

A Phase Separation Inlet for Droplets, Ice Residuals, and Interstitial Aerosol Particles

Libby Koolik^{1,2,3}, Michael Roesch^{1,4}, [Carmen Dameto de Espana⁵](#), [Christopher Nathan Rapp⁵](#), Lesly J. Franco Deloya¹, Chuanyang Shen^{1,5,6}, A. Gannet [Hallar^{6,7}](#) [Hallar^{7,8}](#), Ian B. [McCubbin⁷](#) [McCubbin⁸](#), Daniel J. Cziczo^{1,2,8,5}

¹Department of Earth, Atmospheric, and Planetary Sciences, Massachusetts Institute of Technology, Cambridge, MA 02139, USA

²Department of Civil and Environmental Engineering, Massachusetts Institute of Technology, Cambridge, MA 02139, USA

³[Ramboll, San Francisco, CA 94111 USA](#)

³[Department of Civil and Environmental Engineering, University of California Berkeley, Berkeley, CA 94720, USA](#)

⁴Department of Environmental Systems Science, Swiss Federal Institute of Technology, ETH Zurich, Zurich, 8092, Switzerland

⁵Department of [Earth, Atmospheric and Planetary Sciences, Purdue University, West Lafayette, IN 47907, USA](#)

⁶Department of Atmospheric and Oceanic Sciences, Peking University, [Beijing](#) 100871, [Beijing](#), China

⁶[Department](#) ⁷[Department](#) of Atmospheric Sciences, University of Utah, Salt Lake City, UT 84112, [USA](#)

⁷[Storm](#) ⁸[Storm](#) Peak Laboratory, Desert Research Institute, Steamboat Springs, CO 80488, [USA](#)

⁸[Department of Earth, Atmospheric, and Planetary Sciences, Purdue University, West Lafayette, IN 47907, USA](#)

Correspondence to: Daniel [J. Cziczo](#) (djcziczo@purdue.edu)

Abstract. A new inlet for studying the aerosol particles and hydrometeor residuals that compose mixed-phase clouds – the phaSe sePARation Inlet for Droplets icE residuals and inteRstitial aerosol particles (SPIDER) – is described here. SPIDER combines a Large-Pumped Counterflow Virtual Impactor (L-PCVI), a flow tube evaporation chamber, and a Pumped Counterflow Virtual Impactor (PCVI) to separate droplets, ice crystals, ($\sim 3 - 25 \mu\text{m}$), and interstitial aerosol particles for simultaneous sampling. Laboratory verification tests of each individual component and the composite SPIDER system were conducted. ~~SPIDER was then deployed at Storm Peak Laboratory (SPL), a mountain-top research facility at 3210m a.s.l. in Transmission efficiency, evaporation, and ice crystals survival were determined to show the Rocky Mountains, for a three-week field campaign. SPIDER performance as a field instrument is presented with data that demonstrates its capability of separating the system. The experiments show the SPIDER system can separate distinct cloud elements and interstitial aerosol particles. Possible design improvements~~ particles for subsequent analysis. As a field instrument, SPIDER will help explore the properties of SPIDER are also suggested different cloud elements and interstitial aerosol particles in mixed-phase clouds.

1 Introduction

A mixed-phase cloud has both liquid and ice phases (Korolev et al., 2003; Shupe et al., 2006) with variable number density and mass ratio of liquid to ice particles. Mixed-phase clouds are important factors in aviation and climate (Lohmann, 2017; McCoy et al., 2016; Shupe et al., 2008). In aviation, supercooled droplets can cause aircraft icing and engine power loss (Strapp et al., 2016). In climate, the role clouds play in the earth's radiative budget remains uncertain (Boucher et al., 2013; McCoy et al., 2016). As aerosol particle concentration increases in the atmosphere, liquid clouds may have decreased droplet size and increased spatial and temporal extent (Boucher et al., 2013). This will change the radiative forcing at the top of the atmosphere (cloud albedo effect) as well as the lifetime of a cloud (lifetime effect) (Lohmann and Hoose, 2009; Storelvmo et al., 2008). Mixed-phase clouds are particularly complicated because the partitioning of phases is critical in assessing these effects (Atkinson et al., 2013; Hirst et al., 2001; Korolev et al., 2003; Korolev et al., 2017; Shupe et al., 2006; Tan and Storelvmo, 2019). At present, these effects are difficult to parameterize in models due to a lack of observational data on formation, properties, and phase partitioning (Kamphus et al., 2010; Shupe et al., 2006). This has resulted in a global effort to study these clouds (Abel et al., 2014; Davis et al., 2007a; Hiranuma et al., 2016; Kupiszewski et al., 2015; Lohmann, 2017; Lowenthal

[et al., 2019](#); [Mertes et al., 2007](#); [Patade et al., 2016](#); [Ramelli et al., 2021](#); [Ruiz-Donoso et al., 2020](#); [Schmidt et al., 2017](#)).

The microphysical formation processes of water and ice clouds are generally understood. Droplets form when a critical [saturation supersaturation](#), described theoretically by the Köhler equation, is exceeded. At this [saturation supersaturation](#), aqueous droplets are the favored state and particles that activate are termed cloud condensation nuclei (CCN) ([Lohmann Pruppacher and Hoose, 2009](#); [Wang et al., 2012](#); [Klett, 1997](#)). Ice nucleation is more complex. Ice can form homogeneously, via spontaneous nucleation of ice in a solution droplet, at temperatures below -40°C ([Atkinson Heymsfield et al., 2013](#); [Kamphus 2017](#); [Koop et al., 2010](#); [Korolev et al., 2003](#); [Storelvmo et al., 2008](#); [Verheggen et al., 2007](#); [Wang et al., 2012](#); [2000](#)). At higher temperatures, ice forms heterogeneously through different pathways promoted by ice nucleating particles (INPs) ([Atkinson et al., 2013](#); [Kamphus et al., 2010](#); [Lohmann Hoose and Hoose, 2009](#); [Storelvmo et al., 2008](#); [Tsushima et al., 2006](#); [Verheggen et al., 2007](#); [Wang et al., Möhler, 2012](#)); [Kanji et al., 2017](#)). The specific properties that determine an effective INP remain poorly understood ([Shupe Kanji et al., 2008](#); [2017](#)).

There is also uncertainty regarding the existence of both liquid and solid water in the same environment. The accepted theory is the Wegener-Bergeron-Findeisen (WBF) process, whereby ice crystals, depending on the specific environmental temperature and humidity, grow at the expense of droplet evaporation due to thermodynamic instability ([Korolev, 2007](#); [Pruppacher and Klett, 1997](#)). Ice crystals have a lower saturation vapor pressure than water droplets below 0°C, so the presence of crystals will lower the [relative humidity water vapor content](#) and cause the droplets to shrink or, given sufficient time, evaporate completely ([Shupe et al., 2006](#); [Storelvmo et al., 2008](#); [Tsushima et al., 2006](#); [Verheggen et al., 2007](#)). This effect is often limited by the concentration of ice crystals in the cloud, since ice [crystals are](#) often [less than droplet number more scarce](#) in mixed-phase [clouds cloud than droplets](#) ([Verheggen et al., 2007](#)).

In-situ observations are required to understand the natural efficiency of INP and the microphysical processes of mixed-phase clouds. [Motivated by climate change, estimated to be warming approximately twice as fast as the global average \(Verlinde et al., 2007\), several](#) Several in-situ experiments [to characterize INP \(Hartmann et al., 2020; Irish et al., 2019; Si et al., 2019\)](#) have occurred in the Arctic, where there is a prevalence of mixed-phase stratiform clouds (e.g., 41% of the time in the study of [Shupe et al. \(2006\)](#))). Another common research location has been the Jungfraujoch; ([Hammer et al., 2018](#); [Lacher et al., 2021](#)), a mountain-top site in Switzerland, which has high cloud coverage (37% of the time) which are often mixed in phase ([Kamphus et al., 2010](#); [Verheggen et al., 2007](#)).

Two of the fundamental questions surrounding mixed-phase cloud formation are: (1) what is the ratio of ice to water in a cloud and (2) what are the aerosol particles that act as the CCN or INPs? Currently, there are a variety of instruments that can estimate ice or water content of a cloud ([Abel et al., 2014](#); [Davis et al., 2007a](#); [Davis et al., 2007b](#); [Korolev et al., 1998](#); [Strapp et al., 2016](#)), however, these instruments do not report information about the underlying INPs or CCN.

One technique capable of [capturing separating](#) ice and droplet residuals is the Counterflow Virtual Impactor (CVI) and its laboratory counterpart, the Pumped-Counterflow Virtual Impactor (PCVI). These methods use the [ideaproperty](#) that activated droplets or ice crystals are significantly larger than unactivated, [or interstitial](#), aerosol [partieleparticles](#) ([Slowik et al., 2011](#)). By separating based on mass, researchers can study differences between activated and interstitial aerosol particle. This technique has been used in a large number of studies since the mid-1980s when it was first described by [Ogren et al. \(1985\)](#).

The PCVI uses vacuum-pumped air to form a stagnation plane based on the design of the CVI ([Boulter et al., 2006](#); [Hiranuma et al., 2016](#)). A schematic of the PCVI used in this study is shown in Figure 1. A vacuum pump is used to provide the “pump flow” (PF), while pressurized air is introduced as an “add flow” (AF). AF has also been referred to as the “counterflow”; these terms are synonymous with AF used throughout this work. At the entrance of the PCVI is the “input flow” (IF) and at the terminus is the “sample flow” (SF) ([Boulter et al., 2006](#); [Friedman et al., 2013a](#); [2013](#)). The “effective counterflow” (ECF) is the difference of AF and SF and counteracts the IF to create a stagnation plane that particles of sufficient inertia must cross to be entrained in the SF. The 50% cut size or “D50” describes the [smallest number averaged](#) particle [with size of](#) sufficient inertia to be transmitted through the PCVI with 50% efficiency. The AF to IF ratio can be adjusted to change the D50, reducing or increasing the inertial barrier ([Kulkarni et al., 2011](#); [Slowik et al., 2011](#)).

~~Since the original characterizations, the PCVI has been considered by Boulter et al. (2006) and Kulkarni et al. (2011), the PCVI has been-. A treatment of inadvertent transmission of particles smaller than the D50 as well as droplet and ice crystal breakup was considered by Pekour and Cziczo (2011). The PCVI has been used in conjunction with cloud chambers for several cloud sampling studies (Baustian et al., 2012; Friedman et al., 2013a, 2013; Slowik et al., 2011). A recent advance is the ability to build improve performance and cut costs by building a PCVI using three-dimensional (3D) stereolithography (SLA) printing (Koolik, 2017). 3D printing allows rapid prototyping for complex devices (Jacobs, 1992), making the development of less expensive PCVIs with higher tolerance dimensions possible. 3D printing mitigates costs, decreases build time, reduces misalignment, and allows for rapid and inexpensive tests of potential structural improvements (Koolik, 2017).~~

2 Instrument Theory and Design

~~SPIDER~~The phase separation Inlet for Droplets ice residuals and interstitial aerosol particles (SPIDER) is a vertically-aligned inlet system with three distinct outlet channels for sampling interstitial (or ~~“unactivated”~~“unactivated”) aerosol particles, droplet residuals, and ice crystal residuals (~~Figure~~Fig. 2). It is comprised of three main components: a large PCVI (L-PCVI), droplet evaporation chamber, and PCVI. The droplet evaporation chamber is actively cooled and lined with a series of sensors to provide real-time information on the temperature profile.

A 3D printed L-PCVI was based on the design of the machined Ice-Selecting Pumped Counterflow Virtual Impactor (IS-PCVI) described by Hiranuma et al. (2016). ~~IS-PCVI described by Hiranuma et al. (2016). The flow rates used in this work and those of Hiranuma et al. (2016) are shown in Table 1. To ensure that large droplets and ice crystals were transmitted without breakup, the maximum Weber Number (N_{wc}) was calculated for these flows as 0.3 (see Fig. S1 in the Supplement), which is less than the limit of 10 suggest for the onset of hydrometeor breakup. Details and model results are provided in the Supplement. The L-PCVI was tested over a range of flow conditions that resulted in different D50s (see Fig. S2 in the Supplement). When operated with a 70 L min⁻¹ IF and 7 L min⁻¹ AF (AF-to-IF ratio of 0.1), the IS-PCVI has a D50 of ~9 μm (Hiranuma et al., 2016). By operating the L-PCVI with a 50 L min⁻¹ IF and 11.5 L min⁻¹ an AF (AF-to-IF flow-ratio of ~0.23), 25, for example, the D50 is 20-30 estimated to be in the 10 to 18 μm range. Because droplets and ice crystals are typically 10 μm or larger (Kleinman et al., 2012; Pruppacher and Klett, 1997; Rogers and Yau, 1989), only these activated droplets and ice crystals are large enough to exit greater than the L-PCVI lower cut size and smaller than the inlet cut size will enter SF. It should be noted that Interstitial aerosol and any droplets and/or ice crystals below the L-PCVI cut size will be stopped and transmitted into the PF.~~

~~For the majority of tests described here, the L-PCVI flow rates used in SPIDER were PF, AF, and SF at 55.0, 11.5, and 6.5 L min⁻¹, respectively; the PCVI PF, AF, and SF at 8.0, 2.5, and 1.0 L min⁻¹, respectively. To ensure that large droplets and ice crystals were transmit without breakup, the Weber Number, N_{wc} , was calculated using these flows. Details and model results are provided in the Supplement.~~

Ice crystals and supercooled droplets that pass through the L-PCVI enter the droplet evaporation chamber, which utilizes the WBF process. The chamber can be bare or ice coated and held at -16° C, where the difference in saturation vapor pressure between water and ice is at its maximum. ~~This allows ice crystals to maintain their initial size whereas droplets evaporate during passage. With an ice coating the chamber is, by definition, at ice saturation at -16° C (i.e., ice crystals were stable while droplets evaporated). A bare chamber is sub-saturated with respect to both ice and droplets but with a higher evaporation rate (i.e., more sub-saturated) for the latter. As discussed in the next section, at ice saturation and -16° C, droplets smaller than 25 μm in diameter can be fully evaporated in the chamber while ice crystals are able to maintain their initial size.~~

A PCVI is mounted below the droplet evaporation chamber. For this work a commercial machined PCVI (Model 8100, BMI Inc.), described by Boulter et al. (2006) and Kulkarni et al. (2011), and a 3D printed PCVI, described by Koolik (2017), were both used, and their performance compared. ~~In SPIDER, the PCVI was operated~~As with the L-PCVI, the flow conditions in the PCVI determine the size selection cutoff. For the majority of tests described here, the PCVI flow rates used in SPIDER were PF, AF, and SF at 8.0, 2.5, and 1.0 L min⁻¹, respectively. This results in a D50 of ~5.2 μm ~~in order which is used~~ to reject evaporated droplet residuals and any inadvertently transmitted interstitial aerosol into the PF but admit ice crystals into the SF. Additional flow scenarios have been tested

summarized in Boulter et al. (2006), Kulkarni et al. (2011), and Koolik (2017). The aforementioned maximum N_{we} calculations suggest estimates for the PCVI are also less than 10 (see Fig. S3 in the Supplement), suggesting that droplets and ice crystals are not subject to breakup in either the L-PCVI or PCVI under these conditions.

As noted, the flows in the L-PCVI set the lower size limit of droplets and/or ice transmitted into the SF. When operating SPIDER in the field, it is expected that an additional inlet will be added at the top of the series to prevent inadvertent transmission of debris or snow. In the case of the studies detailed in the following sections, it is assumed that a facility inlet from which SPIDER will sample sets the upper droplet and ice crystal size range. For this work, we base flows on the Desert Research Institute's Storm Peak Laboratory (SPL) inlet, described by Petersen et al. (2019). The SPL facility inlet has an upper D50 of 13 μm aerodynamic diameter with a broad cut size: 75% of particles over 8 μm and 25% of particles over 15 μm diameter are transmitted. Note that this transmission considers spherical particles of unit density (i.e., equivalent to water droplets); ice crystals of larger physical size are transmitted due to their lower density (Petersen et al., 2019).

Using this methodology, SPIDER offers simultaneous sampling channels for interstitial aerosol ~~partiele~~particles, droplet residuals, and ice crystal residuals via the PF of the L-PCVI, PF of the PCVI, and the SF of the PCVI, respectively.

3 Methodology

3.1 3D Component Fabrication

SPIDER incorporates a number of parts that were 3D printed. SLA printing involves the photopolymerization of a liquid resin by a laser in a layer-by-layer process. This printing method was chosen for resolution, surface quality, low shrinkage, and low distortion (Bartolo, 2011; Bhushan and Caspers, 2017; Hagiwara, 2004). There are drawbacks and common errors that occur with SLA, including overcuring (solidified material fails to bind with the layer below it) and time-intensive post-processing (Jacobs, 1992; Wong and Hernandez, 2012) ~~and~~; parts with these errors were rejected before use. The printer used for SPIDER components (Form 2, Formlabs Inc.) uses a 405 nm ~~UV~~ laser to cure specific coordinates in a resin bath to create the part in a layered structure (3D Printing with Desktop Stereolithography). Parts for SPIDER were printed from 'tough resin' (FLTOTL03, Formlabs Inc.) with a 100- μm layer resolution.

~~The following procedure was utilized for creating 3D printed parts: A part was virtually designed using Solidworks. The Solidworks assembly tool was used to verify that multiple parts would fit together as designed. Parts were then exported as a stereolithography file (.STL) and uploaded to the 3D printing software (Preform, Formlabs Inc.) where the part was oriented and printing support structures were added. The contact points attached to the part were small (0.5 mm) to ease removal after printing. From Preform, the part was uploaded onto the printer. After prints were completed, the parts were post-processed following the procedure described by Roesch et al. (2017). Briefly, they were shaken in a bath of isopropyl alcohol (IPA) and then left for approximately 20 minutes to remove uncured resin. Second, parts are exposed to UV light inside a curing box containing the same wavelength as the printer's laser. Third, the support structures were removed and the surface wet-sanded to the final finish. (2017) and Rösch and Cziczo (2020).~~

3.2 Instrumentation

~~Optical~~Two particle counters (OPC-N2, Alphasense) with sizing ranges from 0.4 to 17 μm ~~instruments~~ were installed on the PF of the L-PCVI ~~used for SPIDER performance testing, calibration and PCVI (i.e., the interstitial aerosol and droplet residual channels).~~ A higher resolution optical particle sizer ~~data acquisition: An Optical Particle Sizer (OPS, TSI, Model 3330, TSI Inc.) with an optical sizing range from 0.3 to 10 μm was installed on the PCVI SF (i.e., the ice residual channel). Both instrument types feature a monochromatic light source and a size resolution over 16 bins. The OPC has with a total flow rate of 1.0 L min⁻¹ and an Aerodynamic Particle Sizer (APS, TSI, Model 3321) with an optical sizing range from 0.3 to 20 μm with a total flow rate of 1.2 L min⁻¹ whereas the OPS draws 45.0 L min⁻¹.~~

The evaporation chamber was cooled using a low-temperature cooling bath (Proline RP 1290, Lauda-Koenigshofen). Additional specifications and the operating procedure for SPIDER are included in the Supplement.

4 Verification/Validation Experiments

In order to validate the SPIDER method, individual components were tested in the laboratory to determine performance and/or for comparison to previous studies. Droplets or ice crystals were then sent through the complete SPIDER setup to determine transmission, evaporation/sublimation, and rejection efficiency of each phase. ~~It was not possible to form a mixed-phase cloud within the laboratory; a field experiment, detailed in the next section, was used as a final validation step.~~

4.1 L-PCVI

Hiranuma et al. (2016) described the expected working conditions of the IS-PCVI, the design basis for the L-PCVI, at different flow ratios. For this work, the performance of the L-PCVI was investigated using solid soda lime glass microspheres (Cospheric LLC, $\rho = 2.5 \text{ g cm}^{-3}$) with a diameter distribution of 1 to 50 μm . A size distribution of the soda lime glass microspheres is presented in Figure 3. Aerosol particles were generated with a multi-wrist shaker (Lab-Line Multi-wrist Shaker, Model 3589). A 500 mL Erlenmeyer flask containing the microspheres was attached to the shaker. Aerosol particles were suspended by injecting 5 L min^{-1} filtered air into the top of the flask and setting the wrist to $\sim 750 \text{ rpm}$. The resulting aerosol into one leg of a Y attached to the top of the L-PCVI. The other leg of the Y was attached to a HEPA filter to balance the flows in the system with particle free air. Additional flows through the L-PCVI were controlled with mass-flow controllers (Alicat Scientific, Inc.). The particle size distribution at the outlet of the L-PCVI was monitored with the APS. The SF of the L-PCVI was typically fixed at $\sim 6.5 \text{ L min}^{-1}$ with the APS sampling at 1 L min^{-1} and the remaining $\sim 5.5 \text{ L min}^{-1}$ exhausted through a filter.

The L-PCVI transmission efficiency, defined as the ratio of particle number concentration of the SF to particle number concentration of the IF as a function of the aerodynamic diameter and the product of the enhancement factor (EF). The EF is defined as the ratio of the IF to the SF, the enhancement of particle concentration inherent to a CVI. The transmission efficiency of the L-PCVI was determined by comparing the aerosol size distributions with to without the PF and AF operating. Figure 4 shows the transmission efficiency corresponding to the flow configuration with a constant PF of 45 L min^{-1} and AF of 6.6 L min^{-1} as a function of the aerodynamic diameter (i.e., Scenario C in Table 1). The D50 was determined by fitting a sigmoid curve and was calculated under these flows to be $12.5 \pm 0.1 \mu\text{m}$. The D50 from experiments using different flows configuration can be characterized as a function of the AF to IF ratio analogous to Hiranuma et al. (2016). Hiranuma et al. (2016) described the expected working conditions of the L-PCVI at different flow ratios. The operating ratio of AF to IF in L min^{-1} under laboratory conditions in SPIDER is approximately 0.23, so the D50 of the L-PCVI is expected to be 20–30 μm . The OPC and OPS detect particles up to 17 and 10 μm , respectively, so a characterization of the L-PCVI in the style of the Boulter et al. (2006) characterization of the PCVI was not possible. Instead, measurements over a range of expected D50 was used to show that the L-PCVI rejects particles as described in Hiranuma et al. This is represented in Figure S2 in the Supplement and correlates linearly to the AF to IF ratio (correlation coefficient= 0.85). (2016).

To find the lower bound of the D50, 10 μm polystyrene latex (PSL) spheres were atomized (Roesch and Cziezo, 2020). Particles were first introduced to the L-PCVI with PF and AF off to provide the initial particle size distribution. The flows on the L-PCVI were then turned on. The L-PCVI flow values are given in Table 1. Samples were taken at 1 Hz resolution in both the PF and SF of the L-PCVI.

Figure 3a and 3b show the number concentration obtained in the PF and SF, respectively. The location of the measurement is represented by the circle on the inset schematic. The L-PCVI flows are initially off (represented by “X”), and particles are only measured in the SF. The AF is turned on (represented by the directional arrow) approximately 1,900 seconds into the measurement and the PF 100 seconds later. When the L-PCVI flows are off, a detectable concentration of 10 μm particles is present in the SF; when the L-PCVI flows are on, these particles appear in the PF (i.e., they are unable to cross the L-PCVI stagnation plane and are rejected). We conclude that the cut size of the L-PCVI is larger than 10 μm with the difference in concentration due to the dilution from adding filtered air as AF.

The second experiment replicated performance measurements by Hiranuma et al. (2016) using an IF of 50 and an AF of 7 L min^{-1} (AF/IF ratio 0.14) with a sample flow of 2 L min^{-1} . Hiranuma et al. (2016) report a D50 of 22 μm

for these conditions. To create particles larger than this expected D50, droplets were generated using a commercial droplet generator (DG) (MD-K-130, Microdrop Technologies) with a 50 μm nozzle. The droplet size leaving the nozzle was verified to be 50 μm diameter using a high-resolution camera and image-analysis software. For laboratory conditions and a transit distance of 5 mm from DG to L-PCVI droplets partially evaporate and are calculated to be 40 μm on entrance to the IF; a model of droplet evaporation was considered based on the equations in Lohmann et al. (2016). Details of the model and the results are included in the Supplement. Droplets were created from an ammonium sulfate (AS) solution of 5 g L^{-1} . The residuals size of 50 μm droplets of this concentration after complete evaporation is calculated to be 3.7 μm .

Measurement of droplet transmission and residual size were then performed. The stream of droplets was introduced through the L-PCVI without PF or AF on (Figure 4a). The OPS was connected to the L-PCVI SF with a flow splitter, such that 1 L min^{-1} entered the OPS and 1 L min^{-1} bypass flow to a mass flow controller (MFC) to total the sample flow of 2 L min^{-1} . Particles were detected in the OPS 4.7 μm size bin and extend to the maximum size (10 μm), indicative of transmission and partial evaporation. The L-PCVI flows were turned on (Figure 4b). Particles from 4.7–10 μm continued to be observed, suggesting the droplets were transmitted through the L-PCVI and partially evaporating. We conclude that the particles detected in the SF (Figure 4b) are the result of droplets large enough to pass through the stagnation plane. These experiments bracket the D50 of the L-PCVI between 10 and 40 μm , consistent with 22 μm for Hiranuma et al. (2016). In the Supplement, the results from this work are superimposed on the data from Hiranuma et al. (2016).

4.2 Droplet Evaporation Chamber

4.2.1 Droplet Evaporation Experiments

Droplet evaporation was considered based on the aforementioned equations in Lohmann et al. (2016). From the model (see Supplement Fig. S4 in the Supplement), it is expected that droplets entering the chamber 12.5 μm in diameter or smaller will fully evaporate before reaching the PCVI for chamber saturation supersaturation 0.9 and lower, during the approximately 25 second residence time. Droplets between 12.5 - 25 μm diameter will evaporate if the chamber saturation supersaturation is below 0.5. Droplets larger than 25 μm are expected to partially but not fully evaporate within the chamber; this sets an effective upper limit for SPIDER.

In practice, the relative humidity (RH) supersaturation of the chamber and the L-PCVI AF determine the saturation supersaturation droplets experience. A static SPIDER at -16°C with ice coated walls has a relative humidity supersaturation with respect to ice that is, by definition saturation 1, but $\sim 0.85\%$ RH with respect to liquid water (i.e., the model suggests droplets somewhat larger than 12.5 μm in diameter will fully evaporate). Hygrometer measurements show that dry air from the L-PCVI AF reduces this to $\sim 0.75\%$ RH. At this lower RH supersaturation, the model suggests that droplets 20 μm diameter and smaller fully evaporated in the chamber.

4.2.2 Sustaining Ice Crystals

Ice crystals were passed through the chamber to validate transmission. The L-PCVI was removed and 50 μm diameter droplets generated from a 0.6 g L^{-1} AS solution were introduced directly at the top of the chamber. The chiller was set to -65°C , below the homogeneous freezing threshold of -38°C (Koop et al., 2000). This initiated rapid homogeneous freezing, validated visually. Ice crystals passed down the chamber to the PCVI which was run with flows to achieve a D50 of 2.7–3.8 μm : PF, AF, and SF set to 8.0, 2.5, and 1 LPM, respectively (Figure 5). The resulting particle concentration in the SF of the PCVI, the ice crystal residual flow, is equivalent to the initial concentration of (frozen) droplets. At the initial solution concentration this corresponds to a residual size of AS of 1.4 μm diameter. The residual peak, ~ 3.8 μm diameter (Figure 5), suggests ice crystals pass through the stagnation plane of the PCVI but do not sublimate/evaporate fully at the OPS.

4.3 PCVI

A performance The evaporation as a function of the residence time was tested with aqueous ammonium sulfate droplets. Droplets were generated with a bubble burst generator (“bubbler”) containing 0.1 g mL^{-1} ammonium sulfate. An aerosol flow of 0.6 L min^{-1} was diluted with 0.4 L min^{-1} humidified filtered air to obtain a 1 L min^{-1} SF. This flow was introduced via a 0.5 cm diameter injector at different locations in the chamber to vary the residence

time and the size distribution was recorded with the OPS. The size distribution corresponding to each residence time is shown in Figure 5. The size distribution of the shortest residence time (0.2 s) reflects the initial droplet size distribution. Due to the OPS size range only particles smaller than 10 μm diameter can be directly measured although the distribution reflects decreasing particle numbers larger than this size; a linear extrapolation of the size distribution suggests that particles up to 20 μm diameter may have been present in the flow. Longer residence times indicate droplet evaporation with initially larger droplets becoming smaller and eventually falling below the instrumental size range. In the longest residence time case (28.5 s) droplets have evaporated below $\sim 2.5 \mu\text{m}$ diameter

4.2.2 Ice Crystal Experiments

Ice crystals were passed through the evaporation chamber to validate transmission. Droplets were created using a bubbler containing 0.1 g mL^{-1} ammonium sulfate. A 1 L min^{-1} droplet flow was isokinetically injected at the center of a sheath air flow (5 L min^{-1}). The sheath air flow was cooled using liquid nitrogen introduced via a two concentric tube inlet. The low temperature caused homogeneous ice nucleation of the droplets. Based on observations with the OPS, a broad distribution of ice crystals was formed with a mode size centered between 5 and 6 μm diameter. The evaporation chamber and the outlet were cooled using the low temperature cooling bath set at $-16 \text{ }^\circ\text{C}$. A flow of 6 L min^{-1} , consistent with the operational SPIDER flow (see Section 4.5), was injected at the top of the evaporation chamber. The size distribution at the bottom of evaporation chamber was determined with the OPS; the resulting size distribution (i.e. the size distribution of the sustained ice crystals) is represented in Figure 6. No change in ice mode size or number concentration was observed. This was consistent with the evaporation calculations and indicated minimal or no sublimation of ice crystals under the SPIDER operating conditions.

4.3 PCVI

A validation of a 3D Printed PCVI was performed by Koolik (2017) following Boulter et al. (2006) and Kulkarni et al. (2011). Using a bubble burst generator (“bubbler”) containing a solution of 0.1 g L^{-1} ammonium sulfate, measurements of D50 under various flow scenarios were performed and compared with the OPS. The size distribution of particles generated with the bubbler is represented in the Supplement (Fig. S5). With a constant AF of $2.5 \text{ L PML min}^{-1}$ and SF of $1.0 \text{ L PML min}^{-1}$, the 3D printed PCVI had a working range of IF from 3.9 to $9.2 \text{ L PML min}^{-1}$. The results of the comparison between the 3D printed PCVI and the commercial machined PCVI within this range are shown in Figure 67.

Using the aforementioned SPIDER PCVI flows mentioned in Section 2, a PCVI D50 of $\sim 5 \mu\text{m}$ diameter is expected from the literature (Boulter et al., 2006; Kulkarni et al., 2011). To validate the D50, the SF from the PCVI was compared to the initial size distribution (i.e., for each size bin of OPS data). The transmission efficiency of each bin size was calculated and the data fit with a sigmoid; the D50 was defined as the particle diameter size that corresponded to 50% of the maximum transmission efficiency on the sigmoid. An example of data using the SPIDER operational flows in the last paragraph and the sigmoidal fit corresponding to a D50 of $5.1 \pm 0.1 \mu\text{m}$ is shown in Figure 78. The operational flows used in SPIDER are summarized in Section 2. Additional verification experiments are summarized in the Supplement (Fig. S6).

5 Storm Peak Laboratory Field Campaign

5.1 Storm Peak Laboratory Field Site

The Desert Research Institute’s Storm Peak Laboratory (SPL) is located near Steamboat Springs in north-western Colorado at 40.45°N , 106.74°W , 3210 m above sea level (Borys and Wetzol, 1997). During the winter, SPL is often enshrouded in clouds that contain supercooled liquid and ice crystals, making it an ideal site for sampling cloud condensation nuclei (CCN) and ice nucleating particles (INP) (Friedman et al., 2013b; DeMott et al., 2010). SPL contains a measurement suite for aerosol particles, cloud properties, and meteorological instruments that provide additional information about ambient conditions. This made SPL an ideal site for deploying SPIDER to test its efficacy for sampling mixed phase clouds.

5.2 Field Measurements

SPIDER was deployed at SPL for three weeks during January 2019. This deployment allowed for performance data of the inlet within mixed-phase clouds to complement the individual component, droplet and ice crystal testing conducted in the laboratory and detailed in the preceding sections. Data from January 20th, 21st and 23rd are presented here to show performance in clear, transition and cloudy conditions. SPIDER was connected to an ambient inlet to access aerosol and cloud elements. The inlet has a cut size of $13\mu\text{m}$ for at wind speeds of 0.5 m s^{-1} (Petersen et al., 2019) and the L-PCVI of SPIDER was set to a cut size of $8\mu\text{m}$; these cut points were set to allow transmission of interstitial aerosol particles, cloud droplets, and ice crystals but reduce large wind-blown ice fragments, discussed in detail in the following paragraphs.

As previously described, two OPCs were attached to SPIDER to measure interstitial aerosol particles and cloud droplet residuals. An OPC on the PF from the L-PCVI is termed the interstitial aerosol channel. An OPC on the PF of the PCVI is termed the cloud droplet residual channel. Both of these OPCs suffered intermittent failures during the field study. The scattering signal from a Droplet Measurement Technologies (DMT) Single Particle Soot Photometer Extended Range (SP2-XR) was used to measure ice crystal residuals from the SF of the PCVI (termed the ice crystal residual channel). The SP2-XR allowed for higher sampling frequency and sizing resolution (100 nm – 540 nm) than the OPCs used in the laboratory but with a smaller size limit. Analysis concentrates on SP2-XR data which was available for the entirety of the study.

Temperature during the study ranged from -5 to $-20\text{ }^\circ\text{C}$ and wind speed ranged from 0 – 10 m/s (see Supplement). Background aerosol particle conditions were typically low, with occasional spikes in particle concentrations attributed to new particle formation and local sources (see Supplement). Aerosol spikes were minimal during the three-day period considered here, during which there were two clear periods and two mixed-phase cloudy periods. Data from a cloud imaging probe (CIP) at SPL (Figure 8) and visual verification were used to differentiate clear and cloudy periods.

One potential sampling concern was that wind-blown snow or ice fragments could enter the inlet and lead to incorrect ice crystal residual counts in SPIDER. Our data showed there was minimal / no association between wind speed, which was relatively low during this period, and ice crystal concentrations in clear conditions (see Supplement). The average wind speed during clear periods was 2.2 m/s (max: 5.5 m/s) for period 1 and 6 m/s (max: 8.3 m/s) for period 2 (see Supplement). Wind speeds ranging between 4 – 11 m/s have been shown to lead to fresh snow transport (Li and Pomeroy, 1997), but wind speed had little discernible influence on ice residual concentrations in SPIDER. Previous studies performed at SPL, such as Lowenthal et al. (2019), also found no association between ice crystal concentrations and wind speed. A background of small particles near the SP2-XR detection limit in the ice crystal residual channel not associated with wind was observed during clear conditions (Figures 9a and 9b). This inadvertent transmission is consistent with previous PCVI measurements (Pekour and Cziczo, 2011) and the lab studies and is eliminated from in-cloud data.

During transitions into cloudy periods there was a change in the ice crystal residual concentrations comparable to the CIP and measurements of liquid water content (LWC) (Figure 8). Specifically, the ice crystal residual channel shows higher concentrations during the cloudy period, with an increase between 200 nm – 500 nm that is not observed during clear periods.

These results are consistent with previous studies using phase-separating inlets to measure ice residual concentration and size in mixed-phase clouds (Mertes et al., 2007; Kupiszewski et al., 2016). Mertes et al. (2007) found that super-micrometer aerosol particles dominated ice residual concentrations whereas Kupiszewski et al. (2016) found ice residual particle sizes peaked at 200 nm – 300 nm . Using an ice chamber to nucleate ambient aerosol, DeMott et al. (2010) found that INPs were typically $>300\text{ nm}$ diameter. Our findings suggest an increase in ice residual concentration with increasing residual size from 200 to 500 nm range. This should be considered in the context of the SP2-XR size range which was limited to 540 nm ; conclusions can therefore not be drawn about residual size into the super-micrometer diameter range.

6.4.4 Composite SPIDER Experiment

The composite SPIDER instrument, composed of the L-PCVI, the evaporation chamber, and the PCVI, was tested in the laboratory. The L-PCVI aerosol generation method, described in Section 4.1, was repeated. A 5 L min^{-1} aerosol flow was combined with filtered air. The PF and the AF used in the L-PCVI were 45 L min^{-1} and 6.6 L min^{-1}

respectively. The SF of the L-PCVI, 6.5 L min^{-1} , corresponded to the flow through the evaporation chamber and the IF of the PCVI. The PCVI PF and AF in this case were 7 L min^{-1} and 1.5 L min^{-1} , respectively. While the flow conditions for the PCVI in this test are not identical to the flows tested in Section 4.4., this flow scenario is consistent with the working range of flows in the 3D printed PCVI. In order to determine the overall TE of the system, the size distribution at the outlet was determined with the OPS. First, both AFs and PFs were turned off to obtain the initial size distribution including any losses in the evaporation chamber and connections. All flows were then set to determine the size distribution at the outlet. The TE in this case was calculated as the ratio of the concentration when flows are turned on to the concentration when flows are turned off as a function of the optical size. The TE is then normalized by dividing by the product of the enhancement factor of the L-PCVI and the PCVI. The normalized TE as a function of particle diameter is presented in Figure 9. Similar to the L-PCVI and PCVI, a sigmoid function was fit to obtain the cut-off size. The D50 for the combined system was $4.8 \pm 0.1 \text{ }\mu\text{m}$.

5 Conclusion and Future Work

Laboratory studies have been used to show that both the individual components and composite SPIDER worked as designed. Field experiments were used to extend performance tests into mixed-phase clouds characterized by a suite of instruments at SPL.

In the laboratory verification measurements experiments, each component of SPIDER was isolated and tested to validate that it performed its role in the overall system. Comparisons, wherever possible, were made with previous studies. These included the L-PCVI, the droplet evaporation chamber, and the PCVI. Once each component was verified individually, a test was done to ensure that the combination of instruments also functioned using either microbeads or a combination of droplets and ice crystals. Future laboratory measurements should be conducted to further define the performance of Multiple experiments with different flow combinations were performed to obtain the D50 as a function of the ratio AF to IF for both the L-PCVI and the droplet evaporation chamber. A full characterization PCVI. This allowed for determination of the L-PCVI could better define the flow rates for obtaining an ideal D50 to segregate droplets and ice crystals from interstitial aerosol particle under a variety of ambient conditions. Similarly, it would be beneficial to perform more detailed laboratory experiments on droplet evaporation to determine performance in different cloud environments.

SPIDER was deployed at SPL in Colorado where it was tested during transitions from clear to cloud conditions. As a proof of concept campaign, SPIDER demonstrated a SF for the L-PCVI that it was deployable to a remote field location. Sporadic failures in OPCs on the interstitial and cloud drop residual channels led to minimal performance studies. Using was simultaneous suitable for droplet evaporation and as a high-resolution SP2-XR, we were able to conclude minimal influence by windblown snow during this period and characterize inadvertent transmission similar to previous studies using PCVIs. Data from mixed-phase cloud events exhibited elevated counts of ice residuals that correlated with CIP and LWC measurements. The size of ice residuals, although limited by the detection of the SP2-XR, compared with results from previous field studies. Future field experiments with SPIDER should include more robust / higher resolution OPCs on both the interstitial aerosol particle and cloud droplet residual channel. An OPC with a larger size range would be of use on the ice residual channel viable IF for the PCVI.

Hiranuma et al. (2016) showed that the D50 linearly correlates with the AF to IF ratio (coefficient, $r=0.85$). Our experimental results of the D50 as a function of the AF to IF ratio are somewhat lower than the experimental results from Hiranuma et al. (2016) although we note the difference in 3-D printed and machined L-PCVIs and somewhat different inlet and outlet designs and lengths described previously.

The PCVI was calibrated to define the working flows and to determine its the transmission efficiency. The obtained D50 is $\sim 5 \text{ }\mu\text{m}$, similar to results from previous studies (Boulter et. al., 2006; Kulkarni et al., 2011). The commercial machined and the 3D printed PCVI were calibrated and compared and are in good agreement.

The goal for SPIDER of this work was to develop a comprehensive inlet system for segregating separation and the subsequent sampling of interstitial aerosol and cloud elements in mixed phase clouds. Through these laboratory verification tests and field campaign, we demonstrate have demonstrated that SPIDER is capable of sorting the three components of mixed-phase clouds into distinct channels.

It should be noted that there is not necessarily a one-to-one relationship between droplets and ice crystals and residuals. Droplets or ice crystals can scavenge gas- and particle-phase constituents. Droplets and ice crystals can also undergo breakup or secondary formation processes. The purpose of this work is to detail a means for separation of interstitial aerosol, droplets and ice crystals into three separate channels. The specific cloud properties, such as cloud lifetime, scavenging rates, breakup processes and secondary hydrometeor production mechanisms, at a sampling site will dictate the efficacy of SPIDER to resolve residuals.

Future goals include coupling SPIDER to a particle mass spectrometersspectrometer in order to determine chemical composition. This would of interstitial aerosol, droplet and ice residuals within mixed phase clouds to allow for a determination of the differences between particles that nucleate droplets or ice from unactivated particles. Ultimately, information on cloud nucleation capabilities of various aerosol particles could be compared to laboratory work and integrated into climate models (Korolev et al., 2017; McCoy et al., 2016; Shupe et al., 2008).

76 Author Contributions

MR and DJC conceptualized the SPIDER inlet configuration. LK, MR and CDE designed the experiments, and LK, MR, CDE, CNR, LFD, AGH, IBM and CS, carried them out. LK, MR, CR developed analysis code, LK developed model code, LK and CDE performed data analysis and evaluation. LK, DJC and CDE prepared the manuscript with contributions from all co-authors.

7 Competing Interests

The authors declare that they have no conflict of interest.

8 Acknowledgements

We would like to thank the William and Kumi Martin Foundation and the National Science Foundation (Grants AGS-1749851 and AGS-1749865) for funding. L. Koolik would like to thank the MIT Department of Civil and Environmental Engineering for providing fellowship funding. We gratefully acknowledge Eric Kelsey and the staff of the Mount Washington Observatory for their assistance with the logistics associated an initial proof of concept study. We also thank the other members of the Cziezo Group at MIT for their assistance throughout the development of SPIDER. The authors appreciate the dedication, commitment, and effort of Maria Garcia and Dan Gilehrst towards the deployment of SPIDER at SPL. We further thank Droplet Measurement Technology for the use of an SP2-XR for characterization of the ice residual channel. The Steamboat Ski Resort provided logistical support and in-kind donations. The Desert Research Institute is a permittee of the Medicine Bow Routt National Forests and is an equal opportunity service provider and employer. We further thank DMT for the SP2-XR and NASA for the APS used for characterization of the L-PCVI.

References

- Miller, B. B. and Carter, C.: The test article, J. Sci. Res., 12, 135–147, doi:10.1234/56789, 2015.
Smith, A. A., Carter, C., and Miller, B. B.: More test articles, J. Adv. Res., 35, 13–28, doi:10.2345/67890, 2014.
Abel, S. J., Cotton, R. J., Barrett, P. A., and Vance, A. K.: A comparison of ice water content measurement techniques on the FAAM BAe-146 aircraft, Atmos. Meas. Tech., 7, 3007–3022, <https://doi.org/10.5194/amt-7-3007-2014>, 2014.
Technical Data for Alicat MC and MCR Mass Flow Controllers:
https://documents.alicat.com/specifications/Alicat_Mass_Controller_Specs.pdf, last access: 9 August 2020.
Atkinson, J. D., Murray, B. J., Woodhouse, M. T., Whale, T. F., Baustian, K. J., Carslaw, K. S., Dobbie, S., O'Sullivan, D., and Malkin, T. L.: The importance of feldspar for ice nucleation by mineral dust in mixed phase clouds, Nature, 498, 355–358, doi:10.1038/nature12278, 2013.
Bartolo, P. J. (Ed.): Stereolithography: Materials, Processes, and Applications, Springer US, pp. 340, 2011.
Baustian, K. J., Cziczo, D. J., Wise, M. E., Pratt, K. A., Kulkarni, G., Hallar, A. G., and Tolbert, M. A.: Importance of aerosol particle composition, mixing state, and morphology for heterogeneous ice nucleation: A combined field and laboratory approach, J. Geophys. Res-Atmos., 117, 1–13, <https://doi.org/10.1029/2011JD016784>, 2012.
Bhushan, B. and Caspers, M.: An overview of additive manufacturing (3D printing) for microfabrication, Microsyst. Technol., 23, 1117–1124, <https://doi.org/10.1007/s00542-017-3342-8>, 2017.

[Borys, R. D. and Wetzel, M. A.: Storm Peak Laboratory: A research, teaching, and service facility for the atmospheric sciences., *Bull. Amer. Meteor. Soc.*, 78, 2115–2124, doi:10.1175/1520-0477\(1997\)078<2115:SPLART>2.0.CO;2 1997.](#)

Boucher, O., Randall, D., Artaxo, P., Bretherton, C., Feingold, G., Forster, P., Kerminen, V.-M., Kondo, Y., Liao, H., Lohmann, U., Rasch, P., Satheesh, S. K., Sherwood, S., Stevens, B. and Zhang, X. Y.: Clouds and Aerosols, in: Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change, edited by Stocker, T. F., Qin, D., Plattner, G. K., Tignor, M., Allen, S. K., Boschung, J., Nauels, A., Xia, Y., Bex, V., and Midgley, P. M., Cambridge University Press, Cambridge, United Kingdom and New York, NY, United States, pp. 571–657, 2013.

Boulter, J. E., Cziczko, D. J., Middlebrook, A. M., Thomson, D. S. and Murphy, D. M.: Design and performance of a pumped counterflow virtual impactor, *Aerosol Sci. Tech.*, 40, 969–976, <https://doi.org/10.1080/02786820600840984>, 2006.

DS18B20 Programmable Resolution 1-Wire Digital Thermometer:

<http://datasheets.maximintegrated.com/en/ds/DS18B20.pdf>, last access: 9 August 2020.

Davis, S. M., Avallone, L. M., Weinstock, E. M., Twohy, C. H., Smith, J. B., and Kok, G. L.: Comparisons of in situ measurements of cirrus cloud ice water content, *J. Geophys. Res.*, 112, 1–15, <https://doi.org/10.1029/2006JD008214>, 2007a.

Davis, S. M., Hallar, A. G., Avallone, L. M., and Engblom, W.: Measurement of total water with a tunable diode laser ~~hygrometer~~ ~~hygrometer~~: Inlet analysis, calibration procedure, and ice water content determination, *J. Atmos. Ocean Tech.*, 24, 463–475, <https://doi.org/10.1175/JTECH1975.1>, 2007b.

~~DeMott, P. J., Prenni, A. J., Liu, X., Kreidenweis, S. M., Petters, M. D., Twohy, C. H., Richardson, M. S., Eidhammer, T., and Rogers, D. C.: Predicting global atmospheric ice nuclei distributions and their impacts on climate, *P. Natl. A. Sci. USA*, 107, 11217–11222, doi:10.1073/pnas.0910818107, 2010.~~

3D Printing with Desktop Stereolithography: An Introduction for Professional Users: <https://archive-media.formlabs.com/upload/Intro-sla-whitepaper-04.pdf>, last access: 9 August 2020.

Friedman, B., Ardon-Dryer, K., Carrasquillo, A., Daumit, K., Boulanger, K., Cross, E., Browne, E., Kroll, J., Thornton, J. and Cziczko, D.: CCN closure and composition analysis of droplet-forming aerosol particle, *AIP Conf. Proc.*, 1527, 832–835, <https://doi.org/10.1063/1.4803400>, 2013a.

~~Friedman, B., Zelenyuk, A., Beranek, J., Kulkarni, G., Pekour, M., Hallar, A. G., McCubbin, I. B., Thornton, J. A., and Cziczko, D. J.: Aerosol particle measurements at a high elevation site: Composition, size, and cloud condensation nuclei activity, *Atmos. Chem. Phys.*, 13, 11839–11851, doi:10.5194/acp-13-11839-2013, 2013b.~~

Hagiwara, T.: Current status and future prospects of laser stereolithography, *Proc. SPIE*, 5662, 644–648, <https://doi.org/10.1117/12.596757>, 2004.

Hammer, S. E., Mertes, S., Schneider, J., Ebert, M., Kandler, K., and Weinbruch, S.: Composition of ice particle residuals in mixed-phase clouds at Jungfraujoch (Switzerland): enrichment and depletion of particle groups relative to total aerosol, *Atmos. Chem. Phys.*, 18, 13987–14003, <https://doi.org/10.5194/acp-18-13987-2018>, 2018.

Hartmann, M., Adachi, K., Eppers, O., Haas, C., Herber, A., Holzinger, R., Hünerbein, A., Jäkel, E., Jentzsch, C., van Pinxteren, M., Wex, H., Willmes, S., and Stratmann, F.: Wintertime airborne measurements of ice nucleating particles in the high Arctic: a hint to a marine, biogenic source for ice nucleating particles, *Geophys. Res. Lett.*, 47, 1–11, <https://doi.org/10.1029/2020GL087770>, 2020.

Heymsfield, A. J., Krämer, M., Luebke, A., Brown, P., Cziczko, D. J., Franklin, C., Lawson, P., Lohmann, U., McFarquhar, G., Ulanowski, Z., and Van Tricht, K.: Cirrus Clouds, *Meteor. Mon.*, 58, 2.1-2.26, <https://doi.org/10.1175/amsmonographs-d-16-0010.1>, 2017.

Hiranuma, N., Möhler, O., Kulkarni, G., Schnaiter, M., Vogt, S., Vochezer, P., Järvinen, E., Wagner, R., Bell, D. M., Wilson, J., Zelenyuk, A., and Cziczko, D. J.: Development and characterization of an ice-selecting pumped counterflow virtual impactor (IS-PCVI) to study ice crystal residuals, *Atmos. Meas. Tech.*, 9, 3817–3836, <https://doi.org/10.5194/amt-9-3817-2016>, 2016.

Hirst, E., Kaye, P. H., Greenaway, R. S., Field, P., and Johnson, D. W.: Discrimination of micrometre-sized ice and super-cooled droplets in mixed-phase cloud, *Atmos. Environ.*, 35, 33–47, [https://doi.org/10.1016/S1352-2310\(00\)00377-0](https://doi.org/10.1016/S1352-2310(00)00377-0), 2001.

Hoose, C. and Möhler, O.: Heterogeneous ice nucleation on atmospheric aerosols: a review of results from laboratory experiments, *Atmos. Chem. Phys.*, 12, 9817–9854, <https://doi.org/10.5194/acp-12-9817-2012>, 2012.

Irish, V. E., Hanna, S. J., Willis, M. D., China, S., Thomas, J. L., Wentzell, J. J. B., Cirisan, A., Si, M., Leaitch, W. R., Murphy, J. G., Abbatt, J. P. D., Laskin, A., Girard, E., and Bertram, A. K.: Ice nucleating particles in the marine boundary layer in the Canadian Arctic during summer 2014, *Atmos. Meas. Tech.*, 19, 1027–1039, <https://doi.org/10.5194/acp-19-1027-2019>, 2019.

Jacobs, P. F. (Ed.): Rapid Prototyping & Manufacturing: Fundamentals of Stereolithography, 1st ed., Society of Manufacturing Engineers, in cooperation with the computer and Automated Systems Association of SME, Dearborn, Michigan, United States, 1992.

Kamphus, M., Etter-Mahl, M., Klimach, T., Drewnick, F., Keller, L., Cziczko, D. J., Mertes, S., Borrmann, S., and Curtius, J.: Chemical composition of ambient aerosol, ice residues and cloud droplet residues in mixed-phase ~~clouds~~ clouds: single particle analysis during the Cloud and Aerosol Characterization Experiment, *Atmos. Chem. Phys.*, 10, 8077–8095, <https://doi.org/10.5194/acp-10-8077-2010>, 2010.

Kanji, Z. A., Ladino, L. A., Wex, H., Boose, Y., Burkert-Kohn, M., Cziczko, D. J., and Krämer, M.: Overview of ice nucleating particles, *Meteor. Mon.*, 58, 1, 1.1-1.33, <https://doi.org/10.1175/amsmonographs-d-16-0006.1>, 2017.

Kleinman, L. I., Daum, P. H., Lee, Y. N., Lewis, E. R., Sedlacek, A. J., Senum, G. I., Springston, S. R., Wang, J., Hubbe, J., Jayne, J., Min, Q., Yum, S. S., and Allen, G.: Aerosol concentration and size distribution measured below, in, and above cloud from the DOE G-1 during VOCALS-REx, *Atmos. Chem. Phys.*, 12, 207–223, <https://doi.org/10.5194/acp-12-207-2012>, 2012.

Koolik, L. P.: Characterization of a 3D printed pumped counterflow virtual impactor and an aerodynamic lens concentrator, B.S. thesis, Massachusetts Institute of Technology, Cambridge, Massachusetts, United States, 2017.

Koop, T., Luo, B., Athanasios, T., and Peter, T.: Water activity as the determinant for homogeneous ice nucleation in aqueous solutions, *Nature*, 406, 611-614, <https://doi.org/10.1038/35020537>, 2000.

Korolev, A.: Limitations of the Wegener–Bergeron–Findeisen mechanism in the evolution of mixed-phase clouds, *J. Atmos. Sci.*, 64, 3372–3375, <https://doi.org/10.1175/JAS4035.1>, 2007.

Korolev, A. V., Strapp, J. W., Isaac, G. A. and Nevzorov, A. N.: The Nevzorov airborne hot-wire LWC– TWC ~~probe-probe~~ : principle of operation and performance characteristics, *J. Atmos. Ocean Tech.*, 15, 1495–1510, [https://doi.org/10.1175/1520-0426\(1998\)015%3C1495:TNAHWL%3E2.0.CO;2](https://doi.org/10.1175/1520-0426(1998)015%3C1495:TNAHWL%3E2.0.CO;2), 1998.

Korolev, A. V., Isaac, G. A., Cober, S. G., Strapp, J. W., and Hallett, J.: Microphysical characterization of mixed-phase clouds, *Q. J. R. Meteor. Soc.*, 129, 39–65, <https://doi.org/10.1256/qj.01.204>, 2003.

Korolev, A., McFarquhar, G., Field, P. R., Franklin, C., Lawson, P., Wang, Z., Williams, E., Abel, S. J., Axisa, D., Borrmann, S., Crosier, J., Fugal, J., Krämer, M., Lohmann, U., Schlenker, O., Schnaiter, M., and Wendisch, M.: Mixed-Phase Clouds: Progress and Challenges, *Meteor. Mon.*, 58, 1, 5.1-5.50, <https://doi.org/10.1175/AMSMONOGRAPHS-D-17-0001.1>, 2017.

~~Korolev, A.: Limitations of the Wegener–Bergeron–Findeisen mechanism in the evolution of mixed-phase clouds, *J. Atmos. Sci.*, 64, 3372–3375, [doi:10.1175/JAS4035.1](https://doi.org/10.1175/JAS4035.1), 2007.~~

Kulkarni, G., Pekour, M., Afchine, A., Murphy, D. M., and Cziczko, D. J.: Comparison of experimental and numerical studies of the performance characteristics of a pumped counterflow virtual impactor, *Aerosol Sci. Tech.*, 45, 382–392, <https://doi.org/10.1080/02786826.2010.539291>, 2011.

Kupiszewski, P., Weingartner, E., Vochezer, P., Schnaiter, M., Bigi, A., Gysel, M., Rosati, B., Toprak, E., Mertes, S., and Baltensperger, U.: The Ice Selective Inlet: A novel technique for exclusive extraction of pristine ice crystals in mixed-phase clouds, *Atmos. Meas. Tech.*, 8, 3087–3106, <https://doi.org/10.5194/amt-8-3087-2015>, 2015.

Kupiszewski, P., Zanatta, M., Mertes, S., Vochezer, P., Lloyd, G., Schneider, J., Schenk, L., Schnaiter, M., Baltensperger, U., Weingartner, E., and Gysel, M.: Ice residual properties in mixed-phase clouds at the high-alpine Jungfraujoch site, *J. Geophys. Res-Atmos.*, 121, 12,343-12,362, <https://doi.org/10.1002/2016JD024894>, 2016.

Li, L. and Pomeroy, J. W.: Probability of occurrence of blowing snow, *J. Geophys. Res-Atmos.*, 102, 21955–21964, [doi:10.1029/97JD01522](https://doi.org/10.1029/97JD01522), 1997.

Lacher, L., Clemen, H-C, Shen, X., Mertes, S., Gysel-Beer, M., Moallemi, A., Steinbacher, M., Henne, S., Saathoff, H., Möhler, O., Höhler, K., Schiebel, T., Weber, D., Schrod, J., Schneider, J., and Kanji, Z. A.: Sources and nature of ice-nucleating particles in the free troposphere at Jungfraujoch in winter 2017, *Atmos. Chem. Phys.*, 21, 16925-16953, <https://doi.org/10.5194/acp-21-16925-2021>, 2021.

Lohmann, U.: Anthropogenic aerosol influences on mixed-phase clouds, *Curr. Clim. Change Rep.*, 3, 32-44, <https://doi.org/10.1007/s40641-017-0059-9>, 2017

Lohmann, U. and Hoose, C.: Sensitivity studies of different aerosol indirect effects in mixed-phase clouds, *Atmos. Chem. Phys.*, 9, 8917–8934, <https://doi.org/10.5194/acp-9-8917-2009>, 2009.

Lohmann, U., Lüönd, F., and Mahrt, F. (Eds.): An Introduction to Clouds: From the Microscale to Climate, Cambridge University Press, Cambridge, England, 2016.

Lowenthal, D. H., Hallar, A. G., David, R. O., McCubbin, I. B., Borys, R. D., and Mace, G. G.: Mixed-phase orographic cloud microphysics during StormVEx and IFRACS, *Atmos. Chem. Phys.*, 19, 5387–5401, <https://doi.org/10.5194/acp-19-5387-2019>, 2019.

[McCoy, D. T., Tan, I., Hartmann, D. L., Zelinka, M. D., and Storelvmo, T.: On the relationships among cloud cover, mixed-phase partitioning, and planetary albedo in GCMs, *J. Adv. Model. Earth Sy.*, 8, 2, 650-668, <https://doi.org/10.1002/2015MS000569>, 2016.](#)

Mertes, S., Verheggen, B., Walter, S., Connolly, P. J., Ebert, M., Schneider, J., Bower, K. N., Cozic, J., Weinbruch, S., Baltensperger, U., and Weingartner, E.: Counterflow virtual impactor based collection of small ice particles in mixed-phase clouds for the physico-chemical characterization of tropospheric ice nuclei: Sampler description and first case study, *Aerosol Sci. Tech.*, 41, 848–864, <https://doi.org/10.1080/02786820701501881>, 2007.

[Ogren, J. A., Heintzenberg, J., and Charlson, R. J.: In-situ sampling of clouds with a droplet to aerosol converter, *Geophys. Res. Lett.*, 12, 121-124, <https://doi.org/10.1029/GL012i003p00121>, 1985.](#)

Patade, S., Shete, S., Malap, N., Kulkarni, G., and Prabha, T. V.: Observational and simulated cloud microphysical features of rain formation in the mixed phase clouds observed during CAIPEEX, *Atmos. Res.*, 169, 32–45, <https://doi.org/10.1016/j.atmosres.2015.09.018>, 2016.

Pekour, M. S. and Cziczo, D. J.: Wake capture, particle breakup, and other artifacts associated with counterflow virtual impactation, *Aerosol Sci. Tech.*, 45, 758–764, <https://doi.org/10.1080/02786826.2011.558942>, 2011.

Petersen, R. C., Hallar, A. G., McCubbin, I. B., Ogren, J. A., Andrews, E., Lowenthal, D., Gordner, R., Purcell, R., Sleeth, D., and Novoselov, I.: Numerical, wind-tunnel, and atmospheric evaluation of a turbulent ground-based inlet sampling system, *Aerosol Sci. Tech.*, 53, 712–727, <https://doi.org/10.1080/02786826.2019.1602718>, 2019.

Pruppacher, H. R. and Klett, J. D.: *Microphysics of Clouds and Precipitation*, 2nd ed., Springer Netherlands, Dordrecht, Netherlands, 1997.

[Ramelli, F., Hanneberger, J., David, R. O., Lauber, A., Pasquier, J. T., Wider, J., Bühl, J., Seifert, P., Engemann, R., Hervo, M., and Lohmann, U.: Influence of low-level blocking and turbulence on the microphysics of a mixed-phase cloud in an inner-Alpine valley: *Atmos. Chem. Phys.*, 21, 5151-5172, <https://doi.org/10.5194/acp-21-5151-2021>, 2021.](#)

Roesch, M., Roesch, C., and Cziczo, D. J.: Dry particle generation with a 3-D printed fluidized bed generator, *Atmos. Meas. Tech.*, 10, 1999–2007, <https://doi.org/10.5194/amt-10-1999-2017>, 2017.

~~Roesch~~Rösch, M. and Cziczo, D. J.: Aqueous particle generation with a 3D printed nebulizer, *Atmos. Meas. Tech. Discuss.*, 13, 6807-6812, <https://doi.org/10.5194/amt-13-6807-2020-155>, in review, 2020.

Rogers, R. R. and Yau, M. K. (Eds.): *A Short Course in Cloud Physics*, 3rd ed., Butterworth Heinemann, Burlington, Massachusetts, 1989.

[Ruiz-Donoso, E., Ehrlich, A., Schäfer, M., Jäkel, E., Schemann, V., Crewell, S., Mech, M., Kulla, B. S., Kliesch, L., Neuber, R., and Wendisch, M.: Small-scale structure of thermodynamic phase in Arctic mixed-phase clouds observed by airborne remote sensing during a cold air outbreak and a warm air advection event, *Atmos. Chem. Phys.*, 20, 5487-5511, <https://doi.org/10.5194/acp-20-5487-2020>, 2020.](#)

[Schmidt, S., Schneider, J., Klimach, T., Mertes, S., Schenk, L. P., Kupiszewski, P., Curtius, J., and Borrmann, S.: Online single particle analysis of ice particle residuals from mountain-top mixed-phase clouds using laboratory derived particle type assignment, *Atmos. Chem. Phys.*, 17, 575-594, <https://doi.org/10.5194/acp-17-575-2017>, 2017.](#)

Shupe, M. D., Matrosov, S. Y., and Uttal, T.: Arctic mixed-phase cloud properties derived from surface-based sensors at SHEBA, *J. Atmos. Sci.*, 63, 697–711, <https://doi.org/10.1175/JAS3659.1>, 2006.

Shupe, M., Daniel, J. S., De Boer, G., Eloranta, E. W., Kollias, P., Long, C. N., Luke, E. P., Turner, D. D., and Verlinde, J.: A focus on mixed-phase clouds, *B. Am. Meteorol. Soc.*, 89, 1549–1562, <https://doi.org/10.1175/2008BAMS2378.1>, 2008.

[Si, M., Evoy, E., Yun, J., Xi, Y., Hanna, S. J., Chivulescu, A., Rawlings, K., Veber, D., Platt, A., Kunkel, D., Hoor, P., Sharma, S., Leaitch, W. R., and Bertram, A. K.: Concentrations, composition, and sources of ice-nucleating particles in the Canadian High Arctic during spring 2016, *Atmos. Chem. Phys.*, 19, 3007-3024, <https://doi.org/10.5194/acp-19-3007-2019>, 2019.](#)

Slowik, J. G., Cziczo, D. J., and Abbatt, J. P. D.: Analysis of cloud condensation nuclei composition and growth kinetics using a pumped counterflow virtual impactor and aerosol mass spectrometer, *Atmos. Meas. Tech.*, 4, 1677–1688, <https://doi.org/10.5194/amt-4-1677-2011>, 2011.

Storelvmo, T., Kristjansson, J. E., and Lohmann, U.: Aerosol influence on mixed-phase clouds in CAM-Oslo, *J. Atmos. Sci.*, 65, 3214–3230, <https://doi.org/10.1175/2008JAS2430.1>, 2008.

Strapp, J. W., Lilie, L. E., Ratvasky, T. P., Davison, C. R., and Dumont, C.: Isokinetic TWC evaporator probe: development of the IKP2 and performance testing for the HAIC-HIWC Darwin 2014 and Cayenne-2015 field campaigns, in: 8th AIAA Atmospheric and Space Environments Conference, Washington, D. C., United States, 13-17 June 2016, <https://doi.org/10.2514/6.2016-4059>, 2016.

[Tan, I. and Storelvmo, T.: Evidence of strong contributions from mixed-phase clouds to arctic climate change, *Geophys. Res. Lett.*, 46, 5, 2894-2902, <https://doi.org/10.1029/2018GL081871>, 2019.](#)

Tsushima, Y., Emori, S., Ogura, T., Kimoto, M., Webb, M. J., Williams, K. D., Ringer, M. A., Soden, B. J., Li, B., and Andronova, N.: Importance of the mixed-phase cloud distribution in the control climate for assessing the response of clouds to carbon dioxide increase: a multi-model study, *Clim. Dynam.*, 27, 113-126, <https://doi.org/10.1007/s00382-006-0127-7>, 2006.

Verheggen, B., Cozic, J., Weingartner, E., Bower, K., Mertes, S., Connolly, P., Gallagher, M., Flynn, M., Choulaton, T., and Baltensperger, U.: Aerosol partitioning between the interstitial and the condensed phase in mixed-phase clouds, *J. Geophys. Res-Atmos.*, 112, 1–13, <https://doi.org/10.1029/2007JD008714>, 2007.

Verlinde, J., Harrington, J. Y., McFarquhar, G. M., Yannuzzi, V. T., Avramov, A., Greenberg, S., Johnson, N., Zhang, G., Poellot, M. R., Mather, J. H., Turner, D. D., Eloranta, E. W., Zak, B. D., Prenni, A. J., Daniel, J. S., Kok, G. L., Tobin, D. C., Holz, R., Sassen, K., Spangenberg, D., Minnis, P., Tooman, T. P., Ivey, M. D., Richardson, S. J., Bahrman, C. P., Shupe, M., DeMott, P. J., Heymsfield, A. J., and Schofield, R.: The mixed-phase arctic cloud experiment, *B. Am. Meteorol. Soc.*, 88, 205–221, doi:10.1175/BAMS 88 2 205, 2007.

Wang, B., Lambe, A. T., Massoli, P., Onasch, T. B., Davidovits, P., Worsnop, D. R., and Knopf, D. A.: The deposition-ice nucleation and immersion freezing potential of amorphous secondary organic aerosol: Pathways for ice and mixed-phase cloud formation, *J. Geophys. Res-Atmos.*, 117, 1–12, doi:10.1029/2012JD018063, 2012.

Wong, K. V. and Hernandez, A.: A review of additive manufacturing, *ISRN Mech. Eng.*, 2012, 1-10, <https://doi.org/10.5402/2012/208760>, 2012.

Figure 1: Cross-sectional view of the 3D printed SPIDER PCVI with flows ~~labelled~~labeled. The 3D printed PCVI features the improved conical input nozzle suggested by Kulkarni et al. (2011); otherwise, the design is the same as considered by Kulkarni et al. (2011).

Figure 2: Schematic of SPIDER with its components ~~labelled~~labeled. (a) The L-PCVI (Hiranuma et al., 2016) separates interstitial aerosol from the droplets and ice crystals. (b) Thermocouples report the temperature in the chamber. (c) The chamber is cooled and held at ice saturation to evaporate droplets. (d) The PCVI downstream separates evaporated droplet residuals from ice crystals. (e) The bottom houses electronics and mass flow controllers (MFCs).

Figure 3: Concurrent time series of particles detected in the (a) PF and the (b) SF of the L-PCVI. The flows into the PF and AF were turned on between 1,700–1,900 s. Note that particles were detected only in the SF until the flows were turned on, at which point the particles are counted in the PF. The “X” mark represents no flow through the corresponding chamber of the L-PCVI; an arrow represents flow. The circle indicates where the sample is measured.

Figure 4: Droplet transmission through the L-PCVI under varying flow scenarios. The average number concentration of particles out of the SF of the L-PCVI is plotted against particle diameter for (a) AF and PF off, (b) AF and PF on, and (c) only AF on. Figure 3: Number concentration as a function of the aerodynamic particle diameter of soda lime glass microspheres used for L-PCVI and SPIDER validation experiments.

Figure 4: Transmission efficiency from the L-PCVI as a function of particle aerodynamic diameter (solid circles) fit with a sigmoid (solid line). The triangle represents the size at which 50% of particles are transmitted (the experimental D50). The representative error, $\pm 3\%$, due to instrument uncertainty, is shown on a point close to the D50 and on the D50.

Figure 5: Droplet concentration at the outlet of the evaporation chamber as a function of the optical particle diameter for different residence times.

Figure 6: Ice crystals concentration at the outlet of the evaporation chamber as a function of the optical particle diameter.

Figure 7: \mp

~~Figure 5: Number concentration as a function of particle diameter for ice crystals travelling through the chamber. Ambient air was introduced to the cooled chamber with the PCVI on. Droplets were introduced into the cooled chamber with the PCVI on. The results shown are the difference between these two averages, reflecting the size distribution from ice crystals alone.~~

Figure 6: Comparison of D50 values for [the industrial commercial machined](#) and [3D printed PCVI](#) as a function of IF [in the overlapping range of functionality](#).

Figure 7: Transmission efficiency [from the PCVI](#) as a function of particle diameter (solid circles) fit with a [sigmoid \(dashed line\)](#). The triangle represents the size at which 50% of particles are transmitted (the experimental D50). The representative error, $\pm 5\%$ due to instrument uncertainty as specified by the manufacturer, is shown on a point close to the D50 and on the D50.

Figure 8: Transmission efficiency [from SPIDER](#) as a function of particle aerodynamic diameter (solid circles) fit with a [sigmoid \(solid line\)](#). The triangle represents the size at which 50% of particles are transmitted (the experimental D50). The representative error, $\pm 5\%$, due to instrument uncertainty, is shown on a point close to the D50 and on the D50. [Note that the SPIDER transmission efficiency represents the sequential transmission of the L-PCVI and PCVI.](#)

Figure 9: (a) Cloud Imaging Probe water content over time for particles $30\mu\text{m}$ – $105\mu\text{m}$. Water content was calculated assuming spherical particles and a water density of 1 g/cm^3 . Dashed line at $0.025\text{ }\mu\text{g/cm}^3$ indicates the threshold for cloud presence. (b) Cloud Imaging Probe total number concentration over time where a logarithmic scale is used for clarity. (c) SP2 XR ice crystal residual concentration. Gray shading represents cloudy conditions.

Figure 10: SP2 XR average ice crystal residual concentration. Clear conditions during periods 1 and 2 (a, b) and cloudy conditions during periods 3 and 4 (c, d). Average background concentrations during clear conditions were subtracted from cloudy conditions.