Anonymous Referee #1

This is an excellent paper regarding the development of a new optical freezing array. The aim of this study is to supply the necessary measurement technology for assessing a detailed understanding of heterogeneous ice nucleation in laboratory experiments. Here it is important to distinguish between a singular and a stochastical approach. For the latter, temperature and time are important parameter, thus, the set-up has too meet high demands in accuracy, which has been solved in an excellent way by the new developed freezing array and the related instrumentation for controlling and observation.

As an IN model sample the commercial product Snomax has been investigated. This is good choice since Snomax offers two classes of IN, which are ideal candidates for the testing of the new set-up.

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

Comments:

You might comment on the fact that the volume of your droplets (1μL) corresponds to a diameter of about 1.24mm. This is more than one orders of magnitude larger than the droplets in real clouds at high altitudes (20μm, 4 pL), where freezing processes are currently under investigation. What are the consequences for your results taking the differences in volume into account?

For heterogeneous ice nucleation, the IN concentration per droplet is much more important than the drop volume. The latter is only relevant for experimental reasons, as larger droplets may contain more impurities which may cause ice nucleation at higher temperature when compared to smaller droplets. This is, however, not relevant for the present study since Snomax is such a good ice nucleator at high temperature.

You mention a purging of your cell. What is the flow velocity of the N2 stream? Do you purge during the measurement? If yes, what is the impact on the results? Does it cause heat input? Can droplets evaporate due to the N2 stream, which might transport water vapor out of the cell?

The N2 purge flow does not lead to evaporation of droplets, because the droplets are separated from the gas stream as the sample array is covered by a second glass slide (see Fig.1b). The N2 purge flow was also present during the calibration procedure, hence a potential heat input (if any) is accounted for.

The gray value is a very clever concept which produces excellent results. However, it comes not clear if you work with an inverted gray scale or not?

We do not work with an inverted gray scale, the images shown in Fig.2 represent the actual gray scale. In transmission light-microscopy experiments droplets that freeze become darker as the ice scatters more light out of the light path. Here, we work in reflected-light mode, hence the frozen droplets become brighter as they scatter more light into the light path. We have added the following sentence:

"Since we observe the droplets in reflection mode (not in transmission), in the images liquid droplets appear dark, and frozen droplets appear bright as they scatter light into the observation light path."

When comparing the volume and the respective observed diameter of your droplets, it becomes obvious that there are small differences with the calculated diameter assuming sphericity. Thus, I may conclude that the droplets exhibit a certain spreading on the surface of the support. Could you supply contact angle measurements to quantify this effect and its impact?

We have prepared droplets of fixed volume (1 microliter). For the droplet diameter indicated in Fig.1a we have assumed hemispheric droplets, i.e. a contact angle of 90° (see also Fig.1b), which is supported by contact angle measurements, showing contact angles of 90-100°. However, all freezing analysis calculations are based on droplet volume anyway, and hence the indicated droplet diameter is not important. We have included the following sentence in the revised version:

"Water droplets positioned on the silanized glass show contact angles of about 90–100° (Remmers, 2012). Hence, the shape of the droplets investigated below is well approximated by a hemisphere."

On page 9144, line 23 you mention previously condensed water, which evaporates subsequently to the freezing process and finally freezes on the already frozen droplets. However, you also mention spacers around every droplet, which suggests that every droplet is situated in its own compartment suppressing the infiltration of humidity from outside. Please explain this situation in more detail.

The condensing water originates from the humidity inside each compartment, which in turn is stemming 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 on the cold glass slide. 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."

On page 9149 you explain in detail the fundamentals for equations 3 to 6. However, the simplification from eq. 4 to eq. 5 is difficult to understand. Please explain this step in more detail.

The simplification is now included and explained.

Minor comments:

page 9138, line 14: The abbreviation for ice nucleator (IN) should be introduced

Changed as recommended.

page 9149, line 15: The value of temperature uncertainty might be mentioned in parenthesis.

Changed as recommended.

I recommend this manuscript for publication in Atmos. Meas. Tech. after some minor changes.