Atmospheric
Chemistry
and Physics
Discussions

Open Access

Interactive comment on "Submicron aerosol source apportionment of wintertime pollution in Paris, France by Double Positive Matrix Factorization (PMF^2) using Aerosol Chemical Speciation Monitor (ACSM) and multi-wavelength Aethalometer" *by* J.-E. Petit et al.

Anonymous Referee #2

Received and published: 25 June 2014

The PMFˆ2 approach proposed in this study for PM source apportionment is essentially a regular PMF preceded by factor analysis of ACSM data to provide additional three species, i.e., HOA, p-BBOA, and OOA, for inclusion in the model. Two main issues that need to be addressed better are the PMFˆ2 uncertainty and benefit compared to "conventional" approaches. As has been pointed out in many studies (e.g., Paatero et al., 2005; Chen et al., 2010; Henry and Christensen, 2010), PMF factors represent imper-

fect combinations of sources and the mixing of sources reflect 1) rotational ambiguity and 2) variation of source profiles. It is not true that "conceptually speaking and applied to OA mass spectra, this model only gathers m/z fragments into factors regarding the chemical structures of parent organic molecules..., and thus does not lead to direct information of pollution sources." For example, if one and only one source contributes to ambient HOA and p-BBOA, PMF will not be able to separate them due to a high correlation and instead report a coupled "source factor". The PMF could resolve chemical structures, source profiles, or something in between depending on the rotation (i.e., linear transformation). This is why using priori information as a constraint plays an important role in controlling the rotation, leading to desired solutions. Even so, some mixing of source and molecular information in PMF factors is inevitable. The resolved pBBOA in Figure 4 shows a clear morning-rush hour peak just like HOA, suggesting it is not a "pure" molecular factor as the authors like it to be. For OOA that is dominated by SOA, the chemical structure can never be a constant as resolved by PMF. In short, HOA, p-BBOA, and OOA are far poorly defined parameters than other species commonly used in PMF analysis, including BCwb and BCff. If you weight them too much in the model, uncertainties that multiply through the two-stage PMF process could be enormous; too little, then you beat the purpose of including them. However, there is neither guidance for an appropriate weighting of the parameters nor an adequate evaluation of source apportionment uncertainties in the paper.

Although the source apportionment results compare favorably with some previous studies, the four-factor solution can likely be achieved by conventional PMF analysis using only OA concentrations (regardless of HOA, p-BBOA, and OOA fractions). The four factors will be marked by non-refractory K (wood burning), BCff (traffic), nitrate (semi-volatile secondary aerosol), and sulfate (low-volatile secondary aerosol). OA factions can then be apportioned into the four factors by multiple linear regression. In this particular study, the benefit of including OA factions in PMF is not convincing and in fact it can even distort the results due to uncertainties in the OA fractions. Instead, the paper can focus on how OA fractions from ACSM are allocated into "conventional" PMF

factors to verify the nature of these OA fractions.

Other comments:

1. Page 14166, Line 5: Would it be an issue that ACSM and aethalometer have different size-cut inlets?

2.Page 14166, Line 18: What is the correlation ($r2$) between total BC and m/z 60, thus does BC apportionment really improve the correlation?

3. Page 14166, Line 23: Please clarify if the correlation refers to r or r2. A r of 0.65 (r2 = 0.42) seems to be low.

4. Page 14169, Line 11-12: What is the definition of "enough weight"? How can you quantify the weights of different parameters/species?

5. Page 14171, Line 12: 44% and 5.5% of PM1 or PM2.5?

6. Page 14172, Line 3-7: Higher ambient temperatures typically move partitioning towards the gas phase, and so the high particulate ammonium nitrate during the third period may not be related to the gas-particle partitioning.

7. Page 14172, Line 13: Add "designated to HOA, p-BBOA, and OOA" after "The three factor solution".

8. Page 14175, Line 19: The high correlation between wood burning and OOA seems to indicate that most of the OOA are primary instead of secondary. As combustion itself is a dynamic oxidation process, what is the time scale of conversion for OA to be considered secondary?

Reference:

Chen, L.-W. A., Lowenthal, D. H., Watson, J. G., Koracin, D., Kumar, N., Knipping, E. M., Wheeler, N., Craig, K., and Reid, S.: Toward effective source apportionment using positive matrix factorization: Experiments with simulated PM2.5 data, J. Air Waste

Manage. Assoc., 60, 43-54, 2010.

Henry, R. C. and Christensen, E. R.: Selecting an appropriate multivariate source apportionment model result, Environ. Sci. Technol., 44, 2474-2481, 2010.

Paatero, P., Hopke, P. K., Begum, B. A., and Biswas, S. K.: A graphical diagnostic method for assessing the rotation in factor analytical models of atmospheric pollution, Atmos. Environ., 39, 193-201, 2005.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 14159, 2014.

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper