This is an interesting paper where the authors apply a previously developed model to multiple component ice nucleating systems. They study ice nucleation by snowmax (which has multiple types of sites) and a mixture of snowmax plus nx illite. They con- clude that when mixed with illite the snowmax behaves much like pure snowax, which is important because this was not necessarily known (I mention this again below and I think the motivation for this study needs to be made clearer). The manuscript is rea- sonably well written, but I think the authors should try to be more concise wherever possible. In particular, use of scientific notation would make the manuscript far eas-ier to follow and must be corrected. In addition, I would like to see some discussion over the advantages/disadvantages of this method over singular descriptions. I support publication in ACP once the following specific points have been addressed.

We thank the reviewer for their comments and critiques. They have certainly helped us improve the message of our manuscript and clarified important aspects of the text. The referee's original comments are in italics, and our response follows.

P2., lines 18. How does 'analysis of ice crystal/precipitation sample residues' show that 'droplet freezing can occur at temperatures higher than -12 ˚C'?

In the cited studies, melted precipitation particles were collected and analyzed offline. The analysis typically involved forming droplets from the liquid sample and evaluating their freezing temperatures. Freezing at high temperatures (higher than -12 °C) was observed. We have reworded the text in the revised manuscript to make it more clear how the analysis works. The text now reads (Page/line 2/18-20):

"laboratory studies in which melted precipitation samples were refrozen (Christner et al., 2008; Petters and Wright, 2015; Vali, 1971, 1996) have shown that freezing can occur at temperatures higher than -12 °C."

P2, ln 20-25. 'higher than -12 C. This is a temperature range no investigated mineral samples of atmospherically relevant particle sizes (or other atmospherically relevant non-biological particles) can induce freezing in'. This is incorrect, there is a finite possibility of nucleation at these warmer temperatures. If the authors are referring to a situation in a cloud, are they sure about this? Very rare ice nucleation events seem to lead to secondary ice production, so is conceivable that mineral dust could account for a lot of very warm freezing.

The reviewer makes an excellent point. There is a finite probability of nucleating at higher temperatures given enough surface area of the material or given enough time (stochastic element of ice nucleation). We have reworded the statement to state that no investigated mineral samples of atmospheric relevant particle sizes have shown detectable ice nucleating activity in this

temperature range. In this revised statement, no assumptions of zero probability of freezing are made but the practical finding of mineral dust's activity at these temperatures is highlighted. The revised manuscript now reads (Page/line 2/20-22):

"This is a temperature range no investigated mineral samples of atmospherically relevant particle sizes (or other atmospherically relevant non-biological particles) have exhibited detectable ice nucleating activity in (Atkinson et al., 2013; Cantrell and Heymsfield, 2005; DeMott et al., 2015; Murray et al., 2012)."

P3. In the main para in which the study is justified, some of the concepts introduced by O'Sullivan et al. [2016] should be mentioned. One reason why it is important to study dust-bio INPs is that it is not obvious that the proteins responsible for ice nucleation should retain their ice nucleating ability when adsorbed onto other materials. There is a literature on the subject of binding of proteins to clay surfaces and examples of where the function of the protein is impaired when adsorbed.

The study of O'Sullivan et al. (2016) is extremely relevant to the research questions posed in this manuscript. Text has been added to the intro to describe the contribution of that study on Page/line 3/9-11:

"O'Sullivan et al. (2016) found that ice nucleating proteins from soil can bind onto kaolinite and retain their HIN activity, thus creating an enhanced ice nucleating surface on the dust particle."

The findings of that study will also be discussed and compared with the findings here in the discussion part of the manuscript (see response to final comment below).

Intro. Some discussion of competing models would be useful. For example, how does this approach contrast with that of Knopf and Alpert [2013]. This is a very different approach, what are the strengths and weaknesses? For example, it could be argued that these multiple component models are not based on a physical model since the distribution of sites is empirically fitted to data.

Also, what advantage does this approach have over the commonly applied ice active site density?

In our response to this comment we address the following referee comment as well:

Fig 2. If this data were plotted as nm (i.e. sites per unit mass) would the different curves fall on

top of one another? How does this data compare with literature data for snowmax? Also, does nm capture enough detail to reproduce these fraction frozen curves, or is nm flawed?

There are two parts to addressing these well-posed questions. The first is regarding the comparison between the single component version of this model (developed fully in Beydoun et al. (2016)) and other existing models. Section 3.4 in Beydoun et al. (2016) was dedicated to comparing our model with other existing ones. The n_s framework received very special attention in that paper, with comparisons of data in n_s or n_m shown throughout the whole manuscript. We argued in that work that n_s/n_m treatments to freezing data can lead to inconsistencies whereby the retrieved values don't overlap for different particle in droplet concentrations. We dedicated some of the analysis to the Snomax system in Beydoun et al. (2016) though it was only for four material concentrations and an old sample of Snomax that exhibited differences in HIN activity to the sample focused on in our newer study (see Polen et al. (2016) on Snomax INP degradation over time and storage). In that analysis of n_m spectra of Snomax we did find issues with value overlap, particularly a decrease in n_m for the same temperature as the mass concertation of Snomax in the droplets is decreased.

For the Snomax sample focused on in this study, we had made sure that its HIN activity was consistent with what other groups had reported. A comparison of the n_m values for all material concentrations with other groups is plotted on the right hand side of Fig. 3 in Polen et al. (2016) where it can be seen that the n_m values retrieved from the frozen fractions on the CMU cold plate lie within the envelope of n_m values summarized in Wex et al. (2015). We made sure this is mentioned in the manuscript on Page 8/19-21:

"The Snomax sample in this study is the same as that analyzed by Polen et al. (2016) whose freezing spectrum was shown to be consistent with the compilation of Snomax cold plate droplet freezing measurements summarized by Wex et al. (2015)."

It is typically found that retrieved values of n_m are within the same order of each other, so when plotted on a log scale to capture the full temperature spectrum as is done in Wex et al. (2015) the inconsistencies in n_m practically disappear. This does suggest that the ice active site density approach is not as problematic with Snomax as it is with NX illite or MCC cellulose (see Beydoun et al. (2016)) but depending on the level of detail one is looking to capture, the n_m approach is unable to consistently resolve the frozen fractions equally to the approach presented in this manuscript. It should also be emphasized that n_s/n_m is a diagnostic tool and is not used in a predictive fashion. n_m values are usually retrieved from the frozen fraction curves induced by a particle type and there is no effort to formulate for example an active site density distribution that can be extrapolated to different particle surface areas. In that regard, it is quite different than any approach that attempts to associate a contact angle or a distribution of contact angles to a particle type and attempt to predict the freezing behavior induced by this particle type for different particle surface areas. As for the approach of Alpert and Knopf (2016), a good deal of discussion on the differences between that approach and ours was presented in Beydoun et al. (2016) (see end of Sec. 3.4 in the referenced paper) where we had stated that surface area variability is an alternative approach to explaining the broadening of the frozen fraction curves.

The second part of addressing this comment is regarding the extended multi-component model developed here and its comparison to other approaches. We should have mentioned in the original version of the manuscript that the multi-component approach can be carried out by any of the other models as well. Equation (4), which states that the freezing probability of a droplet containing a mixture of particle types is the product of the independent freezing probabilities, can be developed using other formulations. However, we are unable to speculate about whether alternative treatments can succeed at predicting the behavior of the illite-Snomax mixture in the same way our approach has. We carry out a very distinct mathematical procedure in the random sampling of contact angles that has made it possible to model the transition from narrow to wide frozen fraction curves and capture the levelling of frozen fractions at low Snomax concentrations (see $1x10^{-5}$ to $1x10^{-7}$ wt% concentrations of Snomax in Fig. 2). From a mathematical perspective, the multi-component soccer ball model presented in Augustin et al. (2016) has as many free parameters as the multi-component model we present and therefore may be able to predict the behavior our model does. However, saying to what extent it can succeed would be overly speculative. In Broadley et al. (2012) the assumption of independent freezing probabilities was made to analyze a mixture of kaolinite and illite NX. The assumption did succeed in that study to predict the freezing behavior of the mixture.

Edits and additions to the revised manuscript were made regarding the second part of the response on multi-component models as the first part on the single component version has already been treated extensively in Beydoun et al. (2016). On Page/line 10/21-22 we have added a reference to Broadley et al. (2012) to indicate that the assumption of independent non-freezing probabilities was made there as well:

"It is assumed that every freezing probability is independent of the other, an approach similar to that taken by Augustin-Bauditz et al. (2016) and Broadley et al. (2012) for a mixture of birch wash water pollen and illite NX, and a mixture of illite NX and kaolinite, respectively."

On Page/line 11/17-19, we have added a statement saying the multi-component approach can be taken by any model of HIN activity and used to tests its validity to mixtures:

"It should be mentioned that derivations of and expressions similar to Eqn. (5) can be carried out using any of the other existing HIN frameworks starting from the assumption of independent freezing probabilities induced by each component."

Fig 1. The label (4) is missing from the images of droplets.

Scenario 4's droplet schematic is the same as the schematic on the top right hand corner of Fig. 1a, showing droplets containing dust surfaces larger than their corresponding critical areas. The revised part of the figure caption referring to scenario (4) should clarify this:

"Scenario (4) represents the absence of any biological material (Top right corner of Fig. 1a) 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."

P6. Top paragraph. The multiple component stochastic model described in Herbert et al. [2014] and Broadley et al. [2012] should be mentioned here.

The use of the word "component" in our study is used to describe a type of ice nucleating particle. For example, illite NX is comprised of one component while Snomax is comprised of two components. A single component possesses a distribution of contact angles and thus would be considered a multi-component surface under the definitions in Broadley et al. (2012) and Herbert et al. (2014). We realize now that this can be a source of confusion but hope that the definition of an ice nucleating component can clarify this to the reader. An explicit definition of a "component" in this study has been added on Page/line 7/9-10:

"In this study, a single component is defined as a particle type which can be treated as having one distribution of HIN activity."

P7 What are 'nucleation critical temperatures'?

In the original manuscript 'critical nucleation temperatures' was meant to describe the freezing temperatures induced by a particle's HIN activity. We have removed this term in the revised manuscript and merely use 'freezing temperatures of droplets' instead.

Throughout: The term 'ice nuclei' is used. I urge the authors to use the terminally set out in the Vali ACP 2015 definitions paper. 'Ice nuclei' is used in the context of classical theory in reference to the cluster of water molecules, not the particle on which the cluster is stabilised.

We agree with adopting the terminology suggested by Vali et al. In the revised manuscript we use the term ice nucleating particle (INP) to refer to the ice nucleating entities in Snomax and illite NX and no longer use the term 'ice nucleant'.

P11. Ln 15. Homogeneous nucleation can occur at a much higher T than -38 C. It is not that helpful to consider homogeneous nucleation occurring with a 'limit'.

The reviewer makes a very valid point. The homogenous nucleation temperature is a function of the droplet volume (Vali, 1996). The text has been changed to state that the droplets containing no particle material freeze well above their expected homogenous freezing temperature range. The text has been revised on Page/line 12/5-7:

"The latter process does not strictly proceed through homogenous ice nucleation and causes freezing to happen at a much higher temperature than the expected homogeneous freezing temperature range (Vali, 1996) of the 400-600 μm droplets used in this study."

P12, ln 14-16. Use scientific notation. In fact, use it throughout.

Scientific notation is now used throughout the entire text.

P23. Ln 13-15. Why is this an atmospherically relevant system?

In the original manuscript we described the system of droplets containing a mixture of illite-NX and very small Snomax concentrations because if a mixture of these two components did exist in the atmosphere it would in mostly likelihood contain a much larger mass percentage of dust than biological material. However, this does not make the system atmospherically relevant in the strictest sense of it actually being found in the atmosphere. Therefore, we have removed this description in the revised text.

P26. In the discussion of whether the snowmax-illite system is a 'close proxy to real atmospheric bio-dust mixtures', O'Sullivan et al. [2016] should be discussed. They argue that in soil particles will adsorb ice nucleating macromolecules. Soil borne fungus can shed its ice nucleating proteins into water and these proteins apparently bind to clay particles. This represents a distinct scenario of bio-dust mixtures compared to mixing snowmax with illte.

These are excellent points. We have added the findings of O'Sullivan et al. (2016) to the last paragraph of the discussion. We think this addition complements our findings quite nicely and motivates future work for investigating bio-dust mixtures. The text has been revised on Page/line 24/23-24 and Page 25/1-10:

"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. This is in contrast to the more effective behavior of soil borne fungus investigated in O'Sullivan et al. (2016). The biological macromolecules in that study were shown to bind onto clay and thus partition themselves among the dust surfaces, thereby distributing their HIN activity externally. This difference between Snomax and soil borne fungus should be kept in mind when considering the relevance of dust-bio mixtures in the atmosphere, and future work can attempt to quantify whether the ability of soil born fungus to adsorb unto clay surfaces can render it a stronger INP than Snomax despite its weaker HIN activity."

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