**Atmospheric
Chemistry
and Physics
Discussions**

Interactive comment on "Simultaneous factor analysis of organic particle and gas mass spectra: AMS and PTR-MS measurements at an urban site" *by* J. G. Slowik et al.

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This manuscript takes an exciting new direction in the realm of source apportionment by applying Positive Matrix Factorization (PMF) separately to collocated particle- and gas-phase mass spectral instrument datasets and by tackling the clearly difficult challenge of combining both datasets for simultaneous factorization. I make comments and suggestions for clarification on two main points and make other minor comments and suggestions. This work will be a valuable contribution to the literature.

Main Comments

1. Explanation and Use of C_{PTR}

On pg. 6746, lines 11-14, the text states that the error values for the PTR-MS data are multiplied by C_{PTR}; the C_{PTR} values in Fig. 8 are all > 1. These C_{PTR} values would increase the error values, lowering the relative signal to noise ratio (SNR) of the PTR-MS data. It seems that this would downweight the PTR-MS data relative to the AMS, in the same way that increasing the error values can be used to downweight "weak" variables (i.e., low SNR variables) to have less influence on the fit (Paatero and Hopke, 2003). Are the PTR-MS error values instead divided by C_{PTR}? Then the other discussion of the effect of C_{PTR} makes more sense. It would be helpful, however, if the language in this section was parallel to the labels in Fig. 8 (at present the text discuses underweighting but the figure describes overweighting).

In the discussion of Fig. 8 and balanced solutions (pg. 6755, line 28), you might wish to emphasize the point made in the introduction of C_{PTR} (pg. 6745, line 22-24) that in principle one would expect to have to give extra weight to the smaller PTR-MS dataset (10 *m/z*'s vs. 270 *m/z*'s) by about 27 times, if all *m/z*'s in both instruments have about the same SNR. It may be useful to report the average SNR of both instruments to show that 27 times is too much as presumably the PTR-MS data have somewhat higher SNR than the average AMS *m/z* for the same averaging time. This would clarify the reason for the need to use C_{PTR}.

In Figs. 9 and 10, I would suggest that you remove C_{PTR} from the scaled residuals of the PTR-MS elements before summing to the time series. This should make it easier for the reader to compare to the scaled residual time series of the individual instrument cases in Figs. 1 and 4. If you don't wish to do this, I would suggest adding a note in the caption of Figs. 9 and 10 stating that the scaled residuals and Q/Q_{exp} include the effect of C_{PTR}, even though it is redundant with the text describing the figures. This is also parallel with the removal of C_{PTR} for calculating $\Delta\overline{e_{sc}}$. It would also be very useful to plot the time series of scaled residuals for the separate and unified datasets together in the Supp. Info., so that the reader can better appreciate during which periods and by how much the fitting of the data has changed because of the joint analysis of the

datasets.

Finally, it seems from Fig. 8 that the C_{PTR}=20 solution with 5 factors meets the $\Delta\overline{e_{sc}}$ criterion better than the reported C_{PTR}=10 solution with 5 factors. In choosing the solution with C_{PTR}=10, the authors are not completely following their own method, and this deviation should be justified. I suggest that some of the discussion of the difficulty with the C_{PTR}=20 solution currently in the Supp. Info. be moved to the main text near pg. 6755, line 20 to support the use of the reported solution.

An alternative to the use of C_{PTR} could be to include multiple copies of the PTR-MS data (with the original weights). The number of copies required to achieve a balanced solution might be expected to be similar to the C_{PTR} required for a balanced solution. In this method the robust mode could be used and all of the points would pull with their full weight while limiting the impact of the outliers. It would be helpful to at least mention if this method has been attempted, even (and perhaps especially!) if it has failed. If it has not been tried, I suggest that it is tried and compared with the C_{PTR} method in the revised version of the manuscript.

2. Comparisons of the individual and unified datasets

As a reader I would like to examine the comparison of the spectra and time series of the factors and scaled residuals time series from the individual vs. unified datasets more closely. It would be extremely helpful to include (even in the Supp. Info.) plots of the spectra and time series overlaid, perhaps in the style of Fig. 9 of Ulbrich et al. (2008). This presentation should not replace the current Figs. 10 and 11, though, which are needed to see the unified case results clearly.

The discussion on pg. 6756, lines 9-11 mentions very large increases in scaled residuals in the unified case compared to the individual cases. It seems that the overall fit of the matrices is much worse in the unified case, though additional information was extracted. If this is not the case, this should be clarified. The increase in the values of Q/Q_{exp} should also be given in the text, even if the size of the Q/Q_{exp} insets is

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increased.

Taking the assumption that the fit is less good for the unified dataset, it is then surprising that the reconstruction of the total AMS signal and PTR-MS masses in the unified case (Fig. 12) are close to 1 and very similar to the PTR-MS case only (Fig. 7). Perhaps the residuals (e_{ij}) of the unified case have much larger positive and negative values that still mostly cancel each other?

Requests for More Explicit Math Details

At pg. 6748, line 11, please explain how the two OOA factors were mathematically recombined into one factor for presentation of the mass spectrum and time series.

At pg. 6757, lines 12-15, the manuscript states that "Signal intensity is normalized such that each factor mass spectrum of the individual instrument sums to 1." If the spectrum signals are renormalized, the required multiplicative factor must be applied inversely to the time series so that the product of mass spectra and time series still reconstruct the input matrix. Please explain in more detail how the rescaling was applied.

Further Comments

The text often uses the term "residual" when I think "scaled residual" would be more accurate. It would be helpful to the reader to make this clear distinction.

At pg. 6750, line 24 how was it determined that the signal from the factor was concentrated in short spikes, since the modeled data has been averaged to 15 min? It also seems that by averaging to 15 min. the authors are removing some of the temporal contrast that allows PMF to better separate the factors. Were the datasets run in PMF with shorter averaging times, and was there a reason to settle on a time so much larger than the reporting times of the individual instruments?

It would be extremely helpful to present diurnal average plots of all the factors and tracers (from the individual and unified datasets, even if some or all of them are in Supp. Info.), since the text refers to them often and they are considered as a metric

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used to identify primary emission factors (pg. 6760, lines 7-11).

Reading the discussion of the factors and making comparisons between Figs. 6-7 and 11-12 would be simpler if all of the factors were assigned names. This would be especially helpful at pg. 6762 line 22, where the text refers to "charbroiling and HOA factors" in the unified dataset, but neither $F1_{UN}$ nor $F2_{UN}$ is clearly identified as HOA at pgs. 6757-8.

I support the recommendation by P. Paatero that the rotational freedom of the chosen solutions is explored and suggest that these solutions are included in the Supp. Info. and discussed in the paper.

In the Supp. Info., the figure numbers are incorrect in the text. Similar to the suggestion above for overlaying the factors from the unified and individual datasets, better comparison between the factors from different C_{PTR} cases could be made by overlaying the factors here, too. The comparison of the time series in these cases is complicated by the fact that two things are changing at once (the solutions themselves, and the apparent relative weighting applied by normalizing the changing mass spectra of the solutions). If it would be possible to present the results so that direct comparison change of the solutions could be made, the usefulness of these plots would be increased.

References

Paatero, P., and Hopke, P. K.: Discarding or downweighting high-noise variables in factor analytic models, Anal. Chim. Acta, 490, 277-289, 10.1016/s0003-2670(02)01643-4, 2003.

Ulbrich, I., et al.: Interpretation of organic components from Positive Matrix Factorization of aerosol mass spectrometric data, Atmos. Chem, Phys. Disc., 8, 6729-6791, 2008.

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