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Interactive Comment

Interactive comment on "Ice nucleation by surrogates for atmospheric mineral dust and mineral dust/sulfate particles at cirrus temperatures" *by* C. M. Archuleta et al.

C. M. Archuleta et al.

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General comment:

We greatly appreciate Dr. Hung's expressed concerns and corrections and offer the following specific responses.

Specific responses:

1. There must be some typos in Table 2. The vapor pressures for both equations become negative when T is less than...

Quite right. A lot was missing there, including the exponentials. These will be revised



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esw [mb] = 6.1378 exp(22.542*(T[730;C]/(T[730;C]+273.48))) esi [mb] = 6.1378 exp(179662*(T[730;C]/(T[730;C]+247.15)))

2. Because the TEM image in Figure 2 shows that the aggregate of particle which is not spherical and the authors observe the thin coating of sulfuric acid can affect the ice formation in different behavior based on the mineral core in Figure 3, the authors should discuss if the treated particle is only partially coated by sulfuric acid and the ice nucleation from the treated particles is contributed by both coated and uncoated portions. In such case, is the mechanism of ice nucleation for the treated particles a combination of deposition and condensation nucleation?

Discussion will be added in this regard. We will not speculate further about combinations of mechanisms, since the nature of "deposition" is not at all clear to us.

3. Lines 15-18 in page 3410 are not correct. The nucleation rate should not be dependent on the ice fraction and also the retention time.

We believe that the statements have been misinterpreted. The observable nucleation rates (cm-2 s-1) for a given set of conditions are most certainly dependent on the characteristics of the particular device employed. For example, the CFDC instrument is not capable of easily resolving nucleation rates below about 105. This is simply a matter of statistically detecting small numbers of nucleated ice crystals (with an optical detector) from a population of liquid aerosol particles that is purposely limited to prevent mass transfer impacts on steady-state relative humidity conditions. Concentrations of aerosols are typically a few hundred cm-3. Due to the nature of the flow tube design and detection method (long growth times for nucleated particles and the limitation of nucleation by vapor phase mass transfer), rather small fractions of an aerosol population (of high number concentrations) activate and deplete the vapor phase through growth to cause a large "ice" signal. The role of residence time in defining the range of nucleation rates is less clear for the aerosol flow tube system of course than it is for

the CFDC, where conditions are maintained through the growth region. Nevertheless, the consequence of these experimental factors is that the range of nucleation rates assessed is different in the two studies. This is the essential point being made although this section has been rewritten for clarity.

4. The data points in Figure 8 for Hung et al are not consistent with the original data. Please revise them.

There was a slight error made in reformatting an earlier revision of Figure 8 and we thank the commenter for pointing this out. Four data points for each water activity are now shown in the corrected plot, corresponding to the four mode sizes (50, 120, 150 and 200 nm) of aluminum oxide particles used by Hung et al.

5. There are several points should be explained as the authors compare their results with Hung et al as following:

a. Sulfuric acid vs. ammonium sulfate. Is the heterogeneous ice nucleation from both solutions comparable?

In our laboratory and with the CFDC instrument, we see no statistical difference in the impact of solute on freezing nucleation, at least homogeneously (Chen et al. 2000). If water activity is a factor in freezing, as the data suggest, there would be expected to be similarly little difference for heterogeneous freezing. Nevertheless, we will note the potential for differences, especially since there is a deliquescence limitation for ammonium sulfate.

b. The thin sulfuric acid coating compares vs thick ammonium sulfate coating. In thick ammonium sulfate coating, there is mainly condensation nucleation. In the thin sulfuric acid with non-spherical core, the sulfuric acid solution may accumulation in the concave area and the dust core may not be completely coated. Deposition nucleation could play an important role in the thin coating.

At the point of CCN activation, the particles will have taken up large amounts of water

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and so the non-sphericity is likely only of modest impact. At the point of ice nucleation in these studies, the expected diameter growth factor of particles due to water uptake is on the order of 1.3. At this point, one might expect a full liquid coating on the particles and, possibly, some collapse of the structure of the small agglomerates (see comment to S. Martin on the morphology of agglomerates). We feel that any discussion of the possibility that nucleation by deposition could play is too speculative for inclusion.

c. The authors should discuss the possible impact of the non-spherical shape because it can increase the surface area and further reduce the j values in the results.

Yes, we should and we now do. A conservative estimate of the potential overestimation or underestimation of nucleation rate is a factor of 3 to 5. This will not drastically change any of the conclusions, except to bring the nucleation rate results of the two studies modestly closer. We add details in response to the comments of Scot Martin and tend to agree with him on the fact that the agreement between the two studies is actually remarkably good. Consequently, we will attempt to remove from the paper any comments that could appear unjustifiably judgmental.

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