

Anonymous Referee #2

The paper presents a new tool to study heterogeneous ice nucleation in the immersion freezing mode. The Bielefeld Ice Nucleation ARaY BINARY is an optical freezing array that was thoroughly tested and applied/characterized using Snomax as ice nucleator as described in the manuscript. The ice nucleation process itself is assessed for its temperature and time dependence, thus distinguishing between a singular and a stochastic approach.

The BINARY represents a great extension of already existing laboratory, ice nucleation instrumentation with the capability to improve our understanding on heterogeneous ice nucleation as already shown in this paper using the test substance Snomax for characterization.

In general, the paper content is highly sufficient for a publication in AMT, and the obtained results concerning the instrument characterization and the IN properties of Snomax particles are well elaborated. From the scientific point of view, only minor comments are listed below.

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

Major comment:

I sometimes found the paper hard to read and to follow due to the “jumps” between theory and results. Thus, I would suggest including a section “theoretical background” which separates the theory of ice nucleation and that of the analytical approach from the “results” section. Along these lines, a clearer distinction/separation between these two parts, i.e. IN theory and “math needed for data analysis” could be pursued. Also, IN-relevant data on Snomax would be highlighted more this way, although not being the main task of this study.

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”.

In addition, sentences are sometimes disconnected and structured in a way that impedes the reading flow. A check by a native speaker could be helpful in this way.

A number of minor corrections have been made to the manuscript, which are not listed in detail here, in particular in sections “Temperature Calibration” and “Results”.

Minor comments:

P9138, l17ff: How does the observation of a strong T-dependency of λ emphasizes “the capability of the BINARY device”? This is too vague for an abstract. Be more conclusive concerning this statement, also in the summary. In a similar way, the fact that the value “is larger” than literature values does not explain to the reader the uniqueness of the tool.

That 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.”

P9138, I26: Cziczo and Froyd, 2014, is not the most original paper for heterogeneous ice nucleation. Or do you refer to the introduction of the abbreviation for ice nucleators?

We have added a number of further references for heterogeneous ice nucleation.

P9139, I9ff: “Representing immersion freezing . . .” in models? Still, the authors do not give a clear answer why immersion freezing it is difficult to represent in current models” (see second major comment).

This sentence has been reformulated:

“But the time-dependence of immersion freezing is often poorly represented or not included at all in cloud models, in order to reduce model complexity (Ervens and Feingold, 2012).”

P9140, I7ff: Rename the “above mentioned processes” to clarify to the reader.

This sentence has been reformulated:

“Several instrumental techniques are available for the determination of *temperature and time dependence of heterogeneous ice nucleation* (see, e.g. Murray et al., 2012; Hoose and Möhler, 2012).”

P9142, I12ff: Is the range of 0.5-5 μL exact? What is the typical variation in the 1 μL per droplet you produce?

We have written a separate paragraph explaining the droplet preparation in more detail:

“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 $\leq 3\%$) and positioned individually on the glass surface in each compartment. We note that the use of smaller (e.g. $\sim 0.5 \mu\text{L}$) and larger (e.g. $\sim 5 \mu\text{L}$) droplets is also possible, but only 1 μL droplets were used in the freezing experiments described below.”

P9142, I25ff: Did the mentioned techniques not use double-distilled water for their experiments? This sounds obvious – but is BINARY the first applying this?

Using distilled water is not new of course, but we wanted to describe our droplet preparation procedure, too. This part has been rewritten, see previous comment and text.

P9144, I12ff: Can the condensation of water vapor in the compartments influence the measurement? Please make a statement/reference here and refer to the text afterwards.

The condensing water originates from the humidity inside each compartment, but the droplets formed do not influence the measurements. We included a footnote to explain this in more detail:

“The condensing water originates from the humidity inside each compartment from the vapor pressure of the droplet itself. At the start of the experiment the relative humidity in each compartment is 100% and upon cooling, some of this humidity condenses onto the cold glass slide. These droplets are much smaller than the investigated droplets and, thus, normally freeze at much colder temperature, i.e. at about the homogeneous ice nucleation limit of supercooled water. Therefore, the condensed droplets do not affect the heterogeneous ice nucleation processes in the microliter droplets studied here. Only if heterogeneous ice nucleation is triggered in the condensed droplets (e.g., by a surface impurity/irregularity) they may subsequently seed the larger droplets. This was observed to occur only rarely at the lowest cooling rates and, accordingly, these data points were excluded from the analysis.”

P9144, I25: “Upon heating ...” Please clarify this sentence

This has been reworded: “After a temperature of -10°C is reached, the sample is reheated....”

P9145, I2: “negative peak”. Isn’t that rather a minimum?

Reworded to “local minimum” as suggested.

P9145, I16: The statement why using heating mode could be more comprehensive as I think it is important for the instrument characterization process.

We have reworded the statement which now reads:

“The rate calibration was conducted in the heating mode because superheating of a crystal above its melting point is usually negligible. In contrast, supercooling of a liquid below the melting point often occurs readily and, thus, even minor supercooling would bias a calibration in cooling mode.”

In addition, the text of the remainder of this section describing the calibration procedure was reformulated in a number of places.

P9145, I21: Results in “()” make them appear unimportant

The parentheses have been removed.

P9145, I25: Why is the spraying of the reference material ensuring the detection of the onset of the phase transition?

Reworded: “The onset of melting and the subsequent, almost immediate full melting in such small droplets results in a steeper Δg_v , which is easier to detect and, hence, more accurate than in larger droplets, which often melt entirely only with measurable delay, see Fig. 2.”

P9146, I2: “accordingly” refers to what?

Reworded to “for each calibration substance”: “The threshold value for automatic detection of the phase transitions was adjusted *for each calibration substance*, in particular for the solid-solid phase transitions which involve smaller Δg_v values.”

P9146, I23ff: Please give reference for Snomax IN-activity

We have added several reference of previous ice nucleation studies on Snomax.

P9147, I14: Why using “[]” in the equation?

The parentheses have been removed.

P9148, I1ff: The first paragraph deals with reference results from Turner et al., 1990, the second with own results, and the third switches back to Turner et al. Please consider a rearrangement since the reader expects an answer for the missing Class B immediately, which is given in the third paragraph.

The paragraphs have been rearranged as suggested.

P9149, I16: What do you mean with “rather minute time dependence”? “Rather is a vague word, and the explanation of “minute time dependence” should be more physical.

The sentence containing the term “rather minute time dependence” was removed.

P9151, I25ff: Where do the alpha values of 23.9° and 35.3° come from?

They are the results of fitting the data in Fig.6c to CNT allowing only a constant, i.e. temperature-independent, alpha. The first two sentences of that paragraph were rewritten:

“The lines in Fig. 6c are fits to these data using the framework of classical nucleation theory (CNT), i.e. they represent $j_{het}(T, \alpha)$ fits in which the effective contact angle α was either constant or allowed to have a linear temperature dependence. Figure 6c shows that the measured data are not well described by CNT when using constant effective contact angle values of $\alpha = 23.9^\circ$ and $\alpha = 35.3^\circ$ for Class A and C, respectively (gray lines).”

P9167, Figure4: I assume the Turner classes are indicated by the dashed lines? Please mention this.

We have revised Figure 4 and now show the approximate temperature ranges (also corrected) of the three different classes with colored bars at the top of the figure.

