Review 2 of “Towards parametrising atmospheric concentrations of ice nucleating particles active at moderate supercooling” by C. Mignani et al.

General Comments

I greatly appreciated the detailed responses to my comments, and the subsequent revisions. I have a few issues of concern still, primarily in further discussing the superposition of the D15 parameterization on plots using aerodynamic diameter as the basis, and the *cf* factor applied in using that parameterization. In combination, these two factors will, I believe, bring the parameterization nearly fully in line with the observations under SD conditions. This is as it should be, if the parameterization has any validity. One could imagine that it should fail in capturing all dust INPs because it does not fully capture those active larger than 2.5 microns (aerodynamic), but I have two comments about this conjecture. First, the D15 relationship was developed from both free tropospheric (elevated from the ground) and laboratory data for which there was a strong relation with >500 nm particle concentrations under situations that were totally composed of, or strongly perturbed by, desert dust. In fact, I think that the present study finds the same thing! Note the equivalent correlation coefficient for your linear regressions in SD situations, for INPs versus 0.5 and 2.0 micron aerosol concentrations. This would not be the case if INP concentrations under SD situations were dominated by particles larger than 2.5 microns, would it? This brings me to the second point. The present studies say nothing about the actual size of INPs at the site used in this study. Some investigators have found larger INPs at surface sites, albeit likely because of influences of INPs that are <u>not</u> mineral dust, and partly because the measurements are focused in the near-surface boundary layer where larger particles are always found. Mineral dust is clearly not the major contributor at the site in this paper either, except under SD scenarios. What the current study does show is that in order to obtain a clean relation between aerosol concentrations and INP concentrations under most situations, one must reference a larger particle size for parameterization. That does not permit an assured conclusion that the INPs are always typically larger than 2 microns. You said it yourself that INPs are but a minute fraction of the total aerosol. It could simply mean that other factors enter to populate the smaller size ranges with particles that are not INPs and do not vary in-kind with them. This is another important distinction in my opinion. I feel that you confuse the issue in deciding that larger INPs are the reason that you have to go out to 2 microns to get a good correlation.

Specific Comments

1) Regarding changes made about previous literature on the role of ice formed and growing at temperatures above about -15°C, I have a suggestion. The meaning of the revised statement that “…although other temperatures would benefit from future investigations” is somewhat ambiguous. I think you could say that investigations would benefit from relation of measurements to overall cloud thermal structure, which may at times include lower cloud top temperatures.

2) Related to making clear that D15 is strictly for application on mineral dust dominated populations, somewhere around the introduction of Equation 1 it needs to be stated that the equation will here be applied to all particles at sizes larger than 500 nm. I understand that the concentration parameter is explicitly defined in Equation 1, but I mean that it should be said in

words that although the parameterization is strictly for mineral dusts, it will be applied to all particles. The reason for doing this is to note later that one only expects this parameterization to be valid as related to data under strong dust influences (e.g., SD here).

3) The justification for *cf* = 1 is incorrect, I believe. You say that no calibration factor is required "because INPs were observed in immersion mode (via a drop freezing assay) and not for instance, in a continuous flow diffusion chamber, where…only part of the INPs passing the instrument may become immersed in liquid droplets." This is exactly why *cf* = 3 is needed. If one were comparing data directly from a CFDC to the parameterization, then *cf* = 1 is what you would want to use, as was done in that paper in 2015. If one is using the parameterization in a model and applying it to the dust distribution, or if one is comparing to a method that captures all dust INPs active by immersion freezing, then *cf* = 3 is what one wants/needs to use. It is the full intention of the parameterization, based strongly in the results presented in that paper.

4) Regarding the explanation of the upper bound of concentrations, was dilution not possible, or simply determined not to be desirable? Perhaps say that dilution was not used in order to extend the upper range.

5) Regarding the use of aerodynamic diameter because there is no way to know shape factors and density, I want to stress again that for the purpose of showing the D15 prediction, this paper should be focused on comparison to mineral dust dominated cases. Hence, while I agree with the fact that this would be difficult for application to all unknown particle types, I think there is quite a bit known and often assumed for dust relevant shape factors and densities. So the statement that "If actual particle densities were mostly > 1 g cm^{-2}, our [$n_{0.5}$] would be somewhat higher than if they would have been calculated as physical particle diameters" is unsatisfying. Without showing the size distribution, one does not know how particle numbers fall off with size. Could it amount to a factor of 2? For SD, I think it easily could. For SD especially, I think that shape and density factors are probably fairly well constrained, and a 542 nm aerodynamic diameter could easily be 380 nm. It is simply a misapplication of the parameterization to use aerodynamic diameter in it and compare to aerodynamic number concentrations. If you want to argue that aerodynamic diameter is more suitable for parameterizations, that is another matter. Using a relevant assumed shape factor (perhaps 1.3) and density (perhaps 2.6) for SD in order to correct the parameterization to aerodynamic size and concentration space, this will move the D15 line to the right in the plots. How much? Using your data for SD number concentrations at two aerodynamic sizes and assuming that a linear slope exists between them (possibly not a good assumption, especially for estimated smaller size particle concentrations where the distribution may be steeper), I estimate a 2 factor concentration push of the D15 curve to the right (i.e., 2 per liter where you have plotted 1 per liter).

6) When you consider the above conservative estimate of how the D15 curve needs to be pushed to the right, and the fact that you should be using cf = 3, I would judge that your data for SD episodes are completely in line with D15 in Figure 3. I am less sure how to fix Figure S3, but perhaps that is fixed if the D15 number predictions are fixed. This means as well that the D15 parameterization grossly under-predicts INPs in other standard conditions. This makes total sense to me for situations where biological or other INPs dominate.

Making these changes is simple, in my opinion, and then the D15 comparison only focuses on dust scenarios, and the other significant results in the paper (larger size relation to total INPs needed for this and possibly other sites, and the apparent role of biological INPs) remain unimpeded by this focus on a parameterization that does not account for them.