This is a timely and useful manuscript on aerosol source apportionment modelling with a coupled regional–urban framework. It addresses an important gap by comparing source-resolved model output against several PMF-based observational datasets across a relatively large set of European sites. The multi-dataset setup is a clear strength, and I think the attempt to look at both the regional model performance and the added value of urban downscaling is particularly worthwhile. Overall, the manuscript is carefully done and generally well written, and I appreciated that the authors are fairly open about the uncertainties involved in comparing model results with PMF-derived factors.

In my view, the paper is publishable after minor revision. My comments are mostly about presentation and interpretation rather than any fundamental problem with the work itself. I think the main results are interesting and worth publishing, but a few points could be stated more clearly.

We thank the reviewer for their positive assessment of our work and for the constructive feedback. We have address the suggested refinements, having added additional detail and clarification regarding the matching with PMF factors and urban downscaling throughout the text. Answers to the specific comments are given below in blue. One of the main changes has been that the conclusion section has been reworked to provide a more focused view of our results.

One issue that runs through the paper is the matching between PMF factors and modelled source categories or species. The authors are aware of this and do flag it in several places, which is good, but I think it deserves a bit more weight in how the results are framed. Some of the conclusions look fairly robust, while others seem to depend more strongly on the exact mapping choices that were made. It would help if that distinction came through more clearly, especially in the results discussion and concluding sections.

In the revised manuscript, we have emphasized what the general approach for the matching between PMF factors and modelled sources and species was (e.g., at the start of Section 4.2). To this end, we have also included the motivation behind the merging of certain observationally derived PMF factors; namely, such that the constructed factor can more readily be compared against modelled sources and/or species, making the matching to modelled output species more robust.

We have also added text, for example in the conclusion, highlighting that in our view one of the main challenges is the interpretation of source-specific contributions of primary and secondary OA to PMF factor concentrations. This interpretation is challenging from a PMF analysis point of view, but also from an emission inventory point of view, given that estimates of source-specific SOA production are sometimes included as primary OA (relating to the discussion of CPOA in section 7). Language describing the matching to modelled species has further been clarified in other places throughout the text, also in reply to reviewer 2.

The results also suggest that urban downscaling does improve the representation of some local source contributions, especially road traffic and residential heating. At the same time, the picture is not completely straightforward, since in some cases the downscaling appears to magnify existing biases rather than reduce them. My sense is that the manuscript should be a little more careful here and avoid giving the impression that the added urban-scale treatment is consistently beneficial. It clearly helps in some situations, but not in all, and that comes across in the results.

The more nuanced view of the effects of downscaling has now been highlighted in multiple places throughout the text (e.g., in the abstract, discussion, and conclusion sections), as indeed downscaling can also increase existing biases. Further detail on the impact of downscaling has now also been included in the newly added section 7.1.1, discussing the impact on the model results of the CPOA

wood burning emission split.

More generally, the paper contains a lot of useful detail, but the main message gets somewhat buried at times because there are several datasets, many factor types, and a number of necessary source-matching decisions to keep track of. I think the final sections would benefit from a somewhat tighter synthesis. It would help to focus to three main points: where the model works well, where the main discrepancies remain, and what these mean for future source-specific applications, especially in urban health and policy.

The structure of the conclusion has been adjusted to provide a more focused message. To this end, the first bullet point of the original manuscript (being more of a statement rather than a conclusion), has been merged with the preceding paragraph. This first paragraph now also provides a target for future improvements, i.e. that harmonized PMF protocol applied to both observed and modelled quantities would be beneficial.

The remaining bullet points now highlight strengths (both of the model and of the clear benefit of comparing the model to PMF data), weaknesses (poor model performance for PMF factors related to important emission sources), and the importance of CPOA (tying in with implications for future applications, given the relevance to OP modelling as discussed in the final paragraph).