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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 #1

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Overview: I find this to be an excellent, well written manuscript. This paper is extremely well suited for ACP and will make a nice addition to the literature on atmospheric ice nucleation. The reference list is well thought out and thourough. The data collection method is well presented and the results are of importance at a time when aerosol - cloud interactions are at the forefront of atmospheric science. I have a few comments relating to stating uncertainty and the data presentation but expect these can be rapidly addressed by the authors.

Major Comments:

1) In the Abstract (Page 3724 Line 8; repeated in text, e.g. Page 3726 Line 3): The temperatures (-21 to -56 deg. C) are not the nucleation temperatures but sampling temperatures. The authors have not specified that they are nucleation temperatures but the language used in the paper - for example stating the homogeneous freezing onset immediately after the sampling temperature and using this as the temperature in Figure 4 - incorrectly implies that these temperatures are, at the very least, closely related. This is further confused in the Results (Page 3733 Line 3) when the authors group categories according to freezing mechanism and then use this temperature for Figure 3. Since the sample temperature is really a 'snap-shot' of what the cloud looked like but does not relate information about the history I suggest a clarification that this temperature likely - but does not certainly nor always - relates to nucleation temperature. Put another way there must be a difference between the sample temperature and the temperature at which nucleation occurred. What is it? Can it be either positive or negative? How certain is it? The end of 1st paragraph of 2. Experiment would be a good location. See also Minor Point 1).

2) One of the overriding uncertainties in this paper regards the size spectrum of ice residue. Can the authors add a figure of the total size spectrum for this data? The TEM used is specified by the authors to provide size information (Page 3726). This is of importance is that the authors suggest these data are not in agreement with Fridlind et al. [2004] (Page 3731) in that the ice residue was more like lower-troposphere, not mid-troposphere, aerosol. Later (Page 3732) the authors hedge this conclusion by suggesting a large quantity of small residue below the size cut and/or too volatile to be detected may have been present. This leads to a strangely worded section that first appears to suggest certainty in this data set then leads the reader to wonder if any of the conclusions drawn are valid if 2/3s of the residue is missing. If there are many small ice residuals then the size distribution of sulfate/organic particles should fall off with a 50% cutpoint at 0.07 micrometers (the lower cut of the 'small' impactor). The authors suggest only 1% of the residue is below 0.1 micrometers so this does not appear to be a viable option and could be shown with a size distribution plot. It then must be

assumed that all small particles are highly volatile sulfates/organics, despite the fact that a) one would expect other particles to also be present in this size range and b) sulfate/organic particles were not too volatile to be detected in the ‘large’ and ‘small’ bins. I am left to conclude these data appear to contradict Fridlind et al. [2004] and Seifert et al. [2003]. I would suggest the authors reconsider the implied uncertainty in this section and instead make a stronger argument to show the quality of their data set.

3) Figure 1 and 6th paragraph of Experiment: The authors show FSSP, CVI, and 2DC data using a log scale and use this to suggest crystal breakup (i.e., more residue than original crystals) is not significant. The authors go on to state that if the FSSP over-estimates crystals by a factor of 2 then these data represent 1/2 to 1/3 of the crystal residue. The implication that the uncertainty associated with the FSSP is 2 for this mission is extremely misleading. No error bars are plotted in Figure 1. In truth the response of the FSSP to the ice crystal ensemble in an anvil cirrus, or any ice cloud, is highly uncertain and very likely much larger than 2. To attempt to draw conclusions in this manner, using a log scale and large factors, is not appropriate. No explanation is given for the 2DC appearing in this plot or the meaning of the factors of difference between this and the CVI and FSSP. This section either requires a major rewrite concerning the certainty and limitations of FSSP and 2DC errors and/or modified language when attempting to use these very uncertain numbers to support contentions that the data are or are not in agreement with other studies.

In reading this paper, specifically with respect to the points made in Major Comments 2 and 3, I was left without a clear understanding of how valid these data are and the level of uncertainty. Without reiterating specific points made in these Major and Minor Comments it seems that the authors attempt to simultaneously make firm statements and to agree with all the previous literature to the point of obvious contradiction. I think this would be a superior paper if the authors clearly specified the uncertainty in the measurements (How certain are FSSP counts? What does 2DC date tell us? What material is too volatile in a TEM? What is the size distribution of the ice residue and how does this compare to the suggested 50% cutpoint? What percentage of the ice residue is sampled and with what certainty?) and then draw conclusions that either agree or disagree with the literature in the Summary and not sporadically throughout the text (Are there indications of many small ice residuals from this data or is this speculation? Are there indications of material from the mid-troposphere or, again, is this speculation? How certain is the correlation of ice residue with temperature? Put another way, what is the possible difference between the temperature at the time of sample and when nucleation occurred?).

Minor Comments:

1) In Results (Page 3733 Line 29): The authors suggest these to be the first measurements to show the heterogeneous to homogeneous freezing transition in anvil cirrus. While there is no doubt this is a terrific data set this statement is misleading. As pointed out in the Major Comment 1) the authors have not shown the nucleation temperature, which would be required to unambiguously demonstrate the heterogeneous to homogeneous transition, but instead a sample temperature. Second, as pointed out in Major Comment 2) the authors suggest they may not be able to detect 2/3's of the ice residue in which case they can also not make the argument they could show the heterogeneous to homogeneous transition.

2) In Results (last two paragraphs): The authors seem to imply what is stated in Major Comment 1) and Minor Comment 1) in that the small scale dynamics and the temperature history of the cloud can lead to biases in the data. I believe the trends shown in Figures 3 and 4 are correct but I think these two paragraphs, along with addressing these comments, could make this section much clearer.

3) In Experiment (Page 3726 Line 11): The impactor cut size is specified for a density of 1.7 g / cc. This is a strange choice since impaction is normally specified with an aerodynamic diameter (density unity). While the density of sulfate, ˜1.7, might make sense under other circumstances it does not here because the authors go on to talk

about mineral, industrial, and soot particles to which this density has not relation. I suggest replacing this with aerodynamic diameter and unity density.

4) In Results: Why are 7 samples used in Figure 1 but 11 in the results?

5) In Results (Page 3730 Line 6): The Hudson et al. [2004] references is good but for CRYSTAL please consider the treatment by Jost, H.-J., et al., In-situ observations of mid-latitude forest fire plumes deep in the stratosphere, Geophys. Res. Lett., 10.1029/2003GL019253 (2004).

6) In Results (Page 3733 Line 10): The use of data from a mixed phase cloud seems confusing and out of place in this paper which otherwise deals exclusively with ice nucleation. Is there a reason it is included? If not then it should be eliminated for clarity.

Typographical/Grammatical:

Page 3731 Line 14: ‘This is probably especiallyĚ’ is a strange phrase. Change to ‘This could be especiallyĚ’

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 3723, 2005.