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

Anonymous Referee #2

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Review of, *The SPectrometer for Ice Nuclei (SPIN): An instrument to investigate ice nucleation*

In the submitted manuscript Garimella et al., describe the commercially available SPIN instrument for measuring ice nucleation. To my knowledge the instrument has been on the commercial market for a few years but to date no description of the instrument and its function and capabilities has been available in peer-reviewed scientific literature. While I applaud the large group of co-authors for undertaking the effort of informing the community about this instrument, I do feel that some effort should be made to improve upon the clarity and content of the manuscript before it is eligible for full publication.

The comments of P. DeMott have raised many important issues and at times I reiterate his points for emphasis. As a general comment I do think that if this paper is to be the community reference for SPIN there are many areas where detail must be added. Furthermore, it should be made clear from the abstract onwards what are the 'out-of-the-box' capabilities of the instrument versus contributions that directly result from scientific work and testing. For example, is the machine learning analysis of detector data something that is in part or wholly developed at the user level, or is it an analysis package delivered with the instrument? Given the long list of authors I have no doubt that the current SPIN owners/users have a relatively strong connection, but unless this will form the basis of an organized user group it is not clear that any group acquiring SPIN would have access to and/or benefit from the same community knowledge.

Below I try to summarize items and issues from the general to the specific in the order they arise within the manuscript.

Itemized points:

page 3, line 2-5: Aerosol particles are not **required** for nucleation, rather because they are ubiquitous they assist nucleation. In a system with few enough particles homogeneous nucleation could proceed. The way in which particles facilitate the freezing is by changing the free energy barrier to phase transformation. Classical nucleation theory uses bulk thermodynamics to describe the free energy change as a surface/volume competition that is modified by a kinetic pre-factor. The described “mechanisms" are simply phenomenological descriptions of how this proceeds. Thus some thought should be put into accurately phrasing the first two sentences.

page 3, line 7: The Koop et al. (2000) reference is used throughout the manuscript and also within Figs. 5, 9-10. It should be clear that the Koop theory and curve refers to homogeneous freezing in liquid solutions. Thus, the applicability of that theory to freezing in the atmosphere is not always clear – in their manuscript Koop et al. (2000) ignore geometry and consider only large droplets. A very different curve represents homogeneous freezing from the vapor phase – for examples see Fig. 5 in Murray and Jensen (2010) and Fig. 2 in Thomson et al. (2015). See also the discussion of the relevant figures below.

page 3, line 8-12: Although the IPCC reports are highly valuable and useful references, it seems as though citing the recent reports 2x in the papers primary motivating paragraph (Boucher et al., 2013; Stocker et al., 2014) may ignore primary source literature. Perhaps the points being made by the authors would be better emphasized or bolstered if they were slightly expanded and also referred to primary source material, where INP, mixed phase clouds, and/or cloud microphysics are concerned.

page 3, line 21: “RH’s" is introduced here but not defined until later on page 4. The abbreviation should be defined at first use and used thereafter.

page 4, line 1-4: The sentence, “Development of parallel plate chamber geometry has simplified several technical aspects of the chamber design (e.g., lower chamber weight, less complex machining, and simpler refrigeration plumbing than for the cylindrical geometry) at the expense of edge effects (i.e., deviations from ideality at the chamber edges).", is confusing and relegates seemingly important details to the parenthetical. I suggest rephrasing, removing the parentheses, and adding detail to construct a more holistic picture of what is meant.

page 4: The references to the ZINC and PINC systems and papers raise the question of, how much of SPIN can be understood by referring to the literature that describes these earlier systems? Are there things that can be taken out of those papers that are valid for SPIN? I think it is important to make explicit what is SPIN specific versus universal for parallel plate (vertical) CFDCs and/or CFDCs generally.

page 4, §"Theoretical principles": This section reads more like operational principles/procedures. In its current form it would be better suited to come after the design description. If the preference is to keep it in its place then it needs to be re-written in a more general format. However, I would suggest that the authors both outline the general operating principle and describe a suggested user protocol for typical experiments – not necessarily in the same section.

Throughout this section and the following two the manuscript lacks a clear trajectory and often uses imprecise language. Some effort should be made to more clearly delineate between the theoretical working principle, what *could possibly be* done in SPIN given the intended engineering, and what in fact *is* done in (with) SPIN.

For example,

page 4, line 19-20: "Controlling the temperature and RH is accomplished ...." It seems to me that temperature control has nothing to do with icing the chamber walls. The temperature is controlled by refrigeration and heaters, the saturation condition is established by icing the walls and controlling the temperature gradients. Likewise (line 23), "The two walls are held at different temperatures ...." That is a statement of operational protocol, because within some range it appears you could set the temperatures to whatever you want, they could even be equal. It would be better to say something like, ‘In order to establish the necessary vapor supersaturations for nucleation the ice coated chamber walls are held at different temperatures ...’

page 5, line 6: How well are the particles constrained within the sample flow lamina? Garimella had a presentation at the 2015 AGU Fall meeting suggesting that this is a source of uncertainty (Garimella et al., 2015). That presentation or a manuscript in preparation could be referred to, to say something general about this.

page 5, line 12-end: The effectiveness of the evaporation section is largely dependent on the relevant residence times, perhaps this could be addressed? Isn’t the critical droplet size related to the time a droplet might have to evaporate? Is it important droplets completely evaporate? Or can they simply decrease in size to below a threshold?

page 7, lines 6-10: Both the aspherical morphology and depoloraization associated with ice crystals come from the anisotropy of the ice as a material. Ice crystals have fast and slow growth directions (that lead to their macroscopic morphology, e.g. Cahoon et al., 2006; Wettlaufer et al., 1999) and are also optically birefringent (e.g., Thomson et al., 2009; Lekner, 1991, 1999, and references therein). Both aspects are important.

page 7, line 16: “LabView software is used for instrument control and data acquisition. The SPIN software program consists of several different loops and sub-programs and allows for significant automation during operation. The Control Program starts and stops the other modules, updates the displays, controls the instrument set points, watches the alarms, and otherwise supervises the operation of the entire system." Given the lack of specificity much of this text seems superfluous, it would suffice to say: The SPIN instrument is operated and controlled via a LabView master control program. (or alternatively add pertinent information)

page 8, line 5: What happens when particle counts approach this level? What is ignored? Does this bias results? Is it recommended that a dilution flow is added to keep raw particle counts below this threshold?

page 8, lines 6-15: Is the entire icing scheme fully automated as one could believe from this paragraph?

page 9, line 4: “icing sequence" Quotation marks are generally used to set off material that represents quoted or spoken language. At times they are used to show sarcasm. Here neither appears to be the case – they seem to be indicating LabView program components. If these terms/phrases are being emphasized it would be better to use a different type-setting tool (bold, italics, etc.). The same mistake is made throughout the manuscript, eg., “ice dwell counter," etc.

page 9, line 23 - page 10 line 2: “For the former, diverging the wall temperatures increases the chamber supersaturation,... and ramping both walls..." The uses of ‘diverging’ and ‘converging’ are awkward in this sentence, I suggest rephrasing (for example, ‘increasing the temperature gradient between the walls’). They are correctly used a bit further in the text. Also, the walls are not ‘ramped’. The temperatures are increased and decreased or temperatures are ramped.

page 11, line 12: extra comma after “Rogers"

§3.2 Data processing and methods: Given the audience of AMT this section lacks significant detail and some concentrated effort needs to go into a major re-writing. While machine learning and other data analysis techniques may be known to some, I do not think one can assume they are part of the atmospheric measurement community vocabulary. Detail and references must be added to the section, and if deemed excessive for direct inclusion I suggest that fundamental information now not included could become part of a supplement. For example in Figure 6 data from a GMM-KDE is presented – could a supplement include a step-by-step explanation of the analysis process, perhaps including some idealized system (where the separation is more clearly bi-modal)? These techniques seem to be a powerful tool, but it is hard to appreciate in the limit of the single example and poor explanation.

page 12, line 1: “supervised machine learning" is introduced without a reference. Include a reference so that those unfamiliar with this have a standard text to which to refer.

page 12, lines 3-4: “The also require fewer assumptions to be ...." Machine learning requires fewer assumptions than what?

page 12, line 6: I suggest rephrasing to say, “... historically been analyzed using *post-evaporation section* particle size as...."

page 12, line 10: I suggest rephrasing to say, “... than the ice size and that droplets *above that size* do not survive..."

page 12, line 15: By “efficiently” do you mean completely?

page 13, line 4: The introduction of Kernel Density Estimation should include a reference.

page 13, line 7: The introduction of the 4-d Gaussian mixture model (GMM) should include a reference and likely Mixture and Model should begin with capital letters.

page 13, line 10: “aerosol only" Again a misuse of quotation marks. However, this time the phrase is also left undefined. If this is to be used to indicate a defined procedure/measurement it should be explicitly defined. The same is true of “aerosol + ice" and “ice only."

page 13, line 18: Gaussian kernel support vector machine needs a reference.

page 13, lines 20 –: The final sentence of the paragraph, “Since a condensation ... " is long, awkwardly appended and does not follow from the previous material. Make this an independent paragraph that clearly explains how activated fractions are calculated.

page 14, lines 4-6: “3-class supervised machine learning (bootstrap aggregated decision trees....)" At least two missing references here, 3-class supervised machine learning and bootstrap aggregated decision trees? Also missing is any explanation of why these methods are used versus what is described before and how are they different.

As a general comment to supplement the specific comments regarding the data processing section. Given this is not a computer science journal then one cannot expect that everyone is well versed in machine learning, KDEs, GMMs, etc. As a reader it is very disturbing that much of this seems to be introduced as little more than buzz-words with acronyms. Some effort needs to be made to indicate what in practice is being done at each step. As I understand machine learning is used to predict outcomes – what exactly are the outcomes being predicted? How are the training vectors selected? Are KDEs used to make PDFs of size or of all OPC variables, it is not clear? A 2D GMM-KDE is used for visualization but a 4D used for the actual calculation? Does that mean that the 4D does a better job than we can understand by looking at the figures? Otherwise, what is the difference?

Finally, should the community expect that all SPIN data should be reported in this manner (i.e. As a result of this type of analysis?)? Is this a computationally time consuming process, or is it quick and completely automated? Can (is) such an analysis be implemented in real time, such that one observes the results while running an experiment? What part of the analysis (training data, etc.) needs to be implemented separately for individual detectors on different units, or are all detectors identical to within the uncertainty?

page 18, line 14: What is meant by “validate"? Validate the performance as what? Perhaps this is too strong?

page 19, line 2: Here given what has been presented within the manuscript I take “homogeneous INP" to refer to homogeneous freezing of solution droplets. It is important to make the distinction between homogeneous freezing from solution and homogeneous freezing from the vapor.

page 23, line 20: Kulkarni et al. ref. missing doi (doi missing from many refs)

page 23, line 21: Kulkarni and Kok reference seems incomplete. Is this a report? In a series? Is this document available publicly?

page 26, line 4: The Stocker IPCC reference is missing co-authors or an et. al.

page 31, Figure 5: What are the units of the color bar? “The color scale shows the degree of flow reversal..." What is meant by “degree"? The small subplots can be better connected to the intensity plot. I assume for example in the flow reversal subplot -30 represents the warm wall and -45 the cold wall? This could be indicated in the figure and/or caption. Also, I take the color intensity to represent the maximum flow reversal velocity – is that true? Or is it a mean flow reversal velocity? Also it must be made explicit that the Koop curve represents the onset of homogeneous freezing of liquid solutions (droplets). Given the dashed flow reversal line the phase space that can be explored without flow reversal within the chamber is quite limited. Is this an issue, does it affect the utility of this (and similar) instruments? It would be beneficial to discuss this issue in the text where the flow reversal and figure appear.

Finally, is the plan for this figure to appear in a single column within the 2 column journal? If so the subplots and legend will become very difficult to make out. Perhaps the figure should be redrawn to optimize it for the 1 (or 2) column size?

page 32, Figure 6: Again think about the figure size in 1 or 2 columns. The panels will be very small, and it is difficult to read any of the numbers/axes labels and legends in the current full page format. I had to zoom into the *.pdf many times to recognize that the support vector points were plotted as open circles enclosing data points.

What are the $\log_{10}$(size) units? I have not been able to convince myself of the meaning of these axes. In the upper 3 panels time 2 - time 1 does not look like 0 as it appears in (c). This could be due to rescaling because the intensity shading in (a)-(c) changes. I suggest that an absolute color scheme is chosen (perhaps including a wider color spectrum), such that colors in all panels represent the same probability density.

page 33, Figure 7: Again think about the figure size in 1 or 2 columns.

What does classification accuracy of 99% mean? It seems that some points would be much more certain than others. Does this mean that the total number of each is identified to a 99% certainty level? In all 3 dimensions? The 3D plot shows an area where the 3 colors (particles, ice, droplets) merge. I would assume in this area uncertainty would be high, while far from this uncertainty would be small? In the right-hand panels why does ice fraction equal water fraction at conditions sub-saturated with respect to water? Is this an area of phase space where the droplets and ice cannot be distinguished beyond the level of uncertainty? If so is this a result of small particle sizes? Is there a critical ice (droplet) size to distinguish solid from liquid?

page 34, Figure 8: These panels are probably better arranged vertically for the 2 column format. Is the sigmoid fit used as a scaling factor for results? Is sigmoid the appropriate fit – perhaps the efficiency is unity to a size cutoff, below which the relationship is linear? Without one or more further data points the choice of fit should be supported based on some reasoning. Was the previously presented data (Figs. 6,7) processed using these relationships? I would suggest discussing these issues in the text where Figure 8 appears.

page 35, Figure 9: Increase the size of the legend and labels. Also, again specify that the “expected homogeneous freezing" refers to aqueous phase homogeneous freezing. See my previous references to the Koop article. In such a plot it is difficult to also incorporate uncertainty, but perhaps some indication of the uncertainty range of activated fraction can be given?

page 36, Figure 10: See early comments with respect to formatting, font etc.

page 37, Figure 11: See early comments with respect to formatting, font etc. I wonder if this figure could also be incorporated into the earlier discussion of flow reversal etc.

Comparing with Figure 5 and given that S_{ice}=1.3, then flow reversal should be present in (e). However, I see no evidence of this and the scale incorporates only positive velocities. The two plots are not self-consistent, please explain.

Let me reiterate where I see a need for major revisions.

• The data processing section is difficult to understand and incomplete in its current form.
• Operating procedures and analysis etc. specific to SPIN should be clearly delineated from general discussions of the CFDC principle (eg., How the saturations gradient is established.). Furthermore, I encourage the authors to highlight what they consider to be the ***best use practices*** of SPIN given the current state of understanding. A simple example would be the utilization of an icing temperature of -25°C and time of 5 s – were these observed to somehow lead to optimal experimental conditions, or simply chosen at random?
• Clearly delineate what part of this manuscript represents out-of-the-box SPIN operation/ measurements versus user implemented protocols and analysis. For example, is the water reservoir cooling to 2°C standard for SPIN?
• If necessary re-write the abstract to reflect the scope of the standard versus user enabled capacity of SPIN.

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