Anonymous Referee #3

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.

We thank the referee for the positive evaluation of our manuscript and the following helpful comments.

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 $\lambda \equiv -d\ln(jhet)/dT = 8.7K-1$ 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?

The second part of the abstract has been reformulated as suggested:

"Using different cooling rates a small time dependence of ice nucleation induced by two different classes of ice nucleators (IN) contained in Snomax® was detected and the corresponding heterogeneous ice nucleation rate coefficient was quantified. The observed time dependence is smaller than those of other types of IN reported in the literature, suggesting that the BINARY setup is suitable for quantifying time dependence for most other IN of atmospheric interest, making it a useful tool for future investigations."

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.

We have now described the droplet preparation procedure in more detail. This preparation procedure has been used also for other types of IN, but we refrain from speculating about how we may prepare droplets containing other types of IN in future experiments. The new text now reads:

"Snomax® suspension were prepared by mixing a pre-determined mass of dry material with the appropriate volume of freshly double-distilled water. Individual droplets of 1 µL volume were sampled from the suspension with a micropipette (volume accuracy ≤ 3%) and positioned individually on the glass surface in each compartment. We note that the use of smaller (e.g. ~0.5 µL) and larger (e.g. ~5 µL) droplets is also possible, but only 1 µL droplets were used in the freezing experiments described below."

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 entire second paragraph of the section "Temperature Calibration" was rewritten in many parts.

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.

We do not agree with the statement that the observed small time-dependence is not supported by our data. We provide a detailed analysis of the data to show that we can quantify the very small time dependence of Snomax IN. This is not done, because we think that this small time dependence is important for the atmosphere or atmospheric models, but indeed as mentioned by the referee to show the utility of the BINARY experiment. The paper is about characterizing the experimental setup and we do so by showing that the data obtained with it can be useful for various analyses. The characterization of Snomax and the atmospheric relevance of its time-dependence is of secondary importance. This is why we submitted the paper to Atmos. Meas. Techn. instead of Atmos. Chem. Phys. See also next comment.

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.

We have followed the suggestion by the reviewer: "While this degree of time dependence is probably not of atmospheric importance, we analyze and quantify it in more detail below for two reasons: first, it is interesting from a physical chemistry viewpoint regarding the fundamental process of heterogeneous ice nucleation and, second, such an analysis may help in characterizing the ability and limitations of the BINARY device for measurements of time dependence of IN more generally."

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.

According to Merriam-Webster dictionary the word “significant” can mean both “large enough to be noticed or have an effect” or “very important”. We certainly meant to use it in terms of “large enough to be noticed” and we did not mean to imply that it is very important for atmospheric purposes (see also previous comment). To avoid such ambiguity we have removed the word “significant” in the context of time-dependence throughout the manuscript.

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 sentence has been reworded as suggested: “The above analysis suggests a very small time dependence of Snomax®-induced ice nucleation. Nevertheless, we may… ” Together with the two additional sentences added to the end of the previous paragraph (see text in answer to second-to-last comment).

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.

We do not agree. Fig.9 shows that the effect of time is indeed very small, i.e. temperature is much more important than time dependence. In addition, for constructing this figure we used the nucleation rate coefficient obtained by fitting the experimental data.

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.

We have followed the suggestions by reviewer 2 and 3 and shortened and straightened the "Results" section, by moving large parts into a new section "Theoretical Background for Freezing Analysis".

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 and the Heterogeneous Freezing Nucleation of Supercooled Liquids. J. Atmos. Sci. 1971, 28, 402–409.)

We suspect this comment refers to eq. (1), and the corresponding reference has been included there as suggested. Another reference (Vali, 1994) was added before eq. (3).

The last few paragraphs of Section 4 are especially confusing. What do all these numbers mean?

We have rewritten this part by moving the different lambda-values into the new table 2 in the appendix, making the remaining text more focused and more readable.

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?

We have now included a new sentence with our recommendation:
"We recommend a numerical value of $\lambda = \omega = 8.7\ K^{-1}$ for Class A Snomax IN at the median freezing temperature of -3.8°C (269.4K)."

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.

For us one key result is that it can be shown experimentally that Snomax shows a time-dependence for two reasons: first, because the new BINARY setup shows its ability to be able to measure such a small time dependence. Second, we find it interesting and important from the fundamental physical-chemistry viewpoint that an IN usually regarded as a prime example of a singular IN, does indeed show time dependence, however small it may be. This has been added before the discussion of time dependence, see text at the bottom of page 11.