

The manuscript is rather long for its content, very “wordy”, and many sections are difficult to understand. Also, the writing in places is too sloppy, meaning superficial or stating generalizations without references or convincing proof. I strongly suggest to carefully revise the text and shorten some sections but others may need more information to be better understood as indicated below. For example, section 3.6 on time dependence is very confusing and the mathematical procedure is not clear.

We thank the referee for their extensive and thoughtful comments. They have helped us significantly improve the content of our manuscript as well as the clarity of the message we wish to convey. We have replied to each comment below and revised the manuscript to address the many questions and concerns raised and improved the clarity of the information being communicated.

This manuscript presents an attempt to describe immersion freezing data using a mathematical construct, i.e. by fitting experimentally frozen fraction curves. As stated in earlier works upfront, such as Niedermeier et al. (2010), an active sites concept is not based on a physical foundation or theory. Neither, is the effect of external and internal variability of active sites proven to be a physical concept. The Murray group implied this from fits to data. The scientific value of such (previous and this) approaches will be shown in time. I do not mind this mathematical exercise to somehow describe the experimental data in the lack of a physical model, however, these caveats and assumptions should be stated clearly upfront and the tone of the manuscript changed accordingly. In particular the last third part of the manuscript has to be reworded since it reads as if all the results, effects, distributions refer to something “real” or “physical”, which it does not in absence of a physical model. More careful language would be more appropriate.

We recognize that the original version of our manuscript had been too hasty at times in its assertions about the many concepts presented being physical. In the revised manuscript we have strived to reword much of the content to emphasize that the model presented regarding heterogeneous ice nucleation is a mathematical tool to help describe and interpret the data and derive potentially useful parameterizations. It is not a physical model.

As for the mathematical concept: A distribution referred to as a “g-distribution” is introduced. It is not clear of which kind, but always seems to be a normal distribution function. In principle, this concept is very much the same as the α -PDF, the updated soccer ball model (SBM) or other distribution based fits. The emphasis on continuous distribution values is not clear to me as both α -PDF and the SBM are continuous in a mathematical sense.

The α -PDF and SBM models are similar to our g distribution in that they also entail a distribution of active sites. The α -PDF model assigns a single contact angle to every particle in a population via a prescribed distribution while the SBM model partitions a particle into discrete active sites and assigns these sites contact angles based on a prescribed distribution. The g distribution is closer to the SBM model with the difference being that the g framework does not require partitioning a particle into discrete sites but assuming a continuum of activity.

The text has been revised accordingly, on Page/Line 23/6-22:

“There are other formulations that hypothesize an active site based or multi-component stochastic model such as the ones described in Vali & Stransbury (1966), Niedermeier et al. (2011), Wheeler and Bertram (2012), and Wright and Petters (2013). Vali and Stransbury (1966) were the first to recognize that ice nucleating surfaces are diverse and stochastic and thus active sites need to be assigned both a characteristic freezing temperature as well as fluctuations around that temperature. Niedermeier et al. (2011) proposed the soccer ball model, in which a surface is partitioned into discrete active sites with each site conforming to classical nucleating theory. Marcolli et al. (2007) found a Gaussian distribution of contact angles could best describe their heterogeneous ice nucleation data in a completely deterministic framework. Welts et al. (2012) introduced the alpha-PDF model where a probability density function prescribes the distribution of contact angles that a particle population possesses, such that each particle is characterized by a single contact angle. Wright and Petters (2013) hypothesized the existence of a Gaussian probability density function for a specific species, which in essence is similar to the $\bar{g}$ framework described here. The notable difference is that this probability density function was retrieved via optimizing for all freezing curves, and not independently fitting high concentration freezing curves as we have done here.”

As the frozen fractions curves shift to lower temperatures due to a decrease in surface area and below the critical threshold area as stated here, $\bar{g}$ cannot reproduce the data. However, freezing data can be described when choosing contact angles and calculating $\bar{g}$ values as many times as necessary. The authors are correct that a new distribution for below threshold surface areas is not necessary. (If it were, would it imply that the fit is truly unphysical, i.e. not representing particle properties?) But obviously, drawing as many times as necessary from $\bar{g}$ (which contains all possible contact angle values) to represent the freezing curve does not mean anything physically. One could argue that the number of draws represent just another free “fit parameter”. In general, I am not surprised that data can be fitted with this mathematical construct, but the manuscript must include, state, discuss properly its assumptions. The emphasis to have discovered something “real” in view of these assumptions is incorrect. The effects may all be a result of an assumption that is not known to be true or even applicable. More studies and experiments are necessary.

Presentation of details about the sampling model has been improved in the manuscript and we hope it is now clearer (more information on this is discussed below). The text has been revised on Page/Line 17/6-26:

“To predict the freezing curves of the droplets with particle surface areas lower than the estimated critical area for the systems considered here, the aggregate surface area of the entire particle population within each droplet is modeled as one large surface. A contact angle θ_r is randomly selected from the full contact angle range $[0, \pi]$, and the value of active site distribution g^* for the particle i being sampled for at θ_r is assigned the value of $\overline{g(\theta_r)}$:

$$(g_i^*(\theta_{r,n_{draws}})) = \overline{g(\theta_r)} \quad (16)$$

The g distributions within this numerical model are given an asterisk to indicate that they are discrete distributions.

This process is repeated for a parameter n_{draws} , for each droplet in the array that produced the freezing curve being modeled. n_{draws} is the only parameter that is optimized for so the modeled freezing curves can predict the behavior of the experimental freezing curves. The value of n_{draws} typically ranges from 9 to 65 for the systems analyzed here and is therefore a relatively soft

optimization parameter with small dynamic range. The sampled g^* distributions are normalized with respect to the estimated total surface area for the freezing curve being modeled before being used to compute the freezing probability. Using the sampled g distributions the freezing probability of each droplet is calculated using Eq. (9) and the frozen fraction curve is computed from the arithmetic average of the freezing probabilities:

$$F(\text{below critical area}) = \frac{1}{N} \sum_{i=1}^N P_{fi} \quad (17)$$

where N is the number of droplets in the cold plate array.”

We have removed previous assertions of discovering something “real” with the model being able to fit the data. While the number of draws is just another fit parameter, it actually turns out to be a fairly “soft” optimization parameter varying from 9 to 65 for all the systems considered (additional datasets beyond illite are now analyzed). We hope that the new details and analysis provided will add to the clarity of this aspect of the paper.

I remain confused about the details of the method. It would also be beneficial to show g and the numbers of draws for different experimental data sets to establish this method. Many other questions remain and I mention a few here. It is stated that θ is randomly chosen but does this mean that θ is first sampled from a uniform probability density function, and then $g(\theta)$ is calculated? Does this method of draws also work equally well for above the surface area threshold? Is it correct to say that the g -distribution is not a probability density function from which θ is derived and used in the J_{het} equation, but is it a scaling function or a change from a surface to line integral as stated in the manuscript?

We now explain these details below and in the revised manuscript. A contact angle is first randomly drawn from the full contact angle range. After which the value of the g distribution being modeled at that contact angle is assigned the value of $\bar{g}$ at that randomly drawn contact angle. The process is repeated for n_{draws} . After a few repetitions (on the order of 20 for the illite and cellulose distributions, for example) the sampled g distribution will mimic $\bar{g}$. So one can say that the method does also work for modeling curves above the critical area threshold.

The text has been revised on Page/Line 18/5-8:

“It should also be noted that there is an n_{draws} value for each system above for which the sampled distribution mimics $\bar{g}$. For example, when n_{draws} is 25 for the Illite system the retrieved distribution will produce a freezing curve equivalent to using $\bar{g}$.”

The manuscript does not sufficiently discuss previous work on immersion freezing. On the model side, the authors could test if “subsampling” of an γ -PDF or other distributions (deterministic etc., see e.g. Marcolli or Lohmann group) will result also in a better representation when surface area is changing – likely yes, if sufficient draws are allowed. The water activity based immersion freezing model by the Knopf group also can describe immersion freezing for illite. As far as I recall they do not need to invoke external or internal mixtures to consolidate freezing data obtained from differently sized particles.

We do recognize that a different version of the sampling model can be built around an already existing scheme like n_s . We point to some of the similarities between $\bar{g}$ and n_s in that we think they both represent active site distribution for particle surfaces above the defined critical area. We also do recognize (and have added emphasis on this in the revised manuscript) that is not the first

approach to successfully fit frozen fraction curves for illite for other systems. It is just, as the reviewer points out, different and offers what we think are some valuable insights on how heterogeneous ice nucleation datasets may be exhibiting a surface area dependence that hasn't been traditionally accounted for. Our new compilation of more illite data and its comparison with the previously reported n_s values from different measuring techniques should add value to the manuscript and clarify this message.

The authors use the Broadley et al. data as an “absolute data set” meaning the uncertainty of the data and its implication for the application of this model is not considered. In this study it is emphasized that the nucleation process is stochastic in nature whereas Broadley et al. do not assume this. The Broadley et al. data likely possesses a large statistical uncertainty when stochastic processes are implied. Furthermore, the ice nucleating surface area in each droplet will be uncertain. As stated in figure caption 5, droplets with diameters 10-20 μm were applied. This results in about one order of magnitude uncertainty in surface area. This uncertainty alone would consolidate all curves shown in Fig. 5. In other word, this uncertainty nullifies attempted analysis and proof of the validity of the assumption of internal and external variability and suitability of this parameterization. Again, the presented approach may have some validity but it is very poorly executed by just looking at one data set and not discussing the uncertainties of the data set. Furthermore, the authors mention that they performed cold stage freezing experiments but these data are not shown. Why not making a stronger case, if there is the data?

In the revised manuscript we present additional datasets for illite, cellulose, and Snomax that exhibit a similar trend with decreasing surface area as the Broadley et al. data to make a stronger case for the value of this dependence on surface area and what we think it entails. We agree that there is a surface area uncertainty for any of the freezing curves and acknowledge that it partly may contribute to some of the broadness in the freezing curves. However, this uncertainty would not explain a consistent trend with decreasing surface area but would create a margin of error in temperature over which the freezing curve can lie.

p.1, l. 13-19: The 2nd sentence of the abstract lacks carefulness. Other researchers would claim their parameterizations are consistent with their experimental studies since they describe frozen fraction curves for changes in area, time, etc. There is no clear definition for “consistent” or “comprehensive”, and “freezing properties”? The following sentence then introduces the model with the statement that it uses a continuous function of contact angle and no restrictions on active sites. These statements are somehow misleading. Fact is, the model can reproduce experimental data.

The words “consistent and comprehensive” have been removed and replaced with “well established”. We just want to emphasize that the community has yet to settle on one standard way to describe and report heterogeneous ice nucleation properties.

p.1, l. 26-27: The authors write “the two-dimensional nature of the ice nucleation ability of aerosol particles”. What is the meaning of this? The only way I can make sense of this, is assuming that external and internal particle mixtures are meant by this?

We have removed the reference to internal and external variability in the abstract. It is now introduced and defined later in the text.

p. 2, l. 2-5: This sentence has to be reworded. A distribution cannot be statistically significant.

We have removed all references to “statistically significant” in the revised manuscript to avoid misrepresenting the framework and its interpretation of the data.

p.2, l. 6: “will not” This exemplifies a claim of certainty, when in fact this is based entirely on a model assumption of some active site surfaces. As mentioned above there is no direct experimental evidence for an internal/external active sites.

This sentence has been removed from the abstract. When this conclusion is made later in the paper, we have made sure to indicate that the result is based on our model and not a physical reality.

p. 3, l. 13-14: The results of Vali (2008) do not show there is a strong spatial preference because this could not be directly measured. Vali (2008) might have claimed his experimental results suggest there are active sites in preferential locations (based on mathematical analysis).

This has been reworded to say that based on the model presented by Vali (2008), the experimental results are suggestive of active sites on preferential locations, on Page/Line 3/19-20: “These results suggest that there is a strong spatial preference on where nucleation occurs, supporting a model of discrete active sites.”

p. 3, l. 16-19: The role of time for what? This is very sloppy discussion and does not reflect the community’s concern on this issue besides lacking important laboratory work from Koop, Knopf, Lohmann, and others and field work indicating the important role of time to explain observations. This section has to significantly improve if time dependence is addressed in this manuscript. As it is, the reader is left pretty clueless and cannot do more than accept written statements.

Time dependence is only addressed briefly to introduce the framework and doesn’t comprise an essential element of the message the paper is trying to convey. Our understanding of the current state of knowledge is that heterogeneous ice nucleation is much more strongly dependent on temperature than time (Vali, 2014; Wright and Petters, 2013). As stated in the manuscript, whether the role of time has proven to not merit inclusion in models remains to be seen. It is with our understanding of its potential importance that we have developed our framework to still account for time despite time dependent analysis not being a major focus in this work where we focus on the surface area dependence.

p. 3, l. 20: “completely”? What is meant by this?

This is a typo. “Completely” should be followed by “discarded”. This has been corrected.

p. 3, l. 29 - p. 4, l. 2: This is in principle the repetition of previous sentence describing the findings by Ervens and Feingold. However, here it is somehow generalized: What models? What results? Why are their more drastic variations?

We have reworded the text here to avoid general statements and merely indicate an important finding of Ervens and Feingold (2012). Text has been revised on page/line 4/1-7:

“Ervens and Feingold (2012) tested different nucleation schemes in an adiabatic parcel model and found that critical cloud features such as the initiation of the WBF process, liquid water content, and ice water content, all diverged for the different ice nucleation parameterizations. This strongly affected cloud evolution and lifetime. The divergence was even stronger when the aerosol size distribution was switched from monodisperse to polydisperse.”

p. 4, l. 3: “First principles of classical nucleation theory”. This is a strong claim. I would much doubt that the authors show any derivation from first principles in this manuscript. There is no discussion or derivation of clustering, free energy changes or chemical potentials, capillary approximation, etc.

“First principles of classical nucleation theory” has been changed to “based on classical nucleation theory”.

p. 4, l. 5-8: “accounts for the variable nature of an ice nucleant’s surface and the distribution of ice active surface site ability across a particle’s surface (internal variability), and between individual particles of the same type (external variability).” This must be much more careful formulated. There is no direct evidence for the variable ice nucleating nature of a particle surface or the surface of different particles. This is an assumption the authors make based on previous work that predisposed this assumption into a mathematical fit. Also, on l. 5, ice embryo growth and dissolution is part of classical nucleation theory. This is part of a testable physical theory, but not “proven” to occur. The authors need to recognize that even an ice embryo is theoretical. The existence of a g-distribution is even less so as it serves a mathematical scaling or integrating fitting function, not something physical.

We have reworded this to emphasize that internal and external variability along with the other concepts presented here are modeling tools to describe and interpret the data and present a means to model ice nucleation behavior. They are not physical realities in the strict sense. We have revised the text, on Page/Line 4/11-15:

“The new framework is stochastic by nature to properly reflect the randomness of ice embryo growth and dissolution, and assumes that an ice nucleating particle can exhibit variability in active sites along its surface, what will be referred to as internal variability, and variability in active sites between other particles of the same species, what will be referred to as external variability.”

p. 4, l. 10: “and interpret”. This model cannot interpret the freezing data since it is not based on a testable theory. Its assumptions cannot be proven and a g-distribution cannot be measured. The authors want to interpret freezing as the result of active sites, when in fact they already assume that the presence of active sites result in freezing. This indicates circular reasoning. Although, it is sufficient to say that this approach can successfully describe the freezing data - a valuable result.

Interpret has been changed to "describe".

p. 5, l. 17-19: Reflects a misunderstanding of the authors about CNT. 1. "pure" makes no sense here. 2. CNT does not assume/indicate that ice nucleation occurs uniformly across a particles surface. This formulation considers only an embryo on a surface. 3. A particle surface area is not included in Eq. 2, this is because there is no dependence on particle surface area. Maybe the authors assume that the contact angle is uniform over the entire surface and from this, when applying Eq. 2 over the whole particle surface, infer that ice nucleation ability is uniform across the entire surface. In other words, CNT has never made any assumption of uniformity of particle surface areas, but a single contact angle is only conceptualized by previous studies in the literature. It is not a facet or constrain of CNT. This should also be changed on p. 8, l. 12-14.

The text has been changed to indicate that the stochastic formulation is one that uses CNT with a single contact angle assumption and not that CNT assumes embryo formation is uniform over the surface considered. On Page/Line 6/3-4:

"The simplest stochastic formulation hypothesizes that the nucleation rate is uniform across the ice nucleating particle's surface, i.e. makes a single contact angle assumption."

We have also omitted the reference to CNT on p. 8, l. 12-14:

"The single θ fit has a steeper dependence on temperature a result of the double exponential temperature dependence of the freezing probability in Eq. (4) (J is an exponential function of temperature in itself as can be seen in Eq. (2)) results in an approximately temperature step function."

p. 5, l. 22: Equation 3 can only be formulated assuming that every particle has the same surface area. The authors define A as the surface area of a single particle. Then this A must have an index for each particle? The assumptions for this equation are not clear and are misleading.

It is now indicated that every particle is assumed to have the same surface area A in the derivation of equation (3), on Page/Line 6/10-11:

" A is the surface area of each individual ice nucleating particle (assumed to be the same for all particles)."

p. 6, l. 3-6: "A more realistic approach is to recognize" is a very bold statement. How about "We assume... "?

The text has been modified on Page/Line 6/17-20:

"Given the large variability in particle surface composition and structure across any one particle, which in turn determines the activity (or contact angle, θ) of a potential ice nucleating site, a different approach is to assume that the heterogonous nucleation rate will vary along the particle-droplet interface."

p. 7, l. 1-8: Maybe make clear that these are the authors' definition of internal and external variability. This does not represent text book knowledge and agreed-upon facts.

We have placed emphasis on the concepts of internal and external variability being introduced in this manuscript as part of a new framework.

p. 7, l. 9-11: This is a misleading statement and should be discarded. There is no proof that this approach provides direct insight. The authors are assuming variability without showing that particle surfaces are considerably variable in terms of their ice nucleation ability. Again this is a mathematical construct.

“Direct insight” has been omitted.

p. 8, Eq. 8: J , per definition, is not a function of time but of temperature. Here, this is only the case because via the cooling rate it gives temperature. This is confusing when coming from CNT and not necessary. One could start with Eq. 9.

The symbol for time t has been replaced with $T(t)$ in the parentheses following J since it is temperature that is a function of time and not J .

p. 8, l. 16-21: This is an example, where the authors show no sensitivity that their approach is mathematical only, but use the good fit to make firm statements about the underlying process for which there is no proof/direct observation. In fact, other fit based studies could claim the same. For now, these are non-testable statements and should be avoided.

We have reworded the text here to indicate that internal variability and its impact on time dependence is a mathematical model of what is happening and not a physical interpretation. The claim that evidence of internal variability is captured is discarded. The text was revised on Page/Line 9/11-14:

“The diversity of nucleating ability on the particle surface captured by the g parameter offsets some of the steepness and yields a more gradual freezing curve, more similar to the actual experimental freezing probability curve.”

p. 8, l. 22 to p. 9, l. 6: This section has to be improved. This is too difficult to understand in terms of what has been done mathematically to derive the freezing probabilities. I am left with several assumptions how to proceed.

We have attempted to better describe the details of the modeling exercise done here. We actually run equation (7) for all temperatures for a constant time of 1 hour to assess the freezing probability that results from the hypothetical g distribution retrieved under different conditions. The dotted red line is the modeled freezing probability of the droplets for all temperatures after a waiting time of 1 hour. The text was revised on Page/Line 9/15-23:

“Two droplet freezing probability fits (dotted lines) are also plotted in Fig. 1 under different environmental conditions. Instead of prescribing a cooling rate the freezing probabilities are generated by running Eq. (7) for the entire temperature range with each fit for $\Delta t = 1$ hour. One fit uses the same g distribution used previously, while the additional single θ fit is approximated as a normal distribution with a near zero standard deviation, similar to a Delta Dirac function. The resultant freezing probabilities are then computed and plotted for every T . It can be seen that the g fit retains much stronger time dependence, with the freezing probability curve shifting about 5 K warmer and the single θ curve shifting just 1 K warmer for the 1 hour hold time.”

p. 9, l. 17-22: Again, strong statements for an effect that cannot be fundamentally proven as of yet and that can also be described by other mathematical/physical means. Why not frankly state something like: "These results suggest that ... may ... may ... though previous parameterizations have also been able to describe ...". I assume the authors want to put out this new idea, something to further investigate in the future...

We have added references to similar modeling exercises that have been reported and experimental data showing a stronger role of time than a single theta fit would project. The conclusion of this section has been reworded to emphasize that a multiple theta fit does a better job of fitting the experimental data, be it caused by the broadness in a single droplet's freezing probability curve or the effect of time on freezing. The text was revised on Page/Line 9/24-27 and 10/1-14:

"Wider g distributions therefore yield stronger time dependence due to the partial offset of the strong temperature dependence that the nucleation rate in Eq. (2) exhibits. The result emphasizes that how the active sites are modeled has consequences on what physical parameters (e.g. time, temperature, cooling rate) can influence the freezing outcome and observed droplet freezing temperature spectrum (Broadley et al., 2012). In Fig. 1 a wider g distribution resulted in higher sensitivity to time, which resulted in a shift of the freezing curve to higher temperatures as the system was allowed to temporally evolve at a fixed temperature. This significant change in the freezing probability's sensitivity to temperature is the cause of the more gradual rise in the freezing probability for the system when applying a non-Delta Dirac g distribution. This is effectively enhancing the stochastic element in the particle's ice nucleation properties. The shallower response of freezing probability to decreasing temperature (deterministic freezing) creates a greater opportunity for time-dependent (stochastic freezing) to manifest, as a larger fraction of the droplets spend more time unfrozen. The enhancement of the stochastic element brings about a more important role for time as shown in Fig. 1. The finding of this exercise is consistent with previously published work on time dependent freezing such as those reported by Barahona (2012), Vali and Stransbury (1966), Vali (1994b), and Wright and Petters (2013), amongst others."

p. 9, l. 27- p. 10, l. 1: This text section states that a g distribution is just a probability density function that indicates the numbers of sites with a certain θ . But the text starting on p. 15, l. 8 states that the authors draw θ from a uniform distribution and then calculate $g(\theta)$? So g is not a probability that particles have a certain θ value? Does this mean every θ from 0 to 180_ has an equal chance to be present on the surface of particles, but freezing probabilities are scaled by the integrating factor $g(\theta)$?

In the n_draws method, even though a random contact angle is drawn from a uniform distribution (no preference as to where in the contact angle range of 0 to 180 it is drawn from) the value of g for the particle is then assigned the value of g_bar at the random contact angle value chosen. Once all the random draws are made, the new resultant discrete probability distribution is created from the contact angles sampled from g_bar , and this is then weighted by the surface area of the particle being modeled. This results in a bias for contact angles with higher g_bar values to be represented. Further clarification of the procedure has been added to the text on Page/Line 16/29-30 and 17/1-16 along with a new figure (bottom of Figure 4) that displays a schematic showing the details of this procedure:

"To predict the freezing curves of the droplets with particle surface areas lower than the estimated critical area for the systems considered here, the aggregate surface area of the entire particle population within each droplet is modeled as one large surface. A contact angle θ_r is

randomly selected from the full contact angle range $[0, \pi]$, and the value of active site distribution g^* for the particle i being sampled for at θ_r is assigned the value of $\overline{g(\theta_r)}$:

$$(g_i^*(\theta_{r,n_{draw}})) = \overline{g(\theta_r)} \quad (16)$$

The g distributions within this numerical model are given an asterisk to indicate that they are discrete distributions.

This process is repeated for a parameter n_{draws} , for each droplet in the array that produced the freezing curve being modeled. n_{draws} is the only parameter that is optimized for so the modeled freezing curves can predict the behavior of the experimental freezing curves. The value of n_{draws} typically ranges from 9 to 65 for the systems analyzed here and is therefore a relatively soft optimization parameter with small dynamic range. The sampled g^* distributions are normalized with respect to the estimated total surface area for the freezing curve being modeled before being used to compute the freezing probability. The bottom part of Figure 4 shows a schematic of how g^* is retrieved from $\bar{g}$ using n_{draws} .

p. 10, l. 4- 8: This is very confusing. First somehow one large active site is assumed (summing up surface area) but then it is stated that this active site (which by definition has one nucleation probability) has a continuum of ice nucleation activities.

We have changed the description here and we are no longer referring to the ice nucleating spectrum as one site. It is now referred to as a spectrum of ice nucleating activity, comprised of many sites with strengths and frequencies determined by the Gaussian g distribution. Emphasis on the ascending part of this distribution is given since it is the fraction of the curve that determines the modeled freezing probability. The text has been revised on Page/Line 10/25-29:

“It is therefore sufficient to conceptualize that the particle has a well-defined monotonic spectrum of active sites increasing in frequency while decreasing in strength. The spectrum is modeled as a continuum of ice nucleation activity described by the g distribution, as depicted on the upper right hand corner in Fig. 2.”

p. 10, section 3.2: Why not plot the continuous distributions used in this work including the approximated one and full one (g and $\bar{g}$)? Could be added as a supplement.

We have added a plot showing the g distribution used here and indicated the part of the distribution covered in by the critical contact angle range on the plot. It has been added to Figure 3.

p. 11, l. 12-21 and following: Again, very firm statements on the underlying molecular processes not treated by the mathematical formalism. Statement of active site size is incorrect. CNT does not give size of active site but gives size of a critical ice embryo for given supersaturation. That this somehow, potentially reflects the size of an active site is very speculative and questioned by most recent findings using molecular dynamics simulations (e.g. Cox et al., 2013, Zielke et al., 2015). The fact is that a number can be calculated by integrating Eq. 11, but this is only a result of your assumption of a g distribution. It does not give significant insight.

The estimate of the ice nucleation area provided by this analysis provides useful information that can be compared to other estimates of this quantity, as we have done in the paper. We have

revised the text to clarify that this does not provide a direct measurement of the active site size, on Page/Line 12/1-12:

"Furthermore, the critical contact angle range can be used to estimate a hypothetical nucleating area of the particle – the total active site surface area where nucleation will take place. The nucleation area $A_{nucleation}$ can be estimated as follows:

$$A_{nucleation} = A \int_{\theta_{c1}}^{\theta_{c2}} g(\theta) d\theta \quad (11)$$

For the large ash particle system analyzed in the previous section (Fig. 1) it is estimated that $\theta_{c1} \approx 0.4$ rad and $\theta_{c2} \approx 0.79$ rad. Application of Eq. (11) yields a total ice active surface area estimate of 27 nm^2 . Classical nucleation theory estimates that the area of a single active site is 6 nm^2 (L  nd et al., 2010; Marcolli et al., 2007). The estimated total area of nucleation is therefore consistent with this value and supports the argument that competition between sites along the critical range of θ is taking place. However, the surface area where ice nucleation is occurring remains a very tiny fraction of the total particle surface."

p. 12, l. 25 – p. 12, l. 2: These general statements are incorrect. See general comments above. There are other types of cold stage experiments that apply micrometersized droplets and INPs with surface areas that are atmospherically relevant. Also, this manuscript does not give a fundamental proof that studies using large particles result in erroneous nucleation descriptions. If so, this would have ramifications far beyond the area of atmospheric sciences.

To our knowledge, there isn't a cold plate technique that probes single atmospherically relevant sized particles per droplet. Since cold plate droplets arrays are prepared from particle suspensions, an experiment in which atmospherically relevant particle surface areas (particle count per droplet will still be high) can be conducted. The manuscript does not intend to show that using large particles results in erroneous nucleation descriptions but that there is a particle surface area dependence of ice nucleation beyond the scaling factor used in both the n_s and CNT based schemes. We show evidence of this in our retrievals of n_s directly from the experiments, whereby at low surface area n_s values retrieved from cold plate methods do not overlap in temperature space. The model presented is a mathematical tool that attempts to describe why droplets containing particles with large total surface areas freeze more uniformly than droplets with small surface areas do, for the datasets considered here. We feel that the new datasets added to the manuscript and their discussion demonstrate this variability in n_s as particle concentration and thus surface area is varied.

p. 12, l. 7-9: This is confusing, also due to above issues of definition of variability. The frozen fraction curve resembles freezing of droplets not considering the INPs inside it. The Murray group observes a subset of droplets freezing differently than others, suggesting external mixtures. A few lines above, one large particle in one large droplet is described and here one large droplet with many small particles is considered, but still within one droplet. In fact many small particles should express a larger surface area. The effect of many small cannot be resolved since only freezing of that one entire droplet is observed.

When considering droplets with many particles immersed in them we consider the sum of all individual particle surfaces as one surface area of interest. So when we try to describe these datasets in the context of our framework we treat the immersion as one particle, of which its surface area is estimated using the measured surface area density of the studied sample. We have

clarified this in the revised text, on Page/Line 13/5-7:

"For the application of this model to cold plate data where droplets are prepared from a suspension of the species being investigated, the particle population in each droplet is treated as one aggregate surface."

p. 12, l. 16-18: Poor wording: "threshold of statistical significance". Of a distribution?

We have removed all references to "statistical significance" previously included. Please see our reply to your comment above.

p. 12, Eq. 12: Until now the word 'system' has been something general, but here is there a specific definition to this? What is one system? What is the i th system? Is a single droplet a system, is a single particle a system with active sites, etc.? Be consistent throughout the document.

The use of the term "system" was not consistent in the original manuscript as it referred to both an individual droplet at points and to a species being investigated at other points. The word system now refers to the species under consideration, e.g., illite particles, and it is not used to describe a droplet in the earlier equation derivations.

p. 13, l. 14-22: Reword to express more suggestive nature of results.

We have worked to change the text to suggest that the results are to be interpreted in the context of the mathematical model presented and not in the absolute physical sense. The text was revised on Page/Line 14/7-15:

"Above a certain surface area threshold it is conceptualized that the chance of an ice-nucleating particle surface not possessing the entire range of ice nucleating activity (θ) becomes very small. The model therefore assumes that any particle or ensemble of particles having a total surface area larger than the critical area can be approximated as having $\bar{g}$ describe the actual g distribution of the individual particles. In other words, for large particles with more surface area than the critical area threshold, it is assumed that the external variability between individual particles will be very small such that the particle population can just be described by one average continuous distribution of the ice active site ability, $\bar{g}$."

P. 13, l. 23: Poor wording: "threshold of statistical significance".

Removed, please see above.

p. 14, l. 1: What are high particle concentrations? Whose data are you using here? Should be stated in the beginning of this section. What is a retrieved averaged g distribution?

High particle concentrations are a reference to concentrations that result in total particle surface areas in the droplet greater than the critical area threshold we have identified for that particle system. The structure of this entire section has been changed significantly to make the presentation of the results and the model clearer. The retrieved average g distribution is the g

distribution that creates the best fit of the data using Equation (9). Stating "average" before "g distribution" is unnecessary and misleading and has thus been omitted. The text has been revised thoroughly, on Page/Line 14/16-26:

“To resolve the g distributions of the systems possessing particle surface areas smaller than the critical area the first step is to approximate the critical area. Experiments must start at very high particle surface area concentrations to ensure the number of particles and total surface area per droplet exceeds the critical area. For the illite mineral particle case study considered next, for example, high particle concentrations were those that resulted in total particle surface areas greater than about $2 \times 10^{-6} \text{ cm}^2$. The particle number or surface area concentration is then decreased until the retrieved g distribution (from the measured droplet freezing temperature spectrum for an array of droplets containing particles) can no longer be reasonably predicted by $\bar{g}$. This point can identify the parameter A_c , the critical area of the species under study. A schematic of the procedure is summarized in Fig. 4.”

p. 14, l. 7-31: It seems discussion starts with the right panel of Fig. 4. Why not plotting this one in the left panel? Please add experimental data as well to show model representativeness.

As mentioned in the previous response, much of the organization of this section has been improved, in part to address the referee's suggestions.

p. 14, l. 22-24 and l. 27-30: Your approach is successful, but only due to the assumptions used in simulating the freezing. This does not mean that it actually happens in your sets or Broadley et al., 2012.

In this part of the text we were referring to the success of the n_s scheme in describing the freezing behavior for the high particle surface area experiments. We were not referring to the results of the presented model yet. We hope that the format of the new section will clarify many of this unintentionally misrepresented issues.

p. 15, l. 1-5: This is important. When introducing a new model, it has to be evaluated by different data sets. Why are these results not shown?

New datasets retrieved with our own cold plate system using illite NX, Snomax, and cellulose particle systems and their analysis have been added to the manuscript.

p. 15, l. 6-11: Isn't a running index for $g(\theta_r)$ missing to indicate that the calculation is performed for each individual droplet? Somehow this is missing here and above in the manuscript. In other words g is subsampled to find the contact angle that causes freezing of that particular droplet within the given frozen fraction curve?

A running index i for $g^*(\theta_r)$ has been added to indicate the n^{th} droplet being modeled. An additional index for θ_r has also been added to indicate what the n_{draw} it is being used for.

p. 15, l. 12-19: See general comments above. When subsampling from g distribution (please present) with an arbitrary number of draws it is not surprising to represent the data. If I draw

often enough, I can win any lottery without understanding the nature of the lottery. Can you present how often you draw for different data sets? E.g. a rare active site may have a probability of 10^{-10} . Then you have to draw 10^{10} times...?

The values of n_{draws} for each dataset analyzed in the revised manuscript have been added to the text. The values actually vary from 9 to 65 for all the 3 systems studied here (illite, Snomax, cellulose). A random contact angle is first chosen from the entire contact angle range. Because of the nature of the sampling process, a large number of draws is not necessary for sampling from the very active contact angle range. When a random contact angle is selected, its value at $\bar{g}$ is assigned to the g distribution being generated at that same contact angle. The number of draws required to generate a g distribution similar to $\bar{g}$ ends up being on the order of 25 for the cellulose and illite, and about 70 for Snomax, because enough contact angles have been selected to approximate $\bar{g}$. Note that a new g^* distribution is created using n_{draws} for each droplet for that system. The freezing probability for each droplet in the array is calculated using the new sub-sampled g^* distribution, and Eq. (9). This is followed by using Eq. (17) to compute the modeled frozen fraction. The confusing regarding this method is understandable, and we have revised the text to clarify this, on Page/Line 16/29-30 and 17/1-16:

"To predict the freezing curves of the droplets with particle surface areas lower than the estimated critical area for the systems considered here, the aggregate surface area of the entire particle population within each droplet is modeled as one large surface. A contact angle θ_r is randomly selected from the full contact angle range $[0, \pi]$, and the value of active site distribution g^* for the particle i being sampled for at θ_r is assigned the value of $\bar{g}(\theta_r)$:

$$(g_i^*(\theta_r, n_{\text{draws}})) = \bar{g}(\theta_r) \quad (16)$$

The g distributions within this numerical model are given an asterisk to indicate that they are discrete distributions.

This process is repeated for a parameter n_{draws} for each droplet in the array that produced the freezing curve being modeled. n_{draws} is the only parameter that is optimized for so the modeled freezing curves can predict the behavior of the experimental freezing curves. The value of n_{draws} typically ranges from 9 to 65 for the systems analyzed here and is therefore a relatively soft optimization parameter with small dynamic range. The sampled g^* distributions are normalized with respect to the estimated total surface area for the freezing curve being modeled before being used to compute the freezing probability. The bottom part of Fig. 4 shows a schematic of how g^* is retrieved from $\bar{g}$ using n_{draws} . With the sampled g^* distributions the freezing probability of each droplet is calculated using Eq. (9) and the frozen fraction curve is computed from the arithmetic average of the freezing probabilities:

$$F(\text{below critical area}) = \frac{1}{N} \sum_{i=1}^N P_{f_i} \quad (17)$$

where N is the number of droplets in the cold plate array."

The values of n_{draws} for all systems modeled are now reported on Page/Line 17/24-28:

"The values of n_{draws} for the lower concentration freezing curves for each of the systems investigated here are 21 ($2.02 \times 10^{-6} \text{ cm}^2$), 19 ($1.04 \times 10^{-6} \text{ cm}^2$), and 11 ($7.11 \times 10^{-7} \text{ cm}^2$) for the droplets containing illite; 65 (0.09 wt%), 48 (0.08 wt%), and 23 (0.07 wt%) for the droplets containing Snomax; and 21 (0.05 wt%), 11 (0.01 wt%), and 9 (0.001 wt%) for the droplets containing cellulose."

p. 15, l. 21 and following: Please see general comments on uncertainties of experimental data sets.

We recognize that uncertainty in surface area could result in a significant difference in the predicted temperature range over which freezing would occur for droplets studied here. However, this uncertainty would not explain the consistent trend of broader freezing temperatures as surface area decreases unless surface area uncertainties became larger with decreasing concentration. We do not see why surface area uncertainty would increase with decreasing concentration; in fact we think the opposite is true where at high concentrations the suspensions become less stable due to potential particle coagulation and settling. Physical artifacts under high particle concentrations that lead to coagulation and settling are now discussed in the text for the illite measurements.

p. 17, l. 1: The wording should be much more careful. As is it adds to confusion. What is a curve's behavior? What does it mean to be qualitatively and/or quantitatively captured?

Much of the wording of this section has already been changed in an attempt to clarify the implications of the analysis done. The use of “qualitative” and “quantitative” was unnecessary here. We were simply trying to emphasize that the presented model is able to describe the trend seen in the freezing curves as the surface area of the particles is lowered. We have revised the text, on Page/Line 17/16-21:

“The behavior of the experimental curve is captured using the n_{draws} numerical model in which random sampling from the ice nucleating spectrum dictated by $\bar{g}$ is carried out to predict the freezing curve. The dotted lines in Figs. 5, 6, and 7 are obtained by sampling from the $\bar{g}$ model to successfully predict the behavior of all the freezing curves. The early freezing onsets of the lower concentration systems as well as the broadness in the curves are both captured with the model.”

p. 17, l. 7-9: I thought it is continuous. Why now arbitrarily dividing it in 1 nm² segments? And why this size?

The division of the particle into tiny patches is actually not part of the model presented, but that of an alternative model that is still being developed. We have omitted this sentence.

p. 17, l. 10-30: Again, this is only because of your assumption and does not give any evidence that it actually happens. It is acceptable to state that this paragraph is just your hypothesis and it may or may not be the case.

The revised manuscript stresses that this is a hypothesis and a suggestive mathematical description of the observations. We have removed assertions of a physical reality. We have revised the text accordingly, on Page/Line 18/1-18:

“Perhaps the most notable characteristic is how these freezing curves ascend together early as temperature is decreased but then diverge as the temperature decreases further. The closeness of the data at warmer temperatures (the ascent) is interpreted by the framework as the presence of some rare high activity active sites within the particle population under all the particle concentrations explored in these experiments. At lower temperatures it appears that there is a wider diversity in the activity of droplets that did not contain these rare efficient active sites, and thus there is significant spread in the freezing curve for $T < 242$ K. In the context of the framework presented here this can be attributable to strong external variability of the ice nucleating population, with very strong/active nucleators causing similar freezing onsets for

different particle concentrations at the warmer temperatures, and a lack of strong nucleators explaining the less consistent freezing of the unfrozen droplets at lower temperature. Thus it follows that there is a wider spread in the freezing curves for these droplets, as their freezing temperature is highly sensitive to the presence of moderately strong active sites. This expresses a greater diversity in external variability – the active site density possessed by individual particles from the same particle source. In a later section the claim of more external variability contributing to the broader curves below the critical area threshold is supported with a closer look at the numerical results from the model.”

p. 18, l. 4-6: No, it is the first study that assumes it.

This has been changed to state that this is the first study that models the process in such a manner.

p. 18, l. 20-23: This statement, I feel, is a little unfair. The mathematical description of Broadley et al. (2012) were never designed to fit a global distribution and then fit again for the number of draws for smaller surface areas. As stated above, I don't feel that the authors' procedures are superior, just different.

We do not mean to claim that our method is superior. We were pointing to the difference between using one distribution to describe the freezing data (by drawing from said distribution) and fitting every freezing curve to an independent distribution. The latter approach is treating every freezing curve independently, where the particles in the droplets in the different cases have different active site distributions that are not generated from the same source. We have revised the text accordingly, on Page/Line 22/18-22:

“A similar conclusion along these lines was reached by Broadley et al. (2012) when the authors noted that the best fits to their freezing curves were achieved when the system was assumed to be totally externally variable. That is when each particle was assumed to have a single contact angle but a distribution assigned a spectrum of contact angles to the particle population.”

p. 18, l. 24 – p. 19, l. 13: This section is also too strong in tone. It feels that the authors are dismissing all previous studies as inferior. The only difference between these studies is that different assumptions were made to represent their data. It suffices to say once that the size of active sites are not assumed. The fact that other studies do assume this, does not make their parameterizations any better, worse or less correct.

The tone has been modified here to establish the difference between each methods' approach and not a comparison in the value of each method. We have revised the text accordingly, on Page/Line 23/6-22:

“There are other formulations that hypothesize an active site based or multi-component stochastic model such as the ones described in Vali & Stransbury (1966), Niedermeier et al. (2011), Wheeler and Bertram (2012), and Wright and Petters (2013). Vali and Stransbury (1966) were the first to recognize that ice nucleating surfaces are diverse and stochastic and thus active sites need to be assigned both a characteristic freezing temperature as well as fluctuations around that temperature. Niedermerier et al. (2011) proposed the soccer ball model, in which a surface is partitioned into discrete active sites with each site conforming to classical nucleating theory. Marcolli et al. (2007) found a Gaussian distribution of contact angles could best describe their heterogeneous ice nucleation data in a completely deterministic framework. Welti et al. (2012)

introduced the alpha-PDF model where a probability density function prescribes the distribution of contact angles that a particle population possesses, such that each particle is characterized by a single contact angle. Wright and Petters (2013) hypothesized the existence of a Gaussian probability density function for a specific species, which in essence is similar to the $\bar{g}$ framework described here. The notable difference is that their probability density function was retrieved via optimizing for all freezing curves, and not independently fitting high concentration freezing curves as we have done here.”

p. 19, l. 14: What is meant by multicomponent? Different active sites? In addition, who said that they failed to become a standard? If the authors want this sentence to remain in the manuscript and any other like it, they should write “It is our opinion that multi-component: : have failed: :” Studies by e.g. Hiranuma, Murray and Wex and others do not state that the multicomponent stochastic formulations have failed to become a standard in the way the authors write it.

Multi-component here refers to any formulation that assumes multiple active sites. No one heterogeneous ice nucleation parameterization has thus far succeeded in being a standalone standard, and we have changed the text to reflect this. We think there is a general preference to reporting results from different ice nucleation methods for easy comparison using the n_s framework due to its simplicity and ease of use, but not that this formulation is undisputed and the only one to be used to report heterogeneous ice nucleation results. We have revised the text accordingly, on Page/Line 23/23-28:

" The n_s scheme is now more commonly used to describe and compare cold plate and other experimental ice nucleation data over multi-component stochastic formulations (Hiranuma et al., 2015; Murray et al., 2012; Wex et al., 2015). This is in part due to the necessary inclusion of more variables required by other frameworks (such as prescribing a discrete number of active sites in the soccer ball model by Niedermeier et al. (2011)) than the simpler purely deterministic scheme of n_s ."

p. 19, l. 20: “only”. This method is computationally more demanding than others. The authors admit this on l. 29-30. Why emphasize at this point?

We acknowledge that some computation is required to retrieve frozen fraction curves or freezing probabilities below the critical area. However, this process only needs to be done once, after which the h correction factor can be used to transform the frozen fraction functions below the critical area. We have removed the sentence about this step being computationally cumbersome, as after some consideration we have realized that it shouldn't be considered such.

p. 20, l. 8-10: The word “trivially” should be taken out. It cannot be done yet. One cannot know the distribution of any ice active sites independent of an ice nucleation experiment.

We agree. The word “trivially” has been removed.

p. 20, l. 29 – p. 21, l. 2: The authors do not know what individual atmospheric particles will or will not contain. Under giving assumptions, this is what your analysis suggests.

This conclusion along with others about the nature of the active site distribution on particles below and above the critical area, are meant to be stated in the context of the model presented and not as physical realities. We hope that the changes throughout the manuscript on this general issue will correct this shortcoming and clarify our meaning.

p. 21, l. 28-30: Again, tone: The authors write like a “statistically significant size cutoff” is proven to exist for atmospherically relevant particles. This is far from the case.

We have changed this to state that more studies need to be performed to determine if atmospherically relevant particles exhibit the same trend examined in this paper. We have revised the text accordingly, on Page/Line 26/10-12:

“More detailed analysis studying various atmospherically relevant ice nucleating particles needs to be done to shed light on whether a particle size cutoff corresponding to a critical area threshold can be used to describe the behavior of different species.”

p. 22, l. 5: This statement is too strong and likely just wrong. The majority of the community would disagree with this.

We have changed the tone of this statement to indicate that our findings point to one n_s parameterization not being sufficient to describe all illite ice nucleation behavior, as we have seen the values of this function do not overlap at lower surface areas. Variation in n_s for illite NX was also reported and extensively discussed by Hiranuma et al. (2014). Perhaps one n_s function may be sufficient, but some form of a correction might be needed at low surface areas where we think the actual active site density becomes different between sample surfaces contained in individual droplets.

The crux of our argument is that the surface area normalization assumption that underlies the n_s framework warrants closer inspection and evaluation. The ice nucleation community has essentially been operating under the assumption that the same n_s value will always be retrieved from any proper method, regardless of how large a difference in particle concentration or surface area exists between methods. Inconsistencies in the n_s values retrieved using different methods for the same system (such as illite NX and cellulose MCC) are widely known and discussed in the community. This is often thought to be caused by differences between the methods used, and their method artifacts. Particle coagulation and settling at high particle concentrations is one proposed method artifact, which we also suspect explains our highest concentration illite data. We are suggesting that the observed difference in n_s between methods and research groups may be more fundamental in nature and caused by changes in the distribution of active sites contained in particles sampled in the individual droplets that compose the arrays used in cold plate methods. We have presented experimental data from three systems and two research groups that demonstrate this variability in n_s as particle concentration and surface area are changed, and used our model to interpret and propose an explanation for these effects. While we agree we have not conclusively proven that our interpretation of the causes of these changes in n_s is the correct answer, we do not believe that there is available evidence that disproves our hypotheses. Considering the ongoing issues in reliably determining the concentration of INP and their ice nucleation properties/activity, a healthy debate that considers many possible explanations is warranted. This proposal is the main intent of our central hypothesis and the supporting data and analysis presented. Our discussion of the n_s framework has been revised in the text, and data from our cold plate system for Snomax, illite, and cellulose has been added to the revised paper.

p. 22, l. 10-17: What is the intention of this paragraph? This is too strong in tone. It also discredits all previous work. As stated above, the applied analysis does not allow such firm statements.

The intention of this paragraph is to state that the cold plate technique enables probing a large surface area range which aids in determining whether a single active site density function is sufficient to describe data for all size of a considered particle species or not. Tone has been changed to sound less assertive and more suggestive, on Page/Line 27/4-12:

“Cold plate experimental data potentially provides sufficient information to describe heterogeneous ice nucleation properties in cloud parcel and atmospheric models, however the analysis undertaken here suggests that retrieving one active site density (i.e. n_s) parameterization and applying it to all surface areas can result in misrepresenting the freezing behavior. When samples are investigated, probing a wide concentration range enables the determination of both general active site density functions (e.g. $\bar{g}$) as well as the behavior of the species’ under study at concentrations below the critical area threshold. Once this analysis is undertaken more comprehensive parameterizations can be retrieved as will be developed in the next section.”

p. 22, l. 18-20: Again this holds only under given assumptions.

For the example cases considered here we show that extrapolating n_s to lower surface area does yield errors in say a cloud parcel model. This is supported by the n_s retrievals for the example systems considered. This is discussed in the text, please see comment that follows.

p. 23, 5: “If our assumption are true, then this would have consequences...”

If a cloud parcel model uses n_s values extrapolated from the high surface area freezing curves for the low surface area freezing curves for the example systems considered, the model will neither capture the onset of freezing nor the range of temperatures over which freezing occurs. We have clarified what we are trying to state here, on Page/Line 27/13-30 and 28/1-2:

“The critical area analysis carried out in this paper emphasizes the dangers in extrapolating the freezing behavior of droplets containing a large concentration of particle to droplets containing smaller concentrations or individual particles. Applying a parameterization such as n_s directly to systems below the critical area threshold in a cloud parcel model for example yields large differences in the predictions of the freezing outcome of the droplet population. As the concentration of the species within the droplets was decreased in the cold plate freezing spectra considered here the actual freezing temperature curves diverged more and more from those predicted when the systems were assumed to be above the critical area. This led to significant changes in the retrieved n_s values, as shown in Figs. 4b, 6b, and 7b. The large effects of concentration on the droplet freezing temperature can be directly observed in the frozen fraction curves plotted in Figs. 5, 6a, and 7a. Differences between observed frozen fraction curves and ones that assumed uniform active site density yielded errors in the temperature range the droplets froze over as well as the temperature at which 50% frozen fraction point. Therefore, a cloud parcel model would be unable to accurately predict the freezing onset or the temperature range over which freezing occurs using a single n_s curve obtained from high concentration data. This has important consequences for the accurate simulation of the microphysical evolution of the cloud system under study such as the initiation of the Wegener–Bergeron–Findeisen and the consequent glaciation and precipitation rates (Ervens and Feingold, 2012; Ervens et al., 2011).”

p. 23, l. 20: The previous paragraphs are written in such a way (like a summary and conclusion), that it felt that the paper should finish here. The authors might consider to place some of the said in the conclusions section.

We have incorporated the suggestions of both referees to shorten, reorganize, and clarify the final section and Conclusions of the paper, and appreciate the referee's feedback. This section now read as follows, on Page/Line 27/13-30:

“The critical area analysis carried out in this paper emphasizes the dangers in extrapolating the freezing behavior of droplets containing a large concentration of particles to droplets containing smaller concentrations. Applying a parameterization such as n_s directly to systems below the critical area threshold in a cloud parcel model for example yields large differences in the predictions of the freezing outcome of the droplet population. As the concentration of the species within the droplets was decreased in the cold plate freezing spectra considered here the actual freezing temperature curves diverged more and more from those predicted when the systems were assumed to be above the critical area. This led to significant changes in the retrieved n_s values, as shown in Figs. 4, 6b, and 7b. The large effects of concentration on the droplet freezing temperature can be directly observed in the frozen fraction curves plotted in Figs. 5, 6a, and 7a. Differences between observed frozen fraction curves and ones that assumed uniform active site density yielded errors in the temperature range the droplets froze over as well as the median droplet freezing temperature. Therefore, a cloud parcel model would be unable to accurately predict the freezing onset or the temperature range over which freezing occurs using a single n_s curve obtained from high concentration data. This has important consequences for the accurate simulation of the microphysical evolution of the cloud system under study such as the initiation of the Wegener–Bergeron–Findeisen and the consequent glaciation and precipitation rates (Ervens and Feingold, 2012; Ervens et al., 2011).”

We revised and moved one of the paragraphs from this section to the Conclusions. The revised Conclusions are now as follows:

“Cold plate droplet freezing spectra were carefully examined to investigate a surface area dependence of ice nucleation ability whereby one active site density function such as n_s cannot be extrapolated from high particle surface area to low particle surface area conditions. A method based on the notion of a critical surface area threshold was presented. It is argued that a species' entire ice nucleating spectrum can be confined within a global probability density function $\bar{g}$. For a system, be it one particle or an ensemble of particles, to have a total surface area greater than the critical area is a question of whether the surface is large enough to express all the variability in that particle species' ice active surface site ability. By analyzing droplets containing illite minerals, MCC cellulose, and commercial Snomax bacterial particles, it was shown that freezing curves above a certain critical surface area threshold could be predicted directly from the global $\bar{g}$ distribution obtained from the high particle concentration data alone. The lower particle concentration freezing curves were accurately predicted by randomly sampling active site abilities (θ) from $\bar{g}$ and averaging their resultant freezing probabilities. This framework provides a new method for extrapolating droplet freezing temperature spectra from cold plate experimental data under high particle concentrations to atmospherically realistic dilute particle-droplet systems.

We found that the shifts to colder freezing temperatures caused by reducing the particle concentration or total surface area present in droplets cannot be fully accounted for by simply normalizing to the available surface area, as is done in the ice active site density (n_s) analysis framework. When the surface area is below the critical area threshold the retrieved values of n_s can increase significantly for the same particle species as the particle concentration is decreased. Above the critical area threshold the same n_s curves are retrieved when particle concentration is

changed. Atmospheric cloud droplets typically contain just one particle each. Therefore, this effect of particle concentration on droplet freezing temperature spectra and the retrieved n_s values has important implications for the extrapolation of cold plate droplet freezing measurements to describe the ice nucleation properties of realistic atmospheric particles.

Systems that probe populations of droplets each containing one particle such as the CFDC are unable to probe a large particles-in-droplet concentration range but are powerful tools for the real-time investigations of ice nucleating particles at the realistic individual particle level (DeMott et al., 2010; Sullivan et al., 2010; Welti et al., 2009). The frozen fraction curves produced from such an instrument do not provide enough information to associate the observed variability in ice nucleation ability to internal or external factors. However, future laboratory studies using the critical area cold plate technique we have introduced here (e.g. Fig. 4) will provide new insight into the critical area thresholds of internal variability in ice active site ability for different species. This will produce more informed assumptions regarding the variability in ice nucleation properties observed through online field instruments, specifically when the measurements are made in conjunction with single particle chemical analysis techniques (Creamean et al., 2013; DeMott et al., 2003, 2010; Prather et al., 2013; Worringer et al., 2015).

Atmospherically relevant particle sizes may very well fall below the critical area threshold for an individual particle, at least for some species such as illite mineral particles considered here. Therefore, average ice nucleation spectra or active site distributions such as n_s and $\bar{g}$ may not be applicable for representing the ice nucleation properties of particles in cloud and atmospheric models. However careful examination of the surface area dependence of ice nucleating ability of a species allows more accurate retrievals of active site density distributions that properly encompass this dependence.”

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