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

Anonymous Referee #3

Received and published: 30 August 2013

General comments

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

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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.

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.

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%. 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.

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.

Page 18289, line 16-17

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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.

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% 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 κ must be <0.15 . The fractional size distributions also contain information that could be used to estimate the κ . At the very least, the calculated time-varying kappa could be included so that the reader can gauge how it changes with fractional composition.

Page 18290, line 19

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

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?

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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.

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.

Page 18283, line 10-13

This sentence is unclear and should be re-worded.

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?

Page 18288, lines 7-8

These numbers appear to have been displayed incorrectly. Please correct them.

Page 18288, line 27

Are these the peak densities for March 11 and 15, respectively?

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?

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Page 18306, Figure 7b

On my monitor, there is an orange trace that does not appear in the legend or in Figure 6.

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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ACPD

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