Reviewer comments on "The Fifth International Workshop .... " by DeMott and Co-authors. (Atmos. Meas. Tech. Disc., https://doi.org/10.5194/amt-2018-191)

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Phase 2 of the Fifth Workshop (FIN-02) is the most comprehensive of the ice nucleation instrument inter-comparisons that have been organized since the first one in 1967. Significant progress has been made in recent years in instrument development, in understanding of the processes relevant to ice nucleation and in knowledge about ice nucleating materials. Thus, the paper is of great interest to many scientists. The authors are to be congratulated on the work described.

The paper summarizes the work of a large number of individuals. The long author list makes reviewing the paper somewhat intimidating. How can anything else be noted that such a large group hasn't already considered? On the other hand, perhaps it is harder to make sure that all's in order when so many hands are involved. The reviewer's task is further burdened by having so much material to review. In fact, this reviewer focused more on general aspects of the paper than on detail.

Essentially all in this review addresses what is presented in the paper, not what and how the experiments were done. Most of the instruments participating in the Workshop have already been described in refereed publications. The operating conditions, the selection of aerosol types and the procedures followed during the Workshop have been arrived at after extensive deliberations and with the benefit of experience at earlier workshops. These issues are not addressed here except regarding the consequences of the procedures in interpreting the data obtained.

Broad comments:

1. While it is clear that no classification can reflect perfectly the variety of instruments used, it is worth thinking about how useful is the one applied in this paper. No doubt, the online vs. offline designation has its origin in common references to the instruments in that some can be deployed in the field as a self-standing unit, while the others have essential work done in the laboratory. Thus, 'field' vs. 'laboratory' instruments could also be applied. The current designation has disadvantages. It doesn't inform people unfamiliar with the details of what the essential difference is. Is it the operating principle of the devices? Is it aerosol flow through the device versus samples captured and processed? Portable versus fixed in not necessarily applicable, as a filter processing unit is no larger and difficult to transport as a CFD chamber. A drop freezer can be the smallest of all the devices. Also, field deployment of a device impacting a flow of aerosol on water drops would be an online instrument for which much of the RH discussion about CFD's would be irrelevant. So, while the offline/online distinction isn't all bad, perhaps "direct sampling" versus "post-processing" would be more meaningful. Another useful direction would be to focus on variable RH instruments versus those with liquid water. What characteristics is the most important? Tables 1 and 2 do a pretty good job in listing the main features of the various devices. But the titles of subsections 2.1 and 2.2, and the headings in the Tables could be improved by removing the hesitation in the designations and, preferably before 2.1, explain the reason for the classification. Dry versus wet in the table headings is not the best. My recommendation is to use 'direct sampling' versus 'post-processing' and define in the Introduction how these terms are applied. This is also appears to be the intention with lines 30-32 on page 7. These designations would allow the AIDA chamber to be in the group of instruments with clouds forming on the sampled aerosol, just like in the CFD's. Omit 'portable' as part of the definition. (Apologies for this rambling paragraph.)

2. I am somewhat reluctant to raise this issue, but this paper needs special scrutiny in this regard because it weighs

heavily on the principal results of the paper. Screening of data to be included or excluded is perhaps the trickiest and most sensitive part of the paper. There is a sense of some censoring of the data throughout Section 2. Undoubtedly some judgements had to be made about data validity by some criteria. One can see the attempt to do this, and the authors certainly deserve to be given the credit of proper judgement. However, the first question that comes to mind when looking at the degree of agreement shown in the results is how much screening of the data was done? How many discrepant cases were excluded due to errors or uncertainties? In other words, how much subjectivity entered into the analysis. Some comments on this issue would help the readers gauge the value of the results.

3. It becomes gradually clear as one reads on in the paper, that the focus of the analyses was freezing nucleation, at least as much as it could be assumed to be the dominant mode of nucleation if sufficient supersaturation was achieved to ensure that INPs are in liquid droplets. Perhaps, I missed it, but if this is true, shouldn't it be clearly spelled out in the Introduction, and be part of the goals? Similarly, the size range covered (9/28-32) is a pre-defined constraint and would be best stated up front.

4. Unless I missed it, the promise made at 8/32 to describe the aerosol collection and transfer methods for testing as suspensions are missing from Section 2.4. Problems associated with the method used and the final efficiency of the capture are not found. See also 13/4-5.

5. Some re-phrasing of conclusions such as in lines 6 to 23 on page 13 should be considered. The degree of agreement in the data is remarkable and deserves to be celebrated.. But to say that the agreement extends over 7 orders of magnitude over the whole temperature range is misleading. Except in the center of the temperature range only 4-5 data sets are represented. Thus, in a strict sense, the agreement among all data sets is not quantitative over the whole range. They all coincide in defining a common trend and agree in overlap regions. None of the data sets extend over the whole range. Also, there is a division of the types of instruments for which data are available at the upper and lower ends of the temperature regime. The main novelty in these results is the agreement between CFD type and drop freezing types of measurements in the overlap region. This is a real accomplishment and demonstrates that the two types of measurements can be combined to yield atmospheric measurements over a wide temperature range. The extent to which that is a practical solution for low INP air samples is worth addressing.

6. Only in S.2.1 is the method for calculations of the INP concentrations stated. How were the presented data derived for each device should be part of the description for each one. This also applies to how uncertainty ranges were determined.

7. A suggestion I make reluctantly, because of the amount of re-writing it implies, the authors should consider including a Discussion section, moving some material from Results there, and going a little further in some themes. Having a Discussion section would allow for better perspective on the various aspects of the results (e.g. 14/3-22), and give good opportunities for comparisons with previous publications (e.g. 18/2-28).

8. Parts of the description of results refer to the sigmoid shapes and the central slopes of the INP vs. T curves. I mention here that similar considerations, and a definition for a slope parameter were given in Vali (ACP 14, 5171-5194, 2014.). The rich material in this paper could very usefully add and expand on the analyses in that paper. That may go beyond the scope the authors set for this paper, but in reality it would not be a large step.

9. There are many references to the Supplement. It would be helpful if, in each case, the reference pointed to some specific part of the Supplement.

10. It is regrettable that the ACP (3.1) and AIDA (3.2) sampling results are in different forms, i.e. per air volume and as site density. Was this unavoidable? Considered unimportant? It comes as a surprise to the reader. Having the two different aerosol processing paths provided some assurance of minimizing problems related to each one. Perhaps there were other operational advantages or limitations as well. Yet, one would have like to see the results in the same format. For inter-comparisons the two data sets provide additional support and that is the main goal of the Workshop.

But it isn't clear why the results need to be given in different manner. More careful reading of the paper may reveal the reasons, but perhaps readers (like me) could be helped by a statement of those reasons up front.

11. The focus on measurements of atmospheric INPs in the recommendations on page 22 is well placed. But ice nucleation measurements are also needed for fundamental understanding of ice nucleation and of the nature of INPs. Another important direction that ice nucleation measurements should explore in future workshops is to perrform tests with mono-disperse aerosols.

Specific items:

page/line

3/32 'constrain' INP population???

4/7 The fact that the more general aspects of the project are to be published later is a bit of a problem. A brief description of the three stages would help.

4/10-12 The order of these last two sentences of the paragraph should be reversed.

4/22 What do you mean by 'align'?

4/25-32 Goals are well defined. It would be nice if the conclusions responded clearly to each item.

4/33 -> The intent to define a priori instrument characteristics is somewhat futile. Instruments that participated can be grouped as on-line and off-line but linking that too strongly to dry and wet is too much of a simplification. It would be more effective to describe the methods starting with Fig. 1 and then detail what the instruments do (above water saturation and immersion freezing) and do not do (deposition, condensation-freezing, contact). Indeed, reference to Vali et al, 2015 can help with the definitions.

5/16 The operating principles are in in Tables 1. and 2 to some extent and are described in 2.1 and 2.2. The Supplement is to describe the *detailed implementations* of the basic idea in each device.

Fig. 1 The inclusion of X wt% designation here is unhelpful. The inclusion of this factor complicates interpretation. Indicated range may have been correct for the INPs chosen for test but have no general meaning. There are other materials that cover the whole temperature range with a single wt%. Adding the specific INP for which the graph is valid would add unnecessary detail. I suggest to leave those wt% indications off the graph. Of course, solute effects are ignored here.

6/14 The definition of max RH_w practicable before 'breakthrough' is made difficult by the use of RH_w for many different aspects of the processes.

6/18-20 Is this caveat not in conflict with the goals defined earlier? May be just say that experiments included in the evaluations all stayed below the breakthrough RH_w.

6/31-33 This sentence should start the paragraph to explain why the choice of 102% needs discussion.

6/37-7/1 This is problematic. How is this known? The fraction of aerosol activated as a measure of the RH achieved in an instrument? The nominal RH and actual volume-weighted one differ, as just described in earlier part of the paragraph.

7/3-7/5 Recommendation here is out of place and heavy-handed.

7/7 Shouldn't this 'primary comparison' be defined as a 'goal' up front?

7/11 Quite unclear. Highest vs. maximum. Could the question be said to have been to select the RH point in the scans that the operators thought was most representative for freezing nucleation? Again, this objective should be stated before discussing details of RH variations in the instruments. All those variations are caveats on the validity of the values chosen/used for the comparisons. *In any case, isn't it likely that the activated INP were in drops, due to the high hygroscopicity of the aerosol?* In that case, the INP's don't experience 102% or whatever supersaturation. Clearly, these considerations need to be addressed here.

8/5 Is this because only ice crystals forming from frozen droplets on the surface are counted? How certain is that? Same issue as in previous point.

8/7 " ... limits reduction of ..." is unclear. The influence of vapor competition, and its dependence on INP density on the substrate has extensive literature, and perhaps should not be glossed over so easily. The operational definition used here for making the comparisons deserves more explanation. The concluding sentence on 8/13-15 is unclear - do you mean cases when vapor competition was only inferred to have been present but not evaluated?

8/8 '...were allowed to ..' inserts a hierarchical tone that is unnecessary. An objective tone would be more appropriate. The working arrangements of the workshop are not of interest to the reader. Else, make it clear and explain in the Introduction that there was some sort of checks and balances arrangement in order to improve the final result. I don't know if there was or not. See also comment on 7/3-5.

8/14 "wet suspension" -- ?? wouldn't liquid suspension or water suspension be more direct? Actually, the first two words could well be omitted and the sentence say "Measurements of immersion freezing ...."

8/31 " ... wet suspension groups ...? -- one can deduce what is meant but it could be said better

9/1 "... basic methods ... " may need to be re-considered if changes are made in response to the first item in this review.

9/29 suggest leaving out the word 'mode'

10/3 suggest replacing 'wet dispersion of particles' by something like "aerosol generation from aqueous suspensions" or "particle dispersion from aqueous suspensions" or "dispersion of particles via spraying and drying"

10/32 Was the depletion of aerosol in the APC by sample withdrawal avoided or neglected? Also, how justified was it to assume the decrease in concentration to be valid for all sizes?

12/30 perhaps the intention was to put the words 'basis for" in the sentence, to read : .... primary basis for the comparison of ...

12/31 replace 'most' by 'largest number' ??

13/18-20 The sentence is somewhat garbled.

14/3-22 These comments would be better placed in a Discussion, not in the Results section.

15/12-14 I think it is too uncertain to explain results in terms of the numbers of proteins. Snomax contains full cells and evidence for separation of the INP material (protein) from the cell wall is unclear. Leveling off the INP curves shows a limit in the number of INPs as a fraction of the total number of cells. This is a matter of expression of the INP protein under induced conditions and of the processing of the bacterial culture. Again, the emphasis here should be on the agreement among measurement systems. Interpretation of the shape of the INP curves is a matter for Discussion (see Broad comments 7 and 8).

15/25-> Reference here to first-freezers etc. is confusing without more detailed knowledge of what is being discussed. This type of error analysis for specific devices should be part of the apparatus description (cf. Broad comment 6).

15/32 Supporting Information =? Supplement. Where is the material referred to in this line?

16/10 Why wasn't the fraction expressed as active INP versus total number of potential INP particles introduced? Maybe that is what is meant, but I am not sure from the wording given here.

16/16-17 The conversion to active site density is the same for all measurements, provided particle sizes are known. Why is this introduced here? (cf. Broad comment 10)

20/28-31 Do these two sentences refer to the same or two separate findings?

21 Some numerical values for the errors discussed would be useful.

21/23 'de-agglomeration' has a simpler alternative: 'breakup' and removes the implication that all large INPs are aggregates of many smaller ones

21/28 " ... full immersion of all particles in the same liquid volume .." is too vague to focus on, as other factors like time in suspension may also come into play. Also, are the differences beyond the error bars of the PIMCA-PINC and cold-plate methods?

21/32 why is that need artificial?

22/4 '.. uniformly capture activation ...' is awkward wording