

Interactive comment on “Aerosol measurements at a high elevation site: composition, size, and cloud condensation nuclei activity” by B. Friedman et al.

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We thank Referee #3 for reading the manuscript and providing helpful comments; below we provide responses to each comment individually.

This paper discusses observations from four days in March 2011 measured at the Storm Peak Laboratory, Colorado. The CCN-activated fraction and chemical composition measured using a single particle mass spectrometer are presented, including two days during which new particle formation occurred. The main conclusions from the study is that the CCN-activated fraction qualitatively mirrors the fraction of sulphate-rich

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particles and that freshly nucleated particles can activate at higher supersaturations.

These conclusions are not that surprising, but additional analysis could greatly enhance the findings from this study. In particular, the fractional aerosol composition could be used to infer the hygroscopicity parameter of the non-sulphate-dominated aerosol (see specific comments below). I recommend that this paper be published if the comments below are adequately addressed.

Specific comments

Page 18279, line 1 As demonstrated in your references on lines 5–8 of this page, Köhler theory can be used to predict the CCN-activity of aerosol with both inorganic and organic components.

**The sentence has been revised.

Page 18286, line 17 What does a scatterplot of activated CCN fraction versus the fraction of sulphate dominated particles look like? One would expect that they should be well-correlated since all the sulphate-dominated particles should be activating at all your supersaturations.

**We have plotted the fraction of sulfate particle type for each day studied against the activated CCN fraction, and a positive correlation exists ($r^2=0.76$). Conversely, an inverse correlation exists between the activated CCN fraction and the fraction of organic particle types ($r^2=0.84$). These plots (shown below and included with further correlation plots as suggested by another reviewer) have been included in the paper as figure 8, with further discussion in section 4. Given the activated fraction of CCN and the composition data, the “sulfate” particles are not pure sulfate and do not activate as pure sulfate; instead these particles are representative of a mixture of primarily sulfate with some organic components.

Page 18286, lines 18-19 and Page 18287, line 24 The activated fraction mirrors the sulphate-dominated particles most strongly at supersaturations of 0.22% and 0.42%.

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Sulphate-dominated particles do not affect the activated fraction at 0.12% nearly as much because the larger particles should be less dependent on composition.

**Noted and clarified in text.

Page 18289, line 10 Include the work of Langley et al. 2010, who observed increased CCN at 0.34% supersaturation with growing particles over the Pacific Ocean.

**Included.

Page 18289, line 16-17 Köhler calculations show that pure sulphuric acid particles <50 nm would activate at 0.42% supersaturation, so it would not be surprising that the nucleated particles are contributing to the CCN. In fact, it could explain why the activated fractions peak on 11 March before the sulphate-dominant fraction. On the other hand, the work of Shantz et al. 2012 demonstrated a delay in the CCN-activity of freshly nucleated particles. Is this also the case for your events? It is difficult to determine from Figure 2 at what time nucleation started, a magnified figure of the size distribution would be most helpful. Some discussion relating the size distribution of the particles with the CCN-activity should also be included.

**Nucleation started on March 11 (indicated by growth to 20 nm, likely started before that) at ~11am, and is likely contributing to the peak of activated fraction before the sulfate-dominated fraction peaks. It is difficult to assess if there is a delay in the CCN-activity of freshly nucleated particles, as we do not detect the nucleated particles with SPLAT II until they grow to sizes of at least 80 nm, therefore we do not believe we can reasonably address dependence below 50 nm.

Page 18290, lines 2-4 Why do you assume that the aerosol population is internally-mixed when the SPLAT II measurements clearly show that the aerosol are externally-mixed? There is so much more information available with your measurements. For example, on 5 March 11:00, the sulphate-dominant particles reach a minimum of 40% of the aerosol fraction, which means that all particles >80 nm should activate at 0.42%

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supersaturation. The fact that the activated fraction is approximately 40% would suggest that almost none of the particles in the other classes are activating at this supersaturation, and that their k must be <0.15 . The fractional size distributions also contain information that could be used to estimate the k . At the very least, the calculated time-varying κ could be included so that the reader can gauge how it changes with fractional composition.

**It is true that the particles are externally mixed. Each particle is composed of a mixture of compounds and the aerosol contains particles of different compositions and mixtures. For the sake of simplicity we represent the system as internally mixed. Given the measured compositions, size distributions, and activated fractions we do obtain relatively low κ values than we would expect; looking at error propagation for the κ determination method we used, the calculated κ s have very large uncertainties associated with them. The correlation plots shown above show that we do have correlations for particle type composition vs. activated fraction, slightly less correlation with critical diameter, and less so with κ ; these plots are representative of the large uncertainty in the κ measurements and the error propagation in the κ equation.

Page 18290, line 19 Does the average of 0.2 include all five supersaturations, or only some of them? Please specify.

**The average includes all five supersaturations. This has been clarified in the text.

Page 18305, Figure 6 b and d It is interesting that in the early hours of 15 March, the aerosol is 40% sea salt but the activated fraction is so low. Can you comment on this?

**We do not know why the activated fraction is as low as it is given the measured composition at that time. Concentrations of CCN and total CN were very low ($11 \pm 5 \text{ cm}^{-3}$ and $427 \pm 55 \text{ cm}^{-3}$, respectively). We opine that the low aerosol concentrations and the presence of organics in the particles may contribute to the low activated fraction.

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Technical corrections Page 18279, line 7-8 The two references, Broekhuizen et al. 2004a and Kumar et al., 2003, do not appear in the reference list.

**Changed.

Page 18283 Although it can be deduced from the figures, please specify the range of diameters of the particles that are analysed by the SPLAT II.

**SPLAT II analyzes particles in 50 nm to 3 micrometer size range [Zelenyuk and Imre, 2009]. This has been added to the text.

Page 18283, line 10-13 This sentence is unclear and should be re-worded.

**Clarified.

Page 18286, line 12-13 The reference to "particles larger than 80 nm detectable by SPAT II" makes this sentence unclear. Do you mean that you chose the number of particles > 80 nm measured by the SMPS because that was the lower cutoff of the SPLAT II?

**Yes, that was the meaning intended. The sentence has been clarified.

Page 18288, lines 7-8 These numbers appear to have been displayed incorrectly. Please correct them.

**Corrected.

Page 18288, line 27 Are these the peak densities for March 11 and 15, respectively?

**Yes. This has been clarified.

Page 18303, Figures 3b and 3c It looks like the brown bars in Figure 3b are for Org44_sulf, but the size distribution in Figure 3c is labelled for Org43_sulf. Are they the same? They are not the same; the size distribution in Figure 3c does not include Org44_sulf, as that particle type did not contribute significantly to the overall composition. Page 18306, Figure 7b On my monitor, there is an orange trace that does not

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appear in the legend or in Figure 6.

**The orange trace is referring to the Org44_sulf cluster; the plot has been changed to show the same color shown in Figure 6 for the Org44_sulf cluster.

References Langley, L., Leaitch, W.R., Lohmann, U., Shantz, N.C., and Worsnop, D.R.: Contributions from DMS and ship emissions to CCN observed over the summertime North Pacific, *Atmos. Chem. Phys.*, 10, 1287-1314, 2010.

Shantz, N.C., Pierce, J.R., Chang, R.Y.-W., Vlasenko, A., Riipinen, I. Sjostedt, S., Slowik, J.G., Wiebe, A., Liggio, J., Abbatt, J.P.D., Leaitch, W.R.: Cloud condensation nuclei droplet growth kinetics of ultrafine particles during anthropogenic nucleation events, *Atmos. Environ.*, 47, 389-398, 2012.

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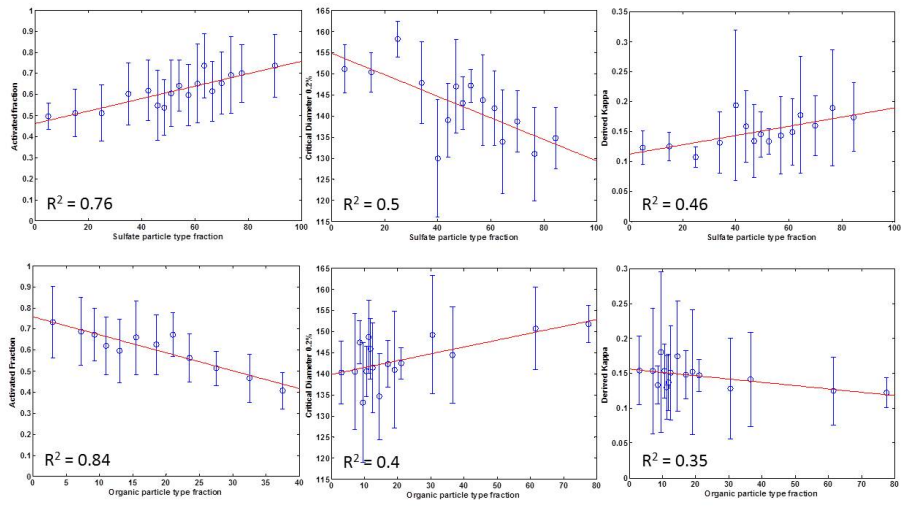


Fig. 1. Correlations between the fraction of sulfate-type (top) and organic-type particles (lower); activated fraction (left panels), critical diameter at 0.2% SS (middle panels), and derived kappa (right)