

Response to Anonymous Referee #2:

We thank the reviewer for his or her comments which are copied below. Our responses and text modifications are shown in bold. Line numbers refer to the manuscript currently under discussion.

The manuscript introduces a parameterization of heterogeneous freezing processes which is based on Classical Nucleation Theory (CNT) and the use of a single contact angle, while allowing for a variation in surface area of the ice nucleating particles (INP) from droplet to droplet. The model is then used to reproduce a suite of different measurements from different groups, and also a sensitivity study is included.

The effect of variations in surface area certainly exists and has to be accounted for. However, I feel that this effect is presented much too pronounced in the present study and the tone of the whole manuscript has to be tuned down. Reasons for this are twofold: 1) The model introduced here only used a single contact angle, while it has been shown in the past, that this produces a much steeper temperature dependence of the ice nucleation process than is observed in experiments (e.g., Zobrist et al. (2007), Welti et al. (2012), Broadley et al. (2012), Augustin et al. (2013), to name only a few). This has been overcome by assuming a contact angle distribution. And while it is not yet known, on a basic level, how ice nucleating sites on INP look like, it is generally believed that they are not all the same in a single sample, particularly not when a mineral dust sample is used, as these usually contain more than one type of mineral. This makes the assumption of a single contact angle implausible. 2) Also, the width of the surface area distributions in the droplets in some of the experiments, as ascribed by the authors of the present study, seems to be much larger than seems reasonable based on the methods used in these experiments. This, together with the use of a log-normal distribution for the surface areas present in the different droplets, seems to overestimate the effect of the surface area variation. This shows in uncertainty ranges given for the different measurements that were modeled by the parameterization, which, in some cases, are excessively larger than the variation in the measurements themselves, with measured values appearing centered within the range.

We thank the reviewer for evaluating our manuscript. We find this comment helpful for improving the manuscript and thank the reviewer for acknowledging that ISA variance should be accounted for. As outlined in our general response and in responses to reviewer #1 and to G. Vali, we have sound and valid reasons for presenting this different approach of analyzing immersion freezing using a physical model and physical observables. Our approach is fundamentally different to other commonly applied approaches. In this regard, we will follow the reviewer's advice and tone down the language where appropriate.

The first point made by the reviewer is that assuming a single (averaged, particle-type) J_{het} value is not plausible. However, the reviewer also admits that using a contact angle distribution is also an assumption and may be equally not plausible. As stated in our general

comment and response to reviewer #1, we do not assume a single contact angle. Our model is in agreement with the concept of an internally mixed distribution of active sites. This means that while there exists a distribution of contact angles or active sites on a particle surface, this variability is small on a per droplet basis compared to effect of ISA variability. Our results show that droplet to droplet variability in ISA likely accounts for most of the variability in the immersion freezing data. This means that droplet to droplet freezing variability due to differences in particle ice nucleation ability may not be the governing mechanism underlying the interpretation of immersion freezing in the experiments of Broadley et al. (2012) and Wight and Petters (2014). We find evidence that the same is true for Herbert et al. (2014), Wex et al. (2014) and Niemand et al. (2012).

The second point is that the width of log normal distributions is unreasonably large. Again the choice of our distribution width parameter, σ_g , is in accord with experimental parameters given by both Wright and Petters (2013) and Broadley et al. (2012). If sufficient information is not given by other authors, inferences regarding whether or not a distribution is too wide or too narrow cannot be made. As discussed in our manuscript, large uncertainties due to too few employed droplets (Diehl et al., 2014, Herbert et al., 2014) could result in different experimental trajectories of the unfrozen droplet fraction, f_{ufz} , being a straight line or curved.

As a further point, it seems that the manuscript becomes unnecessary long by mentioning the dependence on water activity (or relative humidity, RH) as represented in the model. The effect of variation in the surface area was discussed for immersion freezing measurements where measurements were done on diluted droplets, and therefore the occasional remarks or paragraphs dealing with concentrated solutions seems off the main track of the work presented here. This makes this already long paper even longer.

We feel that including a brief discussion of water activity, a_w , dependence is necessary, but as recommended by the reviewer, we will significantly shorten this section. The reason is that $J_{het}(T)$ is taken from the a_w based immersion freezing model (ABIFM) and used in model simulations (Knopf and Alpert, 2013). Although, we only test our model against experimental studies using water droplets, it is equally capable of simulating aqueous solution droplet immersion freezing experiments. Also, the uncertainty analysis (Fig. 9) is supported by the capability of our model to explain data scatter from multiple immersion freezing experiments including over 18,000 pure water or aqueous solution droplets.

p. 13120, l. 4-6: The following sentence will be removed: "Aqueous solution droplets containing IN and having $a_w < 1.0$ will decrease J_{het} for the same T when compared with pure water droplets, an effect captured by ABIFM (Knopf and Alpert, 2013)."

p. 13136, l. 12-14: The following sentence will be removed: "ABIFM is independent of the nature of the solute, and therefore, it can be applied in the exact same way to immersion freezing of pure water ($a_w = 1.0$) or aqueous solution ($a_w < 1.0$)."

Therefore, the paper needs major revisions before it can be considered for publication in Atmos. Chem. Phys. . However, the topic as such is an interesting one, and when following the remarks given above and the more specific ones given below, a publication in this journal might be appropriate.

Sec. 2.1 and throughout the text:

When comparing your model results with data from literature, it is interesting to note that the ranges you calculate in many cases are much larger than the scatter of the data. This might indicate that you overestimate the variability in the ISA variation. You argue with a range of two orders of magnitude in droplet volumes when you derive σ for Wright and Petters (2013). Translation of that to a σ of 9.5 seems pretty much, though, and I would like to see a plot of the distributed ISA. (Typical atmospheric particle size distributions have modes where σ goes up to a maximum of roughly 2.) For other cold stage experiments, particularly when examined droplets all have the same size, the scatter in ISA should be much smaller. Indeed, you use smaller numbers there, but it remains unclear how you derive values for σ in these cases. Additionally, values for σ you use for particles which are size selected with the Differential Mobility Analyzer (DMA) technique are beyond all plausible values, even when a shift or broadening due to the particle non-sphericity is taken into account (e.g., 8.2 for CFDC and LACIS (side note: there is a discrepancy as 8.2 is given in the text while Table 1 gives 7.7)). For spherical particles, a DMA typically has $\sigma < 1.1$. You have to use more reasonable numbers and justify these numbers much better.

The scatter in the frozen and unfrozen fraction data in many cases lie entirely within our model derived 5-95 percentiles. However, the experimental data is typically derived from only a single experimental run or using a limited number of droplets. If these experiments were repeated 1000 times, then 5-95% of the data from all 1000 experiments should fill the 5-95 percentile bounds. Thus, we conclude that a single experiment or employing too few droplets is insufficient to represent an uncertainty. We note that for the case of Wright and Petters (2013) using ~1000 droplets, the percentile bounds closely constrain the data. In the case of Knopf and Alpert (2013) and Hiranuma et al. (2015), our uncertainty estimates are very similar to root mean square errors and the data scatter, respectively, which supports our uncertainty estimates.

On p. 13117, l.18 – p. 13118, l. 2 we have discussed briefly a selection procedure for model parameters when they are not explicitly given in previous studies. However, we agree with the reviewer that a more clear explanation of selecting σ_g is necessary. Also, we describe the procedure for choosing parameters following the ABIFM for calculating J_{het} values. In brief, when a parameter is not directly stated it has to be fitted to experimental data. The fitted parameter is then compared with knowledge of experimental conditions to assess whether or not it is a feasible value. For example, droplet volumes in Broadley et al. (2012) range by a factor of 8, implying that the minimum ISA variability must be a factor of 8. When considering

other factors like variability in INP numbers and surface irregularities, ISA variability must be much more than a factor of 8. As a best fit, we find $\sigma_g=8.3$ and therefore, a reasonable value. This assessment is done for every fitted parameter. We note that many previous studies report only average ISA per droplet, A_{avg} , and neglect information for estimating σ_g , thus for simplicity we set $A_g=A_{avg}$.

p. 13117, l. 25-26: the sentence “For example, if a study reports that 100 droplets were used in an immersion freezing experiment, then $N_{tot} = 100$, or if the average ISA is reported as $7.1 \times 10^{-6} \text{ cm}^2$, then $A_g = 7.1 \times 10^{-6} \text{ cm}^2$. For all studies in which a parameter is not available or easily calculated, an estimate which best reproduces experimental conditions is determined.”

will be changed to,

“For example, if a study reports that 100 droplets were examined in an immersion freezing experiment, then $N_{tot} = 100$. Some previous studies report only average ISA per droplet, A_{avg} , and neglect information for estimating σ_g . If the average ISA is reported as $7.1 \times 10^{-6} \text{ cm}^2$, then for simplicity we set $A_g = 7.1 \times 10^{-6} \text{ cm}^2$. For all studies in which a parameter is not available or easily calculated, it is fitted to experimentally derived f_{ufz} or f_{frz} , and critically assessed whether or not the parameter best reproduces experimental conditions. This applies to J_{het} and the σ_g parameter, the latter of which is not typically considered in previous studies.”

In response to the reviewer’s comments, we have altered our model simulation by constraining the Fluka kaolinite ISA distribution in Wex et al. (2014) to the multiple charge distribution outlined in Wiedensohler and Fissan (1988). We have calculated the probability for particles having multiple charges as a function of particle diameter, $P(\ln D_p)$, at a constant electrical mobility diameter of 300 nm. The distribution $P(\ln D_p)$, is a probability density function from which particle diameters can be sampled in new simulations, IsoCFDC and IsoLACIS. We will include a new supplemental figure, Figure S1, which shows the result of sampling 833 particle diameters from this distribution. Individual particle surface area is calculated assuming spherical particles. Using this new ISA distribution, shown in Fig. S1, a lognormal distribution and parameters μ_g and σ_g are not required in IsoCFDC and IsoLACIS.

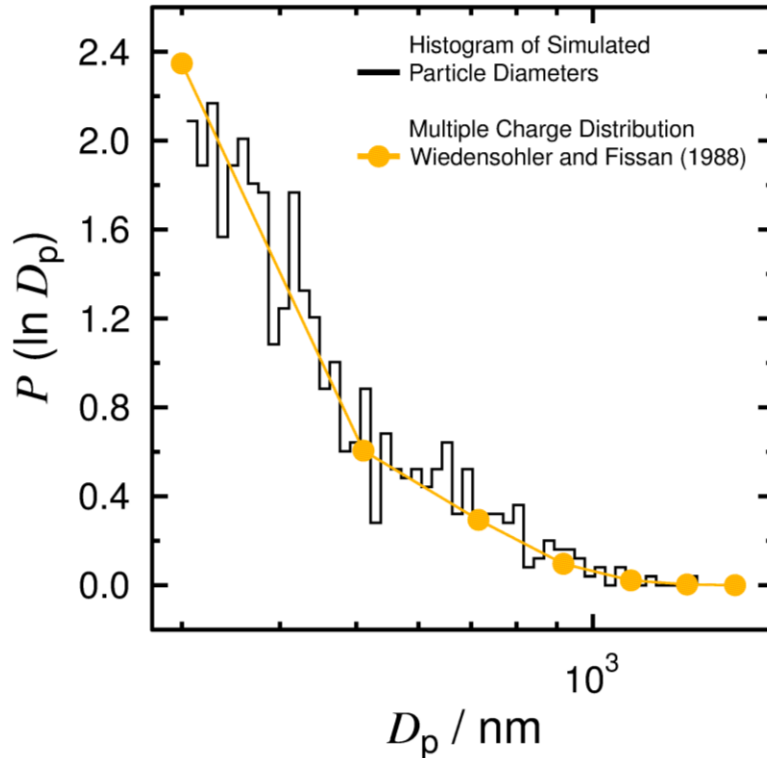


Figure S1: Probability density function, $P(\ln D_p)$, of multiple charged particles with respect to particle diameter, D_p , with a constant electrical mobility diameter equal to 300 nm. Orange circles at 300, 510, 720, 920, 1100, 1300 and 1500 nm represent particles with increasing multiple charges, i.e. 0, 1, 2, 3, 4, 5 and 6 extra charges, respectively. The histogram in black shows the frequency distribution of 833 randomly sampled D_p from $P(\ln D_p)$.

p. 13132, l. 27: “...tendency for greater surface area than assumed. Therefore, a distribution of particle surface area is expected and used in IsoCFDC and IsoLACIS with parameters $A_g = 6.2 \times 10^{-8} \text{ cm}^2$ and $\sigma_g = 8.2$. These values were fitted to experimentally derived f_{frz} .”

will be changed to

“...tendency for greater surface area than assumed. Additionally, particles of larger diameter, and thus larger surface area, may have the same electrical mobility due to the presence of multiple charges. Therefore, a distribution of particle surface area can be expected. Following Wiedensohler and Fissan (1988), the probability for particles having multiple charges as a function of particle diameter, $P(\ln D_p)$, at a constant electrical mobility diameter of 300 nm is shown in Fig. S1. The distribution $P(\ln D_p)$ is a probability density function from which particle diameters are sampled in simulations IsoCFDC and IsoLACIS. Individual sampled particle surface area is calculated assuming spherical particles.”

Also, while a log-normal distribution might capture the distribution of INP surface areas in droplets when these droplets are prepared from suspensions (e.g., for cold-stage experiments), this is likely not the case for those experiments where particles were size selected using a DMA. This makes me wonder how your results would look like if you used a normal distribution, instead of a log-normal one, a topic you might want to address in your work.

We chose to derive the variability in ISA from the multiple charge distribution and not to use a Normal distribution. Please see previous comment.

p. 13113, line 9-13: You may note that the publications you list here are only some of a much larger number.

p. 13113, l. 9-13: “a variety of experimental methods, including the droplet-on-substrate approach (Zobrist et al., 2007; Knopf and Forrester, 2011; Alpert et al., 2011a, b; Iannone et al., 2011; Murray et al., 2011; Rigg et al., 2013; Broadley et al., 2012), oil-encased droplets (Murray et al., 2011; Broadley et al., 2012), differential scanning calorimetry (Pinti et al., 2012), and continuous flow diffusion chamber (Archuleta et al., 2005).”

will be changed to

“a variety of experimental methods, including the droplet-on-substrate approach (Zobrist et al., 2007; Knopf and Forrester, 2011; Alpert et al., 2011a, b; Iannone et al., 2011; Murray et al., 2011; Broadley et al., 2012; Rigg et al., 2013), oil-encased droplets (Murray et al., 2011; Broadley et al., 2012), differential scanning calorimetry (Marcolli et al., 2007; Pinti et al., 2012), and continuous flow diffusion (Rogers et al., 2001; Archuleta et al., 2005; Hartmann et al., 2011; Kulkarni et al., 2012; Wex et al., 2014). These previous studies represent a subset of a much broader selection of experimental methods and designs.”

Paragraph starting at p. 13113: The list of publications you cite in lines 18-20 seems to mostly include studies for which the here mentioned parameters (T , RH , t and A) and their uncertainties are comparably well known. However, it seems to be said here that the respective values are difficult to determine, particularly in the cited publications. The text gives a misleading impression, and rewording is needed. Additionally, the sequence of models you cite is somewhat irregular. The DeMott-parameterizations aims exclusively at deriving INP concentrations, while others use CNT to model frozen fractions (a-pdf, active site, soccer ball model), and yet others omit a time dependence - but these do not appear grouped. This whole section could gain if it were reformulated.

We apologize for the confusion here and agree that this section needs to be reworded to avoid misleading the reader.

p. 13113, l. 14-24: These sentences will be changed to, “The major difficulty with a variety of experimental techniques is how accuracy and uncertainty of T , RH , t , and A

are assessed and how these uncertainties affect extrapolation of laboratory derived ice nucleation parameterizations to atmospherically relevant conditions. Previous investigations have developed state of the art instrumentation and methods to constrain uncertainties (Connolly et al., 2009; Lüönd et al., 2010; Niedermeier et al., 2010; DeMott et al., 2010; Niedermeier et al., 2011; Hoose and Möhler, 2012; Niemand et al., 2012; Rigg et al., 2013; Hiranuma et al., 2015; Vali and Snider, 2015). However, interpreting ice nucleation using empirical parameterizations or models that are fitted to measured frozen fractions and ice crystal concentrations are inherently constrained to the investigated range of T , RH , t , A and concentration of INPs (Rigg et al., 2013; Knopf and Alpert, 2013).”

Please note that the list of previously published parameterizations and models are not intended to be grouped in any order. They are simply relevant examples of parameterizations and models, in which extrapolations are potentially uncertain.

We also wish to use updated heterogeneous ice nucleation terminology following Vali et al. (2014). As such, all instances of the abbreviation ice nuclei (IN) will be changed to ice nucleating particle (INP) or ice nucleating particles (INPs).

p. 13114, line 3-5: Wex et al. (2014) used both, a time-dependent and a time-independent (i.e., such a simple) parameterization, including a freezing point depression, and both approaches described the measurements. It is therefore not correct that this cannot be done with these parameterizations.

We will clarify the main message in this paragraph: A choice of fitting functions and fitting parameters can be made to fit experimental data, but this does not guarantee that a fit can be applied beyond the investigated laboratory conditions. This also applies to Wex et al. (2014) who parameterized their f_{frz} data as a function of T using an approach following Koop and Zobrist (2009), in which a constant temperature offset, $\Delta T_{\text{het}} = \lambda_{\text{het}} T_m$, was used to describe freezing temperatures where T_m is the melting temperature of the aqueous solution and λ_{het} is a fitted parameter. The authors applied both freezing point depression and the singular description, implying that the concentration of solutes modifies the number of ice active sites. This is another example that suffers from the fact that ice nucleating sites are incapable of being measured or characterized.

p.13113, l. 29 - p. 13114, l. 9: “and the soccer ball model (Niedermeier et al., 2011), in which the latter assume that ice preferentially occurs on ice active sites located on the particle surface. According to the singular hypothesis, the number of active sites, $n_s(T)$, is dependent on T only. Furthermore, these parameterizations cannot describe the freezing point depression (e.g. Zuberi et al., 2002; Archuleta et al., 2005; Koop and Zobrist, 2009) and nucleation kinetics (in analogy of homogeneous ice nucleation) observed in immersion freezing experiments where the IN are immersed in aqueous solution droplets (Rigg et al., 2013; Knopf and Alpert, 2013). These limitations clearly

support further analytical efforts to improve our understanding on the governing parameters of immersion freezing.”

Will be changed to

“and the soccer ball model (Niedermeier et al., 2011). For example, Rigg et al. (2013) showed that the single contact angle model, α -PDF model, active site model and singular description cannot describe the freezing point depression (e.g. Zuberi et al., 2002; Archuleta et al., 2005; Koop and Zobrist, 2009) and nucleation kinetics (in analogy of homogeneous ice nucleation) observed in immersion freezing experiments where the IN are immersed in aqueous solution droplets (Rigg et al., 2013; Knopf and Alpert, 2013). According to the singular hypothesis, the number of active sites, $n_s(T)$, is dependent on T only and neglect ice nucleation kinetics. Wex et al. (2014) parameterized f_{frz} data accounting for the freezing point depression using a temperature offset approach following Koop and Zobrist (2009) and using a singular description, i.e. deriving $n_s(T, a_w)$. However, the approach the authors used is solute type dependent (Koop and Zobrist, 2009) and thus, may be cumbersome for atmospheric application where INPs can be associated with a wide variety of solutes. These limitations clearly support further analytical efforts to improve our understanding on the governing parameters of immersion freezing.”

p. 13114, line 17-18: Niedermeier et al. (2010) which you already cite above, belongs to this list given here, too.

This reference is added.

p. 13114, line 25: You could give an estimate of the uncertainty in the surface area estimates already here, based on the literature you cite.

Providing an uncertainty estimate of ISA variability in experimental studies which do not provide sufficient information to derive σ_g would be beneficial. However, analytical formulation of particle surface area or variability is beyond the scope of our work and would suffer great uncertainty due to the lack of quantitative particle sizing information.

p. 13115, line 12-13: Why is it 7 independent studies but 8 different instruments? Please check.

The 7 studies cited are i) Wright and Petters (2013) studying Arizona Test Dust, ii) Broadley et al. (2012) studying illite, iii) Herbert et al. (2014) studying kaolinite, iv) Herbert et al. (2014) studying feldspar, v) Diehl et al. (2014) studying illite, vi) Wex et al. (2014) studying kaolinite and vii) Niemand et al. (2012) studying Saharan dust.

The 8 instruments cited are i) picoliter droplets prepared by oil emulsion (Wright and Petters, 2013), ii) picoliter droplets on a cold stage covered in oil (Broadley et al., 2012), iii) microliter droplets on a cold stage (Herbert et al., 2014), iv) wind tunnel levitation (Diehl et al., 2014), v)

acoustic levitation (Diehl et al., 2014), vi) continuous flow diffusion chamber (Wex et al., 2014), vii) Leipzig aerosol cloud interaction simulator (Wex et al., 2014) and viii) Aerosol Interactions and Dynamics in the Atmosphere chamber (Niemand et al., 2012).

p 13115, line 25: Droplets “will” not necessarily possess different ISA, at least not to the extent you suggest here, so exchanging “will” by “might” is more appropriate.

We disagree with the reviewer on this point. It is a physical impossibility that two droplets can be prepared containing exactly the same ISA. There will always be variability in size, pattern of cracks, edges or surface irregularities, number of molecules, etc. which create differences in surface area.

We agree that the ISA distribution may not be as broad as suggested in our model simulation for some cases. The uncertainty of experimental ISA distribution is unfortunately not well constrained exactly due to the lack of data. However, this does not apply to the studies of Wright and Petters (2013), Broadley et al. (2012) and Niemand et al. (2012), in which we have sufficient support for our employed ISA distribution widths.

p 13116, line 8-14: You are correct that it is often assumed that all droplets contain the same ISA. But I am not convinced that this is necessary in principle. As long as the total available ISA is known, derived parameters as e.g. surface site density, should be the same, no matter if all droplets contain the same ISA or if it is distributed. Otherwise experiments with poly-disperse INP, as e.g. done in AIDA, should result in clearly different surface site densities, when compared with methods which constrain the ISA per droplet to a much narrower range. This, however, is not what is seen in the comparison given in Hiranuma et al. (2015).

Experimental uncertainties are important to consider when answering the question posed by the reviewer. In Hiranuma et al. (2015), experimental data spans ~4 orders of magnitude. We note that considering this uncertainty, all the compiled data by the authors are in agreement despite different scales in time, surface area and temperature. This is also in spite of ISA variability. Therefore, it may be possible that surface site densities, i.e. $n_s(T)$, are clearly different. However, with an uncertainty range of 4 orders of magnitude, $n_s(T)$ from previous studies are in agreement. We find that ISA variability and a time-dependent stochastic freezing process can explain this uncertainty. Using our model simulations, we provide a clear and detailed uncertainty analysis targeting specific ways to reduce this uncertainty further.

And although it is usually not done, it is not true that ISA variability can generally not be resolved from experiments.

The reviewer is correct and we will clarify this statement.

p. 13116, l. 13-14: “Since the ISA variability cannot be resolved from experiments, a droplet freezing simulation must be employed to model ice nucleation for interpretation purposes.”

will be changed to

“Since the ISA variability is not resolved in previous experiments, a droplet freezing simulation must be employed to model ice nucleation for interpretation purposes.”

p. 13118, line 8-12: The wording here seems to suggest that the ABIFM is particular in that it gives parameterizations for $J_{\text{het}}(T)$, which is not the case (see e.g., some of the literature you cite yourself). Please tune this down.

Our simulations are capable of being run using only measurable observables without the need of any fitting parameters. Experimental data parameterized following the ABIFM is used only as a convenient tool (see p. 13119, l. 23 – p. 13120, l. 7). We will clarify this.

p. 13118, l. 10-12: “Knopf and Alpert (2013) compiled and parameterized experimental J_{het} data yielding a continuous function over T called the ABIFM and expressed as,”

will be changed to

“Knopf and Alpert (2013) compiled experimental data which was parameterized as a continuous function over T following the ABIFM expressed as,”

p. 13120, line 12: To avoid confusion, start this sentence with “Two of these test cases, Iso1 and Iso2, have uniform ISA ... “

We will make this correction.

p. 1320, l. 12-13: “For Iso1 and Iso2 having uniform ISA, f_{ufz} (on a logarithmic scale) is linear with t .”

will be changed to

“Two of these test cases, Iso1 and Iso2, have uniform ISA both resulting in f_{ufz} (on a logarithmic scale) linear with t .”

p. 13121, line 3-15: Yes, in your case, the deviation from a log-linear relationship originates in the assumption of a log-normally distributed ISA, where some droplets will have large ISA. The same behavior (i.e., a divergence from a log-linear relationship) was observed already in Niedermeier et al, (2011), only there a variation in the contact angles ascribed to the different particles caused the effect. It will certainly be difficult to determine how much of the observed shape of a curve is due to the existence of a distribution of contact angles or of ISA, but to be complete it has to be discussed here, that not only an ISA distribution causes the observed behavior.

We respectfully disagree with the reviewer. It is not difficult to determine how much of the observed shape of $f_{\text{ufz}}(t)$ is due either to a contact angle distribution or to variable ISA. In fact, this is exactly what is accomplished in our paper. We test if a known (or well defined) ISA variability, explains the deviation of f_{ufz} from a log-linear relationship. Wright and Petters

(2013) and Broadley et al. (2012) give constraints for their ISA distribution, which also fully accounts for f_{ufz} deviating from an observed log-linear relationship. This implies droplet to droplet variability parameterized by a contact angle distribution is likely small.

p. 13123, l. 10: We will add the following sentences, “Droplet to droplet variability in ice nucleation efficiency is typically parameterized with a variable efficiency of sites to nucleate ice or different contact angles (e.g. Niedermeier et al. 2011; Broadley et al., 2012). Droplet to droplet variability parameterized in these ways and employing identical ISA can result in a deviation of f_{ufz} from a log-linear relationship, similar to what is seen in Fig. 1. However, using the known ISA variability (Wright and Petters, 2013), we reveal that the observed deviation from a log-linear relationship can be accounted for entirely by the ISA distribution. This implies that the droplet to droplet variability in ice nucleation efficiency parameterized by a contact angle or active site distribution is potentially unimportant.”

p. 13124, l. 7: We will add the following sentence, “Similar to Wright and Peters (2013), the deviation of f_{ufz} from a log-linear relationship can be completely accounted for by the experimentally constrained ISA distribution”.

What do you want to say with the last sentence in this paragraph? This is not clear to me, please consider rewording or removing it.

p. 13121, l. 14-15: The sentence “Consequentially, any interpretation on the physical process of immersion freezing based on the slope of f_{ufz} is unfounded.” will be removed.

p. 13121, line 26: Here and in other cases, when you use nucleation rate coefficient in your calculations, I would have preferred to get the information about the origin of these numbers much clearer. The information can sometimes be found in the text, but often only much later than I would have preferred it. Please edit the text accordingly.

We have elaborated on our selection procedure for model parameters and revised our text. When unavailable, constant values of J_{het} are fitted to isothermal frozen fraction data. When an experiment using a cooling rate is simulated, the ABIFM is evoked to calculate a continuous function, $J_{het}(T)$. When $J_{het}(T)$ is not available, it must be fitted to experimental frozen fraction data. This is summarized in our revised text of p. 13117, l. 25-26 given above.

p. 13122, line 19: You base the calculation of the surface area on a gas adsorption method (BET), which, however, is only one way to determine the surface area. You might want to stress the fact that surface area is not an unambiguous parameter.

p. 13122, l. 19-20: “Brunauer, Emmett and Teller gas adsorption method (Brunauer et al., 1938).”

will be changed to

“Brunauer, Emmett and Teller (BET) gas adsorption method (Brunauer et al., 1938). It is important to note that surface area measurements are not unambiguous due to the fact that heterogeneous ice nucleation may involve layers of water molecules interacting with surface molecules (Cox et al., 2013). The BET technique is one method to determine surface area and can be used to represent molecularly available surface area.”

p. 13124, line 5-7: Same as said above wrt. p. 13121, line 26: I will not list all of the occurrences, but here again it is not clear to me where the nucleation rate coefficient came from.

Please see previous comments.

p.13124, line 16-18: You mix two things, here: The large uncertainty from the small number of droplets which is examined which you get from your calculation is not related to the different ISA per droplet. The latter depends on how uniformly the experimenter manages to produce the droplets.

The reviewer is correct and we clarify this point. Since sufficient information is not given to derive ISA variability, we infer it from our simulations. When too few droplets are used in a single experiment, measured f_{ufz} vs t is highly uncertain and the corresponding derivation of σ_g from a model simulation will also be highly uncertain. If σ_g was derived simulating a second experimental run, it would likely be very different, resulting in a more linear or curved trajectory over t . We argue that using more droplets or repeating more experiments would reveal the ISA distribution and better constrain σ_g .

p. 13124, l. 14-19: “As previously discussed, repetition of experiments would result in f_{ufz} exhibiting possibly more linear or non-linear behavior with t within the calculated percentile bounds, which results in smaller or larger values of σ_g , respectively. Herbert et al. (2014) assumed that each droplet possessed the same ISA, however, this assumption is not supported due to the large statistical uncertainty from the small number of applied droplets.”

will be changed to

“As previously discussed, a repeat experiment may result in f_{ufz} exhibiting more linear or non-linear behavior with t within the calculated percentile bounds, i.e. within the stochastic uncertainty. Figure 1A shows that a more linear or non-linear relationship of f_{ufz} with t implies a smaller or larger value of σ_g . Herbert et al. (2014) assumed that each droplet possessed the same ISA, however, this assumption is not supported due to the large stochastic uncertainty from the small number of applied droplets.”

p. 13125, line 6-7: Values for nucleation rate coefficients for K-feldspar (microcline) were given in the supplement of Augustin-Bauditz et al. (2014).

Yes, this is correct. However, the J_{het} data of Augustin-Bauditz et al. (2014) span a different temperature range than Herbert et al. (2014) making comparison difficult.

p. 13125, l. 6-7: “Values of J_{het} for feldspar independent from Herbert et al. (2014) to our knowledge do not exist making comparison difficult.”

will be changed to

“Values of J_{het} for feldspar independent from Herbert et al. (2014) in the same temperature range to our knowledge do not exist making comparison difficult.”

p. 13125, line 19-20: As mentioned before, the use of a contact angle distribution does a similar job, so this sentence cannot be kept as it is now.

We thank the reviewer for pointing out this error. Our model demonstrates that ISA variability can account for observations, however, uncertainties are large and so this cannot be said for certain. We will lighten our language.

p. 13125, l. 19-20: “Nevertheless, we can still conclude that immersion freezing is a time dependent stochastic process reconciled only when variable ISA is considered.”

will be changed to

“Nevertheless, a time dependent and stochastic immersion freezing process can reconcile observations when variable ISA is considered.”

p. 13125, line 28 to p. 13126, line 2: The approach used here also uses an empirical parameterization of some kind by assuming large variations in ISA, and as such is not better or worse than other comparable models. Please reformulate.

We disagree with this statement. Large variations in ISA reported in Wright and Petters (2013) and Broadley et al. (2012) are not assumed values, but instead are supported by their experimental results. However, it is true that in the study of Herbert et al. (2014) the distribution is not well constrained. Furthermore, model simulations IsoWR, IsoBR and IsoWR are not empirical at all and instead the parameters are physical and measurable, including N_{tot} , J_{het} , σ_g and A_g . Applying those parameters, heterogeneous freezing and kinetics can be represented without invoking any empirical formulation.

Sec. 3.2: The method comparing apparent and actual values is nice, however, the conclusions again suffer from the fact that the principal assumption was that of only one contact angle (i.e., one nucleation rate coefficient) being present. This all has to be reformulated / thinned out respectively.

We thank the reviewer for commenting on comparing ‘actual’ and ‘apparent’ values. However, we disagree about reformulating our conclusions. Please see our opening comment. We do not make any assumption of using a single contact angle. In fact, previous literature suggests the possibility that contact angles may change as a function of temperature (Zobrist

et al., 2007; Alpert et al. 2011a; Alpert et al. 2011b; Knopf and Forrester, 2011). We point out that the slope of $J_{\text{het}}^{\text{apparent}}$ and $J_{\text{het}}^{\text{actual}}$ are not the same. The reason for the difference in slope is due to ISA variability. As we find that the slope of measured and model derived $J_{\text{het}}^{\text{apparent}}$ are exactly the same, which means that ISA variability can fully explain the observed freezing kinetics.

p. 13129, line 15-16: This goes along the line of my former remark: This new parameterization for a nucleation rate coefficient for feldspar would only be valid if the same ISA variability was used with it. Hence it might be advantageous to not deliver new parameterizations but to describe the effect of ISA variability and its magnitude, all on its own, instead.

It is important to note that the ABIFM parameterization calculates J_{het} , but J_{het} does not depend on INP surface area. Therefore, any ISA distribution can be used with this single parameterization. However, this parameterization is limited due to the range of laboratory conditions. In the temperature range 260-265 K and for $\alpha_w=1$, the ABIFM parameterization for feldspar is valid for $0.078 < \Delta\alpha_w < 0.120$.

p. 13129, l. 16: “a new $J_{\text{het}}(\Delta\alpha_w)$ parameterization for feldspar.”

will be changed to

“a new $J_{\text{het}}(\Delta\alpha_w)$ parameterization for feldspar valid for $0.078 < \Delta\alpha_w < 0.120$.”

p. 13131, lines 17-23: Again, and I know I repeat myself: this could only be said so clearly if your assumption of a single contact angle per substance were correct. This whole passage has to be toned down a lot.

Please see also previous comments. We also feel these sentences can be toned down.

p. 13131, l. 20-23: “This result impacts all immersion freezing experiments conducted as a function of ISA that assume identical ISA, thereby implicitly imposing a surface area dependence on $J_{\text{het}}^{\text{apparent}}$ or $n_s(T)$. However, accounting for the experimental uncertainty and variability in ISA reconciles experimental data.”

will be changed to

“This result could potentially impact immersion freezing experiments conducted as a function of ISA that assume identical ISA, thereby implicitly imposing a surface area dependence on $J_{\text{het}}^{\text{apparent}}$ or $n_s(T)$. Accounting for the experimental uncertainty and variability in ISA may reconcile experimental data.”

p. 13132, line 5: “is not observed unlike” - double negative, always makes comprehension difficult.

p. 13132, l. 5-6 “is not observed unlike previously discussed experiments but instead, the number of ice crystals are optically detected.”

will be changed to

“is not observed and instead, the number of ice crystals is optically detected.”

p. 13133, line 7-8: The comparison done between J_{het} derived in Wex et al. (2014) and in Murray et al. (2011) and Pinti et al. (2012) is not viable. The differences in the derivation of the surface area (BET versus assumption of spherical particles) would need to be accounted for. But even more important, the comparison as done here compares two different kaolinities, data for Fluka-kaolinite from Wex et al. (2014) and for CMS-kaolinite, which is known to be less ice active, as used in the other two publications. Wex et al. (2014) did also include CMS-kaolinite, but the data from that publication you used here is that from Fluka-kaolinite. Accordingly, the mentioning of Fig. 7b on p. 13134, line 17 as an example needs to be removed.

We thank the reviewer for pointing out our error and acknowledge that comparing ice nucleating abilities of kaolinite purchased from two sources, the Clay Mineral Society (CMS) and Fluka, should not be done here. We acknowledge that the ice nucleation ability of the two kaolinite minerals may be different (Wex et al., 2014) and therefore, we will revise the model simulation and corresponding text. The comparison with Murray et al. (2011) and Pinti et al. (2012) will be removed and thus, a discussion of BET versus spherical surface area assumption is not necessary.

The revised model simulations will use the new particle size distribution from Fig. S1, and as a consequence, the parameters to calculate $J_{\text{het}}(T)$ from ABIFM will be fit. New Fig. 7 and Table 1 are given below.

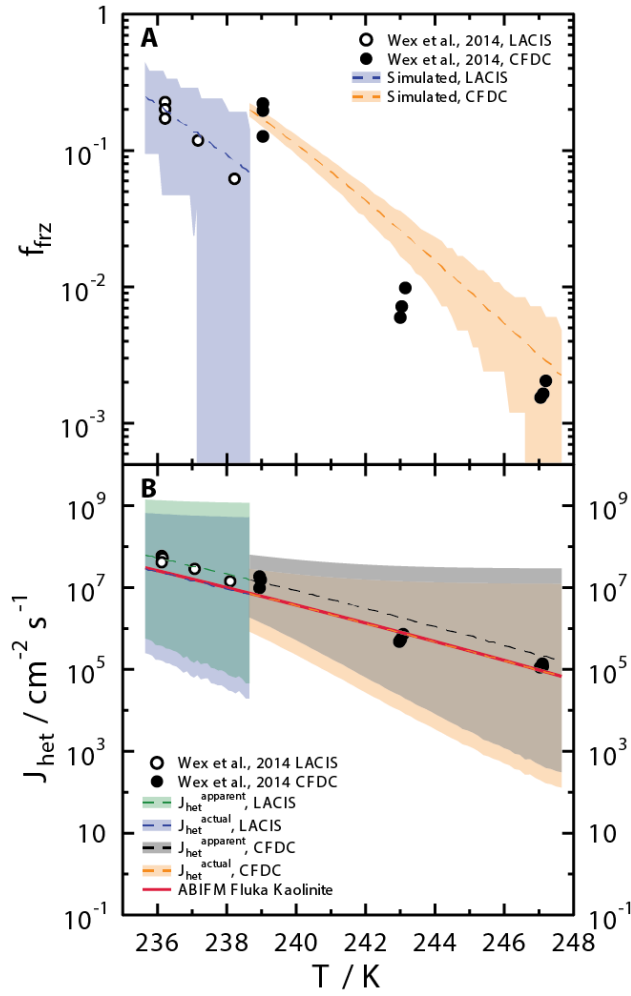


Figure 7

Name	N_{tot}	σ_g	A_g / cm^2	T / K	J_{het} $\text{cm}^{-2} \text{s}^{-1}$	IN Type	Figure	Color
Iso1	1000	1	1.0×10^{-5}	-	1.0×10^3	-	1A	dark green
Iso2	30	1	1.0×10^{-5}	-	1.0×10^3	-	1A	light green
Iso3	1000	10	1.0×10^{-5}	-	1.0×10^3	-	1A	dark blue
Iso4	30	10	1.0×10^{-5}	-	1.0×10^3	-	1A	light blue
IsoWR	1000	9.5×10^{-3}	6.4×10^{-3}	251.15	6.0×10^{-4}	ATD ^a	1B	orange
IsoBR	63	8.3	2.6×10^{-7}	243.3	1.3×10^3	illite	2A	orange
IsoHE1	40	2.2	1.2×10^0	255.15	4.1×10^{-3}	kaolinite	2B	orange
IsoHE2	40	8.5	2.0×10^{-2}	262.15	2.0×10^{-2}	feldspar	2C	orange
IsoDI1	45	4.2	5.1×10^{-1}	255.15	1.3×10^{-2}	illite	3	green
IsoDI2	45	9.1	5.1×10^{-2}	252.15	9.5×10^{-1}	illite	3	orange
IsoDI3	45	1.5	5.1×10^{-1}	252.15	8.3×10^{-1}	illite	3	blue
IsoCFDC	833	MCD ^b	MCD	238.65– 247.65 ^c	ABIFM ^d	Fluka kaolinite	7	blue, green
IsoLACIS	21	MCD	MCD	235.65– 238.65 ^c	ABIFM	Fluka kaolinite	7	orange, black

^aArizona Test Dust. ^bMultiple charge distribution used to define surface area distribution. See text and Fig. S1 for further details. ^cIsothermal simulations were performed at 0.15 K increments within the stated temperature range. ^dValues of J_{het} are calculated from the water activity, a_w , based immersion freezing model (ABIFM) (Knopf and Alpers, 2013).

Table 1

p. 13132, l. 15-16: We will remove the sentence “ J_{het} is taken from Knopf and Alpert (2013) for kaolinite IN.”

p. 13133, l. 5-13: “We find excellent agreement between $J_{\text{het}}^{\text{apparent}}$ and data by Wex et al. (2014). Also, calculated $J_{\text{het}}^{\text{actual}}$ are in agreement with J_{het} using the ABIFM for kaolinite in CFDC and LACIS experiments. J_{het} data by Wex et al. (2014) and data by Murray et al. (2011) and Pinti et al. (2012) disagree by 2 to 3.5 orders of magnitude. This is similar to the difference between $J_{\text{het}}^{\text{apparent}}$ and $J_{\text{het}}^{\text{actual}}$ likely due to A_g being 1.5 orders of magnitude larger than $A_{300\text{nm}}$ and applying a surface area distribution. Furthermore, assuming that the electrical mobility diameter corresponds to the physical particle diameter and being spherical in geometry significantly overestimates ice nucleation kinetics, as demonstrated for IsoCFDC and IsoLACIS.”

will be changed to

“We find agreement between $J_{\text{het}}^{\text{apparent}}$, $J_{\text{het}}^{\text{actual}}$ and data by Wex et al. (2014) when accounting for multiple particle charges predicted by Wiedensohler and Fissan (1988). Furthermore, assuming that the electrical mobility diameter corresponds to the physical particle diameter and the particle being spherical in geometry does not significantly overestimate ice nucleation kinetics within the uncertainty bounds which span 2-5 orders of magnitude. The model input J_{het} represents a new parameterization for Fluka kaolinite where $m=31.32$ and $c=-2.07$ following the ABIFM applicable for $0.220 < \Delta\alpha_w < 0.305$.”

p. 13133, line 21: Wex et al. (2014) also similarly used the time-independent approach (surface site density) and reports that this works almost as well as the time-dependent approach. Therefore this study does not clearly support the necessity of a time-dependent and stochastic treatment of the immersion freezing process.

Here, we wish to conclude that our model simulations based on ABIFM is applicable to ice nucleation studies using a CFDC and LACIS. We will reformulate these sentences.

p. 13133, l. 21-24: “These findings demonstrate that the model simulations are applicable for ice nucleation studies using a CFDC and LACIS, and that data from Wex et al. (2014) support a time-dependent and stochastic immersion freezing process.”

will be changed to

“These findings demonstrate that our new model simulations and the ABIFM are applicable for ice nucleation studies using a CFDC as previously shown by Knopf and Alpert (2013) and additionally LACIS.”

p. 13135, line 3-4: The model simulations indeed fit the AIDA data, but a straight line representing J_{het} (the red line you drew) or likely even a time-independent approach would

reproduce the data similarly well, so I do not see how this further supports the necessity of the quantification of the ISA variability.

Please see also previous comments. Agreement between model and simulation is not insightful without an uncertainty estimate. Within a range of about ~4 orders of magnitude (Hiranuma et al., 2015), both a time-independent and time-dependent approach may reproduce the data. Our model simulations reveal that if ISA variability were considered, then uncertainties could be significantly decreased.

p. 13135, line 10-18: The singular approach is meant to represent averages, so it is correct to say that it cannot capture an increase in scatter in the data due to stochastic effects. But although indeed Fig. 8b shows an increase in scatter as T increases, it does generally not seem to be the case that the scattered data-points in Fig. 8A are captured within the limits of the model. Certainly here, and for other experiments, too, there are measurement uncertainties which are not captured neither by the singular approach, but also nor by your model. This might be worth pointing out, here.

We agree with this point.

p. 13135, l. 12-14: “These observations can only be explained by a stochastic and time-dependent immersion freezing process.”

will be changed to

“These observations can be explained by a stochastic and time-dependent immersion freezing process. We note that other measurement uncertainties may exist which may not be captured either by a deterministic approach or by our model. However, we conclude that stochastic uncertainty is important to consider for future ice nucleation studies.”

p. 13136, line 2, and also p. 13139, line 20 and p. 13140, line 6 and possibly other occurrences: When referring to “stochastic uncertainty” here, what exactly do you mean, besides the influence of time, which is mentioned additionally in some occasions?

We define a stochastic uncertainty as the scatter in the data due to the occurrence of random freezing events. Repeating experiments for a set number of droplets should reveal this data scatter. Alternatively, it can be derived from first principles of statistics in our model simulations. Stochastic uncertainty is visualized by the 5 and 95 percentiles of f_{ufz} , f_{fz} and the upper and lower fiducial limits of J_{het} . The influence of time also affects the stochastic uncertainty as seen in Fig. 1. As time increases, the width of the percentile bounds also increase.

In many instances, we use the terms “stochastic uncertainty” and “statistical uncertainty” interchangeably. Instead, we will replace the term “statistical uncertainty” with “stochastic

uncertainty” to remain consistent occurring on, for example, p. 13117, l. 16, p. 13119, l. 22, p. 13124, l. 19 and p. 13135, l. 26-27.

p. 13117, l. 17: A new sentence will be added which reads, “We define stochastic uncertainty as the scatter in the data due to the occurrence of random freezing events upon repeat experiments as a result of a set number of observed freezing events.”

p. 13121, l. 10-11: “This implies that in isothermal freezing experiments,...”

will be changed to

“In isothermal freezing experiments,...”

p. 13135, l. 27: We will add a sentence which reads “Once again, stochastic uncertainty refers to large or small expected data scatter from observing small or large numbers of freezing events, respectively.”

p. 13139, line 23: In some experiments / some methods, more than 1000 droplets are examined.

p. 13139, l. 23: “...between 10 and 1000 depending on the experiment.”

will be changed to

“...between 10 and more than 1000 depending on the experiment.”

p. 13141, line 1: Using a concentration of illite particles of $10^5/L$ is extraordinarily large and likely not representative for the atmosphere.

The reviewer is correct that this concentration of illite particles is not realistic. However, simulation MPC1 and MPC2 demonstrates a temperature shift in frozen fraction and the concentration of ice crystals. This shift is independent on particle numbers. We also clearly state that we do not simulate any physically realistic cloud. For this reason we will remove the mentioning of Arctic mixed phase clouds to avoid further confusion.

p. 13141, l. 13-16: The following sentence will be removed, “Typical ice crystal concentrations observed in Arctic mixed-phase clouds can range from $0.01-10 L^{-1}$ (air) at temperatures warmer than the homogeneous freezing limit $235 < T < 273.15 K$ (McFarquhar et al., 2007; Prenni et al., 2009; Lance et al., 2011).”

p. 13141, line 12 ff: MPC2 produces ice nucleation at higher temperatures, but not necessarily because of the total variability in ISA that you introduced, but because the droplet with the “best” INP will induce the freezing. In your case these are those droplets which have the largest ISA ascribed, and as the spread in the ISA distribution is large (σ of 5!), you find this shift by 5K. Also, when you then start to discuss Arctic conditions, it surely becomes unrealistic to assume a single substance. For atmospheric conditions, the heterogeneity of types of INP can be assumed

to play a large role (i.e., the occurrence of different nucleation rate coefficients has to be expected). But in the way you treat it, you cover this heterogeneity with an unrealistic large spread in ISA.

Our main purpose here is to give an example how an ISA distribution can influence ice crystal production. Distribution widths between $\sigma_g=1$ and 5 should fall between the red and blue curves, respectively. We agree with the reviewer that there exists a wide variety of INP types with different ice nucleation efficiencies and this is not captured in Fig. 10.

p. 13140, l. 19: We will add the following sentence, “Aerosol populations are highly diverse, but for demonstrative purposes we only use a single INP type.”

p. 13141, l. 16-17: “The model presented here which accounts for ISA variability achieves similar ice crystal concentrations between 251–258 K as demonstrated in Fig. 10b.”

will be changed to

“Figure 10b shows ice crystal concentrations of 0.01 and 10 L⁻¹ (air) at 251 and 258 K, respectively, produced by the simulations. Note that when employing a distribution width between $\sigma_g=1$ and 5, ice crystal numbers and f_{frz} values should fall between the red and blue curves, respectively.”

p. 13145, line 2-3: This is already done in the work by DeMott et al. (2010) and DeMott et al. (2015), which you might mention here.

We meant to suggest that the entire aerosol size distribution should be considered as a source of INPs together with a stochastic, time-dependent ice nucleation process characterized by J_{het} , which is easily parameterized following the ABIFM. We thank the reviewer for pointing this out and will reword this sentence. It should be noted that data from DeMott et al. (2010) and DeMott et al. (2015), only include particles between 0.5-1.6 μm in diameter. It remains unclear if this limitation is of potential importance for reducing the uncertainty in predicting INP concentrations.

p. 13145, l. 2-3: “This implies that field measurements should determine and consider the entire aerosol size distribution as a source of IN.”

will be changed to

“We suggest that field measurements should determine and consider the entire aerosol size distribution as a source of IN for implementation of a stochastic, time-dependent ice nucleation process characterized by J_{het} , which is easily parameterized following the ABIFM.”

Rigg et al. (2013) is missing in the literature-list.

This will be corrected.

Table 1: Again a repetition, but it really needs to be made clearer in the text, where these values you use here come from, particularly σ and nucleation rate coefficients.

Please see previous comments on our parameter selection procedure.

References:

- Alpert, P. A., Aller, J. Y., and Knopf, D. A.: Ice nucleation from aqueous NaCl droplets with and without marine diatoms, *Atmos. Chem. Phys.*, **11**, 5539–5555, doi:10.5194/acp-11-5539-2011, 2011a.
- Alpert, P. A., Knopf, D. A., and Aller, J. Y.: Initiation of the ice phase by marine biogenic surfaces in supersaturated gas and supercooled aqueous phases, *Phys. Chem. Chem. Phys.*, **13**, 19882–19894, doi:10.1039/c1cp21844a, 2011b.
- Archuleta, C. M., DeMott, P. J., and Kreidenweis, S. M.: Ice nucleation by surrogates for atmospheric mineral dust and mineral dust/sulfate particles at cirrus temperatures, *Atmos. Chem. Phys.*, **5**, 2617–2634, doi:10.5194/acp-5-2617-2005, 2005.
- Augustin-Bauditz, S., Wex, H., Kanter, S., Ebert, M., Niedermeier, D., Stolz, F., Prager, A. and Stratmann, F.: The immersion mode ice nucleation behavior of mineral dusts: A comparison of different pure and surface modified dusts, *Geophys. Res. Lett.*, **41**, doi:10.1002/2014GL061317, 2014.
- Broadley, S. L., Murray, B. J., Herbert, R. J., Atkinson, J. D., Dobbie, S., Malkin, T. L., Condliffe, E., and Neve, L.: Immersion mode heterogeneous ice nucleation by an illite rich powder representative of atmospheric mineral dust, *Atmos. Chem. Phys.*, **12**, 287–307, doi:10.5194/acp-12-287-2012, 2012.
- Brunauer, S., Emmett, P. H., and Teller, E.: Adsorption of gases in multimolecular layers, *J. Am. Chem. Soc.*, **60**, 309–319, doi:10.1021/ja01269a023, 1938.
- Connolly, P. J., Möhler, O., Field, P. R., Saathoff, H., Burgess, R., Choularton, T., and Gallagher, M.: Studies of heterogeneous freezing by three different desert dust samples, *Atmos. Chem. Phys.*, **9**, 2805–2824, doi:10.5194/acp-9-2805-2009, 2009.
- Cox, S. J., Raza, Z., Kathmann, S. M., Slatara, B., and Michaelides, A.: The microscopic features of heterogeneous ice nucleation may affect the macroscopic morphology of atmospheric ice crystals, *Faraday Discuss.*, **167**, 389–403, doi:10.1039/c3fd00059a, 2013.
- DeMott, P. J., Prenni, A. J., Liu, X., Kreidenweis, S. M., Petters, M. D., Twohy, C. H., Richardson, M. S., Eidhammer, T., and Rogers, D. C.: Predicting global atmospheric ice nuclei distributions and their impacts on climate, *P. Natl. Acad. Sci. USA*, **107**, 11217–11222, doi:10.1073/pnas.0910818107, 2010.
- DeMott, P. J., Prenni, A. J., McMeeking, G. R., Sullivan, R. C., Petters, M. D., Tobo, Y., Niemand, M., Möhler, O., Snider, J. R., Wang, Z. and Kreidenweis, S. M.: Integrating laboratory and field data to quantify the immersion freezing ice nucleation activity of mineral dust particles, *Atmos. Chem. Phys.*, **15**, 393–409, doi:10.5194/acp-15-393-2015, 2015.
- Diehl, K., Debertshäuser, M., Eppers, O., Schmithüsen, H., Mitra, S. K., and Borrmann, S.: Particle surface area dependence of mineral dust in immersion freezing mode: investigations with freely suspended drops in an acoustic levitator and a vertical wind tunnel, *Atmos. Chem. Phys.*, **14**, 12343–12355, doi:10.5194/acp-14-12343-2014, 2014.
- Hartmann, S., Niedermeier, D., Voigtländer, J., Clauss, T., Shaw, R. A., Wex, H., Kiselev, A., and Stratmann, F.: Homogeneous and heterogeneous ice nucleation at LACIS: operating principle and theoretical studies, *Atmos. Chem. Phys.*, **11**, 1753–1767, doi:10.5194/acp-11-1753-2011, 2011.

- Herbert, R. J., Murray, B. J., Whale, T. F., Dobbie, S. J., and Atkinson, J. D.: Representing time-dependent freezing behaviour in immersion mode ice nucleation, *Atmos. Chem. Phys.*, **14**, 8501–8520, doi:10.5194/acp-14-8501-2014, 2014.
- Hiranuma, N., Augustin-Bauditz, S., Bingemer, H., Budke, C., Curtius, J., Danielczok, A., Diehl, K., Dreischmeier, K., Ebert, M., Frank, F., Hoffmann, N., Kandler, K., Kiselev, A., Koop, T., Leisner, T., Möhler, O., Nillius, B., Peckhaus, A., Rose, D., Weinbruch, S., Wex, H., Boose, Y., DeMott, P. J., Hader, J. D., Hill, T. C. J., Kanji, Z. A., Kulkarni, G., Levin, E. J. T., McCluskey, C. S., Murakami, M., Murray, B. J., Niedermeier, D., Petters, M. D., O'Sullivan, D., Saito, A., Schill, G. P., Tajiri, T., Tolbert, M. A., Welti, A., Whale, T. F., Wright, T. P., and Yamashita, K.: A comprehensive laboratory study on the immersion freezing behavior of illite NX particles: a comparison of 17 ice nucleation measurement techniques, *Atmos. Chem. Phys.*, **15**, 2489–2518, doi:10.5194/acp-15-2489-2015, 2015.
- Hoose, C. and Möhler, O.: Heterogeneous ice nucleation on atmospheric aerosols: a review of results from laboratory experiments, *Atmos. Chem. Phys.*, **12**, 9817–9854, doi:10.5194/acp-12-9817-2012, 2012.
- Iannone, R., Chernoff, D. I., Pringle, A., Martin, S. T., and Bertram, A. K.: The ice nucleation ability of one of the most abundant types of fungal spores found in the atmosphere, *Atmos. Chem. Phys.*, **11**, 1191–1201, doi:10.5194/acp-11-1191-2011, <http://www.atmos-chem-phys.net/11/1191/2011/>, 2011.
- Knopf, D. A. and Forrester, S.: Freezing of water and aqueous NaCl droplets coated by organic monolayers as a function of surfactant properties and water activity, *J. Phys. Chem. A*, **115**, 5579–5591, doi:10.1021/jp2014644, 2011.
- Knopf, D. A. and Alpert, P. A.: A water activity based model of heterogeneous ice nucleation kinetics for freezing of water and aqueous solution droplets, *Faraday Discuss.*, **165**, 513–534, doi:10.1039/c3fd00035d, 2013.
- Koop, T. and Zobrist, B.: Parameterizations for ice nucleation in biological and atmospheric systems, *Phys. Chem. Chem. Phys.*, **11**, 10839–10850, doi:10.1039/B914289D, 2009.
- Kulkarni, G., Fan, J., Comstock, J. M., Liu, X., and Ovchinnikov, M.: Laboratory measurements and model sensitivity studies of dust deposition ice nucleation, *Atmos. Chem. Phys.*, **12**, 7295–7308, doi:10.5194/acp-12-7295-2012, 2012.
- Lüönd, F., Stetzer, O., Welti, A., and Lohmann, U.: Experimental study on the ice nucleation ability of size-selected kaolinite particles in the immersion mode, *J. Geophys. Res.*, **115**, D14201, doi:10.1029/2009JD012959, 2010.
- Marcollì, C., Gedamke, S., Peter, T., and Zobrist, B.: Efficiency of immersion mode ice nucleation on surrogates of mineral dust, *Atmos. Chem. Phys.*, **7**, 5081–5091, doi:10.5194/acp-7-5081-2007, 2007.
- Murray, B. J., Broadley, S. L., Wilson, T. W., Atkinson, J. D., and Wills, R. H.: Heterogeneous freezing of water droplets containing kaolinite particles, *Atmos. Chem. Phys.*, **11**, 4191–4207, doi:10.5194/acp-11-4191-2011, 2011.
- Niedermeier, D., Hartmann, S., Shaw, R. A., Covert, D., Mentel, T. F., Schneider, J., Poulain, L., Reitz, P., Spindler, C., Clauss, T., Kiselev, A., Hallbauer, E., Wex, H., Mildenerger, K., and Stratmann, F.: Heterogeneous freezing of droplets with immersed mineral dust particles – measurements and parameterization, *Atmos. Chem. Phys.*, **10**, 3601–3614, doi:10.5194/acp-10-3601-2010, 2010.
- Niedermeier, D., Shaw, R. A., Hartmann, S., Wex, H., Clauss, T., Voigtländer, J., and Stratmann, F.: Heterogeneous ice nucleation: exploring the transition from stochastic to singular freezing behavior, *Atmos. Chem. Phys.*, **11**, 8767–8775, doi:10.5194/acp-11-8767-2011, 2011.

- Niemand, M., Möhler, O., Vogel, B., Vogel, H., Hoose, C., Connolly, P., Klein, H., Bingemer, H., DeMott, P., Skrotzki, J., and Leisner, T.: A particle-surface-area-based parameterization of immersion freezing on desert dust particles, *J. Atmos. Sci.*, **69**, 3077–3092, doi:10.1175/JAS-D-11-0249.1, 2012.
- Pinti, V., Marcolli, C., Zobrist, B., Hoyle, C. R., and Peter, T.: Ice nucleation efficiency of clay minerals in the immersion mode, *Atmos. Chem. Phys.*, **12**, 5859–5878, doi:10.5194/acp-12-5859-2012, 2012.
- Rigg, Y. J., Alpert, P. A., Knopf, D. A.: Immersion freezing of water and aqueous ammonium sulfate droplets initiated by humic-like substances as a function of water activity, *Atmos. Chem. Phys.*, **13**, 6603–6622, doi:10.5194/acp-13-6603-2013, 2013.
- Rogers, D. C., DeMott, P. J., Kreidenweis, S. M., and Chen, Y. L.: A continuous-flow diffusion chamber for airborne measurements of ice nuclei, *J. Atmos. Ocean. Tech.*, **18**, 725–741, 2001.
- Vali, G., DeMott, P., Möhler, O. and Whale, T. F.: Ice nucleation terminology. *Atmos. Chem. Phys. Discuss.*, **14**, 22155–22162, doi: 10.5194/acpd-14-22155-2014, 2014.
- Vali, G. and Snider, J. R.: Time-dependent freezing rate parcel model, *Atmos. Chem. Phys.*, **15**, 2071–2079, doi:10.5194/acp-15-2071-2015, 2015.
- Wex, H., DeMott, P. J., Tobo, Y., Hartmann, S., Rösch, M., Clauss, T., Tomsche, L., Niedermeier, D., and Stratmann, F.: Kaolinite particles as ice nuclei: learning from the use of different kaolinite samples and different coatings, *Atmos. Chem. Phys.*, **14**, 5529–5546, doi:10.5194/acp-14-5529-2014, 2014.
- Wiedensohler, A. and Fissan, H. J.: Aerosol charging in high purity gasses, *J. Aerosol Sci.*, **19**, 867–870, doi:10.1016/0021-8502(88)90054-7, 1988.
- Wright, T. P. and Petters, M. D.: The role of time in heterogeneous freezing nucleation, *J. Geophys. Res.*, **118**, 3731–3743, doi:10.1002/jgrd.50365, 2013.
- Zobrist, B., Koop, T., Luo, B. P., Marcolli, C., and Peter, T.: Heterogeneous ice nucleation rate coefficient of water droplets coated by a nonadecanol monolayer, *J. Phys. Chem. A*, **111**, 2149–2155, doi:10.1021/jp066080w, 2007.
- Zuberi, B., Bertram, A., Cassa, C. A., Molina, L. T., and Molina, M. J.: Heterogeneous nucleation of ice in (NH₄)₂SO₄-H₂O particles with mineral dust immersions, *Geophys. Res. Lett.*, **29**, 10, 1504, doi:10.1029/2001GL014289, 2002.