EGUsphere, referee comment RC3
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Comment on egusphere-2022-269

Anonymous Referee #3

Referee comment on "*Rolling* vs. *seasonal* PMF: real-world multi-site and synthetic dataset comparison" by Marta Via et al., EGUsphere,
https://doi.org/10.5194/egusphere-2022-269-RC3, 2022

The manuscript presents a comparison between two different approaches of PM1 organic aerosol (OA) source apportionment through the Positive Matrix Factorization (PMF) source-receptor model applied on mass spectra by Aerosol Chemical Speciation Monitor (ACSM): the widely used "seasonal" PMF against the emerging "rolling" PMF. The two approaches are systematically applied on both real-world ACSM datasets (from 9 European sites) and on a synthetic dataset. The comparison shows that the two approaches lead to similar apportionment results, both addressing the quality standards required by the source apportionment protocol. Overall "Rolling" PMF can be considered more accurate, especially in "transition" periods between seasons because it is able to better adapt to the changes in OA sources along the time. Interestingly the application of PMF on the synthetic dataset performed not so well for both the methodologies. This result shows the strong influence of using and selecting anchor profiles to constrain the PMF solutions and encourages the development of local reference profiles to minimize this impact on OA source apportionment.

This is a well-written paper that clearly describes methodologies, analyses and results. Even if the subject looks quite methodological and possibly suitable for more technical journals, actually the manuscript clarifies important and debated advantages/disadvantages of the approaches and can have a large impact on a wide audience of the atmospheric organic aerosol community.

For this reason, I recommend its publication after consideration of some major/minor comments/changes detailed below.

Major General comments:

-The discrepancies between original synthetic values and the PMF outputs on the same synthetic dataset are important and not negligible. For what I understand they originate by the choice of the anchor profiles used to constrain PMF solutions. Considering that it is now very common (and indeed strongly recommended by the AMS-data Source Apportionment protocols) to use constraints to apportion OA primary components, I also believe that this result is somewhat worrying. In particular the fact that POA are underestimated and SOA overestimated with respect to the original synthetic values might indicate that using constrains cannot be always the best option. Although I recognize this alone can be the topic of a specific publication, in my opinion the Authors in Section 3.1 should try to assess the general implications also with respect to previous/future studies applying OA source apportionment protocols. Specifically, the Authors should explain better the reasons for the choice of specific anchor profiles used in the different PMF solutions and possibly show (or at least comment) the results of the un-constrained PMF solutions on the synthetic dataset. More specific comments are below.

-I find the term "truth" to describe the synthetic dataset quite pretentious and misleading. I would suggest to change the name, especially in Figures, using for instance "original synthetic" or something else.

Specific comments

Abstract

P1-P2, L40-46: too general statements, not easy to really understand the importance of the findings and "quantify" them. Sentences like "although the rolling PMF profile adaptability feature has been proven advantageous" or "these results highlighted the impact of profile anchor on the solution" are quite hasty and vague: what this impact is? And the advantages? Although I acknowledge that it is not easy to find a quantitative and synthetic way (suitable for an abstract) to evaluate these advantages/impacts, I would recommend trying to do it by elaborating more (using also some numbers if possible) and / or removing too general sentences.

P2, L46-47: “The results of this comparison….were scarce.”, it is redundant, please remove the sentence and/or integrate with the rest.

P3, L104: what is “o their” meaning? is it a spelling mistake? In general, this sentence is hard to follow, consider to re-phrase it.

P3, L114: I am not a native English speaker but the use of "concerning" as a comparative clause sounds strange to me. Please check here and also in other parts of the text.

P4, L118: again, the term “granularity” sounds quite strange to me associated with timestamps, consider to replace with “size” or other.

P4, L146-148: the last two sentences of the paragraph appear redundant or not clear. Please, consider to re-phrase.

P5, L168-171: the verb of the main clause seems to be missing. Please add it or re-phrase.

P8, L265-266: what about the unconstrained application of PMF? Did you try? How unconstrained solutions perform in comparison with original synthetic values?

P8, L275-280: As already mentioned, the discrepancies reported in this section of the paper are a major issue that deserves more emphasis and possibly more elaborations. The risk is that, considering that the ability to reconstruct even a synthetic dataset is low, someone could question the OA source apportionment protocols and argue that PMF results are in general not robust in apportioning real-world sources, at least the ones using *a priori* POA chemical features. I'm not saying this is true, but in my opinion the Authors should not underestimate the importance of these findings and spend more words to explain what their implications are in applying the OA source apportionment protocols in other studies (past and future). For instance, the analysis of the unconstrained-PMF runs and a comparison with the "best" solutions identified following the protocol could be worth of an assessment or at least comments.

P9, L303-305: here (or somewhere else in this section) a comparison with unconstrained PMF would be very welcome.

P9, L308: "m/z" is repeated.

P11, L357-360: could the higher errors on OOA factors be also due to the fact that SOA sources are changing between seasons? I mean for sake of simplicity in the comparison, SOA components are represented here by only two factors (MO- and LO-OOA) but it is possible that the model (especially rolling application) is able to split SOA in more factors, leaving it the freedom to go to higher number of factors. Could the Authors comment on this?

P11, L360-362: it is well known that BBOA profiles have a higher variability among sites and seasons. Could the BBOA positive whiskers be also influenced by the fact that this variability is better reproduced by rolling PMF?

P11, L364-369: the discussion here is quite misleading: if I understand well, Figure 4 shows the pie-charts without site-specific sources (which are very similar in term of relative contributions), while later on there is a list of differences of ratios probably based on the absolute factor-contributions to the total OA concentrations. In general please clarify why the pie charts are so similar while the ratios reported in the text are so different.

P11, L372-373: what about the discrepancy in Cyprus?

P12, L414-415: "The histogram …is plotted as a histogram", please consider to re-phrase the sentence.

P13, L430: "are always greater for *rolling* rather than for *seasonal*." This is not true for BBOA vs BCwb and for MO-OOA vs SO4.

P14, L474: Figure S8 is reported as Figure S7 in the Supplementary. Check for consistency. Moreover, in this figure I suggest to change the color and/or the thickness of the *seasonal* lines because of difficult reading.

P15, L514-521: I would suggest the Authors to use some of these considerations also to improve the vague sentences of the abstract

Supplementary

P1, L29, right-bottom side: there is a “Sorry” to be deleted.