Referee report on "Mapping ice formation to mineral-surface topography using a micro mixing chamber with video and atomic-force microscopy" by Raymond W. Friddle and Konrad Thürmer

The authors have assembled an experimental setup to grow ice crystals on a sample surface and developed a method to locate where ice forms to investigate the topographical features on the underlying surface by atomic-force microscopy. While the setup could prove useful to study ice growth on surfaces, the intended use to study ice nucleation mechanisms requires a higher vertical resolution to detect small ice crystals and pinpoint the location of ice active sites. In addition, much better control of temperature and relative humidity in the mixing chamber is needed. I think such could be achieved and encourage the authors to improve the setup towards this direction.

Specific comments

Page 1 line 16 Heterogeneous ice nucleation is not limited to temperatures above -36°C. Deposition ice nucleation relevant for cirrus cloud formation occurs at lower temperatures and below water saturation. As the experimental setup described in this manuscript might become useful to investigate deposition ice nucleation, I recommend mentioning it here in the introduction.

Page 1 line 24 The parametrization by DeMott et al., 2010 is not based on size as an ice nucleation property, but simply relates the concentration of INP to the concentration of particles above a threshold size, not implying that only these particles act as INP. This is often misinterpreted, please revise.

Page 1 line 26 Useful parametrizations should capture various situations. Please elaborate and provide references supporting the claim that the mentioned parametrizations are not accurate outside the conditions for which they were developed. Also, surface site density of ice active sites derived from field measurements and laboratory studies have been used to parameterize ice formation in models eg., Vergara-Temprado et al., 2017. This could be mentioned.

Page 1 line 31 Please specify what kind of information microscopy can provide to distinguish mechanisms of ice nucleation.

Page 2 line 3f It is unclear how the 10nm size is derived. Given the resolution of light microscopy, pixel size etc., used in the current setup it seems unrealistic to detect such small objects, making the discussion of framerate and its dependence on temperature and humidity conditions irrelevant. What is the smallest detectable size in the current setup and what is the limiting component?

Page 2 line 15 Clarify how this estimate was made. The resolution is 1.6um? This seems not to be high enough to see growth of 1um crystals. In addition, I calculate at least 10-times longer growth needed to reach this size at this conditions. The mentioned growth rate indicates a RH>>100% and questions the control of relative humidity in the experiment. Ice growth can be used to infer humidity in the specimen chamber (see S3 in Kiselev et al., 2016). I highly recommend a comparison of relative humidity based on ice growth rates and the method used by the authors to determine humidity.

Page 2 line 16f Please elaborate how high-speed AFM can advance heterogeneous nucleation research.

Page 2 line 20 How accurate can the site of ice formation be located with this setup? It is mentioned on page 2 line 1 that the spatial resolution must be on the order of nanometers to locate the ice nucleation site. Please derive the minimum resolved distance for your camera system and verify with a resolution target. A discussion of what accuracy would be desirable in contrast to what can be achieved would be helpful to clarify down to what scale the setup can be sensitive.

Page 2 line 22 Surface features on a feldspar specimen of the size used in this study might not be present on micrometre sized dust particles found at mixed-phase cloud level, and therefore be not relevant for ice nucleation on these particles. I recommend not to emphasise atmospheric relevance.

Page 3 line 1ff Provide a temperature calibration to demonstrate the stability (1°C/hr mentioned in Sec.2.2.), accuracy of temperature control and homogeneity in the mixing chamber. Temperature control is crucial to study ice nucleation and therefore the interpretation of observations made with the setup. Please clarify if temperature is actively controlled or only monitored with the TC-720. Active temperature control is desirable for this type of setup.

Page 3 line 11 Ice and mixed-phase clouds form at a variety of conditions. Ice clouds do not require water saturated conditions. Specify conditions that can be crated in the mixing chamber.

Page 3 line 17f How is frost formation in the mixing column prevented?

Page 3 line 20f Advantages compared to what other technique? What can be learned from using different flow rates?

Page 3 line 23f Please explain why thermal gradients are minimized by that.

Page 3 line 32 Please provide exemplary time series of temperature and relative humidity during an experiment. What is the purpose of switching the wet flow on and off? Could the humidity sensor be used to measure humidity in the outlet flow to verify the humidity in the chamber?

Page 4 line 1 How long does it take to reach steady-state humidity? To vary the humidity in the mixing chamber the flow through the bubbler is adjusted. Does this change steady state? Provide measured humidity after the bubbler as function of flow rate. Another strategy to adjust humidity in the wet flow might be to change the temperature of the bubbler.

Page 4 line 9ff Knowing and controlling the relative humidity (RH) in the experiment is essential for interpretation of results and to infer the ice nucleation mechanism. Calibration of relative humidity should be done much more carefully by eg., using a dew point mirror to measure humidity in the outflow of the chamber. While AH might be useful to determine flow rates of the wet flow, chamber conditions should be reported as relative humidity and temperature. Convert AH to RH throughout the manuscript.

Page 4 line 19 Converting the error in AH_{in} of 0.08g/m^3 to RH gives +/- 18% which is a very high uncertainty for ice nucleation experiments.

Page 5 line 1 AH_{in} reported here and considering the uncertainty given on the last page, relative humidity is equal to RH_w= 85% +/- 18%. Conditions above water saturation are within the experimental accuracy, making the interpretation of the data as purely deposition ice nucleation imprecise. This underlines the point made in the comment above, that control of the experimental conditions is insufficient for ice nucleation experiments. Compare estimated saturation conditions against calculation based on ice crystal growth rate or measure the humidity at the chamber outlet.

Page 5 line 3 Couldn't AFM detect pores on the substrate? What is the horizontal resolution of AFM used here?

Page 5 line 5 "Ice formation" instead of "ice nucleation" would be more accurate.

Page 5 line 12ff What is discussed here is ice growth and not ice nucleation. Inferring ice nucleation mode from this observation seems over-reaching. The two processes (ice growth and ice nucleation) should be separated more clearly throughout the manuscript.

Page 6 line 14 All four humidities applied are high above water saturation (RH=134%, 167%, 201%, 234%). It is surprising to see sensitivity of ice formation on the amount of supersaturation in this high humidity regime other than a change in growth rate. As pointed out in the discussion, different grow rates are a more plausible explanation for the observation than the probability of ice nucleation. The context in which the experimental results are interpreted should be clarified. Is it about ice growth or ice nucleation mechanisms?

Page 7 line 5 Please provide the resolution of the current setup. Is the CCD pixel size limiting the resolution?

Page 10 Fig.2 check if there is a mix-up between e), d). The description in the figure caption seems to be switched. Images show a scale bar of 5um and this seems to be a typical scale how accurate ice formation can be located. In the introduction it is correctly mentioned that ice nucleation occurs on structures with a scale of few nanometres. Features in eg. e) are on a 1000-times larger scale, questioning the interpretation as ice nucleating sites.

Page 10 Fig. 3 replace AH with RH (=167% +/- 18%).

Page 12 Fig. 6 b) replace AH with RH (=134%, 167%, 201%, 234% +/-18%) .

References

DeMott, P. J., Prenni, A. J., Liu, X., Kreidenweis, S. M., Petters, M. D., Twohy, C. H., Richardson, M. S., Eidhammer, T., and Rogers, D. C.:Predicting global atmospheric ice nuclei distributions and their impacts on climate, Proceedings of the National Academy of Sciences, 107, 11 217–11 222, https://doi.org/10.1073/pnas.0910818107, 2010.

Kiselev, A., Bachmann, F., Pedevilla, P., Cox, S. J., Michaelides, A., Gerthsen, D., and Leisner, T.: Active sites in heterogeneous ice nucleation-the example of K-rich feldspars, Science, 355, 367–371, https://doi.org/10.1126/science.aai8034, 2016.

Vergara-Temprado, J., Murray, B. J., Wilson, T. W., O'Sullivan, D., Browse, J., Pringle, K. J., Ardon-Dryer, K., Bertram, A. K., Burrows, S. M., Ceburnis, D., DeMott, P. J., Mason, R. H., O'Dowd, C. D., Rinaldi, M., and Carslaw, K. S.: Contribution of feldspar and marine organic aerosols to global ice nucleating particle concentrations, Atmos. Chem. Phys., 17, 3637–3658, https://doi.org/10.5194/acp-17-3637-2017, 2017.