General comments

This is a timely and well-executed manuscript that evaluates source-resolved aerosol simulations using a coupled regional-urban modeling framework. The use of multiple PMF datasets across a relatively large number of European sites is a clear strength, and the attempt to assess both regional model performance and the added value of urban downscaling is particularly valuable.

Overall, I find the study scientifically relevant and generally well presented. The manuscript addresses an important topic, and the analysis provides useful insights into both model performance and the interpretation of PMF-derived source contributions.

That said, some aspects of the interpretation could benefit from further clarification in order to ensure that the main conclusions are as robust and transparent as possible. In particular, this concerns (i) the mapping between PMF factors and modeled source categories, (ii) the attribution of organic aerosol (especially anthropogenic SOA), and (iii) the interpretation of the urban downscaling results.

These aspects appear closely connected to some of the key conclusions of the manuscript (e.g. regarding the traffic contribution and the added value of downscaling), and a more explicit discussion of associated uncertainties would, in my view, further strengthen the study.

I therefore recommend reconsideration after revision.

We thank the reviewer for their constructive feedback and overall positive assessment of our work. We have address the suggested refinements and clarifications of the text, given by the answers in blue below. In short, we have added additional detail regarding the matching with PMF factors and the effects of urban downscaling throughout the text. The discussion regarding CPOA has further been restructured through the addition of a new section 7.1.1 while the conclusion section has been reworked to provide a more focused picture of the main messages.

Specific comments

1. PMF factor-model mapping

Around lines 214-218 (Section 4.2, introduction to PMF factors) and in connection with Table S3, it may be helpful to provide a clearer and more systematic discussion of how PMF factors are mapped to modeled source categories.

In particular, it would be helpful to clarify which aspects of the results are expected to be relatively robust to reasonable alternative mappings, and which might be more sensitive to these assumptions. This could help the reader better distinguish between discrepancies arising from model performance and those related to structural differences between PMF factors and model variables.

If feasible, a brief qualitative discussion of how alternative plausible mappings might influence the results could further enhance the transparency of the analysis.

We have expanded the introductory paragraph of section 4.2, now providing additional background information regarding the aim and general methodology of the matching with PMF factors and how the structure of the following text aligns with this aim. This more detailed description of the text structure likewise applies to the other PMF datasets. In the discussion and conclusion sections, we now further highlight that the source-specific primary and secondary OA contributions to the relevant PMF factors remains one of the main challenges, both from a modelling point of view and to an extent PMF analysis itself. In our view the most prominent example of this relates to biomass burning CPOA, as discussed in section 7.

While other alternative mapping choices are not discussed in the text (e.g., assigning certain model species or sources to different modelled PMF factors), our view is that an alternative mapping would be more akin towards different choices in the construction of combined PMF factors (e.g., adding the MSA-rich PMF factor to the constructed sea salt factor rather than sulfate-rich factor, since it could also be argued that MSA is derived from oceanic emission sources). A more detailed discussion of the mapping alternatives would then ideally involve modelling all PMF factors individually, and to only then aggregate them into factors that provide a more robust result. However, since the motivation behind the aggregation of certain PMF factors is provided in the text, at least from the point of view of what the model is reasonably capable of distinguishing, our view is that a more detailed discussion of the non-aggregated sectors would not provide critical additional detail. However, in the revised text we now highlight that the MSA-rich PMF factor makes a modest contribution to the final constructed PMF factor mass, such that the impact on the results is limited.

2. Allocation of anthropogenic SOA to road traffic

Around lines 240-248, all anthropogenic SOA is assigned to the road traffic factor. While the manuscript provides some justification for this choice, it might be useful to elaborate slightly further on the rationale.

In addition, it would be helpful to comment on how sensitive the results could be to alternative allocations (e.g. partial attribution to other anthropogenic sources such as solvents or residential combustion). Even a short sensitivity discussion, if available, could help to place the traffic-related conclusions in a broader context.

In the revision of section 4.2.2, we have now elaborated on the choice to assign all aSOA to road traffic. While Weber et al. (2021) note that the biomass burning factor is also suspected of containing aSOA, this is arguably predominantly related to the SOA derived from SVOC pre-cursors from wood burning (following the discussion in section 7), which is by default included as POA in the GNFR C (stationary combustion) emission sector and thereby assigned to the modelled biomass burning factor. In the revised manuscript, we now note that the assignment of aSOA makes little impact on the final results, given that its mass contribution is small (0.3 µg m^{-3}on average). While assigning (a portion of) modelled aSOA to the modelled dust factor may also be an option, given that Weber et al. (2021) note that this factor may contain aSOA (there is indeed a small amount of OC present in the PMF derived dust factor; Fig. 3 of the main text), we refrain from doing so based on 1) the correlation with modelled dust is practically non-existent, 2) the analysis of Weber et al. (2021) is inclusive as to what fraction of aSOA may be assigned to the dust factor, 3) it ultimately matters very little to the model results.

3. Interpretation of the sulfate-rich factor

Around lines 304-309, the inclusion of biogenic SOA and background OA in the sulfate-rich factor is noted. It may be helpful to expand the discussion slightly here.

In particular, this choice may make the interpretation of this factor less directly comparable to a purely secondary inorganic aerosol component. A brief clarification of how this mixed composition should be interpreted when comparing to PMF-derived sulfate-rich contributions would likely improve clarity for the reader.

We have now provided additional motivation for why biogenic SOA (bSOA) is mixed in with the sulfate-rich factor, based on the cited work of Borlaza et al. (2021). Namely, that bSOA and sulfate are both secondary aerosols, having the shared characteristics of being transported over long distances with a comparatively long atmospheric residence time.

In addition, upon reviewing our manuscript, we found the following statement regarding the sulfate-rich factor in the discussion section to be not entirely accurate:

"However, it is somewhat surprising that the average OC contribution of the modelled sulfate-rich factor (0.4 $\mu g\ m^{-3}$), attributable to the contribution from bSOA, agrees well with that from the PMF analysis (0.5 $\mu g\ m^{-3}$). Namely, modelled bSOA shows a distinct seasonal variation, whereas the chemical profile of the sulfate-rich PMF factor is stationary in time. This stationary profile then suggests that in wintertime, the PMF-derived sulfate-rich factor contains OC from other sources, since bSOA is largely absent during that time."

While the above discussion point is still valid from the original sulfate-rich factor point of view, its message gets muddied in our work because we have added the MSA-rich factor to the PMF-derived sulfate-rich factor, and background OA to the modelled sulfate-rich factor. We could expand the discussion to nevertheless put our results in the context of the above statement (which remains valid), but we also feel that the statement does not make an especially important contribution to the paper. In the revised manuscript, we have therefore left this statement out.

As a final note; while the sulfate-rich and nitrate-rich factors taken together represent nearly all SIA (arguably an easier to compare to model quantity), we have opted to analyse them as separate factors in part based on the difference between their intrinsic OP values (although this excludes the contribution from the MSA-rich factor; but these details will be worked out when we aim to simulate aerosol OP at a later stage). The definition of aerosol oxidative potential (OP) has now been included in the introduction section.

4. Interpretation of urban downscaling

Around lines 12-15 (Abstract) and 334-340 (Section 4.3), the manuscript highlights improvements from urban downscaling for road traffic and residential heating. At the same time, the results also suggest that downscaling can, in some cases, amplify existing biases (e.g. for biomass burning).

It might therefore be helpful to reflect this nuance more explicitly in the text, so that the overall message does not give the impression that downscaling is uniformly beneficial.

More generally, the discussion of the added value of downscaling could potentially be strengthened by distinguishing more clearly between different aspects of model performance, such as:

- mean concentrations
- temporal variability
- spatial variability across sites

A clearer separation of these aspects could help to better identify under which conditions downscaling leads to improvements.

A more nuanced view of the downscaled results has now been included in multiple places throughout the text, highlighting that downscaling indeed also has the effect of increasing existing (positive) biases at some sites (both urban and traffic). We now also more explicitly note that the correlation statistics at individual sites are changed comparatively little. The spatial correlation, being calculated from the station averages at each of the sites, by its nature describes the (change in) the overall performance across all sites, with downscaling affecting predominantly urban sites.

5. Residential wood burning and condensable emissions

In Section 7.1, the manuscript highlights the sensitivity of residential wood burning to the treatment of condensable emissions, which appears to be an important aspect of the analysis.

It may be helpful if the authors could elaborate more explicitly on how this sensitivity propagates into the source apportionment results. For example, does it primarily affect the absolute magnitude of the biomass burning contribution, its spatial distribution at the urban scale, or both?

Clarifying this point could help to better interpret the role of downscaling relative to uncertainties in emission representation.

To add clarity to the structure of the text, we have now added a section 7.1.1 "Impact on simulation results", which also adds room for additional detail. To this end, impacts on the relevant statistics between urban and rural stations are now also discussed. Phrasing of the original section 7.1 (and what has now been moved to section 7.1.1) has been adjusted to the new text structure while also being reworded in places to add clarity.

6. Synthesis and comparison across factors

In the conclusions (Section 8), the manuscript provides a comprehensive overview of the results. Given the large number of datasets and PMF factors considered, the main messages could perhaps be made more explicit.

It would be helpful to provide a more concise synthesis that highlights:

- where the model shows robust performance
- where the main discrepancies remain
- what factors appear to drive these differences

In addition, a more systematic comparison across factors using consistent statistical metrics (e.g. bias and correlation, as already partly presented in Fig. 2 and Table S4) could help to facilitate comparison between datasets and source categories. A compact summary table might be a useful way to support this.

I believe that addressing these points would further strengthen the clarity and robustness of the manuscript. The comments mainly concern interpretation and presentation rather than fundamental methodological aspects, and I consider that the manuscript has the potential to make a valuable contribution after revision.

We now highlight that the statistics shown in Table 2 summarize the results for the individual PMF factors for each of the three datasets. We thereby take this table as a condensed form of a comparison across factors/source categories, even though we did not explicitly group factors related specific sources (e.g., traffic) together. A similar synthesis table, but now with the wood burning CPOA split as discussed in Section 7, has also been added to the appendix (Supplementary Table S10).

In part in response to comments from reviewer 1, we have also considerably reshaped the conclusion section. To this end, we now use three bullet points to focus on 1) strong points of the analysis of PMF derived and modelled source contributions, 2) weak points with respect to model performance, 3) which model assumptions strongly affect the weak points (i.e., CPOA).

References

Borlaza, L. J. S., S. Weber, G. Uzu, V. Jacob, T. Cañete, S. Micallef, C. Trébuchon, R. Slama, O. Favez, and J.-L. Jaffrezo (2021). "Disparities in particulate matter (PM_{10}) origins and oxidative potential at a city scale (Grenoble, France) – Part 1: Source apportionment at three neighbouring sites". In: *Atmospheric Chemistry and Physics* 21.7, pp. 5415–5437. DOI: 10.5194/acp-21-5415-2021.

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