

We thank both reviewers for their useful comments and suggestions that helped us to improve the manuscript.

We hereby reply to the questions and comments of Reviewer 2 in detail.

Major comments

- I. 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).

One has to distinguish between the two single-droplet levitation techniques we used. The isothermal (M-WT) measurements can be described by the stochastic approach. For the cooling experiments of M-AL the singular approach, which is also used for cold stage array experiments, can be applied. The droplet freezing in cold stage experiments is usually also considered as completely independent, and Eq. 1 is applied. In our experiments we use the same samples, generate the drops in a similar way as in cold stage experiments. Each drop in M-AL are cooled down similarly, although not identically. But that might be the case on a cold stage, where temperature differences on the surface can result in different drop temperatures. We definitely relate one drop temperature to another. In this way, M-AL and cold stage measurement are comparable in terms of f_{ice} and n_s .

- II. Thinning of the distribution would help elucidate the presence of multiple components as well.

There are many possible sources for the presence of multiple components. One important is the size distribution. Other sources are the internal and external mixture of the sample material, or any physical, chemical, or biological contamination. In summary, in an ideal experimental case, a very pure INP material with thin size distribution would be immersed in a droplet under investigation. Furthermore, the total surface area is a crucial parameter. In order to use the correct value of the total surface area in the calculation, it should be measured directly inside the droplets. That would help avoiding error originating from aggregation in aqueous solutions, for example. Unfortunately, our experimental setup was not sufficient to carry out such measurements. Instead, our instrumentation allows the investigation of the freezing process under conditions that are more realistic, like the free levitation in an airflow, for instance. We believe that our measurements can help researchers making predictions for real atmospheric processes when utilizing results from, e.g., cold stage experiments.

- III. 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.

INAS is a concept emerging from experiments and broadened in the community due to its easy implementation into cloud models. Our aim was to contribute to the justification or disproof of its usage in applications describing cloud processes. Our setup simulates the cloud conditions in a much appropriate way concerning the flow, shape and contact-free levitation. Therefore, we converted our measured data to INAS density. Nonetheless, we also provide results of the heterogeneous nucleation rate coefficients from our measurements under isothermal conditions.

- IV. 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.

Ice nucleation is a stochastic, i.e. time dependent process. The stochastic approach is based on classical nucleation theory and represents a physical description. The singular description is an empirical approach, which was introduced to explain ice nucleation in a simplified manner. The temperature dependence is neglected as it is assumed that critical clusters form on ice-active sites at characteristic temperatures. The singular approach has been used to compare the results from different experimental techniques via the n_s values (e.g., Hiranuma et al., 2015; Wex et al., 2015). Furthermore, the singular description can be easily implemented in cloud models (e.g. Diehl et al, 2015, ACP). If the λ value is small, a large temperature shift is predicted in accord with the stochastic model (s. Theoretical background in the revised version of our manuscript). In the contrary, large λ values result in small temperature shifts, which are in most cases negligible at least in terms of the measurement uncertainties. In these cases, the application of the singular approach is rather justified. Thus, our study helps to elucidate the limitations of the singular approach used by dozens of experimental studies by providing experimental data of λ for a set of INP. We critically revised our paper to avoid any misleading formulation regarding this topic.

- V. 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.

The formulation in the manuscript was probably misleading. We used the term multiple component because Herbert et al. applied this approach. In this context, multiple component may mean internally or externally mixed particles, contaminations, etc., but also the high scatter of contact angles, or the INP surface area variability in the individual droplets. Hence, the effect of particle variability is more important than the time dependence of nucleation. For single component systems the stochastic model has to be applied, whereas for multiple components the singular approach may also be valid. By revision of the manuscript we payed attention to address this issue.

- VI. 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 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.

Please see our reply to the last comment. Of course we cannot completely rule out the effect of our sample preparation or experimental features. That was one reason why we described our procedures in details, and carried out statistical tests on analysis results. Future studies may clarify this.

- VII. 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.

We are not sure what the reviewer’s request is. We can certainly publish the data points and errors shown in the figures before publication, in case the manuscript will be accepted. Publication of raw experimental data on a permanent public repository will follow after final publication.

Minor comments

1. Line 27. This is not the nucleation rate.

Yes, thank you, we corrected it into the nucleation rate coefficient.

2. 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.

Yes, we absolutely agree. Since this issue is well beyond the scope of the present paper, and because we do not possess the instrumental possibility to adequately study this discrepancy between dry dispersion and aqueous suspension techniques, we did not speculate on the reason for it. The main message we want to pass over here is how important the rigorous examination of the limitations of one's measurement technique is (which should actually be evident, but in fact is often not the case). Furthermore, this part served for orientation for the reader that the paper deals with only one type of immersion freezing measurement methods, namely the aqueous suspension technique.

3. Line 90. The stochastic approach is also T-dependent. Please rephrase.

The temperature dependency of the stochastic approach is indicated.

4. Line 106. All sites must be equivalent as well.

Yes, thank you, we corrected the sentence.

5. Line 127. I am having a hard time seeing any difference between this expression and Eq.(3). Is there anything here beyond semantics?

In Eq. (5) time dependency is not included (singular approach), while in Eq. (3) both time and temperature dependency are involved (stochastic approach).

6. Line 177. What is the basis to mix singular and time-dependent processes here? It would seem that they must be mutually exclusive.

We reconstructed the entire section on the theoretical background, and reformulated the approaches in a more consistent and clear way.

7. Line 186. This is not obvious at all. Please state clearly why this model is used, out of the many empirical approaches available.

Please see our reply on the last comment.

8. Line 189. Do you have to repeat the whole analysis if a different nominal cooling rate is used? Atmospheric cooling rates vary widely.

Yes, a cooling rate differing from 1 K/min would cause a different freezing temperature shift. This has to be counted for when comparing measuring devices, but also has to be taken into account in cloud models.

9. Line 274. There is nothing preventing doing this analysis with the time-dependent model. In fact it would be a more rigorous approach.

The primary goal of the current study was the investigation of the temperature shift in cooling rate experiments. The isothermal conditions in M-WT represented the physical basis allowing to interpret the experimental results using the time-dependent model. The M-AL measurements provide INAS densities utilizing the singular approach. Since this instrument exhibits high and varying cooling rates, the implementation of the time-dependent model was first abandoned and we restricted our analysis to the singular approach.

10. Line 311. It is not clear why binning of the results was required.

Binning means in M-AL experiments the counts of individual drops frozen in temperature intervals between $T-0.5$ K and $T+0.5$ K. This was necessary because the measurement setups has a temperature uncertainty of ± 0.5 K.

11. Line 328. How do you go from INAS to the cumulative frozen spectra? What are the assumptions involved?

The sentence was corrected, and now reads “Figure 5 shows the INAS densities computed using Eq. (5) from f_{ice} spectra obtained from M-WT measurements of kaolinite (...)”

12. Line 335. Please clarify this. Time is still involved even if you decide to ignore it.

Yes, time is involved, however, using fixed times of 30 s in M-WT experiments we expressed n_s in terms of f_{ice} . Herbert provided an equation for calculating the time an isothermal experiment needs to reach the same frozen fraction as a cooling rate experiment (Eq. 19 in Herbert et al., 2014):

$$t = \frac{1}{\lambda \cdot r}$$

with r being the cooling rate. Assuming $\lambda=2$ and using the standard cooling rate of 1 K/min, $t = 30$ s. In this approach, the stochastic element is considered to represent the random occurrence of an ice nucleating site somewhere on the INP surface (see Vali, 2014, Eqs. 12 and 13).

13. Line 345. Do these imply that the singular approach is invalid?

The droplet freezing in the M-WT can be described by the stochastic approach. In order to compare cooling rate and isothermal experiments, we took the accumulated data for the total observation time of 30 seconds of the isothermal measurements into account for calculating f_{ice} .

14. Line 403. Still it seems that this would alter the temperature history of each active site.

Yes, we agree. As described in the manuscript, we believe that the huge amount of kaolinite particles, and hence, of the active sites, is so large that the warmer temperature in the drop interior does not play any role in freezing initiation. Furthermore, the temperature history lasts not more than 80 seconds in M-AL experiments, which might be too short for significantly affecting the nucleation ability of active sites.

15. 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.

We decided to move Figure 10 to the Appendix. Our intention was to depict the procedure and help the reader to understand the process, but apparently it confused both reviewers. We added the variables to the list of variables.

The aim of this section was to introduce the procedure for calculating λ , and to estimate whether it differs from ω . This implies whether the INP can be described by single component stochastic approach or not. Unlike Herbert et al., we investigated whether λ equals ω in terms of our measurement uncertainties. In this regard, the conclusions depend on the procedure and instrumentation used.

16. Line 465. If it is not applicable, why are the authors heavily using this model?

The Herbert et al. approach served as basis for our analysis. Since Herbert et al. used constant cooling, their concept had to be modified and adapted to our experiments in which the cooling rate was varying. In addition, we improved the analysis method by introducing the procedure considering data scatter and measurement errors.

We found some noticeable behaviour of our INAS density results from M-AL immersion freezing measurements when compared to other techniques and devices within the INUIT and FIN02 campaigns. We could observe an apparent temperature shift in our results, the shift being different for different materials, and freezing temperatures. We were trying to figure out whether it was a measurement artefact or a phenomenon with a physical basis. This is how we came to the Herbert approach, which adequately modelled our experimental findings.

17. Appendix. There is a lot of non well-behaved data that actually seems way more interesting than Kaolinite.

We chose kaolinite to demonstrate the procedure we used to analyse our experimental data. Kaolinite was also analysed in several immersion freezing studies, as in Herbert et al., for instance. Nevertheless, we agree that there are other interesting data in our dataset, and we are happy that the reviewer agrees with that. We are going to share the measurement data in a scientific repository for other researchers, as mentioned earlier.