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Interactive comment on "Positive Matrix Factorization of Organic Aerosol: Insights from a Chemical Transport Model" *by* Anthoula D. Drosatou et al.

Anonymous Referee #2

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This paper summarizes PMF analysis conducted on OA simulated from a chemical transport model and compared with AMS PMF factors for a single summer month. The authors find similar number and categorization of PMF factors as AMS, and provides further insight for the modeled factors. First, the primary (HOA-like) factor often contains some SOA and biomass OA. Second, two OOA components found are likely more oxygenated and less oxygenated, but are not always separated in volatility space. Finally (among other conclusions), SOA from various sources can be lumped into a single PMF factor. The manuscript is well-written and easy-to-read. Many of the technical decisions (e.g., regarding the PMF error matrix for simulations) seem well-justified, and there are additional insights regarding errors in source apportionment

(mixing of sources or not extracting the source altogether) during periods of minor contributions. There results are of great interest to the atmospheric chemistry community and is therefore recommended for publication with minor revisions.

General comments:

The authors do not seem to explicitly mention this, but PMF appears to have been conducted separately at each site. One reason for applying site-specific PMF to measurements is that anthropogenic or biogenic SOA can be different at each site, but in these simulations they are effectively the same (e.g., a lumped species with C* belonging to anthropongenic POA is chemically the same across sites). However, the site-specific PMF is still desirable here for capturing site-specific proportion of source classes in each factor, and for comparison with site-specific AMS PMF.

The assumption that the components are "chemically different" is mentioned a couple of times, but it is not further discussed. For instance, the lumped species differ in C* or reactivity with OH (depending on source class) so in many cases they are already treated as being chemically different. For the remaining lumped species, it's not unlikely that species from different source classes grouped in the same bin C* are likely structurally different. However, a real chemical difference with respect to gross properties should not necessarily be required by PMF either, as it is possible to use isotope-labeled compounds in the analysis (which source-tagging effectively does here).

Is the simulated OA size fraction used for PMF analysis equivalent to PM2.5 or what might be more directly comparable to the submicron fraction measured by AMS?

Can the authors remark on the fact that the model, which does not include aqueous-phase or condensed-phase chemistry, generates factors which agree on a "high-level" with AMS factors? Is it that condensed-phase processes do not provide sufficient differentiation from covariations fixed by vapor-phase processes? Or could it a limitation of the sites/period studied?

The authors have previously published work integrating 2D-VBS into PMCAMx in the European domain; the model used here is apparently different and referred to as PMCAMx-SR - but the citation refers to a manuscript in preparation so it is hard to understand some of the details. How is this model different and why was the 1-D VBS selected? For this comparison with AMS, comparison of O/C ratios would have been useful to show further correspondence (or differences) between simulated and measured PMF factors (e.g., Aiken et al. 2008, Canagarathna et al. 2015); the authors may wish to add justification for the decision they made here.

Aiken, A. C., Decarlo, P. F., Kroll, J. H., Worsnop, D. R., Huffman, J. A., Docherty, K. S., Ulbrich, I. M., Mohr, C., Kimmel, J. R., Sueper, D., Sun, Y., Zhang, Q., Trimborn, A., Northway, M., Ziemann, P. J., Canagaratna, M. R., Onasch, T. B., Alfarra, M. R., Prevot, A. S. H., Dommen, J., Duplissy, J., Metzger, A.; Baltensperger, U. & Jimenez, J. L. O/C and OM/OC ratios of primary, secondary, and ambient organic aerosols with high-resolution time-of-flight aerosol mass spectrometry, Environmental Science & Technology, 42, 4478-4485, https://doi.org/10.1021/es703009q, 2008.

Canagaratna, M. R., Jimenez, J. L., Kroll, J. H., Chen, Q., Kessler, S. H., Massoli, P., Hildebrandt Ruiz, L., Fortner, E., Williams, L. R., Wilson, K. R., Surratt, J. D., Donahue, N. M., Jayne, J. T., and Worsnop, D. R.: Elemental ratio measurements of organic compounds using aerosol mass spectrometry: characterization, improved calibration, and implications, Atmos. Chem. Phys., 15, 253-272, https://doi.org/10.5194/acp-15-253-2015, 2015.

Others have found instances where the low/high volatility designation of PMF OOA do apply (e.g., Cappa and Jimenez 2010), so the conclusion (line. 512) that the nomenclature is misleading seems to broad - it may be rephrased that statistical separation of OOA by volatility cannot always be assumed?

Cappa, C. D. and Jimenez, J. L.: Quantitative estimates of the volatility of ambient organic aerosol, Atmos. Chem. Phys., 10, 5409-5424, https://doi.org/10.5194/acp-10-

5409-2010, 2010.

Minor comments:

- Should the acronym PMCAMx be defined? For instance, CMAQ (Community Multi-scale Air Quality Modeling System) is typically spelled out when first introduced in a paper.

- Regarding terminology, line 141: "primary organic compounds are all considered to be semi-volatile with C^* ranging from 10^{-2} to 10^6 microg/m^3" whereas Donahue, Robinson, and Pandis (2009) define SVOCs to have C^* ranging between 10^0 and 10^2 microg/m^3.

Donahue, N. M., Robinson, A. L. & Pandis, S. N. Atmospheric organic particulate matter: From smoke to secondary organic aerosol, Atmospheric Environment, 43, 94-106 https://doi.org/10.1016/j.atmosenv.2008.09.055, 2009.

- Figure 7: Is this not more a reflection of the deviation in source apportionment for both measurements and simulation when the source contribution becomes small, rather than error that can be purely attributed to the apportionment from the simulation side (as reflected by normalization to observed values)? Section 3.4 should correspondingly be renamed since "PMF error" can imply many things (error matrix, residual matrix, etc.).

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-802, 2018.