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

Anonymous Referee #2

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The paper presents a theoretical framework for the quantification of aerosol particle diversity and mixing state based on single particle mass spectrometry field data. The approach developed by Riemer et al., 2013, was applied to Aerosol-Time-of-Flight Mass Spectrometry (ATOFMS) data collected during the MEGAPOLI winter campaign in Paris, France 2010. The authors give a detailed description of the newly developed framework and report on campaign-averaged, diurnal and temporal trends in the investigated parameters, namely particle diversity and mixing state.

General remarks:

The analytical framework, data and results presented in the paper of Healy et al. are of importance for the atmospheric science community and especially for those dealing with aerosol mass spectrometry since a new approach is presented to investigate the mixing state and particle diversity from a theoretical perspective. Although the approach is quite complex, needs several data processing steps prior to its application (i.e. a scaling of the single particle counts and clustering), and relies on auxiliary data as that from the Aerosol Mass Spectrometer (AMS), its capabilities as analytical tool are promising based on the results presented. The single particle mixing state and diversity measurements offer insight into atmospheric ageing processes and help to identify aerosol particle sources and their role in air pollution and climate.

The paper is nicely written and well-structured. The detailed repetition/description of the newly developed framework and the mentioned examples help the reader thought the complex topic. The application of the framework to actual data demonstrates the usefulness and capabilities of this analysis approach.

Although the results obtained with the new analytical tool do not directly present many new scientific findings, the paper is well-worth being published in ACP after minor revisions. The manuscript will lay the basis for this analytical framework to be adapted by the scientific community.

Minor remarks:

1) Page 3975, line 22: The “Shannon entropy” should be defined briefly. It might be helpful for the reader to explain in simple words how it is different to the well-known “thermodynamic” entropy to prevent confusion. This, however, can also be done in section 2.3.

2) Page 3977, line 1: What is the definition of “organic aerosol”? It is understandable from the context, but “the mass fractions of . . . organic aerosol” sounds odd. Although it is consistent throughout the paper and used in Healy et. al., 2012, 2013, already, but shouldn’t it be named “organic carbon” or “mass fraction of organic aerosol-constituents” in this context?

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3) P3978, l4: Does the AMS and MAAP uncertainty refers to the measured mass?

4) P3978, l17ff: “mass concentrations used twice in the sentence.

5) P3978, l19ff: Complex sentence – maybe make two out of it?

6) P3979,l6ff: What are the actual mass contributions of potassium, sodium and chloride, and where do you make a cut to not expect a significant impact on particle diversity?

Here, I see a weakness of the paper as it is not clear how the addition (or even subtraction) of another chemical species – even with a small mass fraction – would affect the results. How would the model work for a more complex aerosol composition? Also, the uncertainty associated with the ATOFMS-derived particle speciation, and how this affects the diversity results should be moved and discussed in the main text, not in the supplement. These are important facts to understand the potential of the method, to judge its accuracy and to get an impression on the “natural” variability of the diversity and mixing state.

7) P3986,l27: Why does the ATOFMS not detect cooking-related particles? Explain briefly.

8) P3987,l2ff: Compare to comment 6 and discussion above; Here, the increase in χ can’t be related to either the uncertainty of the method (due to the ATOFMS matrix effects) or the actual change in particle mixing state. Again, a number on the uncertainty of the method would be helpful.

References

Healy, R. M., Sciare, J., Poulain, L., Kamili, K., Merkel, M., Müller, T., Wiedensohler, A., Eckhardt, S., Stohl, A., Sarda-Estève, R., McGillicuddy, E., O’Connor, I. P., Sodeau, J. R., and Wenger, J. C.: Sources and mixing state of size-resolved elemental carbon particles in a European megacity: Paris, Atmos. Chem. Phys., 12, 1681-1700, 10.5194/acp-12-1681-2012, 2012.

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