

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 1 in detail.

Major issues

- I. First, there is insufficient review of previous studies that makes the manuscript unbalanced.

Descriptions of specific previous studies and their main claims using isothermal and cooling rate conditions are not included, but must be in order to be fair and balanced. Herbert et al. (2014) is extensively referenced for its data analysis, however it must be mentioned that they performed isothermal and cooling rate experiments on same particle types. Older studies such as Vali (1994) have cooling rate and isothermal conditions in a single experiment and support a multiple component approach held by the authors. Other studies, such as Alpert and Knopf (2016) and Knopf et al. (2020) come to a very different conclusions than the authors of the manuscript, but also used isothermal and cooling rate experiments. In summary, the literature review is incomplete and must be modified to include these references and any other relevant studies the authors wish to include.

Thank you for this note, we admit that the literature review was unbalanced. We oriented ourselves study to the widely used freezing assays that perform cooling rate experiments and apply the singular approach. In the revised version of the manuscript, we included also investigations where the authors applied stochastic models.

- II. Additionally, assumptions about surface area and the impact of surface area calculations on their conclusions are not stated.

On I. 113, the authors state that in the case of interparticle variability, Eqn 3 cannot be used and J_s is modified (Eqn 9) as a result. Eqn 3 also has surface area, S_p , which has its own uncertainty and variability. What is the estimated error on S_p and how does this error impact their findings? It is important the authors claim that the surface area in each droplet was not directly measured (only calculated from Eqn 4), and so the authors cannot rule out that droplet to droplet variability may be more than they expect.

Along the same lines, the authors choose to vary cooling rate, however, varying surface area could also be done to test for single or multiple components. For example, Hartmann et al.

(2016) changed particle mobility diameter from 0.3- 0.7 μm and found similar n_s values for kaolinite. It would follow that kaolinite would be considered as a single component particle type under sizes relevant for the atmosphere, in contrast to this study in which the authors labeled kaolinite as multiple component. In light of the work of Hartmann et al. (2016), I would ask the authors to include some discussion about changing particle size and the impact on what could be declared as multiple or single component. The authors should state the upper size limit of single particles in their droplets for each type.

The actual concentration of particles in the droplets was not measured. The particle surface areas used for the calculations in the paper were calculated from the particle concentration in the droplet, the droplet volume, and the specific surface area of the INP. We considered all measurement error sources for calculating the propagated error. In order to reduce experimental uncertainty, a homogeneous solution was generated and used for droplet generation. Although efforts were made to unify and standardize the sample generation (also following the suggestions of Hiranuma et al., 2018), we cannot rule out INP surface area variation among the investigated droplets. That can significantly influence the nucleation description (Alpert and Knopf, 2016). There are several sources of error which might increase the surface area uncertainty, like: externally or internally mixed particles, size distribution of the particles, aggregation due to sedimentation and internal circulation. The most appropriate way would be the continuous measurement of the surface area inside each droplet under investigation, but that seems not feasible currently. Furthermore, the ice active site densities may vary on microscopically identical (i.e. size, chemical composition) particles.

We varied the total surface inside the droplets by using different particle concentrations in aqueous solutions. Such experiments resulted in consistent n_s values (see Fig. 5.). Unfortunately, we cannot provide size limits of the particles we used. We used bulk particle samples and in a relatively high concentration. Furthermore, aggregation in an aqueous solution would anyway modify the dry particle size distribution. We discussed these points in the revised manuscript.

- III. There is a fundamental flaw with shifting observed temperatures outside their investigated range.

Applying a temperature shift is fundamentally flawed. For example, the warmest observed temperature that feldspar nucleates in this manuscript is about 263 K. However, the temperature correction is made to 269, which is 5 K warmer than measured. The authors did not measure nucleation at this warmer temperature. This is extrapolating data outside of the observed temperature range. Why don't the authors shift n_s instead? After all, temperature is measured. n_s is not measured, but it is calculated from measurements. This is a caveat that must be claimed and the reader must be warned about using data outside of the authors measured temperature range.

The temperature was shifted to higher values but remained still within the investigated range. The droplet injected into M-AL was adaptively cooling from some positive degrees to below -25 °C. The surface temperature was continuously measured by means of an infrared thermometer. Therefore, all freezing events that occurred at temperatures between 0° and -25 °C were captured. In the case

of the M-WT measurements, the necessary temperature shift was small and within the wind tunnel air temperature variation.

We observed a systematic offset of our data points in intercomparison campaigns (INUIT and FIN02). For some particle types this offset was obvious, and for some it was negligible, i.e. within the measurement error. By seeking for the reason of this offset, first we checked the calculated n_s values and possible errors in the calculation or in the sample treatment. However, since the offset appeared for different concentrations, and also when treating the samples following the experimental protocols of the campaigns, we investigated the shift in the temperature. Finally, we made measurements on the internal temperature of the droplets, before we arrived to the temperature shift caused by the change in cooling rate.

- IV. Finally, it must be acknowledged, that their conclusions about a temperature shift or a particle type being single or multiple component is dependent entirely on their choice of data analysis procedure and not a fundamental property of the types of particles.

Another major concern is that the formulation here and in Herbert et al. (2014) is purely empirical and not based on any physical theory. Therefore, a particle type declared by the authors as a single and multiple component, or having a material dependent temperature correction factor (shift) is also empirical and not a fundamental property. This is a necessary caveat that must be stated. I do not wish to discount the evidence the authors give for either, however, they do need to stress that there is no direct observation of single or multiple components.

We corrected the formulation regarding the classification of particles as single or multiple-component. Instead, we stressed that the freezing behaviour of the particles can be described by a single or a multiple-component approach. Furthermore, we did not aim to develop a new model or framework for immersion freezing, but rather to provide new experimental data to check whether they can be described by the existing approaches, and how the results obtained from two experimental techniques match. The main motivation of the study was the freezing temperature shift observed in our M-AL measurements during intercomparison campaigns (INUIT and FIN02). The deviations often visible in intercomparisons from different experimental techniques are still not clarified. We believe that we can support other experimentalists facing with similar problems, and probably attract attention for this very important issue of freezing temperature shift.

Minor issues

1. There are frequent typos and instances of comma misuse. In addition, there are multiple instances of greek letters being spelled out instead of actually using the symbol. This lack of proofreading is not appropriate for a manuscript submission and shows a lack of care for their own work. I urge the authors to correct this.

We thoroughly reread the manuscript and corrected some typos and erroneously written instances of mathematical and Greek symbols. Although we appreciate the reviewer's opinion but we do not agree with the note on the lack of our proofreading.

2. l. 11-12. In the abstract, the authors write the words "material dependent correction factor". However, it is not claimed what is incorrect. That would be helpful in the abstract. The measured temperature is not incorrectly measured, so please be precise and tell the reader what is really being corrected.

We modified the sentence following the suggestion.

3. l. 20. Please change "The nucleation abilities. . ." to "The ice nucleation abilities..."

Corrected.

4. l. 25-27. This is an inaccurate sentence. n_s is calculated from the total number of nucleation events per unit surface area of the particles, and then it is assumed to be equivalent to the number of sites on the particles. This is a big difference to what is written. The point here is to stress the fact that n_s is an empirical quantity, i.e., defined only by measurements, but assumed to be something that physically exists. No study can know what n_s for a particle is before conducting an ice nucleating experiment, thus it is empirically defined only.

The sentence was corrected following the reviewer's suggestion, and now reads as "This is calculated from the experimentally determined total number of nucleation events per unit surface area of the particles. INAS density is used to represent the number of ice active sites on the particles that are active between 0 °C and the sub-zero temperature "

5. l. 42. Please change "The most. . ." to "One. . .".

Done.

6. l. 44-45. Just because an experiment is inexpensive, easy, and yields many data points, does not make it a standard. Please state these as advantages instead.

We modified the sentence as follows: "Their advantages of inexpensive and easy operation, and the large number of simultaneously measurable droplets offering good count statistics, promoted them for INP characterization experiments."

7. l. 45-47. This is inaccurate. The authors should be well aware of the countless cold stage experiments reproducing homogeneous freezing and expanding homogeneous freezing data sets without any contamination or surface effects. Please remove this sentence.

The sentence has been removed.

8. l. 47-48. This is a bias sentence. According to Budke and Koop (2015) crosscontamination and evaporation was solved in Stopelli et al. (2014) using sealed tubes. If droplets or aliquot volumes are allowed to evaporate or to introduce contamination, then experiments were simple conducted wrong. Contrary to what the authors claim, results by (Budke and Koop, 2015) are not influence by these factors. This sentence must be removed.

The sentence has been removed.

9. l. 49. There is no influence of the supporting surface. Again, previous studies reproduce homogeneous ice nucleation. See Thomas Koop, Ben Murray and Daniel Knopf groups. I will not do the authors literature search. I recommend the beginning of the sentence to change to the following. "We take a step further. . ."

We modified the sentence to the following: "In our study we take a step further to real atmospheric conditions of cloud droplets, and avoid the contact of any supporting surface. The single droplet levitation techniques employed offer experiments ..."

10. l. 63-64. Please remove the redundancy. I read in the previous paragraph and sentences that droplets are freely suspended.

Done.

11. l. 65. What is the "nature" of a hydrometeor to the authors? It is not generated from a bulk solution and pipetted into position. Please remove this term.

We removed the whole sentence.

12. l. 249. It is redundant to say the experimental temperature is kept constant and the experiment is isothermal. Please remove this sentence.

The sentence has been deleted.

13. Figure 2, caption. Should the x-axis be labelled “ T_e ”? Should the inline equation also have T_e instead of T ?

The figure shows the results of the calculation of the approaching time of the surface temperature to equilibrium at different air temperatures. We corrected the figure caption, the axis label and the inline equation accordingly.

14. l. 274. The word “represent” is a little awkward here. I would recommend the sentence to read “Freezing in M-WT experiments was observed under isothermal measurement conditions, and the stochastic approach was applied for data analysis.”

We modified the sentence according to the reviewer’s suggestion as, “Immersion freezing in M-WT experiments was investigated under isothermal measurement conditions, hence, the stochastic approach was applied for data analysis.”

15. l. 279-280. Does this sentence really deserve its own paragraph?

Corrected.

16. l. 350-351. In accord with the major comments. It should be stated here that if S_p varies more than the authors expect, then a single component particle type may be erroneously identified as multiple component.

We carried out careful analysis and considered the measurement uncertainty in order to classify the particle types as correct as possible, but we agree with the reviewer, that the estimated total surface area may vary significantly more than we expected. Therefore, we added the following sentences to this discussion: “In our experiments the total surface area A was estimated from the concentration of the aqueous solution and from the specific surface area. To accurately measure the actual total surface area of INP inside the droplets, which should be taken into account for calculating ω and J_s , is currently not feasible. Therefore, the error of A might be significantly higher than estimated, which would result in a false classification of the INP as single-component.”

17. l. 419-421. When sampling random error, the probability distribution from which numbers are sampled from should be stated. Did the authors sample from a normal distribution? According to the text and error bars in Fig. 5-7, the error is assumed to be normally distributed. The error in n_s in Fig. 9, then is lognormally distributed? Did the authors sample frozen fractions from a normal distribution with or did they sample values of n_s ? Please explain this in the text.

We sampled values from n_s . We did not consider the distribution of n_s but randomly took n_s values falling within the 1σ bounds around the mean n_s value. This might overestimate the λ error.

18. l. 420-421. The authors assume the error bars are 95% confidence intervals. What data is this and how does that relate to the error bars on the previous graphs, which are all 1σ according to the captions? Of course, 1σ is not equivalent to 95% confidence. Their description is inadequate, and sounds like the authors sampled from some distribution, but threw away those values which were sampled beyond the 5 and 95% tail ends. In any case, the description and justification of their random sampling procedure needs to be explicitly written.

Thank you for the note. We used here also the 1σ errors, not the 95% confidence intervals. We corrected the text, and we reformulated the description of the procedure to estimate the error of λ .

19. l. 416-418 and l. 422-423. This is redundant. Please rewrite.

We deleted the unnecessary part of the sentence in line 423.

20. l. 423. Please change the text to read “. . . fitting a linear regression curve to the log-linear graph of randomly sampled data, and subsequently. . .”

This part of the sentence was deleted (s. our reply to the last comment).

21. l. 431-434. The error on ω is washed over in this paragraph. The phrase, “ ω -based temperature shift”, is first used here, however, the authors cannot expect a reader to formulate their own idea exactly how the shift and error on the shift is calculated by themselves. Please include around Eqns (12)-(14) details of how to calculate these temperature shifts. This phrase “ λ -based temperature shift” is used in the list of suggestions at the end of the manuscript, but I am uncertain what is being referred to. I would recommend to specifically define this terminology. Figure 10 is supposed to help with understanding this mathematical flow, however it only adds confusion because it has many undefined quantities that are not even included in the list of variables at the end of the manuscript. These unknown variables I found include T_{cool} , n_s , MAL , $\lambda = (0, 8)$, T_{opt} , $T\omega$. Please state and explain the terminology, variables and equations used in this figure and include them in the list of variables at the end of the manuscript.

The phrase “ ω -based temperature shift” has been removed, and the sentence has been reformulated to explicitly refer to the equation used here.

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.

22. l. 431-434. An addition question about this same paragraph. Is the random sampling of data also used to determine the error on ω in a similar way to λ ? As of now, any equation or description of the error on ω is not clearly stated.

The error of ω is the standard error of the linear fit on the $\ln(R/A)$ vs. T curve. This is now explicitly written in the text.

23. l. 451. Do the authors mean deviations of the “simulated” data points?

Here the corrected data points, i.e. when shifted to higher temperatures are meant. The sentence was rewritten.

24. l. 537-538. There are only two locations in the manuscript where the authors use the phrase “cloud model”, here and in the last line of the abstract. There is no discussion, argument or any information about cloud models in this paper. Therefore, no basis for any suggestion about cloud models is available. Please remove this suggestion, and remove the sentence about cloud models in the abstract.

We removed the sentence about cloud models in the abstract. Nevertheless, in our opinion the freezing temperature shift stemming from the change in the cooling rate is relevant for cloud modelers and is, therefore, presented as a suggestion in Conclusions and Suggestions.

25. l. 541. I am sure the phrase “serve rather for orientation” means something specific to the authors, however this is not clearly defined in the manuscript. Would the authors please explain specifically what is meant by this?

What is meant here is that the apparent temperature shift depends on the specific aerosol sample that is investigated. We want to avoid that any reader would use the temperature shifts tabulated in our paper or in Herbert et al. (2014) “as it is”, because it might vary due to the chemical composition of the sample. Rather, in our opinion cooling experiments should not be conducted using exclusively one cooling rate or without the simultaneous measurement of the nucleation rate coefficient.

26. Figure B2. The labels and legend on the color scale are missing. Also, please state the simulated droplet size.

The figure was corrected by including the labels and the legend, and the drop size is now indicated in the figure caption.