

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

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We would like to thank both Reviewers for their careful reading and thoughtful comments on our manuscript. We have made the changes they suggested and provide a point by point response below with the comment followed by the response. We believe this is a much improved paper as a result.

Reviewer #2

(overview paragraph omitted for clarity)

General comments

1) The introduction of the correction factor c_f for the SPIN CFDC measurements is important for the experimental determination of INP concentrations. It should be men-

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tioned already in the abstract that a mean correction factor of ~ 4 is determined for this SPIN instrument, and that the correction factor is highly variable between 3 and 10. The large uncertainty of individual INP measurements due to the large uncertainty of c_f should be mentioned in the abstract and discussed in detail in section 3.

We agree with the reviewer and explicitly state “We find here variable correction factors from 1.5-9.5, consistent with previous literature values.” in abstract. Not limited by length, we have included additional wording in section 3, per both reviewers suggestions. This is the paragraph starting with “DeMott et al. (2015). . .”

2) The measurements concern immersion freezing experiments but the spreading is likely to be present for deposition freezing experiments as well. Please discuss the influence on INP measurements in the deposition freezing mode. Should the same correction factors be applied?

Please note that a similar comment was made by Reviewer 1. We now explicitly call for depositional regime work in the conclusions. In the introduction we add “In this work we specifically considered effects in the regime supersaturated with respect to liquid water (immersion freezing) but believe these results are also applicable in the sub-saturated regime (depositional nucleation) as well.”

3) While the paper is clearly written in most parts, some parts are imprecise and not well written. This concerns especially the first half of section 3 (pages 8 and 9, Table 1 and Figs 2–5, see specific comments below). The descriptions are not detailed and accurate enough and for several paragraphs it is difficult to extract the main message the authors want to convey.

In response to this point we have re-read and made changes in these sections. The Table 1 caption was re-written in response to this and a point by Reviewer 1. We believe these are more clear as a result.

For example, the description and interpretation of Fig. 4 is only 2.5 lines (p 8, l 3–5),

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and the exact purpose of this Figure does not become clear to the reader. Is it supposed to show that flame changes more or less erratically between 0.1 and 0.8 for conditions that are kept as constant as possible? What does it tell about the reproducibility and uncertainty of the INP measurements?

We have attempted to improve the clarity of this caption with the addition of “This figure illustrates that the ideal condition is not realized and that even within a few % of the nominal total flow of 9.8 lpm the fraction of particles in the lamina is not predictable.”

4) Can effects of thermophoresis be excluded? Do the aerosol particles potentially leave the theoretical aerosol lamina due to thermophoresis?

(re-ordered similar points)

6) The most likely reasons for the observed spreading effect and for the discrepancy between the ideal instrument and the real measurements should be discussed. Are uncontrolled eddy turbulences the main/only reason for the spreading?

The RFR method suggests the location in the chamber where the spreading effects are most highly correlated with. We now expand on this at the end of section 2. While this is consistent with e.g. turbulence in the aerosol injection region we can not rule out thermophoresis. “The reduced RFR subset included 65 variables including wall temperature, flows, and saturation conditions predominantly in the middle and top sections of the SPIN chamber (Garimella et al., 2016); this is the region of the chamber where aerosol is initially encased within the sheath flows. This suggests that turbulence or other small-scale flow features in this region are responsible for the spreading effect in the region where the particle flow is injected into the chamber. However, we can not preclude that other processes taking place in this region, such as thermophoresis, are not also partially responsible.”

5) A particle that moves slightly outside the central lamina but still in the yellow region of Fig 3 should still be activated and growing efficiently. Is the assumption correct that all

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particles that leave the central lamina once (and are therefore counted in the “late” tail of the pulse) are not activated and cannot be measured as INP (therefore necessitating the large correction factors)?

This point regards two different effects. First is the question if particles leave the lamina in which they are supposed to remain. We show that this is not the case in Figure 3-5 and the associated text. Particles that do leave the lamina may still activate but they will only do so for lamina conditions in excess of the “true” activation value for that particle. This is argued in the revised ice growth model in Section 3 and the idealized figures 8 and 9. In the case of Figures 8 and 9 this point is found in the text in the revised last two paragraphs of Section 3.

6) This point was moved up as it related to point 4)

Specific comments

1.) The manuscript switches frequently between the ZINC and the SPIN instrument and sometimes it is unclear which specific instrument is meant (e.g. Fig 2: pulses are shown for ZINC, Fig 3: SPIN results, Fig 4 which instrument? SPIN? (please include instrument name in Figure caption), Fig 5 SPIN, etc.).

Figure captions with instrumental data now explicitly call out SPIN or ZINC.

2.) p 5, l 20: Why did you use 1 second pulses for SPIN and 10 sec pulses for ZINC? Did you measure the CPC in pulse every time and are the blue and red trace in Fig 2 measured for the identical pulse? The blue pulse in panel A seems to be shorter than 10 seconds.

This is now clarified with “In the case of SPIN this was a 1 second pulse while for ZINC a ~10 second pulse was used with automated and manual valves, respectively.” The purpose of the pulse tests is to show the difference in arrival time between CPC in and CPC out. “Under ideal conditions, regardless of duration, this should correspond to an equivalent particle pulse at the chamber outlet. “

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How long is the transfer time through the SPIN and ZINC chambers?

In both cases this is ~10 seconds, given in the referenced papers that start this section, but not directly applicable here.

The example of Fig 2 does not seem to be a typical one: with $\text{flam} = 77.7$ and 76.2% it is much higher than all the values displayed in Figs 4 and 5. According to Fig 5, the most frequent flam is in the range of $10\text{--}15\%$; and the average flam is argued to be ~25% (see comment to Fig 5 below). Please display (also) the measured CN_{out} for such a more typical case.

There is some confusion here, which might have been cleared up with the addition of the instruments to the figure captions. The Reviewer is mixing two instruments and several sets of temperature and conditions. These figures are not meant to be equivalent, they show different regimes. We believe this is now clear with the aforementioned changes.

Does it make sense to present the percentages for flam with a decimal place?

We agree and have revised the precision throughout the manuscript.

3.) p. 6, line 13–19 and Figure 3: the description is not sufficient. In the Figure caption it is stated that the particle distribution is “measured across the chamber”. Is this true? In the text of p 6 it says that “combining the arrival pulse with the shape of the velocity profile the corresponding distribution of particles across the width of the chamber can be determined”. How is this distribution determined in detail? This seems to be a complicated matter to me that would require CFD modeling, etc.? Do you derive a different distribution for each measurement pulse? The term “measured across the chamber” would indicate that CN measurements are made at the end of the chamber at different distances from the cold wall. Please use such a term only if such measurements were actually performed.

In this context “across the chamber” is meant to indicate the position with respect to

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the lamina, not a direct measurement. Also, we do derive a different distribution for each measurement pulse. To clarify this additional text has been added in this location “Combining information from a measured particle pulse and a calculated velocity profile, the corresponding distribution of particles across the width of the chamber for that particular pulse can be inferred (Figure 3). Buoyancy effects on mean chamber flow and mean particle position are accounted for in the calculation of the velocity profiles (Rogers, 1988). The particle distributions are reconstructed by assigning the first detected particles to the maximum velocity position in the calculated flow profile and the assigning peak particle concentration to the calculated lamina position (about which spreading occurs). The particles in the tail of the pulse are assigned positions corresponding to their relative velocities, which are derived from their relative arrival times. Since particles in the arrival tail could fall on either side of position distribution, this ratio is assigned based on matching the exponential fit of the unambiguous portion of the data (between the initial particle arrival and the peak concentration).”

4.) Figure 4: The y-axis should range from 0 to 1. There are only 25 data points shown, the text talks about 30 data points. Are all data points displayed? It is stated that the tests shown in Fig 2 were done at $+20^\circ\text{C}$ (p8, l 3 and p6, l 14). Does this mean that there was no cooling applied and the chamber walls were at room temperature for these measurements? Are these conditions transferable to realistic flow conditions? It would indeed be interesting to see in how far the pulses change between a warm chamber at constant temperature and a chamber operating with the two different cold wall temperatures.

The y-axis of Figure 4 now has a range of 0-1. The text and figure also now both reflect the correct number of data points used in the figure (25). There was no cooling applied and the chamber walls for the tests in Figure 4, and they were at room temperature for these measurements. These conditions were chosen to examine particle spreading with the fewer variables (e.g. thermophoresis in the chamber, increased temperature variability, etc.) than with cooled walls. For comparison, Figure 4 shows data from

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a warm chamber at constant temperature, and Figure 5 shows data from a cooled chamber operating with various temperature gradients.

5.) Figure 5: There seems to be an error in the Figure: The y-axis scale of the panel on the right does not correspond with the histogram on the left. The text claims that the mean of the distribution of flam is at 0.25, in the graph on the left the mean seems to be around 0.15.

With the longer tail at higher values, the mean of the (skewed) flam distribution is 0.25, while the mode is indeed closer to 0.15. We chose to point out the mean for comparison to studies that did not examine the shape of the distribution (e.g. DeMott 2015), where an average value is reported.

6.) p 8, l 10: Here it is reported that values for flam range from 3 to 73 %. Why are values of 76 and 77 % reported in Fig 2? (difference between ZINC and SPIN?)

Yes; as with previous points we believe the caption change in the figures now clarifies this.

7.) Figure 7: The colors are hard to discern. Four colors are shown in the legend, but several other colors are shown in the graph. This is potentially because overlapping colors result in “new” colors? A different representation would be helpful, e.g. show the four probability distributions not as histograms but as line plots.

We agree with the reviewer and have replaced Figure 7 to show line plots instead of bars.

8.) p 9-10: The ice growth model is insufficiently described. What are the assumptions? How are things calculated? I agree with the other reviewer that this needs considerably more discussion.

We agree and refer Reviewer 2 to the points made by Reviewer 1. We have expanded this section by several paragraphs and have included the relevant references. We believe the model is more clear and the manuscript improved as a result.

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9.) p 10, l 13: flam and cf are not equivalent, but rather “inversely equivalent”.

“inversely” added

Technical comments

p 7, l 15: “import” “important”

Corrected

p 7, l 21: “middle-top of the SPIN” → “middle and top section of the SPIN”

Change made

p 11, l 12: “activation curves of droplets. . . “

Corrected

p 11, l 13: “black and blue” “red and blue”.

Corrected

p 11, l 18: “4. Conclusions” (section numbering)

Corrected

p12, l 17: “variability to be conducted”

We believe original wording is correct (‘to’ not needed)

p 27, l 3: “Unlike Figure 6” should read “Unlike Figure 8”

Corrected

p 27, l 15 : “black and blue” “red and blue”

Corrected

Additional References

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