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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Immersion mode heterogeneous ice nucleation by an illite rich powder representative of atmospheric mineral dust" *by* S. L. Broadley et al.

Anonymous Referee #1

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I recommend publication of the paper as it is well-written and well-argued. It would be a valuable addition to the literature were it to be published as-is. That said, I have a few comments, suggestions, and questions that the authors can consider.

First, I think the authors have buried the lede. While the analysis that they've performed is extensive, I think the data they mention but exclude from the analysis is the most interesting aspect of the paper. On pg. 21, the authors discuss the fact that there seems to be a special type of particle present in some of their samples which dominates the freezing behavior if it is present in the droplet. Their estimate is that 1 in 10^5 particles of their powder is this type. They re-affirm this conclusion on pg. 30 (first paragraph, which starts on pg. 29.) The existence of these rare but highly effective IN is an important finding. The fact that 1 in 10^5 particles are of this type deserves to be in the Abstract.

There's another point, which is potentially even more significant (in my opinion), which is not spelled out, but may be lurking in the data and the analysis. The authors show in Figure 3, for instance, that experiments in which they used a low concentration of dust (0.007 weight percent by mass) were consistent with homogeneous nucleation of water over a range of temperatures. Does that mean there were no particles in the droplet or that there were particles in the droplet, but they had no effect on the nucleation rate over the timescale of the experiment? (You could estimate this with a couple of assumptions. I calculate that a droplet with a diameter of 20 μm has a mass of 4.2 ng and an aerosol particle with a diameter of 1 μm has a mass of 1.4x10^{-12} g. (That assumes a spherical particle.) That works out to an average of about 1 particle per droplet for a weight percent of 0.007. That corresponds to (using Poisson statistics) the same probability of having 1 or zero particles in a droplet.) If there are particles in the drop but ice still nucleates homogeneously, that's more important than the existence of the rare, but effective, particles in the powder sample. I would say that the community assumes that if something like illite were present in a cloud droplet, the freezing temperature would be higher than homogeneous. If there are particles present in drops that are nucleating homogeneously, that should be stated in the Abstract.

Pg 4, 1st paragraph: "Atmospheric IN in mixed phase clouds are thought to be..." Not thought to be. The IN **are** mineral dust, soot, etc... And not just in mixed phase clouds, cirrus clouds too, though I suppose you could argue that the ice nuclei in cirrus could also include things like solid ammonium sulfate

Pg. 5, last line of the 1st paragraph: "Additionally, this 'inside-out' contact nucleation was observed to occur at higher temperatures than immersion freezing for the same IN." This has been observed for all kinds of contact nucleation, not just 'inside-out'.

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See, as one example, Fukuta, J. Atmos. Sci., 32, 1597-1603, 1975.

Pg. 8, first full paragraph: The discussion of active sites here is incomplete. While it is true that ice has been observed to form over and over again on specific sites on a substrate, there are other cases in which ice did not exhibit a preference. There's a recent article by Gurganus et al. in *J. Phys. Chem. Lett.* which shows no preferred location for ice nucleation from supercooled water on a silicon wafer. You could argue that there's no preference in that case because there were no active sites on the surface of the silicon wafer (though I think that's stretching the concept of active site pretty thin). There is also work (e.g. Bryant, Hallett and Mason, J. Phys. Chem. Solid, 1960) which showed ice nucleation from the vapor where it did not seem to form preferentially at steps, cracks, edges, etc... (at least from the photos they show). I think the discussion of active sites in this paper is superfluous. You don't even need it for the discussion of the singular model. In my opinion, this discussion muddies the waters and leaving it out would improve the paper.

Page 8, last clause on the page: As a counter argument to the claim that the time dependence of ice nucleation by natural IN is negligible, I would offer a recent observation, documented in Crosier et al., ACP, 2011. Ice formation over such an extended time could be due to something that has a low nucleation rate at those temperatures, but which shows up over the time scales they document. (I realize that the authors are not claiming that time dependence in the atmosphere is negligible, but it wouldn't hurt to provide a counter example to the reference in Pruppacher and Klett.)

Pg 10: acronym, IASSD. This is not used elsewhere in the paper and so no acronym needs to be defined here.

pg. 21: A few possibilities are cited in the second paragraph for the "complex surface area dependence" of freezing in the "high surface area regime". I am not convinced by these explanations. Flocculation is agglomeration of particles in a colloidal suspension. Is the concentration of particles high enough in your droplets for this effect? (Estimate

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the number of particles in the 10% by weight samples, perhaps.) In any case, I would expect that flocculation would change the surface area of the powder in the droplet by a little, but not by much. In any case, if there is a change due to flocculation (or settling out), there should be a time dependence since flocculation and settling out occur over a finite time scale. Was that observed?

equation 15: a physical interpretation is given for b, but not for a. You state that a defines the slope of $\ln J$, but what is that physically? If b is the distribution of nucleation sites, what is a? This is an interesting analysis, but you can fit a lot with two adjustable parameters. A physical meaning for b at least provides a toe-hold for eventually testing the idea, but unless a physical meaning for a is found, equation 15 is just an interesting mathematical exercise. (I would also advocate citing Niedermeier et al., (ACP, **11**, 8767-8775, 2011) in this section of the paper. Their paper also attempts to reconcile the stochastic and singular descriptions of ice nucleation.)

Minor comments

There are frequent references to Pruppacher and Klett, Microphysics of Clouds and Precipitation, 1997. That's a 954 page book. Please provide at least a pointer to the chapter you are referencing (section or pages would be better). Just referencing the book doesn't help much. Someone familiar enough with the field to know where to find the relevant material in P&K probably doesn't need the reference, and someone who doesn't know the field that well, needs more than just a reference to the whole 954 pages.

I commend the authors on their decision to use both color and symbol/line style to differentiate their data. There are folks out there who are red-green colorblind, for example.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 22801, 2011.