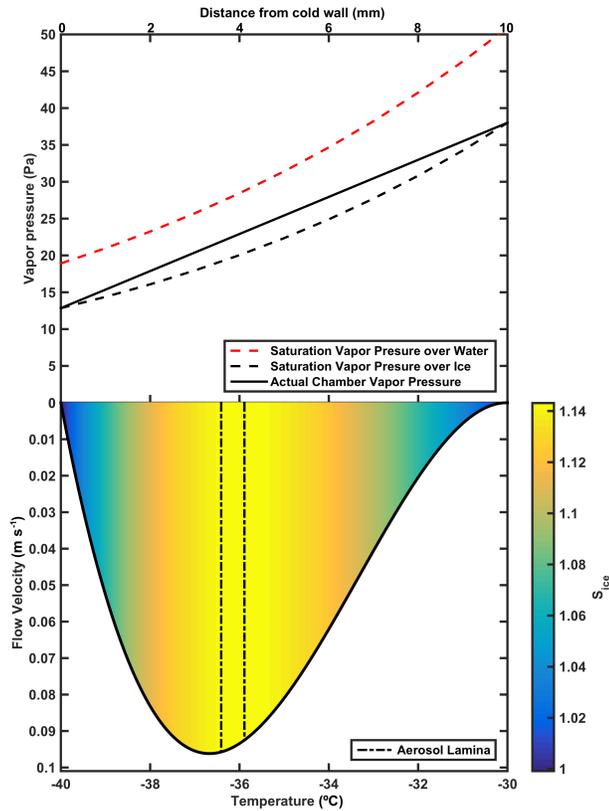
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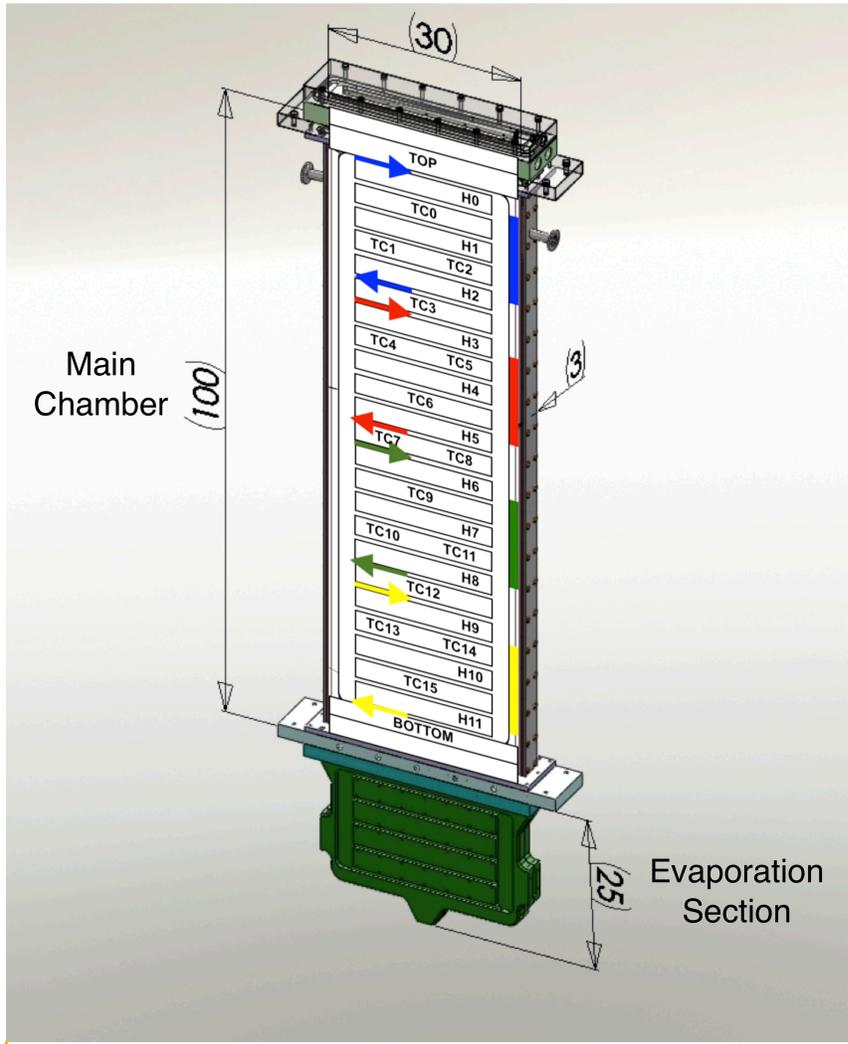
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2 Figure 1. Representation of idealized chamber thermodynamic and flow conditions with chamber  
3 width of 1 cm. The chamber cold wall (left) temperature is -40 °C and chamber warm wall (right)  
4 is -30 °C. The top half of the figure shows the saturation vapor pressures over ice (black dashed  
5 line), over water (red dashed line), and chamber vapor pressure (solid black line) for 10 Lpm  
6 sheath + 1 Lpm sample flow. Note the chamber is supersaturated everywhere with respect to ice  
7 but subsaturated with respect to water. The bottom half of the figure shows the flow velocity  
8 profile with the aerosol lamina given by the black dash-dotted lines. The colors show the  
9 horizontal variation in the ice saturation ratio across the width of the chamber. The asymmetry in  
10 the flow profile is a result of the buoyant displacement of the flow towards the cold wall.

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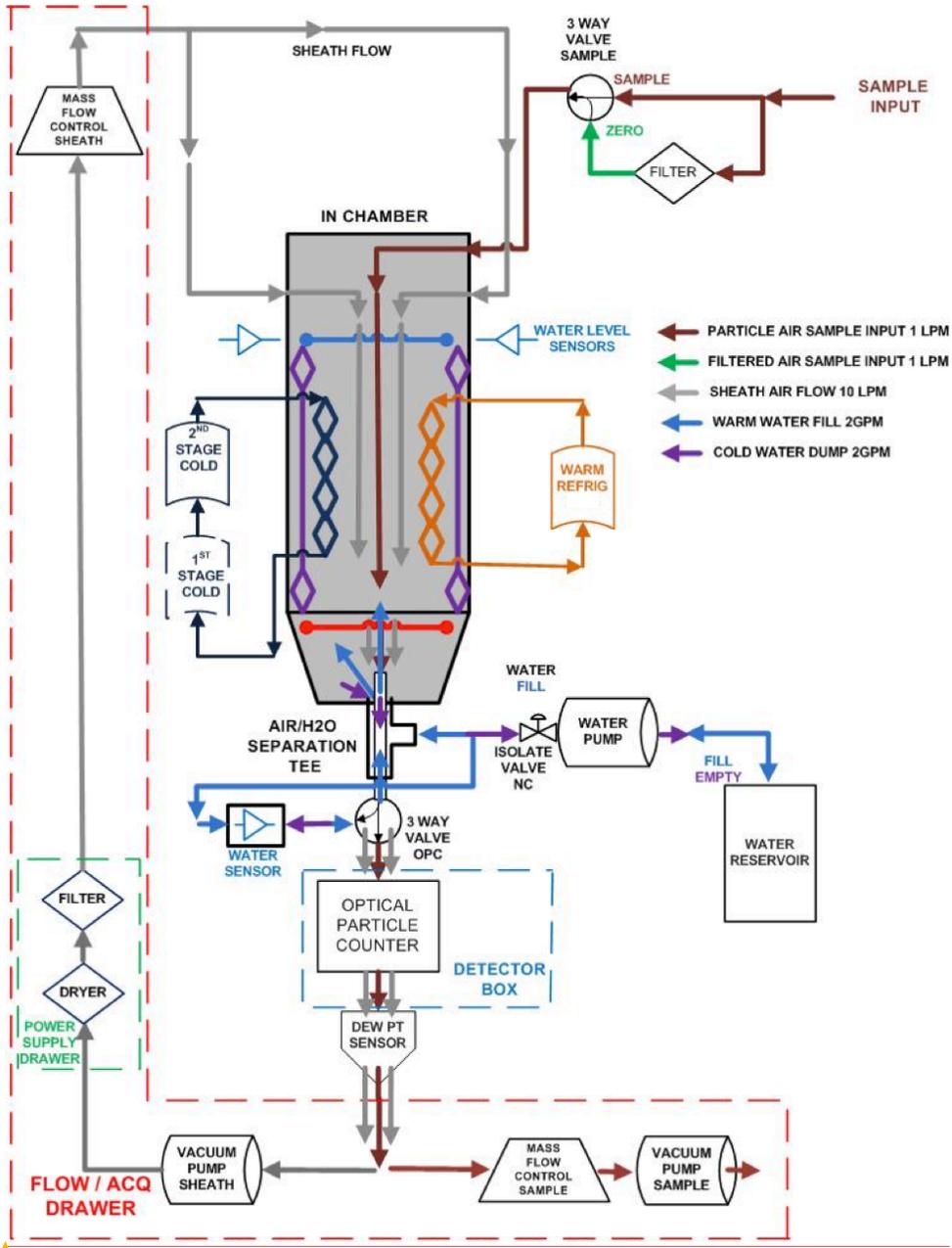


1  
2 Figure 2. SPIN chamber schematic showing dimensions of the chamber with overlaid  
3 thermocouple (TC0-TC15), heater (H0-H11), and refrigeration zone (colored arrows and blocks)  
4 locations on the main chamber. Heater strips span the length of the chamber wall at each  
5 indicated location. The different colored arrows represent different refrigerant paths depending  
6 on which of the four refrigeration solenoid valves are open. All dimensions are in cm.

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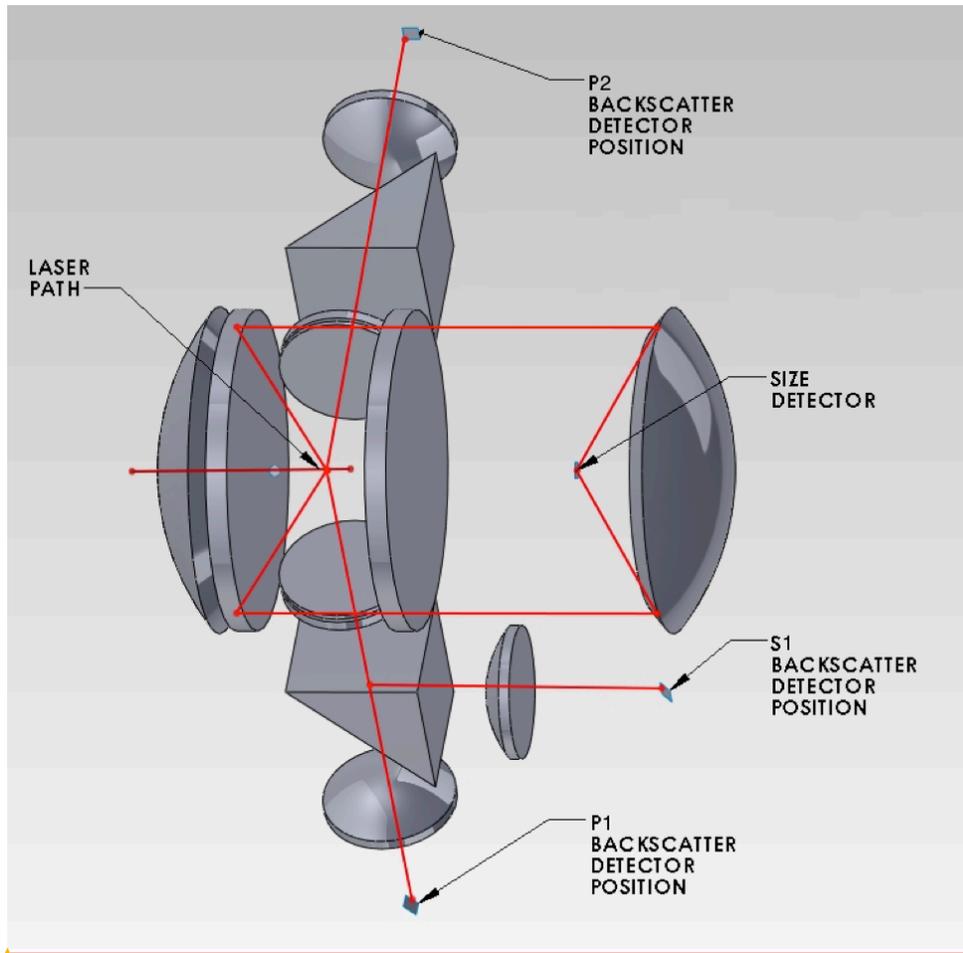
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2 Figure 3. Air and water flow diagram for the SPIN chamber.



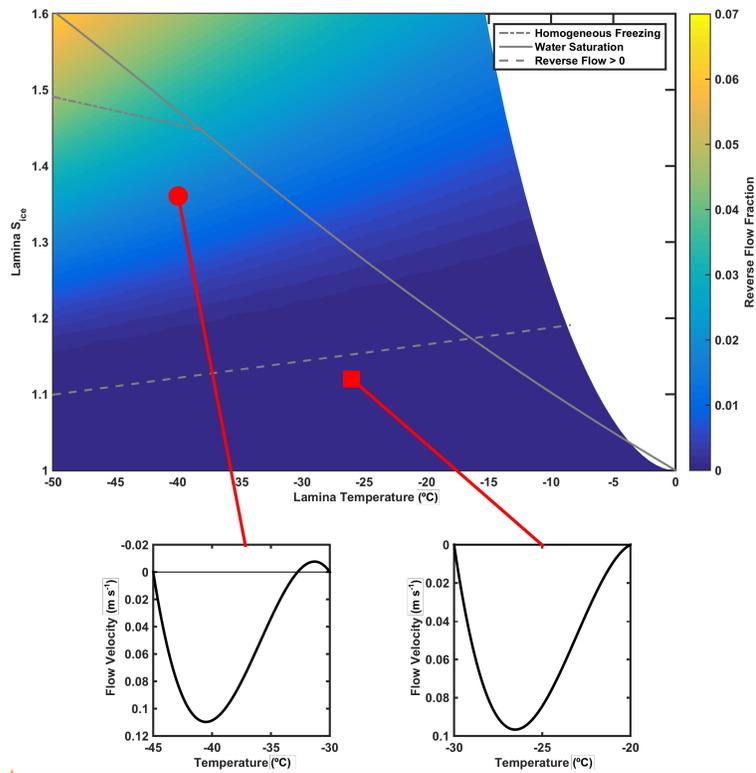
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 2 Figure 4. Schematic optical setup of the SPIN OPC. The laser light is shown entering the  
 3 sampling region, with side scatter rays travelling to the sizing detector, and the backscatter rays  
 4 travelling to the depolarization detectors (see Section 2.2 for details).

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 2 Figure 5. Phase diagram of ice saturation ratio vs. temperature showing the thermodynamic  
 3 conditions accessible by CFDC chambers. The color scale shows the ratio of (upward) reverse  
 4 flow to (downward) normal flow in the chamber (with 10 SLPM sheath flow, 1 SLPM sample  
 5 flow, and 1000 hPa chamber pressure) assuming a negligibly thick ice layer predicted by Rogers  
 6 (1988) with the dashed grey line marking the boundary between zero and nonzero flow reversal  
 7 (see Section 3.1 for details). The solid grey line is water saturation, and the grey dash-dot line  
 8 shows the onset of homogeneous freezing of solution droplets for  $J = 10^{11} \text{ cm}^{-3} \text{ s}^{-1}$  from Koop  
 9 et al. (2000). Two flow profiles are shown as insets: the coldest temperature in each corresponds

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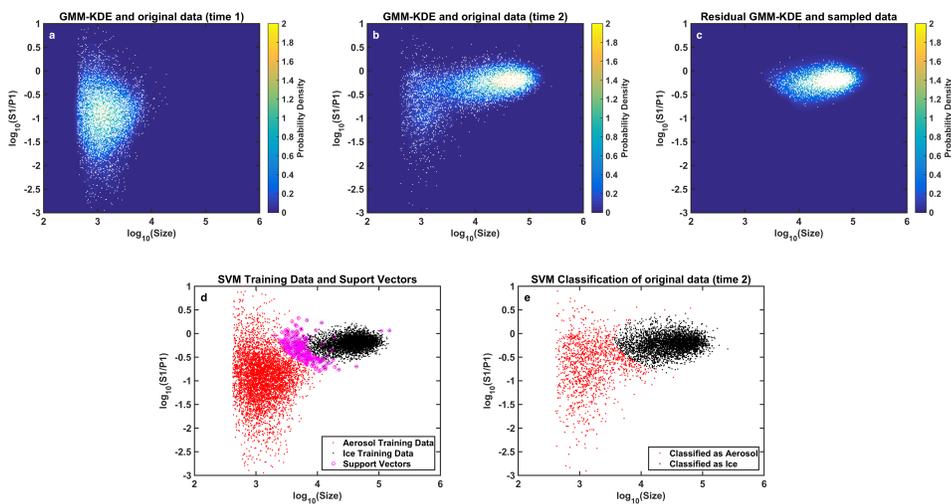
- 1 | to the cold wall temperature and the warmest to the warm wall temperature. Flow reversal occurs
- 2 | along the warm wall in one case (left, red circle) and not in the other (right, red square).
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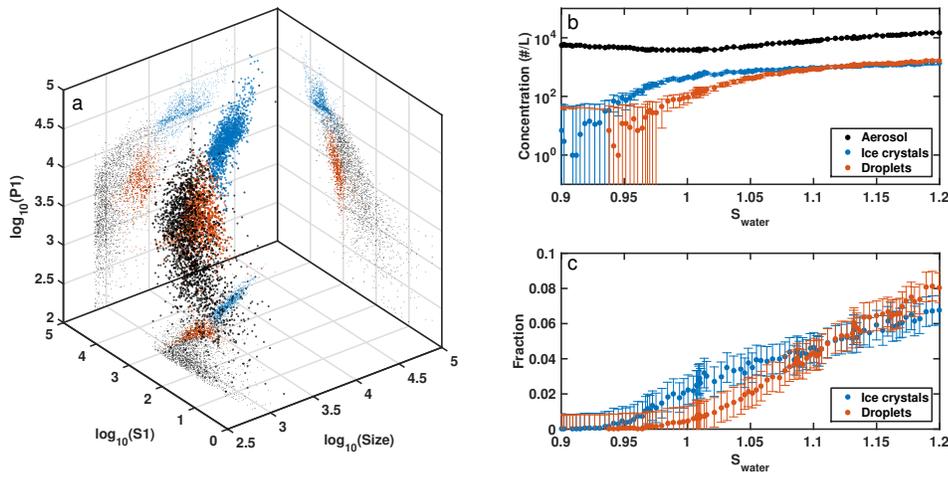
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2 **Figure 6.** Illustration of the supervised ML procedure used to classify OPC data in  $\log_{10}(S1/P1)$   
3 vs.  $\log_{10}(Size)$  parameter space. The units of Size, S1, and P1 are all intensity counts. (a) Data  
4 and GMM-KDE for an aerosol only time interval (time 1). (b) Data and GMM-KDE for an  
5 aerosol + ice time interval (time 2). (c) Residual GMM-KDE (time 2 minus time 1) and data  
6 from weighted sampling informed by the deconvolved PDF. (d) Aerosol data, ice data and  
7 support vectors for SVM training. Cross-validated classification accuracy is 99% in this  
8 example. (e) Data from time 2 classified by SVM as aerosol and ice. See Section 3.2 for details.

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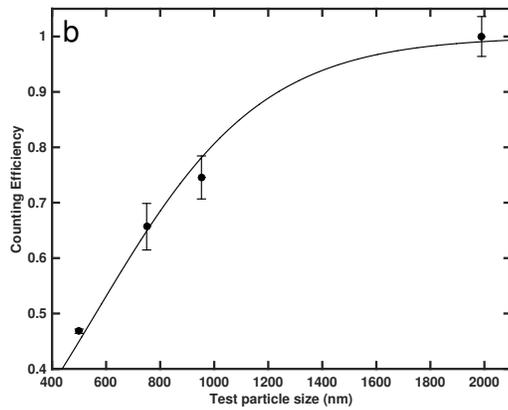
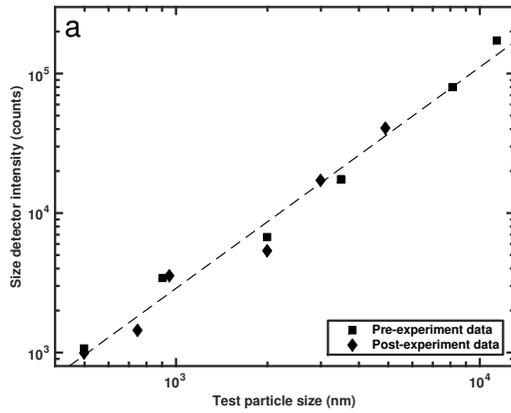
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 2 Figure 7. Application of 3-class supervised ML on an example activation experiment where the  
 3 chamber RH is increased. (a) Aerosol particles (black), ice crystals (blue), and water droplets  
 4 (red) shown in  $\log_{10}(P1)$  vs.  $\log_{10}(S1)$  vs.  $\log_{10}(Size)$  parameter space with classification  
 5 accuracy of 99%. 2D projections of the data are shown at axes limits with smaller markers. (b)  
 6 Concentration of aerosol particles, ice crystals, and water droplets as a function of chamber RH.  
 7 Error bars represent classification uncertainty. (c) Fraction of particles activated as ice crystals  
 8 and water droplets as a function of chamber RH. Error bars represent classification uncertainty.  
 9 Data points with lower error bars below zero indicate that the values are statistically  
 10 indistinguishable from zero.

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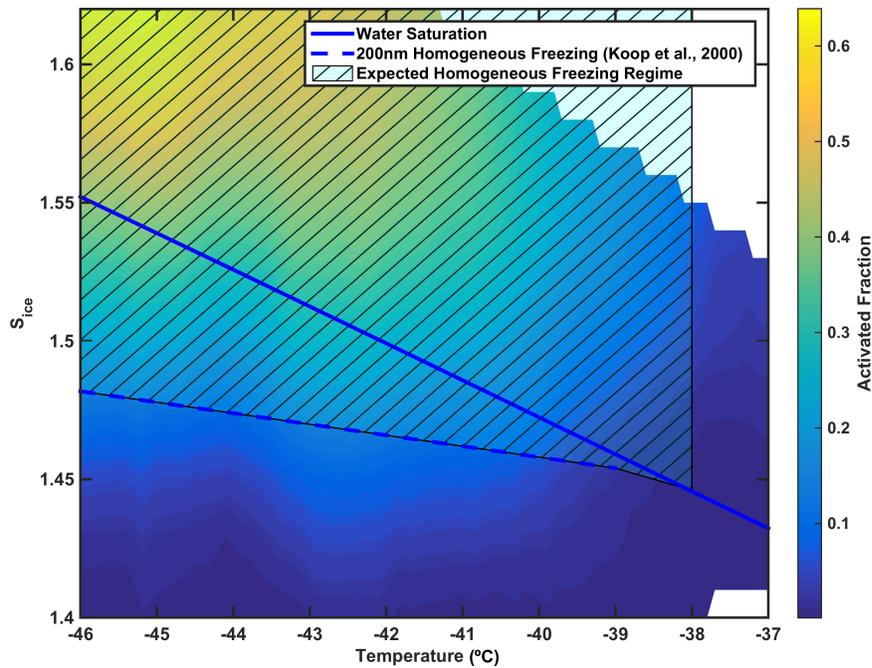


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2 Figure 8. Particle sizing and transmission efficiency of the SPIN detector. (a) Detector intensity  
3 counts versus test particle size before and after the freezing experiments in this study. Dashed  
4 line shows power law fit to the data. (b) Size-dependent counting efficiency of AS particles with  
5 sigmoid fit. Error bars show measurement uncertainty.

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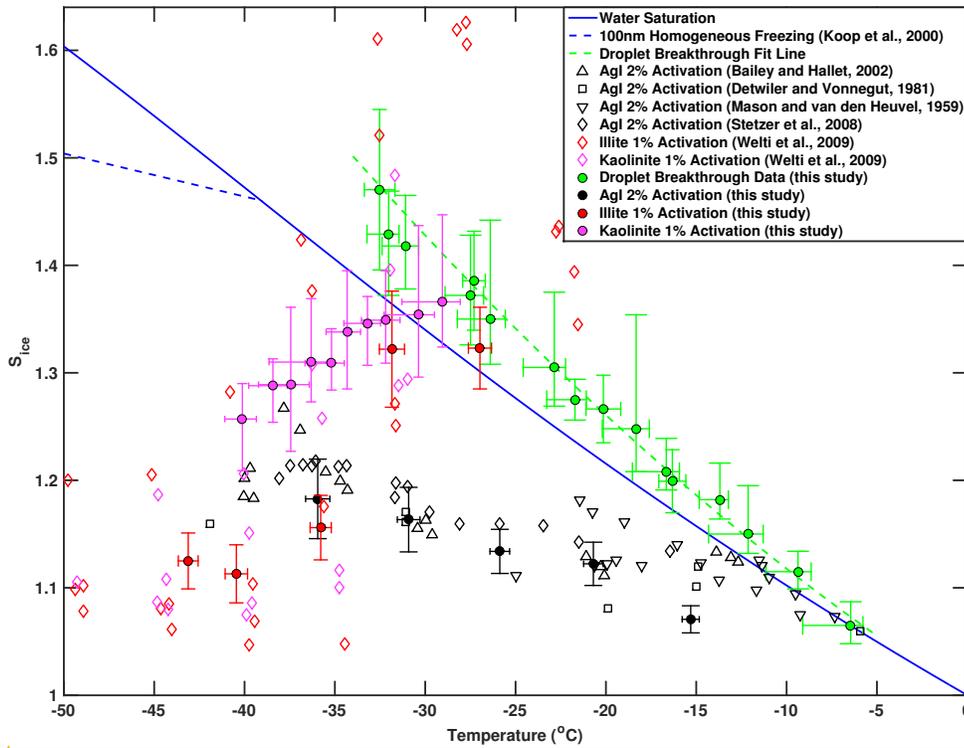
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 2 Figure 9. Experimental homogeneous freezing results. The hatched and shaded area shows where  
 3 homogeneous freezing of deliquesced haze droplets is expected to occur (below -38 °C and  
 4 above the  $J = 10^{11} \text{cm}^{-3} \text{s}^{-1}$  line from Koop et al., (2000)). The color contours show  
 5 interpolated activated fraction of ice crystals as a function of chamber conditions from 38  
 6 experiments (white areas are where no data are present). Typical aerosol number concentrations  
 7 for such experiments are 100s of particles  $\text{cm}^{-3}$ . Typical uncertainties at one standard deviation  
 8 for temperature are  $\pm 1$  °C, for supersaturation are  $\pm 5$  %, and for activated fraction are  $< 1$  %.

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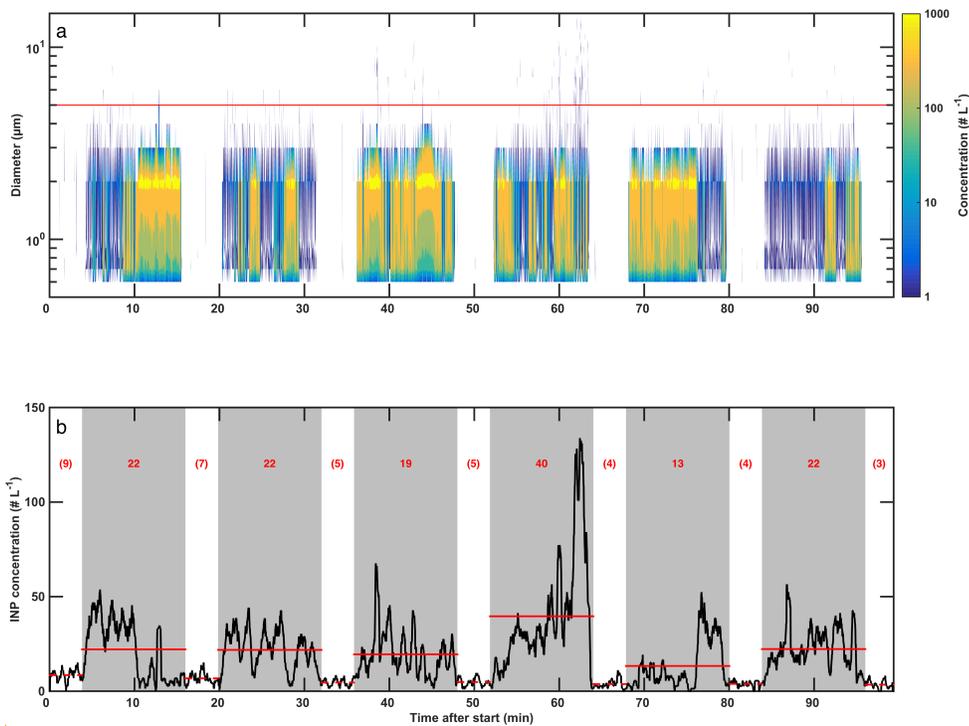
Figure 10. Experimental heterogeneous ice nucleation results and comparison to literature with polydisperse NX illite, AgI, and 500nm kaolinite particles. AS droplet breakthrough data and corresponding quadratic fit line are shown in green. Error bars represent uncertainty in lamina temperature and supersaturation conditions. The homogenous freezing line for solution droplets for  $J = 10^{12} \text{ cm}^{-3} \text{ s}^{-1}$  from Koop et al., (2000) is also shown for reference.

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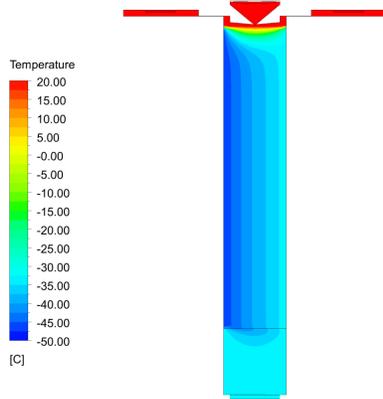
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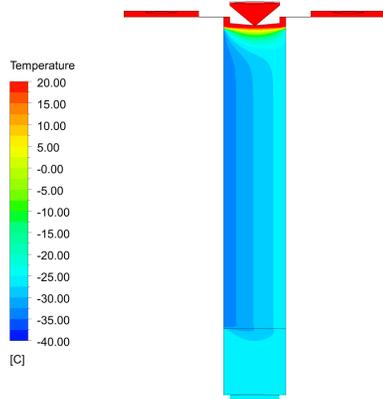
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2 Figure 11. Example field measurement at  $\sim -30\text{ }^\circ\text{C}$  and  $S_{liq} = 1.05$  (reported lamina temperature of  
 3  $-30.7\text{ }^\circ\text{C} \pm 0.2\text{ }^\circ\text{C}$  and lamina  $S_{liq}$  of  $1.05 \pm 0.01$  over the measurement period). (a) The OPC size  
 4 histogram time series for this measurement. The red horizontal line shows the  $5\text{ }\mu\text{m}$  size  
 5 threshold for ice. (b) INP concentration time series using the  $5\text{ }\mu\text{m}$  size threshold. Shaded areas  
 6 show measurement periods and un-shaded areas show filter periods. Solid red horizontal lines  
 7 show average INP concentrations from measurement periods (with the value specified above).  
 8 Dashed red horizontal lines show average frost concentrations during filter periods (with the  
 9 value specified above in parentheses).

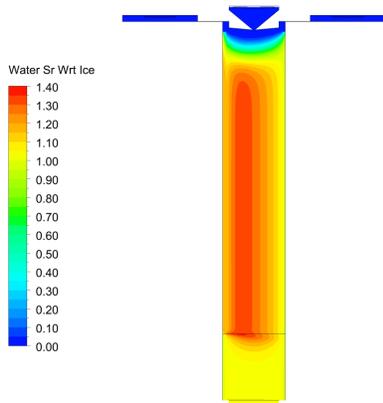
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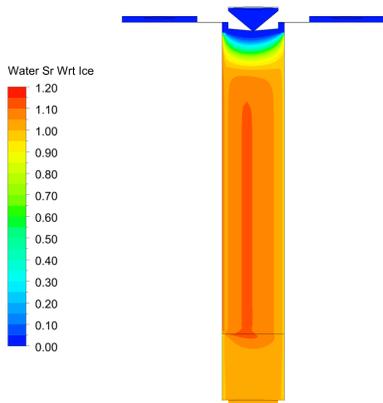
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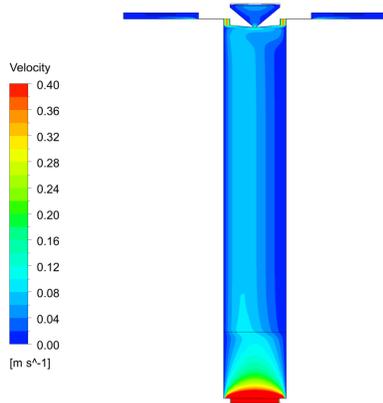
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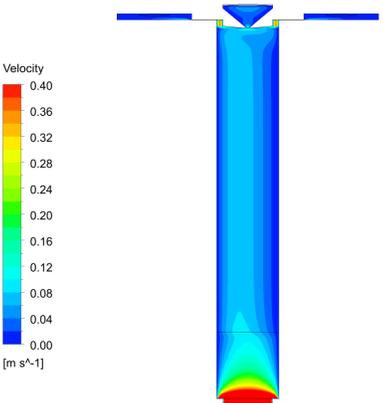
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1 | [Figure 12](#). Fluent simulation results for two sets of chamber conditions in a cross-sectional view.  
2 | Left column shows results for nominal lamina temperature of -40\_°C and lamina S<sub>ice</sub> at 1.3. (a)  
3 | Temperature, (°C), (c) S<sub>ice</sub>, and (e) flow velocity, (m s<sup>-1</sup>). Right column shows results for nominal  
4 | lamina temperature of -30\_°C and lamina S<sub>ice</sub> at 1.1. (b) Temperature, (°C), (d) S<sub>ice</sub>, and (f) flow  
5 | velocity, (m s<sup>-1</sup>).

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