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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Single particle diversity and mixing state measurements" *by* R. M. Healy et al.

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This paper presents the first application of field measurements to the recently introduced entropy measures for quantifying aerosol mixing state. This paper builds on recent work by the authors to develop quantitative methods for single particle mass spectrometry. To my delight, I find this work to be excellent and I highly recommend its publication. I have listed a few minor suggestions below relating to the interpretation of the results.

Specific Comments:

Section 2.2, Page 6: It should be stated what clustering algorithm is used to parse the data (K-means?). How were 10 clusters arrived at? Was it fixed in the algorithm? Also,

why were other clusters not considered for the derivation of the RSFs? In general, it is not clear how or why clustering was used in this study and I think it would be good to briefly state this. What percentage of particles were represented in the 10 clusters? Figure S1: How do the RSFs in figure S1 differ from those presented in Table S1 of Healy et al. 2013. They appear to be quite different.

Page 8, Density assumption for number scaling: I would think that a more accurate estimation of density could be arrived at using the mass fractions. Why or why wasn't this included?

Results, Pg 15: How might the exclusion of aerosol water affect the interpretation of the data from a modeler's point of view? Water is a very important component of aerosol, and one would expect for a physically correct model it should be included. Perhaps this could be briefly discussed. Are there ways of measuring the water content of individual particles?

Results, Pg 12: It is stated that D_i is around 2. What does this mean physically? That the population is typically only composed of two species on average? This might be expected for cation/anion pairs, but I would think that OC or BC would always be associated as well. Wouldn't that make the D_i closer to 3? Or, perhaps this can be explained by the fact that OC is more prevalent in particles than inorganic species? If the values for D_i are compared to D_gamma (single particle diversity, usually always above 3) I think it starts to make more sense. I enjoyed thinking about this.

Results, Page 16, Figure 7: This could probably go without mentioning, but I think it would be good to state if/how this figure was generated from the data. Section 3.4: I suggest moving section 3.4 (before current section 3.1) to the front of the paper before presenting the campaign averaged data. This way the reader fully understands what factors are controlling the average trends.

Page 16. The sentence "Local sources contributed most significantly to aerosol mass concentrations under marine air mass conditions from 28/01/2010-07/02/2010" is po-

tentially misleading considering the coarse mode was not measured. For these periods, what was the PM2.5/PM10 split?

Conclusions, Page 17. The sentence “Aerosol mass spectrometry measurements have been used for the first time. . .” should be modified to state that Single Particle Mass Spectrometry and AMS measurements were used. It is widely understood in the community that AMS refers to the aerodyne AMS, and I think it should be clear that the ATOFMS was used here.

Conclusions: I think it is important to state why the models are undertaking this task. Is it to simplify mixing state for larger scale (perhaps global?) models? One drawback I see with this approach is that the type of species is not explicit in the parameterization. How can one relate mixing state to atmospheric effect (e.g. direct radiative forcing) if the components cannot be "tagged" with composition. Perhaps some discussion of this is warranted.

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