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 #RC3

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The Kulkarni et al, study describes a newly developed operating procedure for investigating the immersion freezing mechanism using continuous flow diffusion chambers. The new method converts the typical nucleation section of such chambers into a "conditioning" section where the aerosol particles are activated into cloud droplets at a fixed temperature where no freezing is expected. Then the particles transition into the newly dubbed "nucleation section" (formerly known as the evaporation section), which is cooled continuously while maintaining ice saturation. The newly developed technique compares well with previously published immersion freezing methods, although it appears to produce higher frozen fractions (within an order of magnitude) than previously observed for several dust species. I find the new method to be well implemented and a nice addition to the ice nucleation measurement community. I support this manuscript for publication and have the following comments:

General comments:

The residence time of the instrument is described as _10 seconds, yet the actual nucleation section is only half of that. This is not that different from traditional CFDCs, however, when the lifetime of the evaporating droplet in the nucleation section is considered, the nucleation time seems closer to _2 seconds (according to the numerical simulations). This should be noted in the text.

Reply: Following sentence is added. The word 'particle' is added to say that total particle residence within the chamber is ~ 10 s.

Section 2.1: …which limits the total **particle** residence time to ≈10 s. The droplet residence and nucleation time within the chamber are a maximum of 6.5 s and 2 s, respectively.

Furthermore, when considering that the droplets evaporate so quickly, is it possible to retrieve some information about nucleation rates based on the observed ice crystal sizes as a function of temperature, as was alluded to for the homogeneous freezing experiments?

Reply: This is another way of expressing INP measurements (Herbert et al. 2014). We know the ice fraction and particle surface area; however, nucleation time is uncertain. These inputs can be used to calculate the nucleation rate (J_{het}). Alternatively, a normalized freezing rate (R/A) can be calculated. We hope to provide the raw data upon request, and this data information would allow readers to calculate these rates.

Herbert, R. J., Murray, B. J., Whale, T. F., Dobbie, S. J., and Atkinson, J. D.: Representing time-dependent freezing behaviour in immersion mode ice nucleation, Atmos Chem Phys, 14, 8501-8520, 10.5194/acp-14-8501-2014, 2014.

Throughout the text, the new method was described as “the new method”. I think it would be nice if the new technique had a name for easier future reference.
Reply: We call this new technique as ‘Modified Compact Ice Chamber’ or ‘MCIC.’ The manuscript is revised, and sentences are revised to incorporate MCIC.

Section 2.1: *Figure 1 shows a vertical cross-sectional geometry of the modified mode PNNL ice chamber,* ***which is now referred to as a Modified Compact Ice Chamber (MCIC).***

Section 2.4: *The immersion freezing efficiency of K-feldspar, illite-NX, Argentinian soil dust, and airborne arable dust particles was measured to test the performance of the* ***MCIC****.*
Section 3: A good agreement with the results obtained from ***MCIC*** *was observed, …*

Section 3: ….4 up to 5 is needed to apply to the CIC-PNNL data to match with the data from the **MCIC**.

Section 4: An alternative method of operating a CFDC-style ice chamber **referred as MCIC** was explored to …

I appreciate that the authors did a thorough evaluation of the instrumental design using CFD and pulse experiments. However, I found the description and justification of the settings used missing, see my comment below.

Although the authors go in depth in their comparison with the dusts tested with previous results, I found the justification for the observed differences to be rather vague. This is especially true when comparing with the observations from the FIN workshop where to my understanding, the same aerosols were being tested at the same time. Therefore it would be nice if the authors expanded on some of the reasoning as to why the results in ns can differ by up to an order of magnitude. For example, is it due to not all particles being activated in other techniques due to lamina issues or perhaps it is due to the conditions that the droplets are evaporating at (warm wall temperature or cold wall temperature) etc.?
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 n_s parameters (e.g. ice crystal detection limit, RH and temperature error limits) could also influence the n_s calculations. 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 ns results.*

Technical and minor comments:

Line 38-39: There is mounting evidence that the traditional view of deposition nucleation, may not be occurring. As referenced in the cited Vali et al., (2015) deposition nucleation has also been referred to as immersion freezing in pores or pored condensation and freezing (Marcolli, 2014). Consider adding pore condensation and freezing as a heterogeneous nucleation mechanism.
Reply: Following sentence is added.

Section 1: *Deposition nucleation has also been referred to as pore condensation and freezing mechanism because it is similar to as immersion freezing but in pores (Marcolli 2014).*

Line 53-54: Consider adding Garimella et al., (2017) as a reference as well.
Reply: Added.

Line 57 and 60-61: Did you test to see if all particles did indeed activate as droplets?
Reply: This was tested by freezing the droplets at and below homogeneous freezing temperatures. See Figure 5b.

Line 77: Are there two sheath flows of 5 lpm of was the total sheath flow 5 lpm? Please clarify.
Reply: There is one sheath flow. The existing sentence is revised.

Section 2.1: The **single** sheath and sample flow rates were 5 and 1 liters per minute (LPM), respectively, …

Line 78: With such a high supersaturation and the required temperature gradient to achieve this supersaturation, how can you ensure that all particles activated as droplets?
Reply: This was tested by freezing the droplets at and below homogeneous freezing temperatures. See Figure 5b.

Line 91-93: Here the temperature gradient between the walls is mentioned and the achieved temperature of -20 C is described in the following sentence. However, it may be worthwhile to specify the supersaturation of the conditioning section here as well (113 % RHw?).
Reply: Following sentence is added.

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

Line 99-102: This should be reworded, consider something like: "The isothermal conditions of the nucleation section is maintained at ice saturation and cooled at a steady rate (0.5_C min-1 100) by a separate cooling bath in order to determine the immersion freezing efficiency of INPs as a function of supercooled temperature"
Reply: Thanks for the suggestion. The sentence is revised as follows.

Section 2.1: *The isothermal conditions of the nucleation section is maintained at ice saturation and cooled at a steady rate (0.5°C min^{-1}) by a separate cooling bath to determine the immersion freezing efficiency of INPs as a function of supercooled temperature.*

Line 102-103: Why does the experiment proceed so far below the homogeneous freezing temperature?
Reply: The experiment could have terminated at the onset of homogeneous freezing temperature (-38 to -39 °C). Cooling below this temperature allowed us to obtain measurements at homogeneous freezing temperature for ~10 minutes. This additional data helped towards quality control and to account for the uncertainty within the temperature. The following sentence is added.

Section 2.1: *This additional supercooling below the onset of homogeneous freezing temperature allowed to obtain freezing data that was used towards data quality control and to account for the uncertainty within the temperature.*

Lines 110-112: Was there any gradient applied to the conditioning experiment during the pulse experiments? I find this unclear in the text. Furthermore, if a temperature gradient was applied in the conditioning section, are there any effects from the ice coating/ moisture from the walls on the buoyancy

profile of the air in the chamber that are missed by doing the test without an ice coating? Also, are there any impacts on the lamina of the chamber when going from the conditioning section to the nucleation section when there is a temperature gradient of 22 C (-20 to -44 C)?
Reply: There was no gradient applied to the conditioning section of the chamber.
Flow conditions across the chamber are laminar (see Fig. 4a). The INP trajectory determined by the various forces (flow conditions and gravity) acting on the particle follows the fluid flow streamlines. Figure 3 shows the steady-state airflow velocity within the conditioning section of the chamber. These results indicate that the chamber conditions do not affect the buoyancy profile of the air. Therefore, particle pulse experiments are also valid after ice coating.

Figures S2-5 show no effect of the temperature gradient between the conditioning and nucleation section temperature on the aerosol lamina within the conditioning section and transitioning zone.

Line 183: remove "either" before "do"
Reply: Corrected.

Line 184-186: Please clarify these sentences. Are the smaller droplets at higher temperatures due to the lower nucleation rate and therefore the droplets evaporate more than at colder temperatures where nucleation is faster?
Reply: The droplet evaporation is observed from -20 till -37.5°C, see Figures S2 – 4. These figures show that droplets evaporate at the entrance of the conditioning section. E.g. Fig S3 c show that water droplet 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).

Line 183: Remove "the" between "of" and "supercooled"
Reply: Corrected.

Lines 191-195: seem to be contradicting each other, consider rewording.
Reply: The sentences are revised as follows.

Section 2.3: *We find good agreement between the experimental and predicted freezing temperatures. These results also show the complete evaporation of supercooled droplets within the nucleation section, because no ice particles are observed above ~≈ 37.5°C, and therefore the freezing results (see section 3) at warmer temperatures (> -37°C) can be ascribed as the heterogeneous freezing of the droplets or immersion freezing.*

Line 200-224: Consider breaking this sentence in two for easier readability.
Reply: The sentence is divided into two sentences for clarity.

Section 2.3: *Higher RHw values enable the encapsulation of all particles that are within and may spread outside (Garimella et al. 2017) the width of aerosol lamina into droplets.* ***In addition,*** *high saturation conditions also help to grow the droplets to the larger size; so, they survive long enough to induce the freezing of droplets within the nucleation section.*

Line 206: Rather than stating “a new mode” perhaps consider stating that it is operated in this specific mode (name the mode).
Reply: We call this new technique as ‘Modified Compact Ice Chamber’ or ‘MCIC.’ The manuscript is revised, and sentences are revised to incorporate MCIC.

Line 223-224: Consider rewording.
Reply: The sentence is revised as follows.

Section 2.4: The region was once covered with basalt lava, but is now built up with loose topsoil – loess.

Line 245-246: Earlier, it is stated that an experiment ends at -44 C yet now the experiment ends at -38, which makes more sense, be sure to be consistent.
Reply: Sorry for the confusion. Although the experiment ends at -44°C, the INP data from -20 to -38°C is only investigated and presented in this study.

References

Garimella, S., Rothenberg, D. A., Wolf, M. J., David, R. O., Kanji, Z. A., Wang, C., Rösch, M. and Cziczo, D. J.: Uncertainty in counting ice nucleating particles with continuous flow diffusion chambers, Atmos Chem Phys, 17(17), 10855–10864, doi:10.5194/acp-17-10855-2017, 2017.

Marcolli, C.: Deposition nucleation viewed as homogeneous or immersion freezing in pores and cavities, Atmos Chem Phys, 14(4),2071–2104, doi:10.5194/acp-14-2071-2014, 2014.

Vali, G., DeMott, P. J., Möhler, O. and Whale, T. F.: Technical Note: A proposal for ice nucleation terminology, Atmospheric Chem. Phys., 15(18), 10263–10270, doi:10.5194/acp-15-10263-2015, 2015.