Response to Referee 1

The referee's original comments are in italics, and our response follows.

This work extends an immersion ice nucleation model to account for mixtures of different aerosol species present within the droplets. Freezing experiments are performed with Snowmax and illite and analyzed using the so-called critical area framework. The authors place emphasis on the influence of external vs. internal variability defining ice nucleation temperatures. They also investigate whether the freezing properties of an illite-Snomax mixture can be described using the contact angle distributions obtained from the individual species.

The correct representation of the immersion freezing of cloud droplets is critical for atmospheric modeling. New frameworks that account for the full range of variability observed in nature are certainly within the scope of ACP. Unfortunately the proposed model seems inconsistent with its own premises and may not lead to a better understanding of the freezing processes. The model assumptions are not clearly stated and the main equations are presented without much justification. The experimental procedure is confusing and lightly described. The paper is also difficult to read and follow. Everywhere there is superfluous information that does not seem relevant and that distracts from the main points of the work. My feeling is that a full overhaul of the paper and even of the underlying model and experiments may be required. At this point the work is not suitable for publication in ACP.

We thank the reviewer for their extensive comments that have helped us improve the clarity and conciseness of our manuscript. We think that much of the reviewer's concerns regarding the model and its expanded form can be addressed via clarifying important aspects in the manuscript under revision and by emphasizing and clarifying the formulation of our original model presented by Beydoun et al. (2016). As repeating what was presented in that original paper in this manuscript is not appropriate, we have strived to direct the referee to the relevant sections of the original paper to address their specific questions, and also clarified this manuscript to make it easier to understand how the model is constructed and used in this study. We note that many of the referee's questions were addressed in the original 2016 paper, while the focus of this new manuscript is not on the model's development and justification, but rather on its application to understanding the ice nucleation properties of Snomax, and Snomax-illite mixtures.

General comments

In general the paper is full with confusing and convoluted statements in places where simple sentences would suffice. The authors should consider reducing its length by removing repeated statements and unnecessary explanations. Also please be more precise. Many ambiguous concepts are thrown out without explanation and just add confusion to the text (e.g., activity, externally-mixed diversity, freezing capabilities, ice nucleation ability, ice nucleation critical temperatures, characteristic temperatures, droplet systems). Although I have tried to point out *some of them, I'd encourage the authors to put a lot more emphasis on simplicity and easiness of reading.*

Thank you for pointing out these issues that reduce the clarity of the manuscript. The wording in many places has been changed to avoid the overuse of terms that describe the same thing. This will be highlighted in the responses to detailed comments below. Heterogeneous ice nucleating (HIN) activity is consistently used to describe the ice nucleating strength of a particle type. External/internal variability are defined in a good amount of detail in Sect. 1.2 and are a measure of how the distribution of HIN activity distributes itself among a particle population. In the revised manuscript these terms have been omitted and replaced with other terms that are now clearly defined and used throughout the manuscript: ice nucleation ability, ice nucleation critical temperatures, and characteristic temperatures. We use the term "freezing temperatures" to describe the temperatures at which droplets freeze.

Please shorten the abstract and emphasize the main points. The first paragraph should be removed.

This suggestion is addressed in the response to the first specific comment below.

The model itself seems inconsistent. Particle sometimes refers to droplets with embed- ded aerosol, sometimes to the aerosol itself, sometimes to the droplets. The procedure used to obtain the distributions and each of the freezing curves must be much more clearly explained. It seems inconsistent that n_draws is treated as a fitting parameter. Normally as n_draws increases g→g however the authors manage to do just the op- posite: higher n_draws produce more variability and a further diverging g* from g. This casts doubt on the consistency of the model. It is also not clear that what is described in the paper is exactly what is done.*

The model assumes that for a given particle concentration, each droplet contains the same particle surface area. This is discussed in detail in the original description of the model published in ACP by Beydoun et al. (2016). Above the critical area, each particle surface is assigned a g distribution equal to $\bar{g}$. Below the critical area each particle surface is assigned a g distribution g^* which is generated from random sampling of contact angles from $\bar{g}$. This process is repeated for each particle surface so that below the critical area particle surfaces exhibit different g^* distributions. In the revised manuscript we have added more clarity and explanation of the original premises of the model but we emphasize the importance of looking at our prior publication by Beydoun et al. (2016) published in ACP for a more extensive explanation and analysis of the model framework. The reviewer states that we have increased variability with higher n_{draws}, but this is certainly not the case. For example, when sampling from the first distribution of HIN of Snomax the text states the following (Page/line 15/24-25):

"The consequent n_{draws} for each frozen fraction curve between the two Snomax critical areas are: 80 for $3x10^{-2}$ wt%, 65 for $1x10^{-2}$ wt%, 40 for $5x10^{-3}$ wt%, 25 for $1x10^{-3}$ wt%, and 6 for $5x10^{-4}$ wt%."

More specific details on this concern are further addressed in the responses to other specific comments below, along with the revisions we made to address these issues.

Everywhere: please use standard scientific notation.

We have switched to standard scientific notation. That certainly makes the manuscript more readable.

There is a complete omission on the effect of variations in the aerosol surface area per droplet in the droplet population. This is contradictory to the main premise of the model where some droplets contain enough material to be above the critical area and others don't. This is a major inconsistency and the authors should explain how it is resolved.

The issue of surface area variability between particles at the same concentration is an important issue that was addressed in the original description of our g framework in Beydoun et al. (2016). We found that surface area variability could not explain the changes in the droplet freezing temperature spectra as concentration was decreased. For a given concentration our model assumes all droplets contain the same surface area. The surface area varies with mass concentration. Thus there is no contradiction to the main premise of the model. We have clarified the explanation of the model in section 2.2 to better communicate the working premises of the model, as follows on Page/Line 9/1-20 and 10/1-11:

"In Beydoun et al. (2016) we started from classical nucleation theory to formulate a framework of heterogeneous ice nucleation which states that any given particle surface can be assigned a distribution of HIN activity, g. g is a continuous normal distribution of contact angles, θ (used as a proxy for the HIN activity at the particle surface-water interface), and determines the probability that a particle induces freezing in a droplet, P_f, at a temperature, T, via:

$$P_f(T) = 1 - \exp\left(-tA\int_0^{\pi} g(\theta)J(\theta,T)d\theta\right) \qquad (1)$$

where t is the time the droplet spent at the temperature T, A is the surface area of the particle, and $J(\theta, T)$ is the heterogeneous ice nucleation rate per unit surface area per unit time as defined in Zobrist et al. (2007). The freezing probability can also be written for a droplet undergoing a constant cooling rate $\dot{T}$ as:

$$P_f = 1 - \exp\left(-\frac{A}{\dot{T}}\int_{T_i}^{T_f}\int_{0}^{\pi} g(\theta)J(\theta,T)d\theta dT\right) \qquad (2)$$

For a large ensemble of droplets containing identical particle surfaces, the freezing probability is equal to the fraction of droplets frozen, F.

We hypothesized that an individual particle type possesses a critical surface area, above which one normal distribution of HIN activity $\bar{g}$ can accurately describe the freezing probability of a droplet containing any particle of that type. Therefore, the freezing probability of a droplet containing a particle with a surface area larger than this critical area can be evaluated using Eqns. (1) or (2) with $\bar{g}$ in place of g and the fraction of droplets frozen, F, of a large ensemble of these droplets N is equal to the freezing probability of each droplet, P_f. On the other hand, when a particle possesses a surface area below that type's critical surface area a random sampling of contact angles to generate a discrete distribution of HIN activity g^* is required for each particle surface in the particle population. In this case the frozen fraction of a large ensemble of droplets, F, is the arithmetic mean of the individual droplet freezing probabilities and can be evaluated using:

$$F = \frac{1}{N}\sum_{i=1}^{N} P_{f,i}\left(g^{*}{}_{i}(n_{draws})\right) \qquad (3)$$

where $P_{f,i}$ is the freezing probability of droplet i and can be evaluated using Eqns. (1) and (2) with g^* used in place of g and n_{draws} is the number of contact angle draws made from $\bar{g}$. The effect of decreasing an INP's total surface area below its critical surface area threshold on the distribution of HIN activity contained in each droplet is illustrated in Fig. 1 by the extensive broadening of the droplet freezing temperature curve when below the critical area. Above the critical area the diversity of HIN activity is essentially eliminated across the particle population; the large available surface area guarantees that the full distribution of HIN activity is contained in each droplet and described by□The creation of the g^* distribution by taking n_{draws} from $\bar{g}$ accounts for this increased diversity in HIN activity when below the critical area threshold."

The authors never seem to consider that classical nucleation theory (CNT) has very significant flaws, and the underlying physics may be wrong. This is particularly true for biological material that does not conform at all with the assumption of a gas-like nucleation process embedded in CNT.

The model uses the mathematical formulation of CNT and adds free parameters to predict heterogeneous ice nucleation behavior. This approach has been undertaken by many groups in the ice nucleation community (e.g. Alpert and Knopf, 2016; Broadley et al., 2012; Niedermeier et al., 2011; Welti et al., 2012; Wright and Petters, 2013) and has proven to demonstrate a good degree of success in modelling the HIN activity of particle surfaces. In all these formulations the traditional CNT framework is modified by incorporating some distribution of contact angles instead of a single contact angle, or a distribution of particle surfaces (the approach of Alpert and Knopf, 2016). In doing so, we are no longer testing freezing behavior against CNT as enough free parameters exist to act as correction factors to the assumed bulk properties embedded in the free energy terms. That is why, despite our model's successful modelling of the HIN activity of Snomax, we never claim that this success gives support or validation to the ability of CNT to predict the freezing behavior of biological material.

To avoid confusion, we have reworded the text introducing the model in Sec. 2.2 (Page/Line 9/2-3):

"In Beydoun et al. (2016) we started from classical nucleation theory to formulate a framework of heterogeneous ice nucleation which states that any given particle surface can be assigned a distribution of HIN activity, *g*."

It is not clear whether illite and Snomax are mixed within the droplets but still remain externally mixed. Did the authors prepare Snowmax/illite particles that were then suspended? Otherwise little can be said on the intermediate range of freezing temperatures.

The mixed suspensions were prepared by mixing the Snomax and illite suspensions, and the droplets were then generated from that mixture. The external mixture exhibited in the frozen fraction curve corresponding to this mixture suggests that some of the droplets do not contain Snomax ice nucleating particles and thus freeze according to the HIN activity of illite. We have added text explaining how the mixed particle suspensions were prepared in Sect. 2.1 on Page/line 8/20-24:

"Suspensions from which the droplets are generated are prepared by mixing a known mass of the sample (Snomax or illite) with a known volume of ultrapure Milli-Q water to form a suspension with a specific weight percentage of the sample in water. The droplets containing a mixture of Snomax and illite were generated from a suspension formed by first creating a 1:1 mixture from the pure illite and pure Snomax suspensions."

Specific comments:

Page 1, Line 6-15. This discussion just distracts from the main points of the paper. It is also irrelevant since the authors do not study the budget and distribution of INP in the atmosphere nor their effect on cloud microphysics.

It is not unheard of in a study on INPs to mention their importance in cloud microphysics and mixed phase clouds. Motivating the work should not be seen as a distraction from the main points of the paper. If the reviewer means to say that such motivating details shouldn't be in the abstract, then we believe that is a matter of personal preference and style. Our understanding is that a generous introductory/motivating paragraph in the abstract is permissible and often desirable. Furthermore, it is ACP editorial policy to encourage longer extended abstracts for their manuscripts, and to briefly introduce the topic and its importance in the abstract.

Page 1, Line 16. The abstract should start around here, clearly enunciating the main points of the paper.

Please see our reply to the previous comment.

Page 2, Line 3. This seems to imply that Snowmax is not active at low temperature, which I find hard to believe. Please clarify

We provide very strong evidence that the freezing happening at low temperatures is due to the presence of illite when analyzing a mixture of Snomax and illite. This is due to the excellent agreement between the part of the frozen fraction curve corresponding to freezing due to Snomax and the frozen fraction curve due to pure illite. Further we (along with many groups before us) have identified a temperature cut off below which no additional HIN activity is induced. Recall that as ice nucleation is a singular event that any ice active sites that can freeze at a warmer temperature will have already done so before the colder temperature is reached. Thus Snomax can certainly cause ice nucleation at colder temperatures, but it will have already induced freezing before this temperature is reached. Please see Wex et al. (2015) Fig. 11 – below -12 C the ice active site density levels off as all the Snomax INP's have already induced freezing.

Page 2, Line 5. This may be inaccurate. It seems that no Snowmax-Illite mixtures were prepared, and they remain externally mixed within each droplet. Please clarify.

Please see our response above on mixture preparations – each droplet was generated from a mixture of Snomax and illite particles. For the droplets generated from a mixture, we can be certain that all droplets contain illite INPs but not all droplets contain Snomax INPs – that is the topic of investigation here. This is deduced from the frozen fraction curves whereby one portion of the curve aligned with a frozen fraction curve corresponding to pure Snomax while the other portion aligned with a frozen fraction curve corresponding to pure illite. The droplets that froze according to the HIN activity of Snomax did contain illite – this is demonstrated in an experiment where the same illite concentration is used to generate droplets containing pure illite and the droplets freeze in a well-defined temperature range (see frozen fraction corresponding to 0.05 wt% illite in Fig. 5).

Page 2, Lines 20-24. This is a very convoluted statement. Please correct it and make it more readable. I'll suggest something like: "Very few studies have focused on this temperature range".

The statement on Page/Line 2/20-24 has been changed and now reads:

"This is a temperature range where investigated mineral samples of atmospherically relevant particle sizes (or other atmospherically relevant non-biological particles) have not exhibited detectable ice nucleating activity (Atkinson et al., 2013; Cantrell and Heymsfield, 2005; DeMott et al., 2015; Murray et al., 2012)."

Page 3, Line 17. The word "new" seems out of place here.

Page 3, Line 17 in the ACPD manuscript doesn't contain the word "new".

Page 3, Line 21-23. This is vague. The statement is complete without the g framework reference.

The *g* reference has been removed from the statement. It now reads (Page/line 3/24 and 4/1-2):

"These mixed particle systems also provide a valuable opportunity to evaluate the ability of HIN models to predict the freezing properties of mixtures using the properties obtained experimentally from the pure components."

Page 4, Lines 3-13. These lines do not seem to meaningfully contribute to the dis- cussion and make the paragraph overly long. Defining internal variability based on a "couple of degrees" Kelvin is also ambiguous. I'd suggest removing the lines.

We agree with the reviewer that those can mostly be removed. The paragraph now reads (Page/line 4/4-18):

"In Beydoun et al. (2016) the concepts of internal variability and external variability in HIN activity were defined and shown to have a dramatic impact on the temperature range a droplet population freezes in. If the same distribution of HIN activity is contained within each particle in an aerosol population (the internal variability), then the range of temperatures over which these particles cause freezing in the droplets they are contained in would be significantly different than if that same distribution was distributed externally (the external variability). Figure 1a illustrates the concepts of internal versus external variability. In the top right-hand panel of Fig. 1a each droplet contains a particle surface with a color gradient representing the distribution of HIN activity. Note that each particle contains the same distribution of HIN – demonstrating full internal variability – and therefore the range of temperatures over which droplets freeze is relatively narrow, as depicted in the bottom panel. As the surface area of the particle surface in each droplet decreases, the HIN distribution becomes externally variable, meaning that each droplet now contains a different distribution of HIN activity, which is shown with the different colors the particle surfaces are illustrated with in the top left hand panel of Fig. 1a. The reduced surface area per droplet leads to a reduction in the freezing temperatures while the increased external variability leads to a broader temperature range over which droplets freeze relative to the larger surface area case where the variability in HIN activity between each droplet was eliminated."

Page 4, Line 17. The range of temperatures is "narrow" only if the distribution is also narrow. It is not necessarily a measure of internal variability.

In the critical area framework, when particle surfaces are above the critical area threshold the variability in freezing temperatures is attributable to internal variability. This hypothesis was fully explored by Beydoun et al. (2016).

Page 4, Lines 19-20. This sentence is out of place.

The sentence has been removed.

Pages 4-5. Line 25. This is a confusing statement. Please split this sentence.

We have clarified the writing and the sentence now reads (Page/line 4/16-19):

"The reduced surface area per droplet leads to a reduction in the freezing temperatures while the increased external variability leads to a broader temperature range over which droplets freeze. This is in contrast to the larger surface area case where the variability in HIN activity between each droplet was eliminated."

Figure 1, caption. Most of this is already explained in the section. Please reduce this wall of text.

The figure caption should explain all schematics used within the figure so it is clear to the reader without having to consult the main text. As this figure is comprised of two parts and each part comprises multiple scenarios, it is not possible for us to shorten the figure caption while still achieving this goal. We have tried to make it a bit more concise by omitting unnecessary explanations of the scenarios the figure depicts. The caption now reads (Page 5):

a) Top, Right: Droplets containing particle surfaces larger than the component's critical surface area exhibiting internal variability (color gradient) in HIN activity. **Left:** Droplets contain particle surfaces smaller than the component's critical surface area and exhibit external variability in HIN depicted by the different colors. **Bottom:** Decreasing surface area leads to higher external variability in HIN activity and thus a wider range over which droplets freeze. **b) Top, Right:** Droplets contain a mixture of mineral dust (brown) and biological (green) particle surfaces, both of which are larger than their component's critical surface area. **Left**: Two hypothetical scenarios of reduced biological particle surface areas are depicted as the concentration of biological INP is reduced. In (2) the biological material distributes itself equally among the droplets and thus remains internally mixed while in (3) the biological material does not further partition itself further and becomes externally mixed. Scenario (4) represents the absence of any biological material and the resultant temperature freezing spectrum is identical to the one for dust lying above its critical surface area shown in the bottom of Fig. 1a. **Bottom:** The predicted impact of the INP concentration and mixing state of the dust-bio mixture on the resultant freezing spectrum of droplets containing the particle mixture.

Page 5. Line 3. This paragraph although somehow relevant is also distracting and a detour from the main points of the work.

We prefer that the reader is exposed to some motivation for the importance of internal/external variability after the concepts are explained and therefore have retained this paragraph.

Page 6, line 10. Does this mean that each droplet has a different distribution of freezing efficiencies?

We prefer the use of the term distribution of "HIN activity" over "freezing efficiency". What this means is that one distribution of HIN activity does not suffice to model the freezing behavior of such a system. The sentence now reads:

"If an aerosol population is chemically diverse and this diversity is externally distributed, then the freezing capabilities of the entire particle population would need to be defined by multiple distributions of HIN activity."

Page 6, lines 8-20. Again this is a very confusing paragraph and it is not clear what it adds to the discussion. Most of this was said in the previous page.

We respectfully disagree with the referee; this paragraph discusses chemical mixing state for the first time in the manuscript. We have therefore retained this section as it adds important new discussion.

Page 7, first paragraph. This is very repetitive. The whole paragraph can be reduced to a single question. I'd say: Is there an intermediate range of freezing temperatures when two dissimilar aerosol populations are present within each droplet?

Reducing the paragraph to a single question such as the one the reviewer suggests would eliminate the explanation embedded on how the freezing temperatures of the droplets containing the mixture relate to internal/external variability of the INP's. This explanation is also that of the right hand panel of Fig. 1.

Page 8, Line 11. Does this imply a freezing fraction uncertainty of 0.02?

If the uncertainty is defined by the resolution of the produced frozen fraction, then yes. This detail is now mentioned in the revised text on Page/line 8/8-9:

"A solution of ultrapure milli-Q water and the particle material being investigated was prepared, from which 40-60 (thus a 0.02 resolution in the retrieved frozen fraction)."

Page 9, Equation 1. The upper limit in the integral should be π. This expression indicates that the "g" distributions pertains to the droplet not to the particle. If the authors assume that each droplet has one particle, please explicitly state it. Also the integral merely represents the average nucleation rate within the droplet.

Yes the upper limit should be π, that typo has been fixed, thank you for pointing this out. The expression pertains to the freezing probability a droplet has if it includes a particle with a *g* distribution of HIN activity. The text now reads (Page 9/5-8):

"g is a continuous normal distribution of contact angles, θ (used as a proxy for the HIN activity at the particle surface-water interface), and determines the probability that a particle induces freezing in a droplet, P_f, at a temperature, T, via:

$$P_f(T) = 1 - \exp\left(-tA\int_0^{\pi} g(\theta)J(\theta,T)d\theta\right) \qquad (1)"$$

Regarding what Eqn. (1) represents, the reviewer is referred to the derivation of this expression in Beydoun et al. (2016), specifically Sect. 3.0.

Page 9 Line 7. This is only true for identical droplets. The authors also seem to neglect *variations in the surface area per droplet.*

That is correct, the model neglects variation in surface area per droplet. This is an effect we considered in our original paper on the *g* framework (Beyoun et al, 2016). The text has been revised to indicate that the frozen fraction is only equal to the freezing probability if the particle surfaces within the droplets are identical to each other (Page 9/13-14):

"For a large ensemble of droplets containing identical particle surfaces, the freezing probability is equal to the fraction of droplets frozen, *F*."

Regarding neglecting variations in particle surface area between droplets, this is explained in Beydoun et al. (2016). This is a relevant quote from Sect. 3.3 of that paper:

"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 (and thus one large particle) and a mean surface area value is assumed for the particle material in all the droplets in the array."

Page 9 line 8-12. Please be precise in the terminology. So far the freezing probability refers to droplets, not particles. The one-droplet one-particle assumption is also implied here.

The freezing probability should refer to droplets while the HIN activity should refer to particle surfaces. The revised text now reads (Page/line 9/15-20 and 10/1-12):

"We hypothesized that an individual particle type possesses a critical surface area, above which one normal distribution of HIN activity $\bar{g}$ can accurately describe the freezing probability of a droplet containing any particle of that type. Therefore, the freezing probability of a droplet containing a particle with a surface area larger than this critical area can be evaluated using Eqns. (1) or (2) with $\bar{g}$ in place of *g* and the fraction of droplets frozen, *F*, of a large ensemble of these droplets *N* is equal to the freezing probability of each droplet, P_f. On the other hand, when a particle possesses a surface area below that type's critical surface area a random sampling of contact angles to generate a discrete distribution of HIN activity g^* is required to produce the freezing behavior of a droplet containing this particle. In this case the frozen fraction of a large ensemble of droplets, *F*, is the arithmetic mean of the individual droplet freezing probabilities and can be evaluated using:

$$F = \frac{1}{N}\sum_{i=1}^{N} P_{f,i}\left(g^*{}_i(n_{draws})\right) \qquad (3)$$

where $P_{f,i}$ is the freezing probability of droplet *i* and can be evaluated using Eqns. (1) and (2) with g^* used in place of *g* and n_{draws} is the number of contact angle draws made from $\bar{g}$. The effect of decreasing an ice nucleant's total surface area below its critical surface area threshold on the distribution of HIN activity contained in each droplet is illustrated in Fig. 1 by the extensive broadening of the droplet freezing temperature curve when below the critical area. Above the critical area the diversity of HIN activity is essentially eliminated across the particle population; the large available surface area guarantees that the full distribution of HIN activity is contained in each droplet and described by $\bar{g}$. The creation of the g* distribution by taking n_{draws} from $\bar{g}$ accounts for this increased diversity in HIN activity when below the critical area threshold."

Page 9 lines 12-15. Please note that for large n_draws g should become g_average. Again the authors mix droplet and particle and it is not clear what the g distribution represents.*

Please see the previous comment regarding mixing the terms particle and droplet. HIN activity always refers to the particle surface while freezing probability refers to the droplet that a particle surface is immersed in. As for the reviewer's comment on the convergence of g* to $\bar{g}$ for large n_{draws}, we refer to Beydoun et al. (2016) where this is clearly stated in Sec. 3.3:

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

Page 10, Line 1. What is N?

In the revised text N is defined on Page/line 9/19. It refers to the number of particle-droplet pairings.

Page 10, line 6. Please specify what is meant by "saturated".

"Saturated" has been replaced with "eliminated" in the revised text.

Page 10, Eq. 4. Please justify this expression. There seems to be a contradiction since the critical surface area is defined per-droplet while here it is used as defined per-species. Please clarify.

The critical surface area was always defined per particle type or species, not per particle-droplet pair. We hope that the clarifications made in Sect. 2.2 now lead to a clearer description of the model premises and assumptions.

Page 11, Eq. 5. This expression only takes into account variation in contact angle, while it *assumes that every droplet contains the same surface area of each species. The model does not seem self-consistent (see main comments above).*

As mentioned in our response to the previous comments, variation in surface area between particle surfaces at the same material concentrations is neglected. Thus in Eqn. (5) each particle type has one value for a surface area.

Page 11, line 11. Change ''chapter" for Section.

The typo has been fixed.

Page 12, line 14. Here and in many other places. Please use standard scientific notation.

We have switched to using scientific notation throughout.

Page 12, line 14. How is the particle surface area distributed in the droplet population?

Please see our responses to previous comments regarding surface area variability between particles at the same material concentration.

Page 13, line 8. Again, the particle surface area inside the droplets is likely non- uniformly distributed and it is not clear what the authors are referring to.

The assumption of one value of particle surface area per material concentration should be clear at this point in the revised manuscript.

Page 13, line 15. Please clarify what the surface area correction is.

The surface area correction was meant to refer to using the same g distribution but with the estimated surface area value for this particular material concentration. The text has been changed from referring to a surface area "correction" to merely saying after accounting for the change in particle surface area (Page/line 13/16-18):

"The g_2 distribution that fits this low concentration freezing curve is a different Gaussian function than the g_1 distribution that fits the first two high concentration freezing curves (after accounting for the change in particle surface area)."

Page 14, lines 4-8. These values seem problematic. In CNT the nucleation rate is a monotonically decreasing function of the contact angle. So the fact that $\mu1 > \mu2$ even though the first population freezes at a higher temperature is contradicting the underlying physics. Secondly, the second distribution is essentially monodisperse, and it can't explain the enhanced variability.

The values are not problematic due to the fact that freezing by distribution 1 is dictated by the rising tail of the $\bar{g}_1$ distribution. The reviewer is urged to read Sect. 3.2 of Beydoun et al. (2016) where this is discussed in detail and the concept of a nucleating area is introduced. In addition, the second distribution is not monodisperse in the strictest sense of the word (a Dirac-delta function) but does have a finite sigma. This is mentioned on Page 19/16-18:

"However, it should be noted that the existence of some broadness in the temperature over which droplets freeze throughout the regime falling below the second critical area does necessitate that $\bar{g}_2$ still has a finite σ, albeit a small one."

Page 15, Line 10-15. This seems contradictory. Shouldn't the random sampling always lead to a different result? In other words what is the variation in F from the randomness introduced by the model?

Random sampling does lead to a different result for each particle surface, but when a frozen fraction is calculated as the arithmetic mean of each freezing probability in a large ensemble the variability is eliminated. We hope this is clearly outlined in Sect. 2.2 of the revised manuscript. A more extensive explanation is also found in Sect. 3.3 of Beydoun et al. (2016).

Page 15, Line 15. It is not clear what the authors mean by "optimization of the dynamic range of ndraws". Please clarify.

We do not say "optimization of the dynamic range of n_{draws}". A dynamic range isn't optimized. The original text says: "This optimization is carried out manually as the dynamic range of n_{draws} is small enough such that a more comprehensive numerical optimization (e.g. a Monte Carlo simulation) was not found to be necessary." Dynamic range here is referring to the range of values of n_{draws} that lead to different frozen fraction outcomes. Again, much of this is explained in detail in Beydoun et al. (2016). Nevertheless, we have reworded this sentence in the revised manuscript to remove any ambiguity (Page/line 15/15-17):

"The optimization of n_{draws} is carried out manually as the range of n_{draws} that results in changes to the modelled frozen fraction curves is small enough such that a more comprehensive numerical optimization (e.g. a Monte Carlo simulation) was not found to be necessary."

Page 15 Line 20. For the smallest concentration: separate draws should always lead to a different outcome. So how reproducible is this?

Please refer to the comment above on reproducibility.

Page 16, Line 5. This is equivalent to saying that g* is changing with composition and contradicts the premises of the model. For $\mu 1 = 0.62$ choosing $\theta>2.20$ does not repre- sent the distribution only its rightmost tail. In fact such a value is not even depicted in Figure 3. Also this value seems way too large for such efficient ice nucleating particles.

We actually choose $\theta > 0.22$, not > 2.20 as was mistakenly stated in the text. Please excuse a very problematic typo, which has been corrected now. The referee is absolutely right that a value of 2.2 for limiting drawing of contact angles represents none of the distribution.

Page 17, Lines 8-12. These statements also signal something troublesome about the model. Drawing 8000 samples from a normal distribution would for all practical pur- poses reproduce the original distribution. In fact at ndraws≈20 an acceptable repre- sentation of a normal distribution is already obtained, even more so for such a narrow distribution. In other words given enough draws a consistent model would always pro- duce g2*≈g2average. So the question is why are the curves so dependent on ndraws? Why is ndraws treated as a fitting parameter?

Yes, given enough draws a consistent model would always reproduce $\bar{g}$. The number of draws however is dependent on the width of the distribution. The reason $\bar{g}_2$ requires more draws than $\bar{g}_1$ is due to the fact that its narrower. As draws happen across the entire range of θ, the narrower the distribution the more draws are necessary to capture the distribution. For $\bar{g}_1, \sigma_1 = 0.05$, therefore the number of draws to reproduce $\bar{g}_1 \approx 3.14/(0.05) = 62.4$. On the other hand, for $\bar{g}_2 \approx 3.14/0.0001=3140$. This is why the number of draws from $\bar{g}_2$ is much larger.

n_{draws} is treated as a fitting parameter since it can create external variability between the modelled particle surfaces, particularly when drawing a small number of times. It's very important to note that the drawing process happens N times (number of particle surface-droplet pairs being modelled) so that a repeatable frozen fraction is retrieved. This frozen fraction is the average of all retrieved freezing probabilities as dictated by Eqn. (5).

Page 18-20. This explanation is very repetitive. Can these two pages be confined to a couple of sentences stating that there is overlap between the two distributions, so that the observed differences are dominated by σ instead of μ?

That is not the only message we are trying to convene in this part of the text. The section concludes with relating the wider $\bar{g}_1$ distribution to the finding of Polen at al. (2016) that the most active ice nucleants in Snomax are subject to degradation and a reduction in their activity over time. Nevertheless, the text has been made more concise and now reads (Page/line 19/19-24 and 20/1-11):

"The second notable feature in the contrast between the two Snomax distributions of HIN activity shown in Fig. 3 is that the mode of the less active $\bar{g}_2$ distribution ($\mu_2 = 0.52$) is actually at a smaller contact angle (i.e. more ice active) than that of the $\bar{g}_1$ distribution ($\mu_1 = 0.60$). This is not contradictory to the resultant freezing curves where the INP's represented by $\bar{g}_1$ cause freezing at several degrees warmer than those of $\bar{g}_2$. The freezing temperatures are actually determined by the ascending tail of the first distribution for the droplets containing high Snomax concentrations (dashed portion of the $\bar{g}_1$ curve shown in Fig. 3). The tail of the broader $\bar{g}_1$ distribution spreads into a lower (more active) contact angle range than the much narrower $\bar{g}_2$ distribution does. Further, the broader $\bar{g}_1$ distribution of HIN activity can be interpreted as being consistent with the finding in Polen et al. (2016) that the more active Type I protein aggregates (which are described by the first distribution, $\bar{g}_1$) are less stable over storage time and refreezes than the Type III protein aggregates (which are described by the second distribution, $\bar{g}_2$). A broader distribution implies that freezing is determined by a wider (more variable) range of HIN activity and thus a wider diversity of the INP's properties. The freezing induced by the more active $\bar{g}_1$ distribution is understood to be caused by rare and very active large proteins aggregates that would arguably be more susceptible to deterioration, weakening, and decay over time. This would produce a wider diversity in HIN activity, which would result in the broader $\bar{g}_1$ normal distribution of contact angles (Fig. 3) that we derived from analysis of the droplet freezing spectra in Fig. 2."

Page 20, lines 15-20. It is not clear how these mixtures are prepared. How is a mixture 1:1 but also 0.1% in illite and 0.00001 % in Snowmax? Are they mixed in the solid phase then diluted? What steps are taken to ensure that each droplet contains a mixture of the two components?

The preparation of the mixture solutions is now outlined in Sect. 2.1. No steps are taken to ensure that each droplet contains both types of INPs. The goal of the study is to explore the extent to which Snomax can partition its INPs so that upon dilution a contribution to freezing remains. It is seen that is not the case by the sharp cut off in freezing temperature between freezing caused by Snomax and freezing caused by illite.

Page 23, Lines 1-15. This paragraph does not seem relevant and it is quite confusing.

The paragraph was meant to discuss an academic thought experiment, in which the amount of illite surface needed to match the freezing capabilities of Snomax is calculated. To avoid distraction from the main points of the paper, it has been removed.

Page 24, Lines 1-5. The authors seem to be describing an externally mixed population of droplets containing Snowmax. For such a case the analysis presented here would not apply since the premise is that all droplets contain both species.

Droplets are generated from a mixture of Snomax and illite particles suspended in water. It is the intent of this study to explore what happens at low Snomax concentrations: does Snomax partition itself continuously and sustain its contribution to freezing? Or does Snomax limit its partioning and the droplets become externally mixed? The questions are asked clearly in Sect. 1.2 and explored in the experiments conducted and analyzed here.

Page 25, Lines 4-5. Please clarify what "doping illite. . . is limited by its ability to partition itself" means. Also only illite is investigated not every dust particle.

"Doping" here refers to Snomax INPs enhancing the HIN activity of the illite surface. Since Snomax is unable to partition its INPs at very low concentrations, the mixture becomes externally mixed and Snomax is unable to spread itself onto all illite surfaces. "Doping" has been replaced with "enhancing" in the revised manuscript.

That is correct, only illite is investigated here that is why the text explicitly mentions that the conclusions are drawn for this proxy mixture only: illite + Snomax.

The revised text now reads (Page 24/18-24 and 25/1-4):

"From the perspective of modelling multi-component heterogeneous ice nucleating particles, it remains a subject of further research to determine how many $\bar{g}$ distributions are necessary to encompass the freezing behavior of mixed particle systems at low temperatures where dust is known to be quite ice active. However, the work presented here supports the notion that two distributions of HIN activity are necessary to distinguish between the dust and biological regimes when illite and Snomax are used as proxies for these two types of INP. Regarding the motivating hypothesis that small biological particles can enhance the freezing capabilities of mineral dust surfaces, it can be concluded that the effectiveness of Snomax enhancing illite or any mineral dust surface is limited by its ability to partition itself and thus allow its HIN activity to be manifested externally. Hypothetical scenario 3, that was introduced in section 1.2 and depicted in Fig. 1b, in which the limited amount of biological material does not distribute itself among all droplets, therefore closely resembles the bio-dust mixture proxy examined here."

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