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Atmospheric Measurement Techniques Discussions

Interactive comment on "BINARY: an optical freezing array for assessing temperature and time dependence of heterogeneous ice nucleation" *by* C. Budke and T. Koop

Anonymous Referee #3

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Budke and Koop provide a quite detailed description and exemplary results from a droplet freezing array system for studying heterogeneous ice nucleation. The use of a polymer spacer, formed by soft lithography methods, to isolate individual droplets for freezing studies of droplet array is a nice contribution to droplet freezing methodology. The details provided on the automated analysis of freezing events was also quite valuable. This topic is certainly of strong relevance to AMT, and will be a valuable contribution to the rapidly growing field of ice nucleation research in the atmospheric sciences. While the new method is sound and valuable, the paper was not very clearly written in many places. My main concern is that the discussion of the role of time-dependent

freezing is rather confusing and muddled. While the data clearly demonstrates a very weak role from time-dependent freezing, the authors then analyze the time-dependent effects to great detail, as if it is a very important factor that must be fully accounted for. The authors should work on making this much clearer, and addressing the other following issues. When these have been addressed this manuscript should be suitable for publication in AMT.

Abstract: I think the abstract will be rather confusing to most readers, even experts. Can the authors explain the main results using less technical language than for example, "For the Class A IN a very strong increase of the heterogeneous ice nucleation rate coefficient with decreasing temperature of $\lambda \equiv -$dln(jhet)/dT =8.7K$-$1 was observed emphasizing the capability of the BINARY device." I do not think the meaning of that will be at all clear to most readers. What "capability" of the device does this illustrate?

Exactly how the droplets were prepared and loaded into each array/well for subsequent freezing analysis is an important detail that is not discussed here. For Snowmax this may be quite simple, but how do the authors propose to do this with more difficult to handle samples, such as mineral dusts, biological particles, or even actual ambient aerosol samples? Some methods for preparing droplets for freezing analysis from a wider range of particle/INP types should be discussed, to complete this new method's development.

I like that there was a detailed discussion of temperature calibration, but found this discussion was not very clear, and overly jargony. (End of Section 3) Could the authors please re-write this section more clearly?

The data clearly demonstrates that Snowmax exhibits a very weak time-dependent component for droplet freezing, as demonstrated by varying the cooling rate. This adds to the growing body of data indicating the small role that stochastic effects play compared to deterministic effects (e.g. Vali, 2004; Wright & Petters, 2013). Yet the authors then discuss at great length the role of time-dependent freezing for their data.

I suspect this is because they want to emphasize the utility of their BINARY system to observe with high sensitivity time-dependent effects. These effects are not supported by the data for Snowmax however, and so the whole discussion is rather confusing. The authors should clarify, perhaps by making it very clear that Snowmax exhibited a negligible contribution from stochastic freezing, and then going into their detailed analysis while making it clear they only do this to illustrate that time-dependent effects can be retrieved from the BINARY analysis.

An example of this confusion in the text: On page 9149, line 13: "At the indicated concentrations the difference between the Tf,50 values at 10 and at 0.1 K min^{-1} is about 0.6 K for both Classes of IN (0.55 K for Class A and 0.64 K for Class C). These 15 values are rather small but they are significantly larger than our temperature uncertainty, implying that we were able to detect a rather minute time dependence for each of the two IN Classes." I do not think a 0.6 K variation in freezing temperature between the extremes of the cooling rates used, 0.1 to 10 K/min, is terribly significant. It is nice that this is beyond the temperature uncertainty, but this weak time-dependent effect could be safely ignored without changing the actual results significantly. But then on the same page, line 18 the authors state: "The above analysis suggests a time dependence of Snomax® induced ice nucleation." Based on the data discussed in the previous sentence, the authors should clarify that only a quite /weak/ time dependence is observed. As written this is rather contradictory, and thus confusing.

This is further illustrated on page 9152, line 9: "We found that changing observation time by several orders of magnitude results in a change of Pi0 from zero to one in a narrow temperature range smaller than 1K. In contrast, at a constant temperature the same change in Pi0 requires a difference in observation time of more than one order of magnitude." I really feel that the authors are dramatically over emphasizing the role of time in the observed freezing rate, which is just not supported by the data, and is contradicted by their analysis. The authors can still illustrate their ability to determine time-dependent effects from their BINARY system without having to misrepresent the

importance for the Snowmax system presented here. Perhaps a new section of "Detection and Quantification of Time-Dependent Freezing Effects" could be added, to make this more of a data analysis methodology focused section.

The data analysis method (Eqn. 3, etc.) looks quite familiar to the method of Vali (1971). If so please cite this as the original analysis method.

Vali, G. Quantitative Evaluation of Experimental Results an the Heterogeneous Freezing Nucleation of Supercooled Liquids. J. Atmos. Sci. 1971, 28, 402–409.

The last few paragraphs of Section 4 are especially confusing. What do all these numbers mean? The authors never clearly stated what their recommended values for an accurate description of ice nucleation rates for Snowmax as a fcn of temperature are, based on their data. How would they analyze/digest their measurements to provide useful descriptions of heterogeneous ice nucleation for modeling purposes? Please synthesize and summarize your analysis so the overall conclusions are key. The lengthy discussions of time-dependent effects, which are not actually important for this dataset, really confuse this discussion.

Interactive comment on Atmos. Meas. Tech. Discuss., 7, 9137, 2014.