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## ***Interactive comment on “Heterogeneous ice nucleation on atmospheric aerosols: a review of results from laboratory experiments” by C. Hoose and O. Möhler***

### **Anonymous Referee #1**

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### **General Comment**

This manuscript is an admirable service to the observational and modeling ice nucleation communities and a welcome addition to the literature regarding ice nucleation studies and parameterizations in general. In gathering and assessing a vast array of data from laboratory studies that present sometimes difficult comparisons, the authors walk the line fairly well of judgment versus considerations based on averages. There are many useful figures. My only minor issue with the paper is that it perhaps goes a little too far in both accepting certain inadequacies and in rejecting certain data sets, driven largely by whether or not the data support the INAS model of ice nucleation. It

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remains to be proven that ice nucleation scales by surface area alone under all circumstances. I suggest that some things simply need to remain as uncertain within current knowledge. I also raise a concern that limitations of instruments to simulate the ice nucleation response to specific water supersaturations has not been taken into account, so some discussions are premature or unnecessary. The manuscript is otherwise very well written. I list a host of specific critical comments below.

## Specific Comments

### *Abstract*

Lines 7: Some definitions are needed already with the appearance of the words “onset” conditions. Perhaps here it simply needs to be in quotations at first mention, as this must have a loosely quantitative meaning, as discussed in the manuscript body at the start of Section 2.

### *3. Qualitative influence of composition on ice nucleation properties*

Section 3.1.1, page 12538, lines 2-3: I would like to suggest that virtually none of the data in the water supersaturated regime in any study using continuous flow diffusion chambers can be attributed in a straightforward manner to specific supersaturation values (high, low or otherwise). This is somewhat of an limitation/artifact of kinetic factors for ice activation detection following droplet activation, freezing, and growth, as discussed by DeMott et al. (2011) and Kanji et al. (2011). One would like to attribute a specific ice nucleation response to a specific supersaturation at one temperature, but the interpretation is for now too nebulous. It is only assured that full condensation has occurred at some higher supersaturation, and then the maximum activation possible at a given temperature in the time allotted is measured. Thus, it is perhaps easiest simply to remove the statement here, and to take care in interpreting special meaning for these data later in the paper.

Section 3.1.3, line 9: This statement could give the impression that some bacteria are

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simply ice nucleation active, but what is being referred to here is that some bacteria (INA) are specially active due to genetics. To group all bacteria in general, for example including those that might possess some IN activity via other means, is somewhat misleading. Some words of explanation are required.

Section 3.1.4: For completeness, this section should mention that some studies have not detected freezing by ammonium sulfate aerosols.

Section 3.1.5, line 10: Why is it assumed that the biomass burning aerosols should be categorized as organic acids and humic-like substances? How does one know the composition in order to characterize them this way? Could these not also possess soot cores?

Section 3.2, pages 12541-12542: I suggest that the variation of mineral dust IN activity with purity is a well enough validated that it could at least be introduced as an issue prior to showing results that are described as for “otherwise comparable conditions.”

Section 3.4, page 12544, Fig. 9: This section and this figure disguise a lot going on with regard to the impact of coatings on ice nucleation. Fig. 9 appears to compare data from different ice nucleation regimes, so it is a bit odd to my view, unless some differentiation of temperature and/or ice nucleation regimes is noted. To contrast coated and uncoated particles, should one not focus the comparison where the interpretation is somewhat clear? For example, only data below water saturation could be compared in order to restrict conditions to where deposition nucleation should be affected (for example, reduced in preference to freezing of dissolved solute coating) in a similar manner. When data from where ice onset occurs in mixed phase cloud conditions are included, the plot now folds in the condensation/immersion freezing mechanisms and so there seems no reason to believe the points would or should fall in the same regime of dependence on ice supersaturation. Also, the referenced Sullivan et al. (2010a) paper demonstrates a different impact of nitric acid reaction within the deposition versus mixed-phase regime, but what is mentioned here is a misinterpretation in my opinion.

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Nowhere in Fig. 2 of Sullivan et al. is it apparent that nitric acid coating elevated ice nucleation within any expected experimental uncertainty. I suggest removing that statement, and to revisit the form and discussion of Fig. 9.

#### 4. *Determining factors of ice nucleation efficiency*

Section 4.1.1, page 12547, line 7: Niedermeier et al. (2011) comparison of LACIS and CFDC methods seems to support the assumption made here.

Section 4.1.1, page 12547, lines 17-22: Four points regarding this discussion –

1) After making the assumption noted above, it is odd to then refer to the freezing events in CFDC's as "supposed" condensation freezing events in line 17. They are either comparable, or "assumed" so.

2) I fail to see the less clear temperature dependence of INAS values for the CFDC's in Fig. 11, taking into consideration all data shown here or available elsewhere. I suspect that some of the appearance of such relates to the type of data chosen or readily available (single active fraction) and the range of RH values used as reference above water saturation (an instrument limitation, as mentioned above). Yet, for ATD, most results are consistent within the scatter of results, as near as I can see. Data from Sullivan et al. is shown at -30C (from Sullivan et al. 2010a or 2010b?). There is data from -25C in Table 1 of Sullivan et al. (2010b) that supports a decrease of about 2 in IN activity compared to -30C, and it is not clear if there is a point for that in Fig. 11a. For desert dusts, I assume that the data used here has been restricted mostly to experiments using monodisperse particles? If that is not the case, then Fig. 6 of DeMott et al. (2011) shows a slope of active fraction for polydisperse Saharan dust that appears to align quite well with the Niemand et al. data shown in Fig. 11b. DeMott et al. also discuss that one needs to consider the RH value used for comparing data from different CFDC's, supporting that maximum active fraction values reported in the water super-saturated regime are probably most appropriate for capturing condensation/immersion freezing. It is not clear what was done in the comparisons shown, but it is worth noting

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the type of data that may be required to come to firmer conclusions.

3) The meaning of the last sentence on “instrumental boundary conditions” is unclear. Please explain.

4) There is another possibility not considered, which is that IN activity does not perfectly scale with surface area at all sizes.

Page 12548, top paragraph: For kaolinite, I really think the different types need to be considered as a source of discrepancies.

Page 12548-125499: Application of the INAS model for bacterial IN seems somewhat contrived. Is it necessary to define a new category of “high ice nucleation activity” biological particles? These are INA bacteria correct? Also, the answer to whether the steep increase of INAS for INA is maintained is already known right? The answer is that Ina proteins are nearly fully active by -12C (Govindarajan and Lindow, Govindarajan, Proc. Nat. Acad. Sci. USA, 85 (5), 1334-1338), and a perusal of the literature indicates that the active fraction for these increases over a number of orders of magnitude down to this temperature. It must eventually level off and I believe that reasons for this are well understood. A demonstration of the leveling off of complete IN activation of such bacteria is shown in the Ward and DeMott (1989) paper referenced in Table 3. I suspect that a great deal more similar supporting data now exists following workshop studies over the last several years, it simply has not been published.

Page 12551: Regarding the issue of time dependence, I am curious to know if the vast database of AIDA chamber experiments might be analyzable in the manner of Eq. (4)? I am not suggesting that it is necessary here, but it would certainly seem appropriate to apply this toward AIDA data as well as to other instruments for which the appropriateness of such an analysis is also not known.

Page 12552, lines 6-7: Is it unclear “why other aerosol particles behave differently” than pure kaolinite, or is it more unclear why they do not? I believe that Broadley et al. offer

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a reasonable suggestion as to why most mineral dust IN are not so purely stochastic in their ice nucleation behavior. Furthermore, Vali has already published much on the nature of most atmospheric IN in this regard. There remains some uncertainty, but there seems little doubt that a purely stochastic description of single atmospheric IN is not applicable. This is covered later in the present paper during the discussion of parameterizations based around classical theory.

Page 12553, Section 5.1.1: The last sentence seems a bit pre-determined. The Niemand et al. parameterization utilized additional data collected in the warm temperature regime not included in previous studies, including a correction to the Connolly et al. data I believe.

### *5. Parametrical descriptions*

Page 12555, section 5.1.3: I was only a bit surprised not to see reference to the recent publication of Curry and Khvorosyanov (Atmos. Chem. Phys., 12, 1151–1172, 2012) regarding parameterizations based on classical nucleation theory. They seem to suggest that single contact angle models work if the size distribution and number concentrations of IN are “appropriately” (however one would know that) specified. Or did I interpret their paper incorrectly, and do you care to comment?

Page 12557, end of section 5.2.1: Indeed immersion freezing cannot be represented on such a plot as Fig. 19. However, the nearly vertical isolines in Schaller and Fukuta (1979) may not represent solely immersion freezing rates, but condensation freezing as a distinct mechanism at the very limit of its occurrence for pure particles instantly exposed to supersaturations. It is not expected that any of the measurements shown in this paper would or could reproduce/corroborate this behavior in any case, as none possess the resolution of supersaturation possible in the small laboratory device used by Schaller and Fukuta. For example, in continuous flow chambers, there is supersaturation variation along the thermodynamic trajectory into the instruments, there is some variation across the aerosol lamina, and there can be kinetic limitations to first

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detection of ice forming in the supersaturated regime. Much of the dependence shown could be an artifact of the methods used. The consequence is that one probably should not overemphasize the meaning of supersaturation sensitivity to ice nucleation in these conditions. Certainly the instruments are not capable of resolving any supersaturation dependence within the resolution of a CCN instrument. Thus, I think that much of this discussion could be removed.

Page 12557, section 5.2.2: If the “Phillips” parameterization is being evaluated for qualitative agreement with the few observations shown, I would not judge that the curves are of a distinctly different shape. They look qualitatively similar.

### Conclusions

Page 12559, lines 17-19: I am not sure what studies or results discussed are being referred to in this sentence on immersion freezing INAS densities.

Page 12560, first point: It is worth pointing out that a few dilemmas exist for meeting this recommendation. First, how does one know the nucleation mode for sure? It is only an educated guess based on a number of factors, unless the method restricts the mode. Single mode instruments have their place, but so do those that allow particles to act naturally. Secondly, something not discussed in this paper is the fact that it is not necessarily the size distribution of the aerosol type in total that matters, but the size distribution of the IN fraction active under a set of conditions. This is not known a priori, unless perhaps methods (always imperfect) have been used to size-select particles, and it is extremely difficult to validate after ice nucleation. Thus, in experiments with a distribution of particle sizes, one is making an assumption of one basic “ice nuclei” population for the aerosol type being used. This assumption may not always be valid.

Page 12560, sixth point: Again, a word of caution, in that deposition nucleation is somewhat ill-defined a priori for any set of particles in a humidity field below water saturation. The mode of action below water saturation must be assumed, and the potential role of contaminants neglected.

## Editorial comments

Abstract, page 12534, lines 2-3: As this sentence is discussing multiple ice nucleation mechanisms, I suggest “the” before “temperature and supersaturation” and ending with “the time dependence of ice nucleation.”

Section 3.4, page 12544, line 25: “have” for “has”.

Throughout the paper: Please add “freezing” after condensation in referring to ice nucleation experiments.

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Interactive comment on Atmos. Chem. Phys. Discuss., 12, 12531, 2012.

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