

Interactive
Comment

Interactive comment on “Investigating the annual behaviour of submicron secondary inorganic and organic aerosols in London” by D. E. Young et al.

Anonymous Referee #2

Received and published: 13 October 2014

In the present manuscript Young et al. present data and source apportionment results from one of the longest AMS measurement campaigns so far. The paper focuses mainly on secondary aerosols. The authors take great care in documenting their PMF analysis, which is commendable. Anyhow, my main concerns concern the PMF. I think that the approach to do the SA over the whole period gives rise to several of the discussed issues: Factor mixing and difference between HR and CTOF. I got the impression that maybe the CTOF OOA2 actually is an overlap of what is LVOOA in summer and aged BBOA in winter. Both usually have (as mentioned in the text) similar diurnal cycles and a relatively large overlap of their spectra. Indications could be: is the correlation of OOA2 with NO₃ larger in summer than in winter (and vice versa for levoglucosan)? Or if one looks at the time series of the PMF residuals is there some

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m/z which have larger residuals in summer and others in winter. Also the fact that the HR PMF does not find a second OOA in winter and no SFOA in summer points in this direction. Was a separation in seasons tried and compared (e.g. division of the data according to average daily temperature)? The presented approach to deconvolve the OOA2 factor may solve that issue to some extent but a seasonwise approach or the application of the increased rotational control provided by the ME2 may improve the solution. In the discussion of the different factor solutions in the supplement an inclusion of the residuals time series instead of just the sum of Q/Q_exp is desirable, if possible.

Some more minor comments:

P 18741 L 26: Not sure if Zhang is a good citation here, I suggest to change to doi: 10.1038/nature13774 or similar.

P 18749 L 5: Mention ACSM in this section? Since it is related and used for long-term measurements routinely.

P 18744 L1+10: As a non British it would be nice for me to know what DEFRA and NERC stands for.

P18745 L18: Can you specify at what current the filament was run and did you estimate the effect on the S/N?

P 18743 L22 + 18751 L9: summarize or summarise?

P18746 L1: I miss a description of the IE? Was an averaged IE used or did you adjust that over time? RIE: there were no significant changes in the RIE over the year? Even after major modifications (e.g. filament, vacuum pump, MCP change)

P18746 L 16: Was it not possible to correct for these step changes with the flowrate and airbeam signal

P18752 L16: As mentioned above splitting into seasons may avoid some problems of factor mixing. Indeed, a split just according to dates may not be the best option but one

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could define better objective criteria like average daily temperature.

Why is information lost by splitting the data? I do not understand this argument. PMF works also well on shorter periods as it was shown in many studies and factor profiles, especially of the secondaries which have different precursors summer and winter may change a bit over time. With shorter periods one could capture that to some extent and gain (not loose) information on annual trends.

P 18752 L 24: No a priori knowledge is needed to run ME2. It is just possible to add additional constraints but not mandatory. Basically the addition of these additional constraints (i.e. factor profiles with some degree of freedom) gives you just a handle to explore more rotations (in a way similar, but much more complex that fpeak).

P18754 L2: Even with your reduce S/N due to the lower filament setting you have much larger S/N than the ACSM?

P18756 L 5-10: I have difficulties understanding this paragraph. Please clarify.

P18764 L9: How can you comment on the change of the oxidation of SOA. The SOA result you get from the PMF only gives you two fixed factors over the whole 1a period. Please clarify.

Fig. 4: It is difficult to see anything in this plot on my printout. Is the OOA2 sometimes negative? PMF results should, as the name says always be positive

Supplement P3: You don't downweight to reduce signal. You downweight to reduce the influence of some signals to the PMF solution in order avoid PMF to try explain noise. Also the modification of the m/z44 related channels is rather a "not upweighting" than a downweighting.

After these concerns have been addressed I recommend publication of this manuscript

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 18739, 2014.