

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 S 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 CFDC 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 RH_w 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.

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, T_{nc} . The minimum droplet sizes shown occur when the temperature is within about 1°C of T_{nc} . 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 n_s 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.

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 T_{nc} value. For such rapid cooling, Eq. 5 from Vali and Snider (2015) with $\xi = 0.3$ 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 cooling¹. 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 n_s versus T data points in Fig 7 flatten out at colder temperatures. As can be seen from the Figs. S2 to S5, the lower T_{nc} 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.

¹There 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 n_s values and the slopes of the temperature spectra. The authors' view of this would make the paper more complete.

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.

10 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.

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

15 line 164: The point about not simulating nucleation is mentioned because of the possible latent heat effect or some other argument?

line 290: The approximation indicated is valid only for $F_{ice} \ll 1.0$