

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.

S. Martin (Referee)

smartin@deas.harvard.edu

Received and published: 9 June 2005

Review prepared by Scot Martin (anonymity waived)

Archuleta et al. provide a thorough experimental account with advanced data analysis on the topic of heterogeneous ice nucleation by mineral dust particles and their surrogates. An important feature of the manuscript is the expression of the freezing results as ice-relative-humidity, freezing temperature, water activity at freezing, and surface-normalized nucleation rates. The authors ensure that their results will be readily understood by a diverse array of scientific camps. Moreover, they are able to make

Full Screen / Esc

Print Version

Interactive Discussion

Discussion Paper

quantitative comparisons to previously published accounts, namely those of Zuberi et al. and Hung et al.

In reading the manuscript, I have the following questions:

1. The primary crystallite size ranges from 19 to 39 nm (page 3995) whereas 50 to 200 nm particles are studied. Therefore, the studied particles appear to be agglomerates, which is supported by the TEM image shown in Figure 2. Should the authors consider addressing how the nonsphericity affects the interpretation of results? In several places in the analysis, spherical particles are assumed.

2. The authors appear to conclude that the results of the noncoated particles are difficult to interpret. Should these results then be omitted from the paper? What added value do these results provide given the uncertainty in their interpretation?

3. It seems that the results of uncoated Asian dust are compared to those of coated synthetic particles. Does this comparison make sense? Shouldn't the Asian dust particles also be coated? The methods section describes that these Asian dust particles are obtained freshly from collected soils, whereas the Asian dust particles over the Pacific and arriving in the USA are coated by sulfates and nitrates. Dust from northern Africa is similarly coated. At the least, if no more experiments are to be done, should these issues be discussed in the paper?

4. The quantitative agreement in j values of this manuscript and those of Hung et al. is actually very, very good (Figures 8 and 9). There are only two orders of magnitude difference, which is very little in the history of nucleation studies (e.g., the many orders of magnitude in the literature regarding rates of ice formation in water). Do the authors want to remind the reader of this?

5. The authors consider several important factors in the comparison of their results with those of Hung et al. One point that is not explicitly considered is the effect of nonspherical & agglomerated particles versus spherical particles (i.e., those of Hung

Full Screen / Esc

Print Version

Interactive Discussion

Discussion Paper

et al.). Do the authors want to mention this point and possible implications, too? For example, agglomerated particles may also provide additional active sites in concave dimensions, which could increase j values.

6. The authors do an excellent job of providing different perspectives on their results (i.e., the expression of their freezing results as ice-relative-humidity, freezing temperature, water activity at freezing, and surface-normalized nucleation rates). Do the authors want to cite a related effort that some readers might find helpful: Hung, H.M., and Martin, S.T., "Apparent Freezing Temperatures Modeled for Several Experimental Apparatus," *Journal of Geophysical Research*, 2001, 106, 20379?

7. I wonder about the comparisons to homogeneous freezing of pure sulfate particles. I understand that the authors want to provide a baseline comparison point between homogeneous and heterogeneous freezing. My first question is if the comparison is based upon the volume-equivalent of the sulfate layer or on the volume-equivalent of the entire particle. Pages 3401/3402 sound one way and the abstract and caption of Figure 3 sound the other way. My second question is, Does either answer to the first question make sense as a comparison point to heterogeneous freezing? It seems to me that a full parcel model with initialization conditions of a scenario (similar to what the authors have done in the past) is necessary to make any meaningful comparison between homogeneous and heterogeneous nucleation. Can the authors clarify these points?

8. Page 3396. Is there any information on the dispersion in CCN activities? If so, does this dispersion have implications on the interpretation of the IN results? It seems that 50% (CCN definition) is being used as a basis to interpret 1% (IN definition), which would only be valid if the CCN dispersion is tight.

9. Page 3405. Should ΔT_{hf} be ΔT_{hetf} ?

10. Page 3405, line 19-21. I do not understand this sentence. Can it be re-phrased or explained differently?

Full Screen / Esc

Print Version

Interactive Discussion

Discussion Paper

11. Page 3406, line 10. Should the functional relationship for J_{hetf} include λ inside the parentheses?

12. Page 3409, eq 4. Should J_{hetf} be $J_{hetf}(A)$ since page 3408 makes the point that the number of active sites and hence J_{hetf} depends on A ?

13. Page 3410, lines 15-18. The sentence as written may explain the difference if observation time (13 versus 60 sec) is the governing explanation (i.e., observation time up leads to sensitivity to smaller j values); however, a 5-fold difference in observation time would not seem to explain the >1000 -fold difference in activation fraction. An alternative interpretation is that a small fraction of the particles have high activity (i.e., $j_{hetf}(A)$), in which case the Hung et al. measurement should capture higher nucleation rates than this manuscript's measurements. The observations are in opposite of this explanation, unless the possibility exists that the particles of this manuscript are different than those of Hung et al. because of the difference between agglomeration and single particles. In this case, the agglomerates would nucleate faster than the single particles, which could explain the differences described on page 3410. Should the authors consider this line of thinking?

14. Table 1. Should the authors consider the effect of water (i.e., equilibration with ambient relative humidity) on coating thickness?

15. Figure 2. Can the dispersion in CCN activity be represented on the figure?

16. Figure 6. Should the "bulk freezing" line instead be called the "bulk melting" line?

17. Figure 8. Why are there two sets of diamonds?

18. Figure 10. Should squares and triangles be distinguished in the figure caption?

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 3391, 2005.

Full Screen / Esc

Print Version

Interactive Discussion

Discussion Paper