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Interactive comment on "The SPectrometer for Ice Nuclei (SPIN): An instrument to investigate ice nucleation" *by* S. Garimella et al.

P. DeMott (Referee)

pdemott@lamar.colostate.edu

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General Comment This paper describes a new ice nucleation device with controlled temperature and humidity conditions in continuous flow. This represents a commercial development based around the continuous flow diffusion chamber concept. A great deal of effort has gone into this characterization, and this paper will likely be of broad interest to the community. I am therefore happy to provide the comments that follow. In my opinion, some key issues for this paper include:

1) This paper is at different times a description of a commercial instrument and at times a record of first developments and studies done by a devoted group of scientists to make this instrument more fully useful. It is difficult to tell where the former ends and the latter begins. Complicating this is a small amount of apparent advertisement that

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enters the discussion and conclusions. Some of this is inappropriate in my opinion. I will point out where in specific comments below. What would be helpful is to be a little more careful to state what the SPIN exactly does versus what appear to me to be developments by the first users and future developments (e.g., full automation of many operations). The instrument is not delivered with all of these capabilities, to my knowledge. Can some statement be made upfront that this paper will include a standard description and describe methods developed by this group to utilize the instrument in more advanced ways?

2) Promotion of "AgI" as a calibration ice nucleant, and the (to me) surprising correspondence with literature data, deserves closer inspection. I spent many years in the engineering and generation of "AgI" ice nucleants. One point of studies prior to my time, in the 1960's, was that AgI possesses some of its active characteristics due to impurities (Corrin et al., 1963; 1967). Hence my use of parentheses above. It is certain that the purity of samples sometimes led to vastly different ice nucleation results in laboratory cloud chamber studies. As a minimum, the way that AgI is generated in this study, its size mode (size dependence is clear in much past literature), and some assurance that this was directly comparable to previous studies is needed.

3) A number of details are suggested for addition with regard to the design and construction of SPIN, since this is the first technical description of the device, coming at a time when there is now a substantial literature record of investigations with these kinds of instruments.

4) I found the validation results using AgI versus RH, as well as the homogeneous freezing results not to take the more ideal form I would expect. Hence, some additional discussion and even a new plot (for homogeneous freezing results) may be needed, along with explanation for why these do not look like (my inference) results shown in Richardson et al. (2010) or DeMott et al. (2009), where direct comparison to predictions of homogeneous freezing are made.

5) Finally, there are many parallels of the current findings regarding reverse flow in the work of Richardson (2009). While that was a dissertation and not a peer-reviewed publication, it is available through Colorado State University libraries and was delivered to some of the authors previously. The figure showing a line denoting the reverse flow regime needs to state the flow rate and pressure for which calculations are made. Other issues brought up in the Richardson studies may be present in the results here in the homogeneous freezing regime. It was found at that time that slowing the flow by half was needed to observe homogeneous freezing ideally. The SPIN may not have this limitation, due to the use of the depolarization detector, but I would be surprised if there is not some issue in differentiation when the ice crystals are only growing to 1 micron sizes amongst a field of liquid particles in the time available.

Some specific questions/comments for potentially addressing are listed below. On balance, these could require a number of mostly minor revisions, excepting the request for an additional figure.

Specific Comments

Introduction

Page 3, Line 7 – The Koop et al. reference is completely about homogeneous freezing. It goes with first part of sentence, but not last, which refers to heterogeneous nucleation.

Page 4, line 2 - Are you certain that lower chamber weight has been achieved in parallel plate designs? My impression is that the SPIN instruments could be heavier than most CFDC's. Can you state a total weight?

Page 4, line 10: I suggest adding Jones et al. (2011) to the list. This CFDC development was actually the first to mimic the original device designed by Rogers (1988), followed later by Saito et al.

Instrument theory and design

Page 4. line 21 - Have you measured this ice thickness as 1-2 mm? We have estimated and reported 1/10 of that thickness in the CSU CFDC's based on liquid meltwater measurements. If truly this thick, how much of the inter-plate distance is filled with ice, and do your calculations account for this? It would thus be useful to also report how wide the plate separation is in the SPIN (here, not only in the Figure caption). A horizontal distance scale on the bottom axis of Figure 1 would be useful, and I would presume that such thick ice layers should be shown (and again, accounted for in the calculations).

Page 5, line 16 – How is the refrigeration controlled in the evaporation section of the chamber? Figure 3 only seems to show cooling each wall continuously at one temperature. What constant temperature are the evaporation section walls held at? That is, at the warm or cold wall temperature?

Page 6, line 17 - How is sheath air drying achieved? Can you comment on whether or not a one-way flow of dry air could replace the re-circulated sheath from in the system shown in Fig. 3? CFDC's have been known to be operated with one-way flow of with recirculation. Are both practically possible with the SPIN? Just asking as a scientific point, not with regard to any perceived deficiency.

Page 6, line 22: The knife edge simply places the sample flow initially in the center of the chamber. The limited range of temperature and supersaturation then depends on maintenance of laminar flow conditions, correct?

Page 6, line 23 – It might be useful to point out that the 4:1 growth to evaporation ratio here is much greater than the 2:1 ratio used by Rogers et al. (2001). But this does bring to mind that you should state all of the dimensions in writing here. Especially, it is important to know the cross-sectional dimension of the actual interior chamber region. It is shown as 3 cm in Fig. 2, but the plate separation is 1 cm correct? So with the ice added, the actual separation is 8mm?

Page 6-7: Not addressed anywhere in this section is how walls have been treated to re-

tain ice. I realize that this is found in publications written about chambers that the SPIN design followed, but it is another small detail that deserves discussion somewhere, as stability of ice surfaces is one of the critical issues for these types of instruments.

Page 7 – Is the OPC quite similar to the DMT CASPOL (Glen and Brooks, 2013) or is it very different?

Page 8, line 5 – Concerning the saturation of counting at 3900 per cc for a 1 LPM flow rate, is there any means to correct for "live time" as done for some multi-channel analyzers?

Page 8, lines 9-15 – The question here is what is present as actual functionality with a SPIN versus what is possible via a user's initiative, and has been demonstrated? . That is, should this say "potentially allowing. . ." and ". . .much of the chamber operation could conceivably be performed remotely"? The impression is given that full remote operation is already possible, but that is not demonstrated in this paper. These things are all possible for any group with some initiative and technical skills (and some have done so), so without thorough demonstration in this paper, I think that such statements should be avoided.

Methodology

Page 8, line 23 – Can you say what type of dewpoint sensor is used, its accuracy, and how well can it resolve a dewpoint temperature of -40 °C? This typically requires an advanced sensor.

Page 9, line 1 – When you say the chamber is cooled to icing temperature, you mean both walls and both wall sections (growth and evaporation) cooled uniformly I assume?

Page 9 – This might be another place to mention how the walls are treated to be "wettable," and what they are made of, materially.

Page 9, lines 6-7 – Is this "dwell counter" time the total time of water in the chamber or the total time that that water remains prior to it being quickly removed? In other words,

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how long is water actually in the chamber? The time mentioned is quite short for other CFDC's, yet the ice thickness mentioned is much thicker, so just looking for details that users may wish to know.

Page 9, lines 15-17: "This background concentration (typically a few counts per liter) sets the lower detection limit of INP and must ideally be < 0.1

Page 10, line 5: Doesn't this describe the same effect described above as the "background"? It is not clear. Also, please add that this is uniformly more frequent at higher supersaturations "for the SPIN." Such a generalization may not apply to all CFDCs for this process that is still rather poorly understood, and may vary depending on the method used to obtain wall wettability.

Page 10, lines 6-8 – The statement that frost background of the same order as INP number concentrations means that the INP data will include a significant artifact is understood, but this is likely quantifiable. It certainly does not require that the frost be only 0.1

Page 10, lines 11-12 – It is useful and could be important to report what a typical ramp rate is in terms of cooling rate or d(RH)/dt.

Page 10, line 13: How long are the typical filtered air periods in order to provide a sufficient measure of background frost numbers?

Page 11, lines 13-15 - This seems a variation in the Rogers approach, which may be worth noting. Rogers did not consider along-wall differences, just average wall temperatures in locating the lamina and the flow profile. I am not sure how one uses this information, since the lamina position and velocity profile calculation usually requires assumption of a single value along the walls. What is actually reported then to associate with an INP measurement (for sample conditions and residence time)? Richardson (2009) did consider along wall temperature differences, but he needed FLUENT simulations to resolve the importance of this factor in defining and conditions

and affecting freezing.

Page 13 and Figure 6 – While much of this comment is editorial, I place it here because I feel that this figure requires better explanation. Can the scales and labels be made larger? Please explain the "Size" units or otherwise please explicitly remind of the form of the size parameter. If the standard definition, I could not get the magnitudes correct, or else the wavelength is different than listed for the laser. Also, if the lower size threshold is 0.4 micron diameter, these figures really show particles growing to two orders of magnitude larger in size within the SPIN? Perhaps better, why not show this figure with actual geometric particle size? Finally, has this exercise been done for specific size ranges of INPs, and what are the RH conditions associated with part b (should be stated)?

Page 14, Figure 7 – This figure may bear more discussing as well. It may be helpful here to reiterate that the evaporation region reduces “droplet” fractions. Otherwise, one may wonder why full droplet activation, or anything close to it, is not achieved in this experiment at higher supersaturation, One wonders anyway, since once droplet breakthrough occurs, it seems unusual not to see all droplets retaining sizes well above the OPC lower threshold size (e.g., DeMott et al. 2015). Is this feature due somehow to droplet evaporation after exiting the SPIN and entering the detector region (i.e., due to heat transfer)? The supersaturation seems too high to think that the AgI is not fully activating as droplets at some point. On another matter, when you say validation accuracies, what is the basis for validation? One also would like to know if there is any particular lower ice number concentration value for which this machine learning procedure is easily used. For example, data are shown at INP number concentrations as low as 1 per liter in Fig. 7. Are these values already corrected for background frost? The method seems to be most reliable for INP number concentrations of several 10s per liter. For INP concentrations in much of the mixed-phase cloud regime, how long would one need to integrate ambient data to reliably use this method if INP number concentration is say even 4 per liter? Would this become problematic when droplet

breakthrough occurs and nucleated ice concentrations are quite low? Any statements on the useful range of this otherwise elegant method would be appreciated. Finally, is it possible to speak about the use of a constant particle size that could distinguish ice versus aerosol and drops, perhaps as a function of temperature and RH? Just looking at the data in Figures 6 and 7, it seems possible to use this information for many instruments with similar residence times.

Page 14: How likely is it that AgI is in exactly the same form and size as for other studies referenced in the following results? It surprises me, considering past cloud seeding related literature regarding AgI activation, and how even minor contamination can lead to disparate results.

Results and comparisons to literature

Page 14, line 19 – Is this validation or calibration?

Page 15, lines 4-6 – The use of ammonium sulfate here is a classic calibration study for existing CFDCs, but it does lead me to ask about the expectation that sulfate will act only for homogeneous freezing in the lower temperature regime. Heterogeneous ice nucleation activity has been reported for crystalline ammonium sulfate aerosols (Abbatt et al., 2006). It might be important to state why this is not expected in this case. Also, for size selection, can you say the proportion of multiply charged particles? This could affect the homogeneous freezing transition, which I suggest next requires more detailed explanation and discussion.

Page 15, lines 8-9 – The broad sweep of Figure 9, which is nice as a first inspection based on a large suite of measurements, nevertheless suggests to me that the instrument is not reproducing conditions of homogeneous freezing as clearly as observed by some other CFDCs. This transition and the definitive statements made in this section bear some quantitative inspection. What would a line drawn vertically in the figure (e.g., a single experiment) indicate for the active fraction of particles as a function of RH? How would it compare to predictions based on water activity theory

for freezing? This is a standard on which to compare, such as shown in DeMott et al. (2009), Richardson et al. (2010) and some other past literature. Stating clear agreement with Koop et al. (2000) suggests the need for such a presentation. Furthermore, does the figure show false detection of ice at temperatures warmer than homogeneous freezing?

Page 15, lines 15-21 - I must say that it is not sufficient simply to say that the homogeneous freezing behavior is "captured" by the SPIN, versus requiring much more detailed inspection in future studies. As I said above, it does not look ideal (activated fractions do not increase orders of magnitude as shown in other CFDC studies of homogeneous freezing), but this is difficult to tell in this mapped figure. A detailed comparison will be needed, and this suggests an additional figure. What are flow rates and residence time in the chamber? What is efficiency of ice detection below water saturation (i.e., are ice crystals predicted to grow to sizes that will be easily distinguishable via size or polarization)? Noting the discussion in Richardson (2009) and Richardson et al. (2010), especially the need to reduce flow rates in shorter-length CFDCs such as SPIN in order to detect (at least by size) the presence of ice crystals growing above about 1 micron at temperatures below -40C, I wonder if this complication is present in any of the data shown here (i.e., explaining the need for high supersaturations to realize high activated fraction)?

Page 16, line 3 – Editorially, I suggest removing "significantly" here. A few percent above water saturation is not much in order to detect immersion freezing (DeMott et al. 2015 and references therein). It seems that the depolarization detector will be required in many cases. Hence my earlier question about the lower level of sensitivity for ice amongst a field of many liquid particles using this method. It would seem useful to design the instrument with a longer evaporation region.

Page 16, line 7 – AgI aerosols only provide a benchmark if they are carefully produced in exactly the same manner based on a great deal of past literature. Is agreement expected or fortuitous?

Page 16, lines 16-20 – I think that this statement should be omitted. It is not true. Other portable INP chambers have simply switched to two-stage refrigeration systems to achieve lower temperature conditions in the past. See, for example, DeMott et al. (2003) (already referenced here) and Prenni et al. (2007), ground-based and aircraft examples of using a refrigeration system to extend to lower temperatures. It really is not a difficult change for instruments that can be flexible with the configurations used.

Conclusions

Page 18, lines 2 to 5 – Here I suggest referring to agreement of your results with Richardson et al. (2009), who quantified uncertainties in a similar manner.

Page 18, lines 7 to 9 – Considering comments above, I consider this a point that is as yet unproven.

Page 18, line 14 – demonstrate, not "validate", in my opinion.

Page 18, lines 16 to 19 – This is a judgement statement that I consider inappropriate for a scientific article. The introduction of this instrument commercially is an important milestone. Nevertheless, the lowered barrier involves a financial investment that is about 4-5 times the cost of a CCN instrument. An individual building an instrument could do so cheaper, and in the process obtain the invaluable experience and investment that my colleagues here have demonstrated is needed to understand and effectively interpret such measurements. It is certainly true that the availability of a commercial instrument could "potentially" lead to an increase in temporal and spatial coverage of INP measurements. This remains to be seen.

Page 18, lines 21 to 23 – The full performance of the depolarization detector would seem to be worthy of additional study. A comparison of ice detection versus other instruments and a standard such as a cloud chamber simulation would be useful for the future.

Page 19, paragraph 1 - The discussion here should reflect any responses to my ques-

tions and comments above. Again, utility has been demonstrated, not validated quantitatively.

Page 19, lines 15 to 17 – I suggest striking the statement regarding "SPIN's availability. . ." in preference to something to the effect that "The commercial availability of such a device may allow for increased coverage of INP measurements that will help constrain. . ." In preference to some of this material overall, I wonder if you could say what future needs for demonstration and validation remain (I would say that quantitative intercomparison is needed), and what challenges may remain?

Figure 5 - By "degree of flow reversal" do you mean the fraction of the flow that is reversed? You need to state a total flow and pressure for these calculations right? What would it be at 300 mb, or something more typical of cirrus sampling conditions? Also, it could be useful to superimpose the lamina position on these flow figures.

Editorial notes

Page 3, Lines 15-19 – I understand the flow here in saying that CFDC's have been used for both laboratory and field studies, but to be clear, such instruments were designed not to simply detect the conditions for ice nucleation in the laboratory, but to quantify the number concentrations active under a variety of conditions far beyond onset conditions or for specific nucleants. And they were designed for field measurements specifically, from the start. So the statements here are not historically accurate. I suggest, "Several types of instruments have been developed to measure the efficiency of heterogeneous nucleation of cloud droplets and ice crystals. Many of these have applicability for measurements in the laboratory, as well as intended application for field observations."

Page 9, lines 21-22 – Just to advise that use of the term "time dependence" could be confusing, considering stochastic freezing time dependence that is not easily assessed with this type of instrument, and is not what is being referred to here. How about "temporal changes" of INP concentrations?

Page 11. Line 17 – The Kulkarni and Kok reference seems incomplete. Is it a report? Is it freely available?

Page 17, lines 9-10: Are the top 13 thermocouple temperatures then averaged for reported SPIN conditions?

Figure 1 - Re: see above comment on plate separation, in that it could be useful to add a horizontal distance scale under temperature.

Figure 9 – can the typical number concentrations of ammonium sulfate be stated in the figure caption?

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