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Atmospheric Measurement Techniques Discussions

Interactive comment on "Cleaning up our water: reducing interferences from non-homogeneous freezing of "pure" water in droplet freezing assays of ice nucleating particles" *by* Michael Polen et al.

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In this manuscript Polen et al. describe a study of non-homogeneous ice nucleation in 'pure' water samples and a variety of substrates. They then go on to make a sequence of suggestions for groups using droplet freezing assays. This is a highly valuable manuscript and it will help both newcomers to the field and established groups improve their droplet freezing techniques. I agree with the authors statement that individual groups have a great deal of knowledge on this subject, which isn't necessarily made publically available. The comments I have made below are made in this spirit. I support the manuscript's publication and list a few comments which I hope the authors will use to further strengthen their manuscript:

1.) Handling blank experiments vs. ‘pure’ water control experiments. In the list of recommendations, I encourage the authors to include a recommendation that experimentalists conduct full handling blank experiments in addition to ‘pure’ water control experiments. By handling blank experiment I mean putting the water used in the experiment through the full process that the water containing a sample had been through. In Vergara-Temprado et al. (1) we demonstrated that this was critical. The ‘pure’ water control experiments were lower than the handling blanks (see Figure 2). The experiments where droplets were loaded with black carbon froze at a similar temperature to the handling blanks, but were above the pure water control. We consequently reported limiting values. In Fig 12, is the control experiment a handling blank?

2.) Abstract: The word ‘plagued’ is perhaps a bit strong. It is a limitation.

3.) The authors refer to homogeneous nucleation at -38 C and the ‘homogeneous nucleation limit’. This creates the impression that homogeneous nucleation has a well-defined limit where it occurs. It is volume and time dependent and this matters. For example, a 1 um droplet cooled at 1 K min-1 will have a freezing probability of 0.5 at -33.5C (according to Koop and Murray (2)). Furthermore Herbert et al.(3) found that enough cloud droplets started to freeze in a cloud to start to affect cloud properties at around -33 C or so (depending on the homogeneous parameterisation), even though 50% of droplets would only freeze homogeneously at around -38 C. I would like to see the introductory sentences adjusted to be less definitive about the when homogeneous nucleation becomes important. Also, at ln 51, define the ‘homogeneous nucleation limit’ as, for example, the T at which 50% of 10 um droplets are expected to freeze on cooling at 1 K min-1. 4.) Ln 34-35. Vergara-Temprado et al. (4) could be cited here, this paper clearly shows a sensitivity of clouds to INP. 5.) Ln78. Replace ‘steals’ is not the best choice of words. 6.) Ln93-97: Tarn et al. (5) also used microfluidic technology to study heterogeneous freezing. One of the objectives was to see if the oil and surfactant influenced nucleation. They measured ice nucleation (ns) for a range of materials and compared to literature. The results suggest that these technologies can be used to make droplets and study heterogeneous nucleation.

7.) Ln 214. ‘correct for’, replace with ‘account for’. This isn’t a correction.

8.) Fig 2 and discussion. These images are similar to those shown in Fig 4 or Whale et al. (6), mention that the new findings are consistent with what was previously found. Also, note that it is relatively easy to see if this is a problem.

Also, just for this discussion: we have found that this becomes a more significant problem when doing experiments in a humid environment. We solved this problem by improving the design of our ul-NIPI chamber to make it more air tight.

9.) ‘Particle sedimentation out of the droplet’. I don’t think this is very likely. They may sediment to the bottom of a droplet, but are unlikely to sediment across an interface. Emersic et al. provide no evidence that particles can sediment out of a droplet.

10.) P 9-10. We have also compared HPLC water and compared to water from MiliQ systems. We have found that the quality of HPLC water is also variable. I would recommend that whatever the source of water, the experimenter should demonstrate its quality and do the experiments to test the quality at sufficient intervals. I also agree that water from MiliQ machines can be highly variable, but in our experiments if the machine is well-maintained then the quality is systematically high, although we too have had periods when the quality was much lower.

11.) Fraction frozen plots throughout. I think it would be valuable to show the theoretical homogeneous nucleation curve. I would suggest using Koop and Murray (2) since in this paper the authors attempted to constrain classical theory in a physically plausible way, which gives more confidence in the values at higher temperatures (relevant for ul sized droplets) where there is very little or no data.

12.) Substrate dependent nucleation: Mention the result of Price et al. (7) (Figure 4) where it was shown that a Teflon substrate produced lower freezing temperatures when compared to a salinized glass surface. 13.) Ln 713: On the topic of K-feldspar being sensitive to water. We explored this in Harrison et al. (8) and showed that BCS 376 (a standard feldspar which is available for anyone to buy) only degraded by ∼1 C over 16 months in water. Other feldspars, in particular those exhibiting hyperactivity, are more sensitive to time in water.

14.) Also, I think Vali's comment on this paper is really valuable, and it would be useful to record this approach in the literature (either in Polen et al. or Vali could consider a short not in AMT setting this out formally?).

References: 1. Vergara-Temprado J, et al. (2018) Is Black Carbon an Unimportant Ice-Nucleating Particle in Mixed-Phase Clouds? J. Geophys. Res. 123(8):4273-4283. 2. Koop T & Murray BJ (2016) A physically constrained classical description of the homogeneous nucleation of ice in water. J Chem. Phys. 145(21):211915. 3. Herbert RJ, Murray BJ, Dobbie SJ, & Koop T (2015) Sensitivity of liquid clouds to homogenous freezing parameterizations. Geophys. Res. Lett. 42(5):1599-1605. 4. Vergara-Temprado J, et al. (2018) Strong control of Southern Ocean cloud reflectivity by ice-nucleating particles. P. Natl. Acad. Sci. USA. 5. Tarn MD, et al. (2018) The study of atmospheric ice-nucleating particles via microfluidically generated droplets. Microfluid. Nanofluid. 22(5):52. 6. Whale TF, et al. (2015) A technique for quantifying heterogeneous ice nucleation in microlitre supercooled water droplets. Atmos. Meas. Tech. 8(6):2437-2447. 7. Price HC, et al. (2018) Atmospheric IceâĂŘNucleating Particles in the Dusty Tropical Atlantic. J. Geophys. Res. 123(4):2175-2193. 8. Harrison AD, et al. (2016) Not all feldspars are equal: a survey of ice nucleating properties across the feldspar group of minerals. Atmos. Chem. Phys. 16(17):10927-10940.

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