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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Comparative Study On Immersion Freezing Utilizing Single Droplet Levitation Methods" *by* Miklós Szakáll et al.

Anonymous Referee #2

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1 Introduction

In this work the authors use single droplet levitation techniques to analyze the freezing behavior ice nucleating particles (INP). Both isothermal and non-isothermal experiments are performed over a wide range of materials and conditions of atmospheric interest. The authors perform a detailed analysis of their experimental conditions, and of the ice nucleation behavior of the different materials. As extensive, single-droplet ice nucleation data are scarce, I find the data set is of interest to the atmospheric community. The analysis methods are however confusing and not appropriate for the type of experiments performed. They must be thoroughly clarified before the work could be published.

2 Main Comments

The analysis method defeats the purpose of carrying out single-droplet experiments. The authors interpret their results in terms of a previously developed model, appropriate for cold-stage type experiments, where the number of frozen events is counted out of a droplet population. It is certainly possible to relate the single-droplet and the population experiments (using a number of assumptions that must be clearly stated). But one wonders whether this is the best use of the single-droplet data. In the latter each of the analyzed freezing events is completely independent, and expressions like Eq.(1) (which is the same as Eq. 5) are not directly applicable. Instead one may expect that the statistics follow the usual Poisson distribution and be analyzed as such (the difference would be notorious when analyzing the non-isothermal experiments). Thinning of the distribution would help elucidate the presence of multiple components as well. Without attempting a more fundamental analysis, in terms of the actual statistics of single droplet events, it is hard to see what is new in this work.

The authors invest considerable effort to describe their experiments in terms of the active site density (INAS). However their results scream on a different direction: that the INAS approach is not appropriate to parameterize ice nucleation. Clearly time-dependency, particle-to-particle, and droplet-to-droplet variability must be accounted for. Looking at their results, one would expect the authors to call for a reanalysis of all of the ice nucleation data reported over the last decade. Instead, they go through considerable length trying to force the data into a flawed description of ice nucleation. This is a disservice to the scientific community and only works to perpetuate existing biases in the description of ice nucleation.

The suggestion that isothermal experiments should be analyzed with a time-dependent model whereas a time-independent model should be used in experiments with varying temperature is wrong. If the authors are trying to elucidate the fundamental nature of ice nucleation, this should be independent of the analysis method. As mentioned above the time-independent formulation is at best a crude approximation hence and a time-dependent formulation should be emphasized.

There is nothing in the way the analysis is conducted that would suggest the existence of multiple components in the analyzed Kaolinite and sample. Fitting to a highly empirical model is not a sufficient condition, and given the results it suggests limitation of the assumed empirical model rather than a fundamental feature of the nucleation process. The authors suggestion that two distinct INP types would lead to a uneven distribution of nucleation efficiencies in the droplets (hence $\omega \neq \lambda$) is not supported by the isothermal experiments since this would also lead to two different slopes in Figure 6, and likely to a departure from the Poisson-type behavior.

It is also not clear what the authors mean by multiple component. The empirical correlations obtained in the Herbert et al. (2014) assume a normal distribution of the “b” parameter which amount to a collection of different nucleation sites. (hence different components?) The λ parameter seems more related to the way the INP are dispersed in the droplet population hence it is just a feature of the experimental setup.

The authors state that they would make the data available upon request. This is appropriate during the review process. However to allow independent scrutiny the data supporting the plots must be placed on a permanent public repository before final publication.

3 Other Comments

- Line 27. This is not the nucleation rate.
- Line 32. Maybe another reason is that in the dry suspension method there is one particle per droplet, while in bulk measurements many particles are immersed within the same droplet. Hence there maybe slight differences in the environment around each active site.
- Line 90. The stochastic approach is also T-dependent. Please rephrase.
- Line 106. All sites must be equivalent as well.
- Line 127. I am having a hard time seeing any difference between this expression and Eq.(3). Is there anything here beyond semantics?
- Line 177. What is the basis to mix singular and time-dependent processes here? It would seem that they must be mutually exclusive.
- Line 186. This is not obvious at all. Please state clearly why this model is used, out of the many empirical approaches available.
- Line 189. Do you have to repeat the whole analysis if a different nominal cooling rate is used? Atmospheric cooling rates vary widely.
- Line 274. There is nothing preventing doing this analysis with the time-dependent model. In fact it would be a more rigorous approach.
- Line 311. It is not clear why binning of the results was required.
- Line 328. How do you go from INAS to the cumulative frozen spectra? What are the assumptions involved?

- Line 335. Please clarify this. Time is still involved even if you decide to ignore it.
- Line 345. Do these imply that the singular approach is invalid?
- Line 403. Still it seems that this would alter the temperature history of each active site.
- Line 409. I find this section very hard to follow. In fact I can't make sense of Figure 10. The authors go through a lot statistical dredging to try to fit the Herbert et al. (2014) model. The conclusions seem very dependent of the procedure used. There is hardly anything fundamental that can be extracted, especially not about multiple components.
- Line 465. If it is not applicable, why are the authors heavily using this model?
- Appendix. There is a lot of non well-behaved data that actually seems way more interesting than Kaolinite.

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