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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Uncertainty in counting ice nucleating particles with continuous diffusion flow chambers" *by* Sarvesh Garimella et al.

Anonymous Referee #1

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General Comment Overall this a well written and very useful paper, if a bit finely focused only on the effect of lamina entry and spreading as a source of uncertainty. Nevertheless, the paper does suggest that this is a major factor of uncertainty in INP measurements with such devices. Two things stood out strongly to me as needing some attention. First, no description of the ice growth model was given, including equations and assumptions on condensation coefficient for ice growth, habit, or at what size particles were started for growth. Second, no demonstration data is given to provide confirmation the errors predicted by the particle transfer studies using ice nucleation data collected in the chambers. Are there any conditions for which higher RH operation is possible in order to confirm that activation appears as predicted in the SPIN instruments (i.e., data consistent with a cf of 2.6-9.5 times)? Past data seems to exist for ZINC in workshops. Can any of these data be utilized? This might make a nice

addition, although I realize that determining a reference RH value could be problematic. Showing an RH scan could at least be useful.

Minor revisions are recommended in general. Specific questions/comments for potentially addressing are listed below.

Specific Comments

Abstract

Line 13: suggest "instrument theory of operation"

Line 14-16: The sentence ends oddly following the "and". This is a product of the machine learning or the combination of flows? Should there be separate sentences here?

Introduction

Page 3, line 20 – Concerning the need for accurate and unbiased measurements, the need exists, but it may or may not be achievable. Why not frame it as "assessing the accuracy and bias of such measurements"?

Page 5, line 3: The referenced study of DeMott et al. (2015) did not only discuss the effect of aerosol spreading outside the lamina, so perhaps this should read "discussed the effect of aerosol "spreading" outside the lamina and other possible factors that together result in a low bias. . .". This is the first mention of the hypothesis that it is the lamina spreading that is primarily responsible or most important.

Methodology

Page 6, lines 6-8: Is there a reason that this ZINC test is done at such a low temperature below that of the mixed phase cloud regime where one thinks of the need to exceed water saturation. Was the thinking just to cover a broad range?

Page 6, lines 14-17: Is there any reason to test different sizes of particles for transfer?

For example, would there be any difference expected for 100 nm versus 1000 nm for whatever phenomenon is responsible for spreading outside of the lamina or whatever leads to undercounting?

Page 6, lines 18-19: In creating a lamina profile of particle arrival times, it is not clear how you decide the position of a particle coming from either side of the central lamina.

Page 7, lines 20-21: What thermodynamic variables besides wall temperatures are meant? What other thermodynamic variables are available in the upper part of the chamber. Why not provide a list of all "features" in a supplemental table?

Page 7, line 22: Where the lamina is “initially” encased within the sheath flows?

Results and Discussion

Page 8, line 3: Both walls at -20 °C and with what uncertainty?

Page 8, line 7: Ice saturation ratio calculated in this case for average wall temperatures?

Page 8, line 18-19: The non-ideality in DeMott et al. was reported for a specific aerosol. Issues of response of aerosol to the chamber conditions is not discussed herein, and one can imagine that it is different for Snomax than for dust particles. Additionally, it could depend on particle hygroscopicity and so forth. Hence, that work was not an expansion on the work of Tobo et al. It regarded a different aerosol entirely, and a point made was that it was not clear if corrections translated to any aerosol. Since this is what is promoted in the present paper, you may wish to make this point.

Page 9, line5: You could perhaps say more here. For example, that the cf could depend on instrument control factors, so would require RFR analysis for any given CFDC.

Page 9, lines 7-9: Since homogeneous freezing is a clear rate process, you would expect a distribution of ice crystal sizes even in that case, no? This could depend on the exact temperature and the nucleation rate at that temperature. It will likely

be monomodal, but I would not expect it to be monodisperse unless you can state the temperature and nucleation rate and show that all particles would be expected to freeze within a very short time after entry into the chamber. I think this paragraph needs some rewriting.

Page 9, lines 16-18: As above, would particles freeze instantly? Even considering their adjustment time in RH and T to chamber conditions that must amount to a second or more?

Page 9, line 23: It is not intuitive in consideration of expected ice crystal growth rates that 4 micron ice particles are expected to be the product at -40 °C. Is it due to the high supersaturation? What is the lamina residence time in ZINC? How large are particles assumed when they freeze? What is the condensation coefficient? What growth rate equation is used? Are spherical ice crystals assumed? There is much missing here in order to evaluate the statement made.

Page 10, lines 8-9: See questions above. How could one possibly know so well the sizes of ice crystals expected if you do not know the deposition coefficient for ice well enough to know what to specify? Are edge effects in the OPC the only explanation of ice crystals less than 3 microns in size? That size could represent a many second or more growth time at -40 °C. And one must assume spherical ice crystals to claim to know the sizes very well.

Page 10, line 14: Does a perfect immersion freezing nuclei imply that they are fully dilute at a condition of a water activity of 1? Most CCN are not activated at 100 percent RH, but immersion freezing can happen before activation if the temperature is cold enough. I think it may have to be stated as a highly idealized and possibly unrealistic example.

Page 11, lines 2-3: A number of 10 percent of aerosols freezing in ambient air is taken as realistic for ambient atmospheric conditions? Under what conditions? Again, I think one can say this is for demonstration purposes, not meant to simulate a real case

except one that might be found in a laboratory or at very low temperatures such as for cirrus parcels.

Page 11, line 12: “of droplets”

Page 11, line 16: perhaps “we propose due to the primary importance of the particle spreading effect.” Otherwise you are interpreting another study that discussed multiple processes potentially at play.

Conclusions

Page 11: A general comment noted in the above summary comments. It was surprising that no actual ice nucleation data are shown in this paper to support that the spreading effect is realized in the same manner as reported by DeMott et al. (2015).

Page 12: Regarding the comment about instrument geometry, you may wish to say what you mean. For example, some instruments are parallel plate, with lamina edges, while others are cylindrical in design.

Page 12, lines 4-6: There was little exposition given to the idea that small scale flow features at the point of aerosol injection and sheathing are responsible for the observed spreading of particles outside of the lamina. Fluid dynamics simulations might be advised in the future. Perhaps you should say that the only reason for non-ideality explored here was related to sample injection methods. A full analysis might also include particle compositional variability as well, since many INPs are thought to be relatively hydrophobic. One might also ask how the noted behaviors impact "deposition" nucleation?

Page 12, lines 11-14 and beyond: I suggest some revision to the statement here. CFDCs have needed to be operated at higher RH than expected values to inspect immersion freezing. However, an RH value of say 102 percent is not non-physical for Cu clouds, and 106 percent may not be either in wave clouds or very strong elevated convection. Of course, this discussion might be easily resolved by saying the super-

saturations are higher than expected for immersion freezing of most particles, rather than stating realism for the atmosphere. Furthermore, S-liq = 1 is not the threshold for immersion freezing. It can be higher or lower in dependence on particle hygroscopic properties, particle size, and temperature, and data in the literature demonstrate this.

Page 12, line 13: INP number concentrations.

Table 1. Please explain some terms better. For example:

1) Lamina saturation at TC3 means using the temperature difference across walls at this elevation to calculate the saturation profile there? Likewise TC4, etc...? 2) What does total volume flow at mass flow controllers mean? Where else do you know it? 3) Probably need to explain the heater concept, since some CFDCs do not heat their coolant.

Figure 1. If you were to draw the sample to scale, would it represent such a small fraction of the flow cross-section?

Figure 3 caption: Don’t particles also migrate to higher and lower temperatures?

Figure 4 and caption. Why such a narrow range of total flow, or if constant, why does it vary so much? Why would that be important and why would it even vary? There is a need to state conditions for which these data are collected, that it is for 1-sec pulses, etc...

Figure 9 caption: S-liq > 1.07 is the droplet breakthrough point for what SPIN temperature, or is it uniform? Also, I do not really get what is shown in panel b as a “composite of black and blue traces” from panel a. Why not just say that what is observed by an OPC is shown in panel b?

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