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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

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We thank all the reviewers for their thoughtful comments. Several reviewers pointed out the same criticisms of the paper: the difference between sampling temperature and ice nucleation temperature, and how uncertainty in the particle concentration and sizing is addressed. While these are fundamental limitations of the data set itself, we are revising the paper to treat them differently, as described below. We have also revised most of the figures and added an additional table in response to referee’s concerns. Their comments are discussed below, in the order submitted to ACPD.

Response to Referee 1: Major Comments

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1) The reviewer and others are correct that the temperatures used in the paper are the sampling (environmental) temperatures, and not necessarily the nucleation temperatures of the ice that we sampled. The fact that the colder environmental temperatures do exhibit different nuclei types suggests that a relationship with nucleation temperature exists, but it is true that this relationship is uncertain and difficult to quantify. For example, the coldest anvils were from well developed cells where the ice-forming air must have been exposed to cold temperatures in order to reach these levels. Anvil ice sampled at warmer temperatures may have been from smaller cells where air never reached the homogeneous freezing temperatures, but may have also sedimented from above in larger storms. This question could possibly be addressed through detailed analysis of anvil evolution using radar data (when available) or complex model studies which are beyond the scope of this paper. In order that others might study this further, we have added a table (Table 2) that includes sampling dates and times which are cross referenced on Figure 4. We have also changed "nucleation temperature" to "environmental temperature" where appropriate, and added a brief discussion of their possible, but uncertain relationship.

2) We thank the reviewer for suggesting that there may be additional useful information in the sizes measured by electron microscopy and collected by the impactor. We have looked thoroughly at this data and feel that there are a number of sources of potential error in determining size distributions by this method, and for this reason, choose not to add size distributions to the paper. However, we will provide a summary discussion here.

There are at least four types of error in determining size distributions from the impactor/microscopy measurements: 1) using a 2-dimensional size as it appears on the grid, 2) possibly undercounting small particles due to the tendency to choose larger particles that are more visible to the human eye, 3) undercounting small particles due to their greater likelihood to be vaporized by the vacuum environment and/or the electron beam, and 4) bias in sizing due to preferential sorting of particle sizes as a function of location below the jet itself. The first error can be corrected for to some extent through composition-dependent spreading factors such as determined by McInnes et al. (1997). When we apply these to our data for sulfates, organics and salts, still less than 2% of the observed particles are smaller than 0.1 microns diameter. The distribution peaks at about 0.22 microns and shows little variation as a function of temperature. It is possible that additional corrections are necessary for crustal and metallic particles that may lie flat on the grid, and so appear larger than their equivalent spherical diameter. This additional type 1 error, as well as the type 2, type 3 and type 4 errors mentioned above, are not readily quantified. For this reason, we are not comfortable with including size distributions determined in this way in the paper. However, since most of these errors create a potential bias toward undercounting small particles, the possibility that we are missing particles smaller than 0.1 microns cannot be discounted.

3) It is true that the number concentrations given in Fig. 1 are highly uncertain, especially those from the FSSP. We really don't know the true values, and uncertainty in concentration of small ice crystals in cirrus is one of the greatest handicaps we face in working with these data sets. New instrumentation that avoids the potential ice crystal breakup problem is greatly needed and is becoming available. The point of Fig. 1 is to show that there is no strong evidence that the number of residual nuclei sampled on the CVI microscope grids was enhanced due to crystal breakup. We have added estimated error bars to Fig. 1 and reworded this section, as suggested by the reviewer, to make this point more clear. We have also discussed the 2D-C data in more detail. The other questions at the end of this reviewer's comment are discussed throughout this response.

Response to Referee 1: Minor Comments

1) We have reworded this statement to reflect the uncertainty in the data.

2) We have re-arranged this section to hopefully make the discussion clearer.

3) We have included both the aerodynamic unit density diameter, as well as the diam-

eter for a larger density typical of some of the particle types we sampled.

4) This counting process was very time consuming, and as a result, was not done for all samples but for a subset. If we had done the count on more samples, we would have had to do chemical analysis on fewer samples, which we felt was less desirable. This was not explained in the paper (our oversight), but is now briefly discussed in the Figure 1 caption.

5) We have added the Jost et al reference; thank you.

6) We feel that the data from a primarily liquid cloud at much warmer temperatures provides a contrast and would be of interest to many readers. It illustrates an interesting point that in the atmosphere at low levels, droplet nucleation occurs primarily on soluble material, at mid levels ice nucleation occurs primarily on insoluble material, while at high levels the nucleation agent switches back to soluble material. Since inclusion of this sample was also questioned by Referee #3, we have discussed its significance further in the text. We have also removed this sample from the average compositions shown in Fig. 1, and eliminated the reference to it in the Summary and Conclusions.

7) Done, thanks.

Response to Referee 4

1) This comment is similar to Major Point 3 of Referee #1. We agree with the reviewer that the factor of 2 FSSP uncertainty may be too small, and have reflected a larger uncertainty in Figure 1. This and the inclusion of the recent Yin et al (2005) paper mentioned has changed our discussion of the comparison with Fridland's results, along the lines of that suggested by this reviewer.

2) We have now included this 0.1 microns threshold in the abstract.

3) The reviewer is right that the difference in size distribution with sampling altitudes, and the tendency of the Citation to sample the lower and middle parts of the anvil, may cause our results to differ from those predicted by the model. Based on earlier

comments, this section has been modified to de-emphasize the model/measurement comparison.

4) You are correct; this is an interesting and unexpected result that has been emphasized in the text and added to the abstract.

5) There was substantial sample-to-sample variability, as is implicit in Figure 3. We have also added a statement to this effect.

6) The trend is more clear in the small residuals (right plot of Fig 4) than in the large residuals (left plot) for reasons we have speculated at in Section 3.2. We have reworded this section slightly to make this clearer. It certainly would have been nice to have samples between -21 and -35C to fill in this region.

Citation Note: We have added the recent Yin et al. paper, which is very applicable to our results, thank you.

Response to Referee 3

1) This comment is similar to Major Comment 1 of Referee #1 (see that response). While we do not feel we have enough information to specify an error bar for sampling temperature vs. nucleation temperature, we have changed "nucleation temperature" to "environmental temperature" and modified the text accordingly.

2) Thank you for the additional interesting references. We have included the Zuberi et al. (2002) paper in the final manuscript where it applies to some of our results. We have chosen not to include individual laboratory studies in the manuscript's introduction, but focus on experimental results instead.

3) We are not sure we completely understand this comment. We have added the lower cut size of the CVI (5 microns diameter) to the paper. The upper cut size is less well defined, since in order to measure total ice water content, we do not employ a "waste flow" for larger particles such as that employed in the Ström and Cziczo studies. Crystals larger than approximately 50 microns aerodynamic diameter are expected to

impact or possibly breakup up inside the CVI inlet, with only partial collection efficiency for their nuclei. However, since the majority of ice crystals are smaller than this upper limit (c.f., Fig. 1) for all cases used, this should not bias our results, which are based on number concentration.

4) See response to Referee #1, Minor Comment 6.

5) We have modified our conclusions to reflect the uncertainty in the measurements.

Response to Referee 2

The particle size information has been added to the abstract.

The FSSP size range is approximately 3-54 microns diameter and the 2D-C measures crystals larger than about 30 microns. This information should have been included, and it is now in the final version of the manuscript.

Including error bars in Fig 1, as suggested by some of the reviewers, makes the discrepancy between the various instruments substantially less. See response to Referee #1, Major Comment 3 and Referee #4, Comment 1. Unfortunately this major source of uncertainty will have to be resolved with future work.

We did not see a large difference between ambient and CVI residual characteristics for the reasons discussed in the manuscript. We can, however, compare the percent of residual particles expected to be non-volatile with the Siefert et al. (2004) results. They found, in the Northern Hemisphere, that 10-30% of cirrus residual particles were non-volatile, with little dependence on temperature. Our cirrus data did show a temperature dependence (small residuals, Fig. 2), with 27-53% crustal and industrial residuals (median 44%) at $>$-38C and 6-30% crustal and industrial residuals (median 18%) at T$<$-38C. Since most of the salts we sampled are also expected to be non-volatile, including salts would increase the percent of non-volatile residuals to $>$50% in both temperature regimes. This is probably not relevant for the Siefert et al. results, however, since they specifically excluded convective cirrus cases where surface sources of salts might be important. Since salts are soluble (and so can act as homogeneous freezing nuclei) yet also are non-volatile, the thermal volatility technique may not be suitable for distinguishing heterogeneous from homogeneous freezing nuclei in anvil cirrus.

References

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