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Interactive comment on "Revisiting the differential freezing nucleus spectra derived from drop freezing experiments; methods of calculation, applications and confidence limits" *by* Gabor Vali et al.

Anonymous Referee #2

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The derivation of the differential ice nucleus concentration metric (k(T)) is a valuable analysis method for droplet freezing assays commonly used to assess the concentration temperature spectrum of INPs. Droplet freezing assays have become widely used over the last few years, and are largely analyzed using the cumulative INP temperature spectrum (K(T)) also originally derived by Vali 1971. The differential form focused on here has some nice advantages compared to the common cumulative form, such as providing a more robust way to separate the contribution from background freezing interferences, and could see wide applicability throughout the growing ice nucleation

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Discussion paper

community. The differential form is not straight-forward to employ however, and the insights provided here will be of great value to those that wish to perform this analysis. I suspect that these issues are likely a cause for the lack of adoption of the k(T) metric by the ice nucleation community. Vali's efforts to clarify the use and utility of this metric will go a long way to help make this sort of quantitative analysis more widely practiced. The manuscript is well-written and easy to follow overall. I have a few comments and suggestions for revisions that might further improve the paper, which is certainly well within the scope of AMT.

This may be beyond the intent of this manuscript, but it seems that a very useful discussion regarding the important role that droplet size/volume and particle concentration play in accurately retrieving both K(T) and k(T) spectra and thus the INP(T) concentration spectrum would be extremely valuable to the ice nucleation community. Might these factors play a role in the reported discrepancies in n_s/K(T) from different methods of the same, as proposed by Emersic et al. (2015) and Beydoun et al. (2016)?

Page 2, line 2: These spectra also provide measurements of INP concentrations versus T.

Page 2, line 15: It would be good to also discuss the roles that droplet size and particle concentration play in the ability to retrieve information on INPs active at colder Ts.

3/7: This is an interesting and important point that I think is often overlooked by the growing number of groups using droplet freezing assays. It would be useful to elaborate more or this and further justify the notion that the approximation of one INP per droplet being responsible for the freezing event is valid when small delta-T values and large values of N are used. Please cite any key references here so more information on this can be readily found.

3/12: 10% error in what quantity? The approximation below in Eqn. 2?

4/20: Should mention that delta T is not just the temperature interval between successive droplet images, as might be commonly and erroneously thought. It appears to be a free parameter that must be adjusted for each dataset, as discussed further in the manuscript.

Section 7: It seems that a common way in which droplet freezing data are analyzed is to compare the freezing properties of one particle sample with others and determine if they are similar or different, or if some sort of processing or aging (heat, H2O2, acid, etc.) changes the freezing properties of the sample in a distinguishable manner. The use of confidence intervals applied to the k(T) differential analysis would seem to provide a quantitative way to determine if one particle sample distributed amongst an array or droplets has a freezing spectrum that is statistically similar or different from another. It could also be used to determine if data is significantly above the background freezing noise of the system. Could you please add a paragraph or more of discussion on these topics to this section? I think it will help to highlight the many different important types of analysis that can be done using these methods, and the utility of the derivative k(T) metric.

There are numerous little typos and misplaced words throughout the manuscript. Some of these may have been corrected since the original submitted abstract. I did not list them here but please proofread carefully to catch all of these.

Cited References

Beydoun, H., Polen, M. and Sullivan, R. C.: Effect of particle surface area on ice active site densities retrieved from droplet freezing spectra, Atmos. Chem. Phys., 16(20), 13359–13378, doi:10.5194/acp-16-13359-2016, 2016.

Emersic, C., Connolly, P. J., Boult, S., Campana, M. and Li, Z.: Investigating the discrepancy between wet-suspension- and dry-dispersion-derived ice nucleation efficiency of mineral particles, Atmos. Chem. Phys., 15(19), 11311–11326, doi:10.5194/acp-15-11311-2015, 2015.

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