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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 #1

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1 Overview

The manuscript submitted to *Atmospheric Chemistry and Physics* titled "Comparative Study On Immersion Freezing Utilizing Single Droplet Levitation Methods" by Szakáll et al. presents an ice nucleation study of a variety of different types of particles using two methods, a vertical wind tunnel (WT) and acoustic levitator (AT). The WT gives approximate isothermal conditions and the AT has a cooling rate. The authors claimed if a particle type was single or multiple component by comparing values of ice active surface site densities, n_s, of these isothermal and cooling rate experiments, and following the derivation of Herbert *et al.* (2014). They conclude that freezing temperatures should be shifted with respect to derived n_s values. In addition, there is a list of sug-

gestions for further study. This study has value to the *Atmospheric Chemistry and Physics* community providing new n_s values for a variety of particle types, and parameters to describe their experiments following a single or multiple component approach. The methodology combines careful and well designed cooling rate and isothermal experiments, which is certainly relevant for evaluating time and temperature dependence for ice nucleation. There is clear uncertainty and statistical analysis for their conclusions. Unfortunately, there are major issues that must be resolved before I can recommend publication. First, there is insufficient review of previous studies that makes the manuscript unbalanced. Additionally, assumptions about surface area and the impact of surface area calculations on their conclusions are not stated. There is a fundamental flaw with shifting observed temperatures outside their investigated range. 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.

2 Major Issues

- 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.

- On l. 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.

- 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.

- 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? Afterall, 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.

3 Minor Issues

- 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.

- 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.

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

- 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.

- l. 42. Please change "The most..." to "One...".
- 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.
- 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.
- l. 47-48. This is a bias sentence. According to Budke and Koop (2015) cross-contamination 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.
- 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..."
- l. 63-64. Please remove the redundancy. I read in the previous paragraph and sentences that droplets are freely suspended.
- 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.
- l. 249. It is redundant to say the experimental temperature is kept constant and the experiment is isothermal. Please remove this sentence.

- Figure 2, caption. Should the x-axis be labeled "T_e"? Should the inline equation also have T_e instead of T?
- l. 274. The word "represent" is a little awkward here. I would reccomend the sentence to read "Freezing in M-WT experiments was observed under isothermal measurement conditions, and the stochastic approach was applied for data analysis."
- l. 279-280. Does this sentence really deserve its own paragraph?
- 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.
- 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.
- 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 explicit written.
- l. 416-418 and l. 422-423. This is redundant. Please rewrite.

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

- 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 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 suppose 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_{ω}. 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.

- 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.

- l. 451. Do the authors mean deviations of the "simulated" data points?

- 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.

- l. 541. I am sure the phrase "serve rather for orientation" means something spe-

cific to the authors, however this is not clearly defined in the manuscript. Would the authors please explain specifically what is meant by this?

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

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