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Atmospheric Chemistry and Physics Discussions

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

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We thank the reviewer for his/her very thorough, competent and constructive review of the manuscript which has lead to substantial improvements of our text. The reviewer's comments are included below in italics.

*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 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.*

We have followed the reviewer's suggestion and have put "onset" into quotation marks at the first two appearances in the abstract.

*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.*

We thank the reviewer for these insightful remarks. We have decided not to omit this point here, because it has frequently triggered questions from the audience when presenting this plot. Therefore the sentence was modified and more information was added: "Some data are also reported at significantly water-supersaturated conditions (e.g., Koehler et al., 2010). This may be due to kinetic limitations and humidity inhomogeneities within the continuous flow diffusion chamber (CFDC) instrument (Petters et al., 2009; DeMott et al., 2011)."

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

These sentences have been reformulated as follows, taking into account suggestions by Dr. C. Morris: "Only a small number of bacterial strains and fungal species has been identified as ice nucleation active (INA) at high subzero temperatures. It has been shown that these possess an ice nucleation active protein located in the outer cell wall which has structural similarities to the crystal lattice of ice (Govindarajan and Lindow, 1988; Kajava and Lindow, 1993). Even among these INA bacteria, only a small fraction of all cells actually nucleates ice at temperatures roughly higher then $-10°$C (Hirano and Upper, 1995)."

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

We have added the following sentence: "In contrast, Chen et al. (2000) observed ice formation on crystalline sulfate particles only at relative humidities higher than those required for homogeneous freezing." In Table 4, Hung et al. (2002) was added, and the table caption was changed from "Laboratory experiments with ammonium sulfate particles" to "Laboratory experiments with solid ammonium sulfate particles".

*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?*

We didn't mean to imply that biomass burning particles consisted (only) of organic acids and humic-like substances. This last figure of ice nucleation "onset" conditions simply assembled all those particles which could not be clearly fitted into one of the other categories. We have modified the title of Figure 5(c) and the figure caption and now explicitly name biomass burning particles.

*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."*

We have added a sentence at the beginning of this section:"For pure minerals, the chemical composition and the crystalline structure can be determined more accurately at least in theory. For example, Mason (1960) tried to relate the ice nucleation abilities of different minerals to their lattice structure. It should be noted that impurities in the naturally occurring minerals impede the interpretation of the results (see discussion below)."

*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.*

A couple of experiments (Cziczo et al., 2009; Koehler et al., 2010; Chernoff and Bertram, 2010) have demonstrated that at least at some of the investigated temperatures, deposition nucleation was completely suppressed and ice could only be activated when water saturation was approached, i.e. most probably via condensation freezing. It was our intention to include these results in our figure, which made it necessary to combine deposition nucleation and immersion/condensation freezing into one plot. At the same time, we can not be fully sure about the ice nucleation mode in particular when ice formation is observed below, but close to water saturation, which makes a classification ambiguous. We have therefore modified Fig. 9 such that it now shows the saturation ratios with respect to water instead of the saturation ratios with respect to ice. In experiments with coated particles which required water saturation, supersaturation or humidities close to water saturation, a transition to the condensation/immersion freezing mode is a likely explanation. In addition, the temperature ranges of the experiments are now all noted in the legend. The discussion in section 3.4 has been revised to describe the new figure: “The saturation ratios with respect

to water for activation of a specific fraction of particles at otherwise similar conditions (temperature, particle size) are reported. Most measurements shown here report higher saturation ratios for coated particles than for uncoated ones, i.e. a deactivation of mineral dust by surface treatments. Where the saturation ratio with respect to water is above or close to 1 for the coated particles, this implies that at the given temperature the treated particles could not or only to a minor fraction be activated in the deposition mode, and that instead condensation of liquid water was required before ice formation.”

*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. 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.*

This statement was referring to the higher activated fractions for nitric acid exposed particles at $RH > 98\%$ in Sullivan et al. (2010a), Fig. 1a and b. However, the reviewer is correct that this is within the experimental uncertainty, and we have revised this statement: “Sullivan et al. (2010a) found suppression of deposition nucleation by nitric acid coatings on the nucleation ability of ATD particles, but no apparent effect above water saturation.”

*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.*

We have added this reference: “This assumption is corroborated by a comparison to flow tube experiments (Niedermeier et al., 2011a).”

*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.*

This was a language problem on our side. "supposed" has been replaced by "assumed" in the revised manuscript.

*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.*

We agree that this is probably due to the type of data that were available to us. We were referring to the fact that the data from the three listed references, if examined individually, did not show a temperature dependence of the estimated INAS densities. E. g., the data by Kanji et al. (2011) show up on a horizontal line in Fig. 11a, because they were all reported for the same activated fraction of 0.1% in Kanji et al. (2011), their Fig. 3. "Some of the assumed condensation freezing events in continuous flow diffusion chambers (Welti et al., 2009; Koehler et al., 2010; Kanji et al., 2011) appear to exhibit a less clear temperature dependence of the estimated INAS densities (Fig. 11a, b), because based on the available data, these could only be derived for selected activated fractions."

*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.*

The data in Fig. 11a were based on Sullivan et al. (2010a). We have replaced them as suggested by the data for -30 and -25C from Sullivan et al. (2010b) and modified the legend accordingly. Thanks for the hint.

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

The data by DeMott et al. (2011) (for RH$_w > 105\%$) have been added to Fig. 11b. Furthermore, the following sentence has been inserted at the end of the previous paragraph: "In CFDCs, the activated fraction at a given temperature increases further when the relative humidity is increased to several percent above water saturation, possibly because of the dilution of impurities on the particles surfaces in larger droplets (DeMott et al., 2011). This can explain some of the spread in the data discussed below."

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

We were referring to the increase of IN activation above water saturation. This sentence has been replaced by the discussion added in response to the previous point.

*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.*

These are valid comments. We have inserted the following two sentences: “Furthermore, impurities in the dust samples and physical or chemical processing can be a source of the observed discrepancies, as discussed in section 3.2. Finally, it is also possible that different size sections of the investigated samples exhibit different ice nucleation properties. ”

*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?*

We agree with the reviewer that the new definition of a category of “highly ice nucleation active bioaerosols” is not necessary, as these are exclusively INA bacteria. The relevant sentences have been reformulated.

The aim of the application of the INAS density concept to INA bacteria and other bioaerosols was to provide a quantity which is directly intercomparable for different aerosol species, without the complication of size. In our opinion, this intercomparison is particularly illustrative for INA bacteria, which are significantly more active than mineral dust not only because of their size.

*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 theWard 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.*

We have reformulated the last paragraph of section 4.1.1 and have added a reference to Govindarajan and Lindow (1988): “The INAS density for dust and biological particles does not necessarily retain this steady increase at decreasing temperatures (although a number of experiments with one aerosol type found similar exponential temperature dependencies of $n_s$, Niedermeier et al., 2010; Murray et al., 2011). In particular, it has been observed that $n_s(T)$ for INA bacteria levels off at temperatures lower than $-10°$C (Govindarajan and Lindow, 1988; Ward and DeMott, 1989). Furthermore, it should be noted that several measurements show large deviations from the average values of $n_s(T)$, and that a difference by one or two orders of magnitudes has a large impact if applied for the calculation of atmospheric IN concentrations.”

*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.*

We will certainly explore this idea in future studies.

*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 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.*

We agree that it is rather a surprise that the kaolinite particles studied by (Murray et al., 2011) can be fitted well with a single-component stochastic model than that other dusts can't. This sentence was removed. The whole paragraph was rewritten as follows: "In extensive early studies (Vali and Stansbury, 1966; Vali, 1994, 2008), it was shown that immersion freezing exhibits time-dependence, but that the observed time dependence is weaker than expected from classical nucleation theory (see Appendix A1). Recently, further in-depth analysis of the influence of time on the activated fraction were conducted by Murray et al. (2011), Broadley et al. (2012) and Welti et al. (2012). In their experiments, the cooling rate or residence times could be varied by more than a factor of 10. Their results are summarized in Fig. 16, along with two experiments by Niedermeier et al. (2011b) and DeMott (1990). While Murray et al. (2011)'s and Welti et al. (2012)'s results indicate significantly more ice nucleation in experiments with slower cooling rates, the other studies do not confirm this behaviour and instead show sometimes more, sometimes less ice nucleation when more time is available for freezing. It should also be noted that the spread in the data entering these calculations is large. For none of the experiments, the ratio of ice fraction or INAS densities in the slow versus fast experiments converges towards the ratio of the cooling rates or residence times, as would be expected if nucleation was described well by a temperature-dependent nucleation rate with one value for all particles. Nevertheless, Murray et al. (2011) found that such a single component stochastic model fits their data best, in particular if constant-temperature experiments are considered. Welti et al. (2012) could best fit their results with a distribution of contact angles."

*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.*

We meant to imply that this parameterization also agrees with data which it wasn't based on, e.g. the data by Zimmermann et al. (2008). However, these other measurements are few and the estimated INAS densities have large uncertainties. Therefore this sentence has been modified as follows: "Niemand et al. (2012)'s parameterization included measurements in a wider temperature range and extends up to $-12°$C." The data by Connolly et al. (2009) have now been corrected by a factor of 10 as described in Niemand et al. (2012).

*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?*

Curry and Khvorostyanov (2012) and the earlier publications by V. Khovorostyanov and co-authors have developed a CNT-based description of "deliquescence heterogeneous freezing", which could also be termed deliquescence followed by immersion freezing. We had originally not included this parameterization in the appendix (though it was cited earlier in the text) for several reasons. First of all, our article is meant to be a review and as complete as possible summary of heterogeneous ice nucleation experiments, but not a complete overview over existing parameterizations (there are a few more which would have to be mentioned in that case). We have therefore chosen to briefly summarize and intercompare only those parameterizations which are frequently used to fit results from laboratory experiments. Second, it is not straightforward to apply this parameterization to immersion freezing experiments where the particles have not become immersed by deliquescence and condensation in an air parcel where they interact with and deplete the environmental vapour concentration, but in instruments where the supersaturation is prescribed or where the particles are suspended in the droplets via other pathways. Third, the parameterization includes several additional free parameters (e.g. the insoluble mass fraction and the droplet radius) than just the contact angle, making it difficult to produce plots equivalent to our Fig. 23 or Fig. 21.

To clarify our intentions, the following sentence in section 5.1.3 was reformulated: “Some of the different formulations of classical nucleation theory which have been used to fit laboratory measurements are summarized in Appendix A1.” In section A1, Curry and Khvorostyanov (2012) is now briefly discussed and their formulation of $\Delta F_{diff}$ is added to Fig. 22. The new text reads: “Khvorostyanov and Curry (2004) provide a unified treatment of deliquescence and condensation freezing, in which the energy for homogeneous germ formation has a different form than Eq. A2 and requires knowledge about additional parameters (see also Curry and Khvorostyanov, 2012). Their prefactor $A_{imm}$ is identical to Zobrist et al. (2007), but the activation energy has a different temperature-dependent form (Khvorostyanov and Sassen, 1998):”

$$\Delta F_{diff,Khvorostyanov} = 0.694 \times 10^{-19}\mathrm{J} \times (1 + 0.0027 \times (T_c + 30)) \quad (1)$$

$$A_{imm,Khvorostyanov} = A_{imm,Zobrist} = n_{1,w}\frac{kT}{h} \quad (2)$$

*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 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.*

Thanks for these insightful remarks. As suggested, we have removed the last sentence of section 5.2.1, and have reformulated the second-last one as follows: “Schaller and Fukuta (1979) measured nearly vertical isolines for condensation freezing above water saturation. The CFDC measurements included in Fig. 19a have a large uncertainty in the maximum supersaturation seen by the aerosol particles, such that this regime is poorly resolved.”

*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.*

We agree that the parameterization by Phillips et al. (2008) captures the major features of the experimentally determined isolines, but the ‘kink’ at -45C in Figs. 20b and 20c kept us from calling this a ‘good agreement’. We have changed this sentence to: “The resulting $\tilde{n}_s$-isolines for dust/metallic aerosols agree qualitatively well with the observations, while the $\tilde{n}_s$-isolines for black carbon and organic aerosols deviate moderately from the typical shape identified in Fig. 19.”

*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.*

This referred to the parameterizations by Connolly et al. (2009), Niedermeier et al. (2010), Lüönd et al. (2010) and Broadley et al. (2012) shown in Fig. 17a. The reference to this figure was added.

*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.*

These are very important points, which will hopefully be addressed by future studies. We hope that our well-intended recommendations will point the community to work on and discuss these issues.

Based on the reviewer's input, we have attenuated our first recommendation to make allowance for methods which can't meet them in principle. Furthermore, we have added an item about polydisperse versus size-selected measurements, and have added the recommendation to compare single-mode and multi-mode methods to the fifth (now sixth) item.

- "All experiments should as far as possible determine and document the size distribution of the investigated particles, the activated fraction, if known the nucleation

mode, and the residence time or cooling rate."
- "Experiments with polydisperse particles should report the total surface area to relate the ice nucleation activity to this parameter as shown in this manuscript. In particular for particles with complex composition like mineral dust, additional size-selected measurements are required to investigate whether the INAS density is constant over the whole size distribution."
- "Further coordinated experiments with different instruments and identical samples (as in DeMott et al., 2011) are recommended to resolve instrument-dependent differences. In particular, such intercomparisons should strive for closure between single-mode methods (e.g. droplet freezing assays) and instruments covering multiple nucleation modes (CFDCs, cloud chambers)."

*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.*

For the empirical concept of INAS densities and the application of this type of parameterization in models, no further knowledge is required about whether or not liquid layers form on the particles' surface before ice formation. We have therefore reformulated this sentence: "The concept of INAS densities should be extended to the analysis of experiments below water saturation (deposition nucleation/immersion freezing of concentrated solution droplets). "

*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.*

Corrected.

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