

Interactive comment on “A new method to quantify mineral dust and other aerosol species from aircraft platforms using single particle mass spectrometry” by Karl D. Froyd et al.

Robert Healy (Referee)

robert.healy@utoronto.ca

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Froyd and coauthors describe an approach to quantification of dust and specific aerosol species (organics, sulfate) using single particle mass spectrometer (SPMS) measurement data combined with concurrent scattering-based particle counting measurement data. The latter are used to provide accurate particle number-size distributions in bins as a starting point for the analysis. The approach involves first using mass spectral information to infer density and shape factor for each measured single particle. These properties are then used to convert the aerodynamic diameter of each particle to its respective volume equivalent diameter. Once all of the single particles are binned

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into volume equivalent diameter bins, the fractional contributions of particle ‘classes’ in each bin are calculated. As the total particle number in each bin is already known as a function of time from the supporting particle counting measurements, here the SPMS data are only used to produce the breakdown of particle compositions within each size bin. Single particle and ensemble sulfate and organic mass concentrations can then be calculated. Total dust mass concentrations are also estimated. Although applied to aircraft datasets in this case, the approach could be extended to ground-based field studies for other SPMS instruments. The uncertainties associated with the method, including particle counting measurement uncertainty, SPMS counting statistics, the use of binned diameters, and assumptions around particle shape factor and density are carefully considered and laid out in detail. This manuscript represents a roadmap for future SPMS users that aim to use their single particle data in a more quantitative way. Although it is a little long, the content here is certainly useful for future applications of the method. I have only minor comments below.

Apart from the uncertainties listed in the Appendix, one issue is particles that are not efficiently ionized (or ionized at all) by the SPMS, because these classes will be absent in the analysis. Differences in ionization efficiencies for different particle classes, or absent classes, will affect the relative counts and fractional contributions of each class in each bin if it is assumed that all particle classes are detected with equal efficiency. Are there known particle mixing state impacts on relative ionization efficiencies for the PALMS instrument? If so these should be discussed and an estimation of the impact of this phenomenon on the quantification uncertainty would be useful.

Page 2, line 25: Nitrate, ammonium and potassium have also been previously quantified for particle classes and for single particles using similar approaches for ground-based measurements (Healy et al. 2013, 2014). Those applications also share the similarity with this work of taking concurrent particle counting measurements as the representative total number-size distribution rather than using size-dependent detection efficiency curves to work up from SPMS counts. It should be noted that that dataset

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was less challenging for quantification applications however, because only the submicron distribution was considered and crustal/sea salt contributions were minimal in that case. The assumption of equal detection efficiency for all mixing states was also taken in that work, but the spread in relative sensitivities observed for quantified species for each hour of the measurement period indicated that matrix effects associated with mixing state do impact quantification accuracy, at least for ATOFMS measurements.

Page 4, line 8: also 50% for 3.2 μm ?

Page 4, line 33: Are the negative spectra used in any way?

Page 15, line 15: Nitrate can be quantified using the approaches laid out here.

Page 16, line 8: “with decreasing altitude”

Page 17, line 19: Fig. 10 not included

References

Healy, R. M., Sciare, J., Poulain, L., Crippa, M., Wiedensohler, A., Prévôt, A. S. H., Baltensperger, U., Sarda-Estève, R., McGuire, M. L., Jeong, C.-H., McGillicuddy, E., O'Connor, I. P., Sodeau, J. R., Evans, G. J., and Wenger, J. C.: Quantitative determination of carbonaceous particle mixing state in Paris using single-particle mass spectrometer and aerosol mass spectrometer measurements, *Atmos. Chem. Phys.*, 13, 9479-9496, <https://doi.org/10.5194/acp-13-9479-2013>, 2013.

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