Dear Reviewer,

Thanks for providing these comments to further improve the manuscript. Apologies for the delayed response, the last few months have been challenging during this pandemic. Please find below the reply to your comments. These comments are also used to revise the manuscript.

Thanks,

Gourihar Kulkarni

Anonymous Referee #RC2

Referee comments on "A new method for operating a continuous flow diffusion chamber to investigate immersion freezing: assessment and performance study" by G. Kulkarni et al. 2020

In the submitted manuscript Kulkarni et al. describe a new method for operating a Continuous Flow Diffusion Chamber (CFDC) and show both system modeling results and results from testing using various experimental test aerosol and some ambient air sampling. I find the manuscript generally well written and presented. The idea for the new CFDC operation principle is original and enticing. This idea potentially expands the operational range of CFDC instruments and could be a significant contribution to the community. However in its current form the submitted work lacks clarity in some key areas. Some additional work also needs to be done with respect to the figures, where either interpretation is difficult and/or mistakes appear to have been made with labeling in the main text etc

Nucleation Temperature and Crystal Growth: I think the primary question that the authors must clarify is related to quantifying the ice nucleation temperature and ice crystal growth within the evaporation (now nucleation) section of the CFDC. The authors have done a nice job of trying to model the droplet growth in the 'conditioning' section, but have **not shown analogous results for modeling the crystal changes in the evaporation section** (I recognize they posit that given the saturation condition is RH_{ice} = 100% there are no changes – but consider comment below). My interpretation of Figure 4 and many of the Supplemental figures is that if the system behaves as modeled then liquid droplets quickly evaporate within the nucleation section – on approximately the same time scale as the temperature and RH fields equilibrate. This suggests that the nucleation occurs in this transition region and that the fixed nucleation section temperature in fact controls the gradient between the two sections **but does not necessarily represent the actual nucleation condition**. **What size water droplet must nucleate into ice in order to grow to reach the quoted 3 _m OPC cutoff for ice?** If there is a lower bound on this value then one might interpret before **what point along the droplet evaporation curves ice must form**. Likewise it would be interesting to **understand the range of potential ice crystal sizes** depending on at what point entering the chamber a droplet nucleates. Clearly at the warmest temperatures the gradients between the two chambers are weaker and thus the constraints on thermodynamic forcing will be better, but at the colder **temperatures I remain to convinced that the nucleation occurs at the equilibrated chamber conditions.**

Reply: The evaporation section conditions are constant. This section is maintained at constant temperature and RH_{ice} (=100%) conditions (Fig. 4a). We expect no change in the ice crystal size.

Correct, the temperature within the transition section (varies from conditioning section to nucleation or evaporation section) does not correspond to the equilibrium nucleation section temperature (e.g. Fig. S5

b). Freezing occurs at various temperatures that range from the conditioning section temperature (~ -20 °C) to the nucleation section temperature (e.g. -30°C) conditions. The temperature uncertainty across the aerosol lamina and nucleation section are ± 0.9 and ± 0.4°C, respectively. Here, we have used temperature uncertainty across the nucleation section as the temperature uncertainty within the ice fraction. The ice fraction is defined as the cumulative fraction of the droplet frozen, and it is reported at the coldest section of the chamber (i.e. steady state nucleation section temperature). See supplementary section Text S1. Following sentence is added.

Text S1: *The freezing temperature (T) is defined at the steady state temperature of the nucleation section, and the freezing temperature uncertainty is assumed to be similar to the uncertainty across the nucleation section (± 0.4°C).*

CFD simulations (e.g. Fig S3 c) show that water droplets of size greater than 2 µm in radius will mostly contribute towards nucleation of ice. Droplets smaller than this size are exposed to subsaturation conditions, and they evaporate quickly (< 1 sec; see Fig S3 b). It should be noted that as nucleation occurs in the order of a few ms (Holden et al. 2019), the droplets smaller than 2 µm might also contribute towards nucleation of ice. However, the contribution of these smaller droplets of less than 2 µm is very small (see Fig. 5a).

Holden, M. A., Whale, T. F., Tarn, M. D., O'Sullivan, D., Walshaw, R. D., Murray, B. J., Meldrum, F. C., and Christenson, H. K.: High-speed imaging of ice nucleation in water proves the existence of active sites, Sci. Adv., 5, eaav4316, https://doi.org/10.1126/sciadv.aav4316, 2019.

Further evidence already included: on page 6 the authors state that, “ice particle size measured by the OPC can be representative of the size of the droplet while freezing.” However, all simulations of droplet growth suggest maximum droplet sizes between 2 and 2.5 _m. Figure 5 shows peak OPC concentrations from about 3.57 to 5.02 (diameter) which more-or-less corresponds to the peak predicted particle sizes, and those droplet diameters only occur immediately in this transition region and **not within the equilibrated portion of the chamber**. However, also to consider is that, although the equilibrated chamber represents RH_{ice} = 100%, as long as **droplets do exist ice particles can grow due to scavenging...to what extent? Perhaps this is minimal? Will the droplet evaporation go back to the walls?**

Reply: Figure 5a shows ice crystal sizes and their respective concentrations at different temperatures. As mentioned above, droplets of size less than 2 µm in radius may contribute towards the total ice crystal concentration, but their fraction compared to the total concentration is very small.

Flow conditions across the chamber are laminar (Fig. 4a). The droplets and ice crystals follow particle trajectories determined by the various forces (flow conditions and gravity) acting on the particle. It appears that these particles have insufficient inertia to cross the gas streamlines (Fig. S5; see five INP trajectories), such that scavenging of droplets by ice crystals can be ignored. Correct, the water vapor from the droplet (during evaporation) might go towards the wall. Also, some of the vapor might exit the chamber.

Below I present an itemized list of additional thoughts and comments as I came to them in the text, which I hope helps to further contextualize my thoughts.

Itemized Scientific and Editorial Suggestions:

Specific Suggestions by Page and Line Number (page, line):

_ (1,26) enough to say sampled from 'am ambient aerosol inlet'. The location etc. is described later.
_ (2,42) replace toward with for
_ (2,50) percent
_ (2,50) CFDCs also
_ (2, 61) particles are activated not 'all aerosol'. Remember the strict definition of aerosol is the gas, particle mixture thus activation of all aerosol seems strange.
_ (3, 68) the Compact Ice...
Reply: All the above comments are addressed.

_ (3, 70) thermally isolated or insulated? How much thermal contact do the 2 sections actually have?
Reply: Corrected, they are thermally isolated. The two walls are not in contact with each other, but they are separated by double-layered insulated gasket.

_ (3,78) Here begins the use of many symbols _, =_, etc. which continues throughout the manuscript in an ill-defined manner. I presume most often these are being used to indicate approximately, for which I suggest _. Although definitions are a bit muddled the use of similar to _ to many, including me, denotes an order of magnitude (-ish) approximation. I am sure the authors intention is to convey a more approximate value than that in many of their uses here and throughout.
Reply: Corrected. The ~ symbol is replaced with ≈ symbol.

Here also the RHw is indicated as 106%. Later in the numerical modeling section 2.2 a RHw of 113% is chosen and this value also seems to be chosen in the experimental descriptions that follow. I am left confused, why these differences?
Reply: The RHw = 106% corresponds to the CIC chamber (the original chamber, but not the modified chamber or MCIC). The RHw = 113% corresponds to the modified CIC chamber (MCIC).

_ (3, 80) An OPC
Reply: Corrected.

_ (3,93) Please also include here the saturation condition that results from the choice of temperatures – it would be nice to also have the value in terms of ice saturation.
Reply: The saturation (water and ice) conditions for these conditions are shown in Figure 3. We added the following sentence to address this comment.

Section 2.1: *The resulting water and ice saturation conditions are shown in Figure 3.*

_ (4, 103) 'The choice of steady-state cooling....' I think the manuscript would benefit from a longer discussion related to the cooling rate. The empirical choice of cooling rate as being satisfactory is supported by the filled symbols on Figure 6, which I understand were measurements made with the chamber at static conditions. However, did the authors try any other cooling rates? Do they have any evidence of what a maximum cooling rate might be? I think any additional information that might have been gathered with regard to the operational limits would add value to what the authors have done.
Reply: Some exploratory work with different cooling rates is explored. The ice fraction results of ambient sampling showed a negligible difference between the cooling rates.

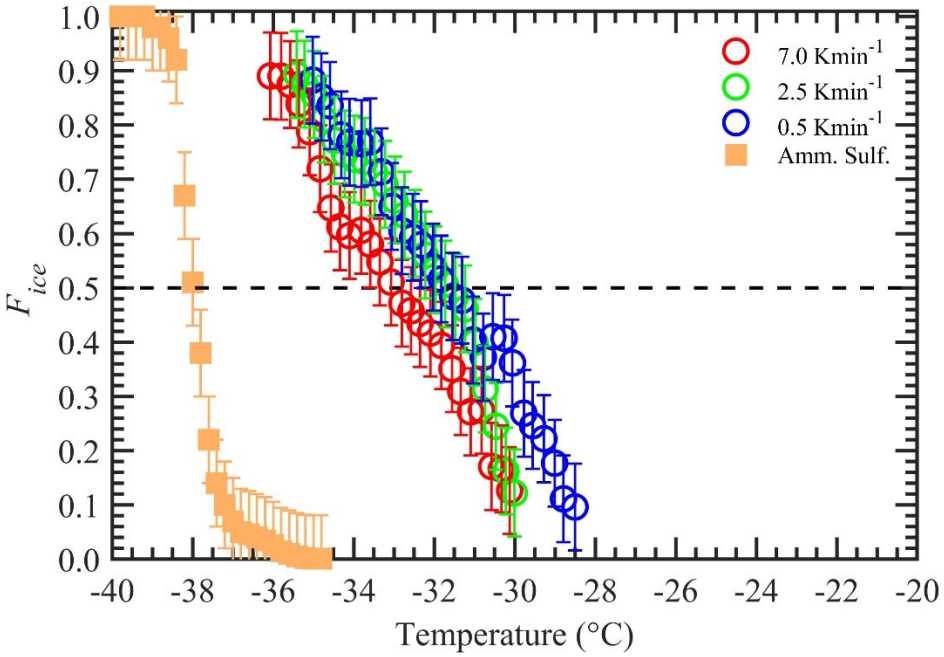

Figure S8: The F_{ice} of airborne arable dust species as a function of temperature and nucleation section cooling rates. The cooling rate of 0.5 Kmin^{-1} was used in this study.

The figure is added to the supplementary section. The following text is added to the manuscript.

Section 2.1: *The implications of higher cooling rates towards INP measurements were also explored.*
Section 3.0: *The experiments with higher cooling rates (2.5 and 7.0 °C min^{-1}) had also a negligible effect on F_{ice}* of airborne arable dust species *(Figure S8).*

_ (4,110-113) More clarity is needed with respect to the pulse experiments. The pulse duration is quoted as 10.5 s. In Fig. 1 of the supplement the dashed line is used to indicate the limit after which particles are considered to be outside of the lamina. However, if the pulse duration is 10.5 s and the residence time is _ 10s shouldn't pulsed particles continue to arrive until 20.5 s? This would presumably significantly alter the 16% number in the text. How is my understanding deficient?
Reply: The data is shown when CPC starts recording the particle counts. The below sentence is added to the figure caption of figure S1.

Figure S1*: The data is shown only when CPC started recording the particle counts.*

_ (4,113-116) Final sentence of this section seems to be better suited to introduce the following section.
Reply: Corrected.

_ (5,133-134) such a geometry; I am confused by the end to this sentence. "...it was coupled with energy and viscous heating to enable the species..." I think this needs to be reworded. What was coupled exactly? Is energy conservation meant? Please clarify this sentence.
Reply: Corrected. These sentences describe the viscous model used to model the flow and droplet trajectories. The sentence is revised as follows.

Section 2.2: *The viscous model – the standard RNG κ – ε turbulence model was used. This model treats velocity fluctuations better than other turbulence models for such a geometry. This turbulence model was used in conjunction with species transport modeling capability such that the effects of smaller eddies of fluid motion are better captured.*

_ (5,141) I found the relevant information is S.1 not S1, but this appears very far into the supplement. It would be useful to order the supplement in an order that corresponds to how it is referenced in the text.
Reply: Sorry for the inconvenience. It should read S.1. To avoid the confusion, we rename it as Text S1.

The order of Text S1 and S2 is rearranged.

More notes with regard to S.1: What is meant with e_1 and e_r? The use of 'environment' is confusing. I think e_1 represents the far field vapor pressure, while e_r represents the equilibrium vapor pressure at the surface. Similarly the temperature terms should be precisely defined. Furthermore, the D_v term introduces another temperature T and pressure p that seem to have the same definitions as T_1 and e_1. Please use uniform notation and be clear.
Reply: Corrected.

Finally r_0 is the initial radius of the droplet, but by my reading, for the purposes of this manuscript r_0 has been set to equal the dry aerosol particle diameter. However, we know that at deliquescence (DRH) any soluble aerosol particle will have a sharp transition terms of growth factor (GF). For example at DRH the GF for NaCl jumps suddenly from 1 to _ 1:6$_1$. How is this discontinuity accounted for? Even for mineral surfaces one would expect the r_0 to be potentially, importantly different when it is completely coated in bulk water versus when it is dry or just has adsorbed water present.
Reply: The r_0 sizes are already CCN sizes. We repeat the sentence already described in section 2.2.

The potential INPs are assumed (i.e., sub-saturated particle growth is ignored) to activate to droplets because they are greater than cloud condensation nuclei sizes (Seinfeld and Pandis, 2016) and grow as long as RHw is increasing or remains constant.

_ (5,143) Figure 2 is referred to but I believe the intent is Figure 4a perhaps?
Reply: Thanks. Yes, it is Figure 4a. The typo is corrected.

_ (5, 154) Figure S2-5: I found myself spending a lot of time digesting these figures and wonder if the authors should revisit what in fact **is best to include in the main text**. Perhaps they **might hybridize some current figures to add some detail to the main text** that only appears now in the supplement.
Reply: An example is already included in the main text. See Figure 4b. A reference to other supplementary figures is included in the figure caption

I would also suggest that in Figures S2-S4 the authors choose different color maps for time and RH.
Reply: Thanks for the suggestion. This is tried but gets overly complicated to interpret the results. The choice of similar colormap is justified because then it is easy to compare the low and high values using consistent colors.

With 2 color maps and an offset perhaps panels b and c could potentially be combined. Even if not flipping between figures would be easier if the color maps differed.

Reply: Addressed above.

Figure S5 is missing a legend. Also in this figure the red droplet radius points seem problematic. Firstly, they seem to show a discontinuity at the chamber transition that none of the other curves indicate.

Second, one would intuitively expect their values to perhaps lie between the black and pink, but also the red temperature seems to be lower than the black as it gets close to the transition. Why does the particle further from the cold wall have a colder temperature than that which is closer? I find that a clear explanation of this figure, and especially the reason the red points stand out is lacking.
Reply: The legend is like in Figure S4. The following sentence is added to the caption.

Figure S5: *The plotted data line style and marker symbol are similar to the legend described in Figure S4.*

_ (6, 167) Table S1: replace very small with _ X.
Reply: Corrected.

_ (6, 169) evaporating droplet
Reply: Corrected.

_ (6, 175) 200 nm? Why not use 300 nm to match the simulations? Perhaps a comment on this choice would be useful.
Reply: The choice was based on the optimization of two factors: number concentration and monodisperse size. This size allowed us to generate the maximum number of monodisperse particles. Generating smaller sizes produces multiple charge particles, whereas generating larger size particles produces fewer particles. The following sentence is added.

Section 2.3: *The choice of this size allowed us to generate the maximum number concentration of monodisperse particles.*

_ (6,177) space betwen RHw and conditions
Reply: Corrected.

_ (6,180s) **See my comment** above with regard to the **OPC spectra and interpretation** based on water droplet size predictions. Also, as a reader it became confusing that the authors switched from discussing droplet radius to droplet diameter when they begin discussing OPC data. **I suggest** that one dimension is chosen and all discussions and figures converted to this for consistency.
Reply: The comments regarding ice crystal size and the relationship between droplet and ice crystal size related to the nucleation section temperature are addressed above.
We revised Figure 5a such that Yaxis shows the particle units in radius, and it is now consistent with the other figures.

The following figure is added.

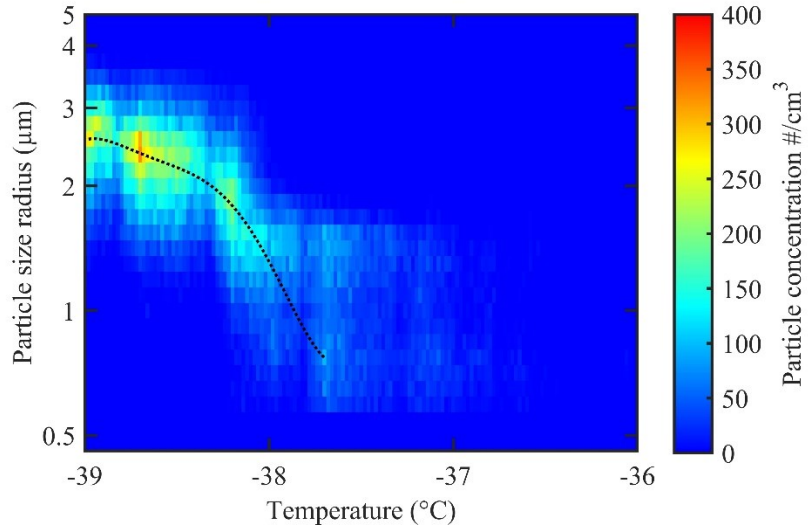

Figure 5: Homogenous freezing of water droplets containing one wt. % ammonium sulfate solution. (a) OPC classified ice particle concentrations as a function of ice crystal diameter at different temperatures. Warm and cold walls of the conditioning section are maintained at -9 and -27°C, respectively.

_ (6, 183) How are ice particles of 2.0 _m observed, when previously it was stated a cutoff of 3 _m is used to select ice? Is this a result of my radius versus diameter confusion?
Reply: The cutoff of 3 µm is defined in diameter. The new sentence in section 2.1 reads as

Section 2.1: *… certain size-threshold (≈3 µm **in diameter**).*

We also revised the cut size definition in section 2.3. The new sentence in section 2.3 reads as

Section 2.3: *… we observe ice particles of size ≈ 2.0 µm **in diameter**.*

_ (7, 200) "It can be seen that RHw values close to 113% are required before all the AS particles are activated to droplets." I believe the observation is of ice, not of droplet activation. The DRH of ammonium sulfate is _ 82% and weakly dependent on temperature. Thus all ammonium sulfate particles should activate at much lower RH. The value of RH here is what is needed for them to grow to a size, subsequently freeze, and remain big enough to be measured as ice.
Reply: Agree. The sentence is revised as follows. The original sentences that follow this sentence discuss the importance of high RHw conditions.

Section 2.3: *It can be seen that RHw values close to 113% are required before all the AS particles are activated to droplets **and measured as ice crystals** (Figure 5b).*

_ (7, 207) Here again another size 400 nm mobility diameter particle is used, perhaps a word as to why this choice was made, relative to the 200nm or 300 nm used in other contexts in the text?
Reply: Following words to the existing sentence are added.

Section 2.4: Laboratory measurements showed that the contribution of double and triple charged particles was less than 7 and 3%, respectively, **which also justified the choice of 400 nm size particles.**

_ (7, 211) 7% and 3%

Reply: Corrected.

_ (7, 222) was once .....is now
Reply: Corrected.

_ (7, 225) I suggest the authors stick with SI units – mph to m/s.
Reply: Corrected.

_ (8, 232) particles were also collected.... Were the same particles collected on the SEM films after the CFDC or was this sampling run in parallel?
Reply: It was run in parallel. Highlighted words are added, and the existing sentence is revised as follows.

Section 2.4: **In parallel to INP measurements,** the particles were collected on a carbon type-B film (Ted Pella Inc.; 01814-F) for scanning electron microscopy-energy dispersive x-ray spectroscopy (SEM-EDS) analysis to better understand the size distribution and composition of these airborne dust particles.

_ (8, 252) froze at the highest
Reply: Corrected.

_ (9, 263) See previous comment related to temperature ramping.
Reply: This comment is addressed above. See new figure Figures S8 caption.

Figure S8: *The F_{ice} of airborne arable dust species as a function of temperature and nucleation section cooling rates. The cooling rate of 0.5 Kmin^{-1} was used in this study.*

_ (9, 265) allows a comparison with other....
Reply: Corrected.

_ (9, 271) But citations in order from earliest to latest.
Reply: Corrected.

_ (9, 278) Here error in n_s is mentioned but does not lead to any uncertainty plotted in Figure 7. In addition to the error bars plotted from other studies it would be nice to have error bars plotted for this study.
Reply: The errors are plotted but they are invisible in the figure. For example, for K-Feldspar species, the n_s value at -22°C is 0.1083x10^{12} (m^{-2}) and the error is 1.613x10^{7} (m^{-2}).

_ (11, 339) Perhaps the authors could spend some more time attempting to explain why their results seem to be systematically high relative to the other studies (Figure 7). Are there good physical explanations for this?
Reply: In addition to the different measurement methods that might have led to this discrepancy (already discussed in the main paper); it is also possible the experimental uncertainties from different ns parameters (e.g. ice crystal detection limit, RH, and temperature error limits) could also influence the ns calculations. The following sentence is added.

Section 3: *The experimental uncertainties (e.g. ice crystal detection limit, RH, and temperature error limits) from these methods could also influence the n_s results.*

_ (conclusion) From the conclusions I am missing a discussion of whether other existing CFDCs could employ this technique. What for example are the physical constraints in terms of evaporation section length? Given the published geometries of instruments like ZINC[2], SPIN[3] etc. could these instruments hope to run using the operational mode introduced here? Alternatively, if new chambers were being designed what features should be introduced or geometry utilized to enable operation in both traditional and this new mode? Recommendations to the community would strengthen the paper.

Reply: Yes, other CFDC's could employ this new method. Based on CFD results (Fig.S3-5), the minimum evaporation/nucleation length required is 0.2 m. Implementing a separate refrigeration system to independently cool the nucleation section, the new operation mode can be adapted. For a new chamber geometry, the length of the conditioning section can be increased such that droplet size can be increased. This feature is useful such that the lifetime of the ice layer can be increased because higher RHw = 113% is not needed.

The following sentences are added to section 3.

Section 3: *Our results can guide design considerations for future CFDC-style ice chambers. The length of the conditioning section can be increased so that higher RHw would not be necessary to activate all the particles to sufficiently large droplet sizes (≈ 2 µm in diameter). This design feature could help to increase the lifetime of the ice layer. Based on CFD results (Fig.S3-5), the minimum evaporation/nucleation length required is 0.2 m. Also, implementing a separate refrigeration system to independently cool the nucleation section, the presented new operation mode can be adapted.*

_ (Figure 1) Can basic chamber dimensions be included, space appears plentiful.

Reply: We added the following sentence to the figure caption of Figure 1.

Figure 1: *The length of both the conditioning and nucleation section is 0.45 m. The width of the chamber is 0.15 m. The gap between warm and cold walls is 0.01 m.*

_ (Figure 2) Why include temperature from when initial cooling began? Why not just the shaded region, or shaded plus rewarming?

Reply: This is shown to give an idea of temperature time-series from the beginning of the experiment.

_ (Figure 5a) See previous comment with regard to radius versus diameter. Also, why the arbitrary scale? Is this a result of OPC binning? Can scale be changed to be linear? This plot is very hard to interpret in its current form.

Reply: Figure 5a is revised, please see above. A new figure is added that shows the Y-axis in particle size in radius units.

The scale is fixed. It was plotting typo. Adopting a linear scale makes the figure difficult to analyze. The new figure is clearer.

_ (Figure 6 caption) Suggest a change in text: Other solid square markers represent data collected when the chamber was operated in a steady-state temperature mode (instead of steady cooling).

Reply: Thanks for the suggestion. The sentence is revised.

Summary:

I have enjoyed reading the submitted manuscript and find that this is an intriguing new idea. In order to recommend the manuscript for publication I think the authors **need to state more convincingly** that

they constrain the conditions for the observed nucleation. Furthermore, I think the conclusion would be significantly enhanced by describing **whether or not other existing CFDC systems** could run or test run such a mode of operation. I also encourage the authors **to conduct a round of editing** to ferret out small mistakes that I found numerous enough that not all could be included here.
Reply: Thanks for these comments. The ice fraction is defined, see Text S1. The design recommendations for future CFDC chamber development are described in section 3. English editing was performed.

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[1] Castar`ede, D. and Thomson, E. S. (2018). A thermodynamic description for the hygroscopic growth of atmospheric aerosol particles. Atmospheric Chemistry and Physics, 18(20):14939–14948.
[2] Stetzer, O., Baschek, B., Lueoeond, F., and Lohmann, U. (2008). The zurich ice nucleation chamber (zinc) - a new instrument to investigate atmospheric ice formation. Aerosol Science and Technology, 42(1):64–74.
[3] Garimella, S., Kristensen, T. B., Ignatius, K., Welti, A., Voigtl ¨ander, J., Kulkarni, G. R., Sagan, F., Kok, G. L., Dorsey, J., Nichman, L., Rothenberg, D. A., R¨osch, M., Kirchg¨aßner, A. C. R., Ladkin, R., Wex, H., Wilson, T. W., Ladino, L. A., Abbatt, J. P. D., Stetzer, O., Lohmann, U., Stratmann, F., and Cziczo, D. J. (2016). The spectrometer for ice nuclei (SPIN): an instrument to investigate ice nucleation. Atmospheric Measurement Techniques, 9(7):2781–2795.