Review

for the manuscript:

Immersion mode heterogeneous ice nucleation by an illite rich powder representative of atmospheric mineral dust

by

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submitted to
Atmospheric Chemistry and Physics - Discussion

The authors present in this paper immersion freezing measurements with NX illite particles. This material is characterized thoroughly and it is well motivated that this material is a good surrogate for atmospheric dust particles. The measurements described here are extensive in the variations of parameters like wt% of material within a droplet or cooling rates which is very helpful to constrain theoretical approaches or parameterizations to fit the data. In the following sections a variety of models and approaches are tested with the so called multicomponent model being extensively discussed in all possible consequences. While this is a thorough work I consider the amount and choice of material presented here as too broad for a single paper. In the end, I am missing a clear take home message because all model approaches discussed here have their weaknesses which however the authors correctly address. In summary, the amount of models/parameterizations seems a bit overwhelming to me while other approaches which have been shown to describe ice nucleation data well are not considered here. I recommend the paper for publication but with revisions and possibly considering a re-organization of the manuscript or splitting it in two parts, one with a focus on measurements and material characterization and one which focuses on parameterizations and models.

Some detailed comments:

Abstract:

p. 22802, line 8: Please add a short not about the usage of ATD as reference material for IN studies.
-, line 10: Heterogeneous nucleation <u>in the immersion mode</u> by NX illite...
-, line 14: … in terms of <u>their</u> ice …
-, line 16: … than assumed in <u>a</u> parameterisation …
-, line 26: … is assumed <u>that</u> there …

1 Introduction:

p. 22803, line 8: … of solid particles <u>termed ice nuclei (IN). These IN</u> are rare …
-, line 21: … about a third of <u>all</u> IN ….
p. 22804, line: 3: … condensation of <u>liquid</u> water …
-, line 18: ….was found <u>to</u> be ….
-, line 19: … found <u>that</u> there was …

2 Theoretical background:

This section is very clear and a good summary of the concepts needed to understand the folowing data analysis. I only suggest to use A instead of S as a symbol for surface area in formulas 4, 6, 7, and 13, 14 because A is normally used for areas and S is often used in CNT for saturation ratios and can therefore be misinterpreted.

3 Experimental

p. 22811, line 25: It would be helpful to also give a typical size (mean and variance) of the droplets used. How does this and the wt% concentrations translate into a number concentration of particles/droplet? Are there experimental conditions met, where it is likely that on average less than one particle is present in a droplet? See also my comments on sections 4.1 and 4.3.

4 Results and discussion

In general I have been confused many times with the experiment numbers. I recommend to re-name them to either numbers+letters (e.g.1a) or put at least a dash between roman number and extension (vi-a). But personally I think it is much easier to read a scheme like 1a or 1-a.

4.1 heterogeneous freezing temperatures
The fact that some droplets with low wt% concentrations of illite froze homogeneously brings up the question if in these experiments some of the droplets did not contain a particle at all. Please discuss this possibility as already suggested for section 3. Fact is that surface area is not distributed continuously but in discrete amounts (by adding particle by particle into a droplet). This is especially relevant if you come close to an average concentration of one particle per droplet (please consider a poisson statistical distribution here) Maybe this might explain some of the observations here (and in section 4.3). This is also a point to consider when the data is interpreted with atmospheric relevance in mind. In the atmosphere, normally exactly one particle is present in one droplet, here this can vary largely.

4.3 High/low surface area regime.

I am not so happy with this rather arbitrary distinction between low and high surface area regimes. If this is something real a possible scientific reason for this distinction should be discussed. One of them might be related to the points I made before regarding the statistical number concentration of particles within one droplet. On the other hand, it should be tested, that the change in median freezing temperature and its leveling at higher concentrations might not be explained and described by a continuous function in agreement with predictions made by the CNT based models in this study. In other words, are these two regimes really two different regimes or just different regions in a continuously changing function?
p. 22818, lines 15-20. I am not sure if the data in Figure 4 supports the conclusion that only in the lower regime the data is surface area dependent. It may also be surface area dependent in the other regime just less pronounced, especially when considering the error bars.

4.4 model fits

In figure 6: Do the error bars for surface area consider the statistical implications (poisson distribution of individual particles) discussed above?

4.5 isothermal experiments

no comments for this section.

4.6 multicomponent model

Th discussion in this section is very interesting and I see that this approach may be used to fit experimental data well. However, it comes without a good connection to the other approaches discussed earlier and without a connection to the theoretical section 3 as far as I can see. The question comes up because other authors have used approaches where a distribution of contact angles is assumed. Since this is a very lengthy part of the paper with alone 7 Figures – which are relevant and interesting indeed – I just question if this isn't too much material for one paper. If the authors intend to promote and characterize this model then I would make this more clear from the beginning or possibly put this material in a companion paper.

5 parameterization for models

Given that so much effort in this paper is put into the previously described models I am surprised to see here again another approach to fit the data of the presented experiments.
p. 22826, line 4-6: I cannot follow the argument that the empirical multicomponent model of section 4.6 should be the most physically accurate one. If so the authors should describe more clearly how this model is related to the physics of nucleation which are described very clearly in the theory section of the paper. I hope I am not overseeing something obvious here.

6 conclusions

This sections highlights that each model discussed here seems to only fit well with a certain fraction of the data. Since the authors discuss some models used in other studies I cannot follow the motivation to selectively ignore those models which fit the data best in those studies (e.g. distribution of contact angles and active site model by e.g Marcolly et al. 2007 and Lüönd et al. 2010) while introducing the multicomponent model as a model with similar assumptions but without the physical basis (they are fully based on CNT). A comparison between these models and the multicomponent model presented here would have been much more interesting and could also be better motivated since the "simpler" models have already been shown to not fit well to experimental data sets in several studies.