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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.

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General comment:

We appreciate the comments made on the potential utility of this paper and wish to acknowledge the reviewer's concerns regarding details of aerosol generation. There is always a fine balance between providing detail of this type and maintaining readability. We respond below to the specific comments and note where changes to the manuscript will be made with regard to these.

Specific responses:

1. *Was a neutralizer installed in front of the DMA? Fig.1 does not show it explicitly. If the aerosol is not neutralized, I would expect a significant number of multiply charged, larger particles of the same mobility at a given DMA voltage setting. Therefore, a significant number of also larger particles would have entered the CFDC along with those of the pretended size. The TSI atomizer is known to produce highly-charged aerosol (Forsyth et al., 1998).*

A charge neutralizer is a standard component of the TSI 3071 DMA system used. Where air enters the instrument there is a Kr-85 source. This will be stated explicitly now in the text. Nevertheless, it should also be understood that multiply-charged particles are a concern even with a neutralizer. This factor was considered for this study, as fully documented in Archuleta (2003), the thesis from which this paper derives. We now reference this document and will provide documentation in the text about the predicted percentage and sizes of multiply charged particles for selected monodisperse sizes. We also offer to provide the size distribution for the Asian dust particles. As discussed below, most supermicron particles are removed during atomization and prior to the DMA. Although the original size possessed a number mode above 5 microns, this large size mode was effectively eliminated by impaction in advance of the DMA. Further, the small size mode of the dust was centered below 0.05 microns and thus multiply-charged particles were less of an issue for this sample than for the manufactured metal oxide aerosols. For the Asian dust sample in particular, "50 nm" particles has 95/5/«1 percent singlets/doublets/triplets (50, 72, 90 nm), "100 nm" particles had 86/13/1 percent (100, 149, 191 nm), and "200 nm" particles had 94/6/«1 percent (200, 317, 426 nm). We will list this information in the revised paper, and discuss in the results section why the singlets still provide the predominant contribution to ice formation.

2. *Asian dust aerosol is typically poorly water soluble (Maxwell-Meier et al., 2004; Mori et al., 2002). The authors used an aqueous suspension of the dust to generate the aerosol. After evaporation of the droplets leaving the atomizer one would expect a bimodal size distribution (similar to the dispersion of PSL suspensions). The first mode is*

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expected to be somewhere below 100 nm particle diameter due to particles composed of the soluble material from the dust (CaCO3+CaSO4 internal mixture). The second mode is expected to represent particles consisting of a non-soluble core of one or two dust particles partially covered with CaCO3+CaSO4. The size of this mode is rather hard to predict because it depends on the properties of the dry dust powder and the properties of the suspension (degree of disaggregation etc.). Therefore, the way the aerosol has been produced induces further changes to the size dependent chemical composition. First, this could explain the observed hygroscopic growth factors of 1.155, 1.125 and 1.05 for 50, 100 and 200nm particles, respectively (p.3402, 10). Smaller particles contain water soluble material and grow under increasing RH conditions whereas larger particles are basically insoluble and grow very little. Second, the question arises to what degree the ratio of soluble material to mineral oxides, which might affect the IN properties, has been 'authentic' in these experiments. So it would be useful to provide more details about the aerosol generation and characterization, if available, or caution the conclusions in view these aspects.

We are happy to provide additional details regarding aerosol generation that are of concern to the commenter and potentially to other readers. We first note that what one expects and what one obtains in generating aerosols from solutions can be two different things. In this study, we clearly were dealing with an aerosol distribution that was mono-modal below 300 nm and dominated by insoluble species. A new figure will be added to show the size distribution of the "Asian" dust as produced by the generation system. We will also include a transmission electron microscope (TEM) image of a typical particle natural dust particle as generated and discuss the elemental spectra via EDS. The natural sample originally contained a primary size mode at about 15 microns, but was subjected to an impactor to remove particles above 10 micron before being recollected and sent to our laboratory. Additionally, the atomizer design and an impactor not shown in the original schematic served to effectively remove large aerosols. The net result was a size mode at about 35 nm, with an order of magnitude fewer particles at a size of 200 nm. The HTDMA data clearly support a difference in

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the soluble content, increasing with a decrease in size in this mode of particles. That is why we reported this result and this was simply accepted as a fact resulting from the generation method. There is not a great deal of soluble matter with the original samples and its dilution by nearly 100 times in distilled water is the reason that not much soluble matter remained with the natural dust samples after generation. We now quote the soluble weight percent as inferred from the HTDMA results (50 nm - 5 percent, 100 nm - 4.0 percent, 200 nm - 1.5 percent).

We intended to be careful to state that the size-dependent composition was likely modified by the generation procedure, but we will now add additional words to this effect. In these first studies of ice nucleation by natural dust samples, completed nearly 3 years ago, we were not greatly concerned about the "authenticity" of the particles as regards their exact soluble composition. Further, our experiments and those of others we reference suggest that the exact soluble content is not a critical factor as regards ice nucleation. Soluble content affects primarily water uptake of the particles, but not their water activity, and it appears that the latter quantity is the more relevant parameter. Obviously, the inferred size of the insoluble core is an issue, but the highest weight fraction soluble content for the 50 nm particles still represents less than 5 percent of particle mass. We did not infer strong differences in the core compositions as a function of size from our TEM studies (not detailed here), but we note the larger separation of activation results versus size for the natural sample versus the manufactured particles, so we are now careful to note that this size dependence may reflect compositional differences versus size that do not exactly reproduce what is found for airborne dust. We have begun to address these issues in more recent experiments using dry-dispersed particles.

Archuleta, C.A.: *Ice nucleation by surrogates for atmospheric mineral dust and mineral dust/sulfate particles at cirrus temperatures*, M.S. Thesis, Dept. of Atmos. Sci., Colorado State University, Fort Collins, 108 pp., 2003.

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Interactive Comment

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