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Interactive comment on "A new multi-component heterogeneous ice nucleation model and its application to Snomax bacterial particles and a Snomax-illte mineral particle mixture" *by* Hassan Beydoun et al.

Anonymous Referee #1

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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.

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.

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

The model itself seems inconsistent. Particle sometimes refers to droplets with embedded 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 $g^* \to g$ however the authors manage to do just the opposite: 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.

Everywhere: please use standard scientific notation.

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 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.

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.

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.

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

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

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.

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".

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

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

Page 4, Lines 3-13. These lines do not seem to meaningfully contribute to the discussion 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.

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.

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

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

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

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

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

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.

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?

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

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.

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

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.

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.

Page 10, Line 1. What is N?

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

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.

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).

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

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

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

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.

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

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.

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?

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

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

Page 16, Line 5. This is equivalent to saying that g* is changing with composition and contradicts the premises of the model. For $\mu1 = 0.62$ choosing $\theta>2.20$ does not represent 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.

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 purposes reproduce the original distribution. In fact at ndraws≈20 an acceptable representation 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 produce g2*≈g2average. So the question is why are the curves so dependent on ndraws?

Why is ndraws treated as a fitting parameter?

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?

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?

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

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.

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.