Atmos. Chem. Phys. Discuss., 11, C3527–C3530, 2011
www.atmos-chem-phys-discuss.net/11/C3527/2011/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Elucidating determinants of aerosol composition through particle-type-based receptor modeling" *by* M. L. McGuire et al.

Anonymous Referee #2

Received and published: 18 May 2011

This article describes the application of positive matrix factorization (PMF) to pre-clustered single particle mass spectral data in order to resolve local and regional sources of particulate matter in Southern Ontario, Canada. Local and regional scale meteorological data and models have been used in combination with the temporality of the factors obtained and the particle mixing state provided by the mass spectra, to apportion the various single particle types to their respective sources. A nine-factor solution was observed to be the most robust, with most factors assigned to local-to-regional or regional scale transport of particulate matter emitted on both sides of the Canada-US border. Dust particles containing nitrate were found to be the most important local source, while significant regional sources include biomass burning associated with forest fires in the Canadian Prairies and anthropogenic carbonaceous emissions from urban areas in Canada and the US. While PMF has been applied to single particle data in some previous studies, this work represents the most methodical and comprehensive approach thus far, and provides a benchmark study that will be useful for others attempting source apportionment using single particle mass spectral data. Choosing the most reasonable number of factors is essential, because, as has been demonstrated here, the iterative addition of factors which appear to artificially split the existing factors can sometimes be justified by examining particle mixing state and the extent of chemical processing during transport. The authors also performed detailed meteorological analysis, which demonstrates the value of using high-resolution trajectory models where available to identify local-to-regional sources of particulate matter.

Specific comments:

Page 9836, lines 13-15. Considering this statement, were size-resolved variables explored for the different clusters? For example, does splitting the existing clusters into size bins lead to variables with different temporality. This is possibly beyond the scope of this manuscript but may be interesting for bimodal clusters with input from more than one source.

Page 9841, line 13. The authors mention that reducing particle cluster numbers after running ART-2a through manual recombination resulted in unsatisfactory PMF solutions. Does this suggest that this subjective approach, commonly used for ATOFMS studies, should be reconsidered, or even avoided? Typically higher vigilance factors are used, resulting in higher cluster numbers and thus encouraging manual recombination. A few lines on this would be of interest to other ATOFMS users.

Page 9841, line 26. This section needs to be expanded, or moved to the supporting information and covered in more detail. Size-dependent transmission efficiency problems associated with the aerodynamic lens are typically “scaled up” using concurrent scanning mobility particle sizer or aerodynamic particle sizer measurements, and this

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was described in detail in the companion paper cited in the manuscript (Jeong et al., 2011a). It is surprising that the scaled particle counts do not lead to equally robust PMF solutions, considering that the raw ATOFMS size distributions are skewed due to the transmission efficiency curve associated with the lens. What about volume estimates or mass estimates, what are the PMF solutions like? The unsatisfactory PMF solutions should be included in the supporting information, along with the authors' opinion on why the scaled datasets do not work as well, or at least give the same results. This will definitely be of interest to the ATOFMS community. The scaled size distributions could also be used in Figure S2 and in the particle size mode descriptions in section 3.3.

Page 9864, lines 16-30. Alongside the recommendations provided in this section, it would be interesting to gauge the authors' opinions on the inclusion of other online measurements (e.g., EC, OC, PM mass, NOx etc.) in the PMF analysis and how this might affect the results.

Minor comments:

Page 9834, line 16. Remove apostrophe

Page 9837, lines 21-24. Please rephrase. The ATOFMS does measure refractory aerosol components.

Page 9848. The use of both R and R2 values is confusing. It may be best to use one or the other for continuity.

Page 9849, line 3. Remove apostrophe

Page 9853, line 4. Replace "m/z's" with "m/z".

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Page 9857, lines 14-16. Rephrase this sentence, possibly "air masses which passed over the Canadian Prairies 48 h previously".

Page 9860, lines 19-21. Rephrase this sentence

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Page 9860, line 27. Would “uptake” be better than “deposition”?

Page 9877, Figure 3. The C1 contribution to the Long Range Transport is difficult to see here, perhaps shift the x-axis. The third factor is named “Primary Organic” here while it is simply “Organic” elsewhere.

Page 9882, Figure 8. “Primary Organic” is used again here. The authors should stick to one label.

Supporting Information, page 3, line 2. Rephrase possibly “as their chemical composition and temporality were definitive”

Supporting Information, page 3, line 11. Remove apostrophe

Supporting Information, page 3, line 18. Rephrase possibly “precluded their assignment as fresh or unprocessed particles”

Supporting Information, page 4, line 31. Superscript +

Supporting Information, page 5, line 25. Remove apostrophe

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