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 #RC1

Referee comments on "A new method for operating a continuous flow diffusion chamber to investigate immersion freezing: assessment and performance study" by Gourihar Kulkarni, Naruki Hiranuma, Ottmar Möhler, Kristina Höhler, Swarup China, Daniel J. Cziczo and Paul J. DeMott

Overview:

This paper is a useful addition to the literature on INP measurements in general and to the many reported uses of CFDC instruments in particular. A new mode of operation for a CFDC-type instruments is proposed and evaluated in the paper. In this mode, immersion freezing measurements over a range of temperatures are obtained with steady cooling rather than in the more customary mode of single temperature or step-wise cooling. What is called the evaporation section for many CFDC instruments is changed to nucleation section in this paper.

The proposed method puts emphasis on the temperature dependence of INP activity whereas much of the CFDF literature deals with the dependence of nucleation on humidity, although there is a large range of types and operating modes of CFDC instruments (cf. Hiranuma et al. 2015, with Supplement). The question of the relative importance in these chambers of activation via deposition or freezing is sidestepped in the current paper. It is also set aside in these comments because of the general view that immersion freezing is dominant in most cases.

From an operational point of view, the evaporation of the drops at the low RHw of the nucleation section avoids the possibility of droplets being counted at the outlet. This avoids one of the common problems with CFDC instruments.

The authors have done a number of tests to support the results presented and examined some potential error sources. However, probably because the approach is new, additional questions arise and some aspects of the measurement method require further scrutiny.

Reply: Thanks for the reviews and feedback.

Exposure time and temperature:

This issue can be addressed principally on the basis of the simulations presented in Section 2.2 of the paper and in the Appendix. According to these calculations droplets rapidly decrease in size at the same time as the temperature adjusts to the temperature of the nucleation chamber, Tnc. The minimum droplet sizes shown occur when the temperature is within about 1 C of Tnc. Furthermore, the comparison in Fig.

S5 shows that variations in the entry position of the aerosol do not add further errors. From these results it would follow that all droplets reach the set temperature of the nucleation sections within errors comparable to other instrumental uncertainties.

However, the simulations are for ideal laminar flow. **To what extent is this actually the case? How much extra spread is caused by deviations from the ideal flow and by polydisperse INP sizes?** Larger drops might evaporate later but the temperature they reach would not differ from the set value. But, if there are droplets that evaporate faster than the simulated values, these would have a higher minimum temperature of exposure and that would lead to underestimates of the final results. Since Fig. 7 shows that derived ns values are higher than those reported in other papers for three out of the four samples tested, it appears that there is no major problem in this regard.

Reply: Agree, these simulations are for ideal laminar flow. To quantify the particles outside of the lamina, pulse test experiments using monodisperse particles are performed. These results are presented in Figure S1 in the original manuscript.

The following text and figure are added to the revised manuscript.

Section 2.1: *Simulations (see below) are performed to investigate the sensitivity of polydisperse particles. The particle residence time of three different monodisperse particles (0.3 µm, 1.0 µm, and 2.0 µm) traversing the chamber is calculated. Results show that the residence time of these particles is similar indicating monodisperse size pulse experiments are also applicable to other size particles.*
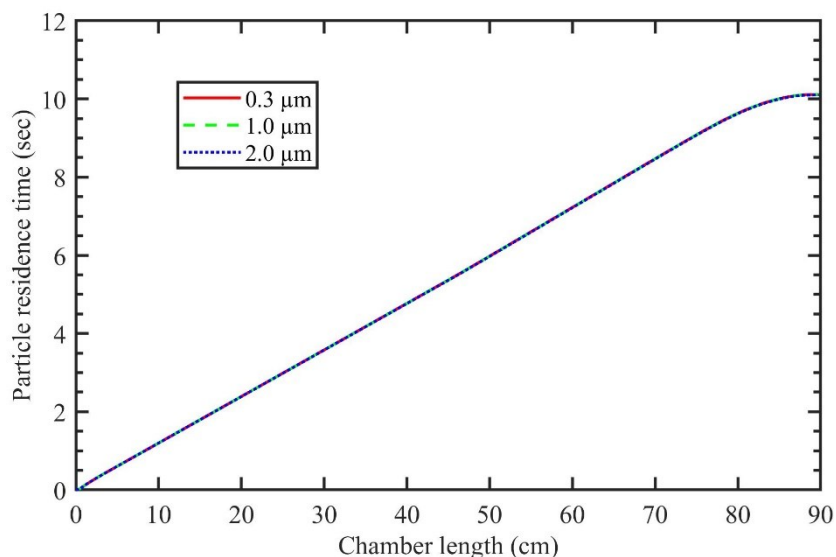
Figure S1: Particle residence time of different size droplets within the chamber.

More importantly, the short exposure time of INPs to the coldest temperature necessitates consideration of the time dependence of nucleation. Cooling rates of the droplets when entering the nucleation section approach 10°C sec[-1] for the lowest Tnc value. For such rapid cooling, Eq. 5 from Vali and Snider (2015) with ξ = 0.3 indicates a 2°C shift toward colder temperatures compared to a 1°C min[-1] rate of cooling, i.e. the same activity would be observed with 2°C additional cooling[1]. With that correction, **the current data in Fig. 7 would have to be represented by points shifted to the right** to bring the comparison on the same basis as the other data, although the exact cooling rates associated with each data set from the literature would have to be considered as well.

The tests with constant temperature of the nucleation section (lines 274-281 and solid squares in Fig. 6) do not address the point raised above. This is because the 0.5° C min[-1]cooling rate is negligible compared to the rapid cooling of the drops on transition from the conditioning to the nucleation section.

While there is no a priori reason for assuming that activity has to rise exponentially, it is also worth considering **whether rapid cooling in these experiments may explain why** the slopes of the ns versus T data points in Fig 7 flatten out at colder temperatures. As can be seen from the Figs. S2 to S5, the lower Tnc is, the faster the cooling is and thus **larger corrections (moving points to higher temperatures) would be necessary to normalize the data** to a fixed cooling rate.

[1]There is no empirical evidence to support the use of the equation for cooling rates 600 times over the reference value, but there is no other basis at this time to make a better estimate.

From the above it follows that the rapid cooling occurring in the transition from the conditioning section to the nucleation section **influences** both the magnitudes of the derived ns values and the slopes of the temperature spectra. **The authors' view of this would make the paper more complete**.

Reply: We appreciate these comments. Previously, time-dependent immersion freezing framework (e.g. Vali and Snider (2015), Herbert et al. (2014)) had been formulated to understand time dependent nature of ice nucleation. The framework allows to correct the shift in temperature towards colder temperature for a given change in cooling rate. The cooling rate constant ξ depends on the nature of the INP population, and this constant varies from ~0.15 to 1.6 (Table 2, Herbert et al. (2014)). In this work, we investigated various test species (K-feldspar, airborne soil dusts from the arable region, illite-NX, and Argentinian soil dust), and the ξ values for each of these species for the droplet conditions (size and one INP per droplet) that are used in this work are unknown. Therefore, the application of such an empirical relationship to correct for the shift in temperature because of the droplet cooling rate is not possible currently. These parameters can be obtained by conducting immersion freezing tests using direct processing (e.g. CFDC style instruments) and post-processing (e.g. BINARY style instrument) in parallel.

It should be noted that our experiments shown as solid squares in Fig. 6 do indicate the minimal influence of rapid cooling on ice fraction or n_s values. In this experiment, the nucleation section is held constant at one temperature, and while the droplets are transitioning from the conditioning section to the nucleation section, the droplets undergo rapid cooling. Please see the discussion in section 3.

We added the following paragraph to acknowledge the possibility of influence on reported temperatures because of rapid cooling.

Section 3: *Further, the time-dependent immersion freezing framework (e.g. Vali and Snider (2015), Herbert et al. (2014)) suggests that the rapid cooling of the droplets could shift the cumulative ice fraction towards the colder temperature based on the cooling rate and particular INP material constant. However, these input parameters for the time-dependent model are not available currently to quantify the temperature shift for the present experimental conditions. Future studies that involves collocated direct and post-processing INP instruments would be needed.*

Sensitivity and error analysis:

The paper states that it is possible to execute three test cycles before icing problems. It also states (line 256) that full temperature spectra were acquired in about 30 minutes. However, information about the **input aerosol concentrations** used in the tests wasn't readily found in the paper. The temperatures of the

tests for the airborne dust were restricted to -28°C and colder. It would be useful **to know more about sample concentration** (in terms of active number at test temperatures), and **sampling duration requirements versus statistical counting errors**. Perhaps this sort of analysis formed the basis for the accuracy estimates indicated on lines 263-265 of the paper but it is unclear if that is the case.
Reply: We added the following sentence.

Section 2.4: *The input aerosol concentration of all four INP species varied from 100 to 800 # per cubic centimeters, and the sampling duration was ~30 minutes.*

The statistical counting error is considered by calculating the standard deviation of the ice fraction measurements. This is discussed in the original manuscript on line 247.

Minor points:

line 21 and other places: Is arable dust a soil science definition? Perhaps the meaning of the term could be clarified for the context used here. Desert dust? Top soil? Agricultural dust? A detailed description of the sample is given on lines 240 on but the term is used already in the abstract and is frequently used in the paper prior to the definition.
Reply: We modified the definition. The revised definition (in bold) reads as follows.

Abstract: *The performance of the MCIC was evaluated using four INP species: K-feldspar, illite-NX, Argentinian soil dust, and **airborne soil dusts from an arable region** that had shown ice nucleation over a wide span of supercooled temperatures.*

Abstract: *… during the second phase of the Fifth International Ice Nucleation Workshop (FIN-02) campaign, and **airborne arable soil dust** particles were sampled…*

Section 2.4: The arable soil dust is defined as follows.

<u>Airborne soil dust from the arable region</u> or shortly airborne arable dust particles *were sampled at the PNNL sampling site during a regional windblown dust event.*

line 62: "sequence' might be better here than "spectrum".
Reply: Corrected.

line 164: The point about not simulating nucleation is mentioned because of the possible latent heat effect or some other argument?
Reply: CFD simulations described in section 2.2 do not involve droplet freezing (or nucleation of ice) simulations. This is not performed because the objective of numerical simulations was to better understand the flow behavior and their impact on droplet dynamics (growth and evaporation). The following sentence added to section 2.2 clarifies the goal of this section.

Section 2.2: *At the entrance of the nucleation section, the temperature and RHw profiles can be unsteady, and to better understand the flow patterns of these profiles within the transitioning zone, and its impact on droplet behavior, numerical simulations using computational fluid dynamics (CFD) are performed.*

line 290: The approximation indicated is valid only for Fice << 1:0
Reply: Correct. This assumption of approximation is mentioned in the main text.

Section 3: *The approximation is valid for ice fraction << 1.0.*

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