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# ***Interactive comment on “A comprehensive laboratory study on the immersion freezing behavior of illite NX particles: a comparison of seventeen ice nucleation measurement techniques” by N. Hiranuma et al.***

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This paper is the result of a large effort in organization and in execution. It represents a significant step in clarifying the power and the limitations of laboratory ice nucleation studies. It also adds considerable new information about the ice nucleating capacity of

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the mineral illite NX. The authors are congratulated on conceiving and carrying out this work.

The main accomplishment of this investigation is to show that many different measurement methods can be used to arrive at a quantitative evaluation of the ice nucleating ability of illite NX. Using the same sample of the mineral and performing measurements with the instruments located at their home bases is a useful alternative approach to the inter-comparison workshops with co-located instruments. Discrepancies among the various measurements in this intercomparison were about the same magnitude as those found for simultaneous measurements with a dust sample in the 2007 workshop (DeMott et al. 2011). Here a larger number of instruments were involved, with a greater diversity of operating principles, so the comparable result represents a success and perhaps even some advantage. It is worth noting that the results represents a substantial improvement over the long term; the scatter was much worse in the results of the 1975 workshop (Vali, 1976).

However, the results also demonstrate fairly serious limitations. Discrepancies of about two orders of magnitude in the derived measures of ice nucleating ability indicate that comparisons of data obtained in different experiments - past and future - will have to be compared with that sort of variability in mind. Furthermore, measurements of the abundance of INPs in the atmosphere or in other systems have to be accepted with similar possible error ranges.

The approach of using a sample powder distributed to different locations has its own difficulties, principally that of ensuring sample stability. It could be expected that a mineral powder is fairly stable but that is not absolutely certain. The effects of oxidation, humidity changes, radiation, aging, vapor adsorption, etc., cannot be separated from differences that arise due to variations in measurement techniques. Tests conducted with the suspensions to diagnose changes in composition (last paragraph on page 22055) is a step in the right direction and shows the possible importance of such tests.

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What do the results say about the success of this endeavor? First, the greater degree of agreement among the measurements with suspensions shows that those methods have greater control and fewer uncertainties than the tests with dry aerosols. The downside to the drop-freezing tests is that the background noise level is relatively high, restricting measurements to temperatures above  $-20^{\circ}\text{C}$  or  $-25^{\circ}\text{C}$  at best. Second, the scatter in the results for dry aerosol methods is due to diverse operating principles on which the measurements rely. These uncertainties are difficult to surmount. Third, the results support the notion that the frequency of nucleating sites per particle is proportional to the surface area of the particle for illite NX and similar materials.

Regarding the success of the analyses in terms of  $n_s$ , the criteria for that claim are not clearly stated. One could argue that the scatter of measurements are a combination of the instrumental variations and of incomplete fulfillment of the assumptions of the analysis. Can the authors state what they consider the proof of adequacy of the  $n_s$  analysis? The size-sorted results? Also, could they explain what is meant (22090/15) by “uniform distribution of active sites for available  $S_{\text{total}}$ ”? Independence of site density from particle size? How well is that proven?

The overview of the results in Fig. 6 is not as informative as should be. This graph is valuable in demonstrating the overall trend of the results. However, the author might consider also displaying the results in terms of the ratios of the individual measurements to the geometric mean of all the data across the temperature range covered. That type of display would provide a clearer depiction of the data for evaluating trends with respect to each measurement technique. Also, it would be useful to see results presented separately for the suspension measurements and for the dry aerosol measurements.

The influence of sample size is neglected in the analysis. Weighting data points by error ranges resulting from sample sizes would have been useful.

22059/Eq. (1) Since analysis of the data is being conducted with the time-

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independent singular approximation, it is somewhat misleading and needless to introduce  $J_{imm}$  in Eq. (1). This rate is not used in subsequent steps and there is no definition of what values of  $t$  are used for the different experimental methods. I recommend deleting the middle part of Eq. (1).

There seems to be another problem with Eq. (1) in that it is unclear whether the logarithms is taken over both bracketed terms or only the first one.

Is the equation dimensionally correct?

The value of writing Eqs. (1) - (3) in terms of size bins isn't really useful for this paper, since no size-resolved data are presented and neither were the measurements performed in a size-resolved manner.

Could the authors address what uncertainties arise due to shape assumption, conversion to BET and DLS surface area?

22063/7-10 The authors state that the "... effects of impurities upon ice nucleation activity cannot be evaluated ..." and that the impurities may be responsible for variations in ice nucleating efficiency at various temperatures. The underlying assumption here is that there is a specific temperature of activity associated with each component or impurity. If that is what the authors mean evidence need to be presented. Since that claim is made in the literature only for illite NX, the generalization here made is questionable.

22063/13 It is unclear what special advantage illite NX has as a reference material over other minerals or other materials. The scatter in measured ice nucleating ability by different methods counters this statement.

22066/8-10 What would it have meant if the results showed different  $n_s(T)$  spectra for different mass concentrations? Dilution of samples with clean water is not normally

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expected to change the derived spectra. The statement here is a confirmation of that expectation not a new result.

22066/14-17 Do the values given represent a cut-off size or the center of a narrow band in sizes?

22067/5 Typo in  $n_s(T)$

22067 What is meant by 'effective' surface?

22067/15 abbreviate pL and nL as in previous paragraphs

22068/22 Size-independence is a significant finding and deserves more detailed description (limits if validity, degree of agreement . . .) In Fig. 4 what does "AIDA size selected" refer to?

22069/21 What discrepancy is being referred to?

22073/16 The title of Section 3.3 is not a good reflection of what is actually described.

22073/17 I would have found it useful to have Figure 6 ahead of the detailed presentation of the results from each instrument. Discussions refer to differences from the overall trend, etc. which are not readily perceived from Figs. 4 and 5.

22073/21 Typo:  $in n_s$

22073/22-27 It is unclear to me whether these statements refer to the overall trend

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or some group of data sets.

22074/5 Aren't the numerical values of the slopes negative?

22074/10 Since the fraction of active sites is reflected by the absolute values of  $n_s$ , it is unclear what the authors want to express here.

22074/14-20 There appears to be some repetition here.

22074/27 A possibly significant point is being made here - the amount of scatter in suspension measurements versus dry aerosol measurements - but this is masked by the larger number of the latter type of data. The authors could examine this difference in a rather simple way and it would be very useful to have that analysis presented in the paper.

22076/11-14 A resounding conclusion is stated here only to be qualified in lines 14-17, with more analysis promised. This is confusing. The reference to uniform distribution is not supported by any specific result.

22076/19 Grammar issue: the past tense in this sentence conflicts with the reference to the section to follow and the next sentence which uses the present tense.

22077/2 Typo: space missing between in and  $n_s$ .

22077/2 What does shifting of activation temperatures mean?

22077/5 So-called T-binned data presentation does hardly deserve to be used as section heading. It is a fairly standard procedure.

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22077/13 Typo: space between 'r' and 'for'

22077/20 Grammar: past tense used here is out of sync with the rest of the writing

22077/21 Don't the values of  $Hor(max - min)$  and  $Ver(max - min)$  depend on where those are taken? Are the values indicated in the graphs picked for particular reason? Are these the maxima within the gray bands for each value?

22078/1-3 What is the reason for expecting the results here given?

22078/3-7 The valuation of  $Ver(max - min)$  is too limited. Only the point of its highest value is commented on. It would be useful to provide more information about its numerical value across the entire temperature range.

22078/7-10 What meaning do the authors attach to  $Hor(max - min)$ ? Clearly, the numerical value of  $Hor(max - min)$  is much larger than any temperature measurement error. Is the authors' interpretation related to variations in the activity of sites between one or other measurement method? If so, what reasons can be given for such changes? If  $Hor(max - min)$  is just a reflection of the spread along the abscissa, it does not merit the introduction of a new parameter.

22078/17 Please clarify what you mean by pronounced freezing and differences. In fact, the intention behind this whole sentence is a bit vague.

22078/19 This paragraph mixes past and present tense wording.

22078/23 The distinction drawn for experimental methods using dry aerosol inputs as 'working on a particle by particle basis' is vague. Doesn't the evaluation of suspension measurements also assume that each nucleating site is located on a different particle?

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The authors are hinting at a subtle point which is not explored in detail and is poorly expressed by what is said. The main difference, in my view, is that suspension methods run into background problems at cold temperatures and that dry aerosol methods lack sensitivity (sample volume) at warmer temperatures.

22078/29 Freezing efficiency is not defined.

22079/4-11 While it is easy to agree with the general point being made here, the meaning of many parts of this paragraph is quite vague. What is meant by systematic uncertainty, absolute standard technique, . . .? I think that what is said in this paragraph would be better placed in the Introduction.

22082/23 Was the SBM fit obtained using the LACIS data points or to the straight line shown in Fig. 9?

22085/5 Are particles removed from the filter with full efficiency in the washing process? If that is not sure, it should be mentioned as a potential explanation of the observed discrepancy.

22085/7 Description of this method for FRIDGE is missing in the Supplementary Methods.

22090/23 This paragraph is rather confusing, specially the first sentence.

22091/22 Could you clarify what is meant by ‘temperature change is the major driver of immersion freezing’?

22092/1 What is the connection of this sentence to the previous one?

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## References:

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