Atmos. Chem. Phys. Discuss., 12, C4833–C4836, 2012
www.atmos-chem-phys-discuss.net/12/C4833/2012/

Atmospheric
Chemistry
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Discussions

Interactive comment on "Factor analysis of combined organic and inorganic aerosol mass spectra from high resolution aerosol mass spectrometer measurements" *by* Y. L. Sun et al.

Anonymous Referee #2

Received and published: 19 July 2012

General comments:

This paper presents the re-analysis of an existing AMS dataset previously presented by the same first author, using PMF on the whole high-resolution mass spectrum rather than simply the organic peaks in isolation, which is the common practice. This produces a larger number of factors than the previous analysis, featuring many inorganic peaks. While some of the results are expected (identification of ammonium nitrate and ammonium sulphate), this analysis gives some interesting insights into the role of organic nitrates and amines and this technique will probably prove useful to others in the future.

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This paper stops short of over-analysing the results, so no major advances in atmospheric chemistry are gained and most of what is said could be considered incremental on previous works. However, the new results in themselves do represent novel observations of particulate composition (particularly with respect to the organic nitrates and amines) and tantalising hints as to the roles of some of these hard-to-measure species, so these in their own right make this paper worthy of publication in ACP (rather than a technical journal).

The paper is well-written overall, although I do have a few reservations (see below). But given that taken at face value, the results are hard to argue with and the authors provide sufficient information for someone to repeat the analysis, none of these should be considered potential showstoppers. I therefore recommend this be published subject to minor revisions.

Specific comments:

The authors neglect to provide a time series of the basic AMS data products or the derived factors. While some of these are available in previous publications, they would be highly informative and should be given here.

Not enough detail is given as to why a solution with 8 factors with an fpeak of 0 was chosen. The authors need to explicitly give their reasons for why the other potential solutions were rejected. They should also have tested the numerical stability of the solution through seed variation or bootstrapping.

The authors seem to have been very selective in their treatment of the different factors when attempting to recreate size distributions. A general concern is that species that vaporise on slower timescales (tens of milliseconds) may not be adequately represented in pTOF space, which distorts the relative contributions of various components. It could be that this is happening here, so for the sake of general technical interest and for those attempting this technique in the future, they should go into more detail in the supplementary material regarding the problems encountered. It would also be useful

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to know how much normalisation the different factors required.

P13308, L21: Given the long timescale for gas-phase production of sulphuric acid, it would not necessarily be expected that a pronounced lunchtime peak would be manifested. There may be other explanations, such as the formation of organosulphates. Also, the phrase “the sulfate in the rest of OA factors shows a pronounced noon peak” should be reworded as “the other OA factors containing sulfate showed a pronounced noon peak” because the diurnal profiles only show the behaviour of the factor as a whole, not the sulphate in isolation.

Section 3.1: Given that sulphate and nitrate give strong signals at completely different m/z channels and their lack of correlation extends to periods where each of the species approach zero independently of the other (seen in Fig. 1 in Sun et al., 2011c), PMF assigning these to distinct factors isn’t so much an observation as a mathematical certainty. Therefore, this separation does not contribute anything new to the science beyond what can be seen with a simple time series and statements such as “PMF analysis results clearly indicate their different formation and evolution processes” massively overstate the significance of this result. The lack of correlation between sulphate and nitrate is already well-documented and fairly well understood, but what is perhaps more scientifically significant is the organic matter associated with them.

P13315, L16: The authors should explain what they mean by ‘similar aging properties and internally mixed characteristics’.

P13332: More detail should be given on the smoothing used and the reasons why this was used.

Technical corrections:

P13304, L4: The time zone should be defined relative to UTC

P13308, L23: The phrase “These results suggest the different sources of sulfate in SO4-OA and other factors” does not make sense. Suggest rewording.

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P13310, L13: It should be noted that conventional wisdom considers a portion of sulphate to be primary, however the adoption of low sulphur fuels has all but eliminated this.

P13313, L21: Replace ‘re-look’ with something more formal.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 13299, 2012.

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