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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Chemical characteristics of ice residual nuclei in anvil cirrusclouds: evidence for homogeneous and heterogeneous ice formation" *by* C. H. Twohy and M. R. Poellot

Anonymous Referee #4

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This manuscript provides an interesting analysis of anvil ice residual nuclei and implications for ice nucleation processes in deep convection. New information contributing to the understanding of ice nucleation is presented. The manuscript is well written and very suitable for publication in ACP. However, I would like the authors to consider the following comments.

1. A key question in terms of the implications of the results is what fraction of the total ice crystal residual population is missed by the impactor/electron microscope analysis. The concentrations of particles detected is shown in Figure 1. Would it be possible to

provide an estimate of the uncertainty in these numbers? The comparison with FSSP ice concentrations in Figure 1 and discussion on page 3728 suggest that the fraction sampled is somewhere between 1/2 and 1/6, depending on possible overestimation of ice concentrations by the FSSP. There has been considerable debate recently about crystal breakup resulting in anomalously high concentrations reported by FSSP-type instruments. I think it should be acknowledged that the FSSP ice concentrations could be exaggerated by more than the factor of 2 suggested by *Field et al.* [2003], with the possible implication that the current analysis includes residuals from most ice crystals present. If this turns out to be the case, then the results here imply that most of the ice crystal nuclei come from the boundary layer, in contrast to the *Fridlind et al.* [2004] modeling results. You might comment on this possibility in the discussion of how these results compare with the *Fridlind et al.* [2004] conclusions on page 3731 (26-29) and page 3732. Note that the conclusion reached by *Fridlind et al.* [2004] that mid-tropospheric aerosols entrained into convective plumes are an important source of nuclei for anvil ice crystals also hinged largely on the FSSP (and similar light-scattering probes) measurements of ice crystal concentrations and size distributions. Hence, our understanding of what aerosol sources (and what types of aerosols) dominate ice nucleation in deep convection hinges, unfortunately, on the accuracy of FSSP-type instruments in ice clouds.

2. Along the same lines, an important issue discussed in the manuscript is that residuals smaller than about 0.1 μm are not sampled. I suggest mentioning this threshold in the abstract.

3. Page 3732, lines 16-19: I would think that the contrast between the measurements (mixture of anvil ages) and model results (fresh anvils) might only make the aerosol source discrepancy worse. Those crystals that survive in the anvils should be the smaller ones that nucleated later, presumably by homogeneous freezing on sulfates entrained into the plume above the boundary layer. However, the measurements in a mixture of anvil ages generally suggest a larger fraction of crystals nucleated on

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boundary-layer aerosols than the model results did. Perhaps this discrepancy relates to a bias toward sampling of the lower parts of the anvils by the Citation. The lower parts of the anvils would generally be more heavily populated with large crystals nucleated lower in the convective updrafts, allowing time for growth and aggregation to the larger sizes.

4. The finding that about a third of the residuals are salts is generally consistent with *Cziczo et al.* [2004] and strikes me as a somewhat surprising result. These results suggest that sea salt is perhaps a more important source of homogeneous freezing nuclei (compared to sulfates) than has previously been recognized. A comment to this effect might be worth including in the manuscript, and the fact that a third of the residual particles are composed of salts should perhaps be mentioned in the abstract.

5. The percentages in Figure 2 presumably represent averages from the 14 residual samples and 4 ambient samples. How much sample-to-sample variability was there?

6. Last paragraph on page 3733 (discussion of Figure 4 left panel): I don't see the transition at about -35 C to homogeneous freezing as being quite so clear as the text suggests. There is one data point at about -21 C dominated by insoluble nuclei, then several points at T < -35 C with soluble/insoluble ratios near unity but no apparent temperature dependence. Without data points between -21 and -35 C, it is difficult to conclude that the transition occurs at -35 C.

Citation note: You might also cite *Yin et al.* [2005] who also found that entrained aerosols are important in deep convection.

References:

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