Anonymous Referee #2, 03 Jun 2025

Reviewer comment: *'The manuscript details the use of a non-reactive tagging method of tracking primary organic matter (OM) using the model COSMO-MUSCAT to investigate the contribution of residential heating to OM during the winter of 2021, at 3 measurement stations across central Europe. The findings indicate that the modelled OM is underestimated at these sites, which is mainly attributed to the current under-representation of wood combustion SOA. The study is well framed, and conclusions are adequately presented. I recommend the publication of this work, before the authors clarify and accommodate the following questions/recommendations.'*

Author's response:

We thank the reviewer for the careful review of our manuscript and constructive comments and suggestions to improve it. We have modified the manuscript accordingly as outlined below.

Reviewer Comment:

1.) L6:8: "Although the magnitude and temporal changes of the model results mostly agree with total OM values at two measuring stations, it appears to underestimate measurements at a site in the central Czech Republic." It would be better to mention earlier in the introduction that there was 1 measurement site in Germany and 2 in CR to avoid confusion.

Author's response:

Good point, we added this information in the abstract.

Author's changes in manuscript:

Line 4 and 5:

'The model results are compared with winter measurements from one site in Germany and two sites in the Czech Republic, where solid fuels are commonly used for heating.'

Reviewer Comment:

2.) L18: " wider societal costs associated with it". interesting point but would benefit from stating the wider societal costs. The values can be presented here to underline the magnitude of the costs and losses.

Author’s response:

We added an estimation of the cost of air pollution:
‘A report by the World Bank Group (2022) estimates that the societal cost of ambient fine particulate matter pollution in the Europe and Central Asia region reached 4.6% of gross domestic product (GDP) in 2019. This estimate reflects the economic impact of PM2.5 related health outcomes, including premature mortality, morbidity, and lost productivity’

Author's changes in manuscript:

Line 19 ff:

‘A report by the World Bank Group (2022) estimates that the societal cost of ambient fine particulate matter pollution in the Europe and Central Asia region reached 4.6% of gross domestic product (GDP) in 2019. This estimate reflects the economic impact of $PM_{2.5}$ related health outcomes, including premature mortality, morbidity, and lost productivity.’

Reviewer Comment:

3.) L71: “The second main wind direction is East (about 17% of the time), with dry continental air masses influenced by long-distance transport from Poland, Belarus, Ukraine, Slovakia and the Czech Republic (Spindler et al., 2001, 2012, 2013).”. Is this during summer or winter?

Author’s response:

The given percentages are referring to the whole year, not only to one season. We changed the sentence accordingly.

Author's changes in manuscript:

Line 79 and 80:

‘Easterly winds occur 17% of the time throughout the year, bringing dry continental air masses affected by long-range transport from Poland, Belarus, Ukraine, Slovakia, and the Czech Republic (Spindler et al., 2001, 2012, 2013).’

Reviewer Comment:

4.) L135: Add the values of splitting profiles in the appendix.

Author’s response:

We have added the values to the appendix. For simplicity, we only provide tables for the GNFR C values.

Author's changes in manuscript:

Tables A1 and A2 have been added to the appendix. Table A1 provides the splitting factors for particulate matter, while Table A2 provides the splitting factors for non-methane volatile organic compound (NMVOC) emissions.

Reviewer Comment:

5.) L163: wouldn't it benefit to use a 2-d VBS method over the 2 product Odum parameterization?

Author's response:

This is a valid point. Implementing the VBS method would enhance the model because it accounts for chemical ageing, unlike our current two-product approach. While incorporating VBS into COSMO-MUSCAT would be complex, it is worth considering for future work. However, this falls outside the scope of this revision.

Nevertheless, we believe that the two-product approach remains suitable for our study. By categorising products into lower- and higher-volatility classes, the approach covers a broad range of SOA products and allows for easy adaptation to new experimental data.

Author's changes in manuscript:

None

Reviewer Comment:

6.) L196: add a table in the appendix detailing the main tagged species

Author's response:

We have added a table to provide an overview of all the species that have been tagged.

Author's changes in manuscript:

Table A3 lists all the species that were tagged in this study.

Reviewer Comment:

7.) L208-209: Since gas phase species and aerosol chemistry is not considered, can the authors quantify how much the SOA will be under-predicted based on the lack of these processes in the model.

Author’s response:

Our new tagging approach currently only analyses passive tracers (i.e. non-reactive tagging), so SOA is not explicitly evaluated by source region or sector. However, for the given winter scenario we primarily attribute anthropogenic SOA to the other combustion sector (GNFR-C) as the precursor AVOC are mainly emitted from this sector. SOA from anthropogenic sources contributes between 13 - 20 % to the mean total OM mass at the three stations during the investigated wintertime period. Similarly, biogenic SOA contributes 25 - 45 % to total OM on average. Gas-phase processes and aerosol chemistry are generally implemented in COSMO-MUSCAT, the sentence this comment refers to has been revised for better clarity. SOA formation itself is represented via the SORGAM module, which is active in the tagging simulations. However, due to the nonlinearity of SOA formation, it is not possible to directly tag SOA species.

Author's changes in manuscript:

Line 224 and 225:

‘However, gas phase chemistry and aerosol chemistry are not considered at present within the tagging algorithm.’

Reviewer Comment:

8.) L249: is there a comparison of simulated and measured boundary layer height?

Author’s response:

Following the other reviewer's suggestion, we have changed this paragraph and removed the citation of Stern et al. (2008) at this point. Consequently, the comment regarding the boundary layer height has been removed.

Author's changes in manuscript:

Line 262 ff:

‘Im et al. (2015) analysed the performance of multiple models in simulating $PM_{2.5}$ concentrations as part of the AQMEII model intercomparison project. They found that most models systematically underestimated $PM_{2.5}$ at rural stations, with biases ranging from -2%

to -60%. The COSMO-MUSCAT model performed relatively well, showing a bias of -24.82%. However, all models struggled to capture wintertime levels, underestimating concentrations by more than 50% across all regions.'

Reviewer Comment:

9.) L253-254: provide value for the "slight difference".

Author's response:

Good point, we changed the sentence to give more information:

Author's changes in manuscript:

Line 268 ff:

'The snow event on 7–8 February led to a decrease in $PM_{2.5}$ concentrations in Melpitz by approximately 10 µg m^{-3}. In Frýdlant, a slight decrease of around 4 µg m^{-3} was observed after the event, while in Košetice, concentrations even increased by about 4 µg m^{-3}, indicating limited overall washout effects.'

Reviewer Comment:

10.) Figure 4: simulated PM2.5 and AMS PM1 are not directly comparable. for e.g if we consider Nitrate how can one interpret high model PM2.5 conc and low AMS PM1 concentrations. Both the values could be in similar if we consider only modelled PM1. Also, 4d is comparing PM1, PM2.5 and PM10. Are the PM1, pm2.5 and PM10 masses correlated? It would perhaps be better to compare, relative or normalized nitrate, sulphate, OM and EC concentrations if one must compare PM1, PM2.5 and PM10 conc.

Author's response:

We thank the reviewer for this important feedback. We agree that a direct comparison between modelled $PM_{2.5}$ and observed PM_1 (from AMS/ACSM) introduces some uncertainty. However, we believe that the comparison remains valid and informative for multiple reasons.

We acknowledge the findings of Poulain et al. (2020), who noted that ACSM may underestimate total sulfate concentrations when the PM_1 fraction of $PM_{2.5}$ mass falls below 60%, as often occurs in winter when particles grow beyond the submicrometer range due to processes like ammonium nitrate condensation. Under such conditions, a portion of sulfate and nitrate mass may reside in particles larger than 1µm and thus be missed by ACSM. This implies that the $PM_{2.5}$ concentrations of sulfate and nitrate may be somewhat higher than indicated by the ACSM data. Nevertheless, the magnitude of these species remains relatively small at all sites, and their potential uncertainty does not substantially affect our main finding, that the model's underprediction of total $PM_{2.5}$ mass cannot be attributed to a underestimation in inorganic aerosol. Nevertheless, nitrate and sulfate are typically predominantly found in PM1 (Zhang et al., 2023).

We therefore believe that the qualitative comparison between modelled $PM_{2.5}$ and measured PM_1 remains informative. The model does not explicitly resolve particle size distributions, and the $PM_{2.5}$ output for these species primarily represents the accumulation mode.
For EC, our model only considers primary combustion sources, which are known to emit particles almost exclusively within the PM_1 size range. Observations at Melpitz by Poulain et al. (2011) suggest that more than 90% of eBC mass in PM_{10} is actually in the PM_1 fraction. Therefore, comparison of the different size classes should be reasonable. This also applies to the organic aerosol component: organics are mainly distributed in the submicrometer size range throughout the year (Poulain et al. 2020), making the comparison between modelled $PM_{2.5}$ OM and AMS PM_1 OM reasonable, especially in winter.

We added this argumentation to our results evaluation in the manuscript.

Poulain, L., Spindler, G., Birmili, W., Plass-Dülmer, C., Wiedensohler, A., and Herrmann, H.: Seasonal and diurnal variations of particulate nitrate and organic matter at the IfT research station Melpitz, Atmospheric Chemistry and Physics, 11, 12 579–12 599, https://doi.org/doi:10.5194/acp-11-12579-2011, 2011.

Poulain, L., Spindler, G., Grüner, A., Tuch, T., Stieger, B., van Pinxteren, D., Petit, J.-E., Favez, O., Herrmann, H., & Wiedensohler, A. (2020). Multi-year ACSM measurements at the central European research station Melpitz (Germany) – Part 1: Instrument robustness, quality assurance, and impact of upper size cutoff diameter. Atmospheric Measurement Techniques, 13, 4973–4994. https://doi.org/10.5194/amt-13-4973-2020

Author's changes in manuscript:

Line 299 ff:

'The AMS/ACSM may underestimate total sulfate and nitrate concentrations in winter, when particle growth shifts part of the mass beyond the PM_1 range (Poulain et al., 2020), though these species are generally predominantly found in PM1 (Zhang et al., 2023). Given their relatively small contribution to total $PM_{2.5}$ at our sites, it is unlikely that secondary inorganic aerosols are responsible for the discrepancy between the predicted and measured $PM_{2.5}$ aerosol mass concentrations.'

Line 310 ff:

'Although differences in particle size cut-offs must be considered when comparing observations and model results, Poulain et al. (2011) found that around 90% of the mass of elemental black carbon (eBC) in PM_{10} is contained within the PM1 fraction. Comparing across these different size classes should therefore be reasonable.'

Line 329 ff:

‘The discrepancy between Sunset and AMS/ACSM observations may partly arise from the different particle size ranges each instrument targets: Sunset samples $PM_{2.5}$, while AMS/ACSM captures only PM_1. However, since organic aerosol is predominantly found in the submicrometer size range throughout the year (Poulain et al., 2020), the impact of the size cut-off on the comparison is expected to be minor. This is further supported by observations in Frýdlant, where both PM_1 (online) and $PM_{2.5}$ (offline) Sunset data are available and show only small differences.’

Reviewer Comment:

11.) L297: “The model underestimates the OM concentrations in Košetice (RMSE: 6.48 µg m^{-3}) while for Melpitz and Frýdlant the overall fit is good (RMSE: 1.17 and 2.01 µg m^{-3}).” doesn’t this negate the earlier claim that the OM is underestimated in the simulations?

Author’s response:

This is correct for AMS/ACSM measurements, but not for those taken using the sunset filter. We have improved the results section and provided a more detailed explanation for each device.

Author's changes in manuscript:

Line 324 ff:

‘Across all three stations, the comparison to the Sunset data show a systematic underestimation by the model, with large negative NMB values: -73% in Melpitz, -79% in Košetice and -67% in Frýdlant.’

Line 334 ff:

‘AMS/ACSM instruments are particularly well suited for capturing temporal variability, due to their high time resolution. The Sunset instruments provide an estimate of the total carbonaceous mass and are useful for assessing the magnitude of concentrations. It uses the same filters as the gravimetric reference method, allowing a more direct comparison to total $PM_{2.5}$ mass and offering a more complete picture of the aerosol burden. In Melpitz and Frýdlant, the model aligns reasonably well with AMS/ACSM observations, with RMSE values of 1.17 and 2.01 µg m^{-3} and NMBs of –8% and +18%, respectively. Correlation is also relatively strong in Melpitz (R = 0.60), but lower in Frýdlant (R = 0.19), where the model fails to capture diurnal variability. The model underestimates the OM concentrations by AMS/ACSM in Košetice (RMSE: 6.48 µg m^{-3}; NMB: –74%) and also does not fully reproduce the diurnal variations (R = 0.39) (see Fig. A2 in the Appendix).’

Reviewer Comment:

12.) L310-315: Since the winter values are heavily meteorological dependent one must mention if the weather patterns during the said years matched 2021. From the description above the year 2021 seems to be an odd one considering the long Sahara dust events and the cold periods. I would suggest making such comparisons to more tangible SOA concentrations, which can then present a broader picture of a trend in SOA underestimation.

Author's response:

It is true, that the direct comparison with previous years is not directly possible, but still it might give some valuable information about the study sites. We therefore would like to keep the values in the manuscript, we added an explanatory sentence to make the differences clear.

Author's changes in manuscript:

Line 363 ff:

'For our study period we found Sunset Filter values ranging in average from 5.06 $\mu g\ m^{-3}$ in Melpitz to 7.74 $\mu g\ m^{-3}$ in Košetice, exceeding typical values reported for previous years. This suggests a strong influence of meteorological conditions on the overall concentration levels.'

Reviewer Comment:

13.) L 345: Public power contribution. This is interesting. One would expect higher contributions from public power at Košetice especially in the cold period as the air masses is stagnant. Can you explain why is the contribution of public power low during the cold period?

Author's response:

Yes, we would assume that the relevant power plants are not close enough. Overall, Public Power's contributions are low at all stations. For Frýdlant, the proximity to the Turów power plant can be seen in the changing country contributions.

Author's changes in manuscript:

None

Reviewer Comment:

14.) L 358-359: Melpitz cross border transport. Is this during the cold or warm period? Looking at Figure 7, it appears that Košetice and Frýdlant have larger cross-border (Poland) contribution to public power than Melpitz. Also, it appears that at Melpitz the cross border contibutions is more in the warm period but at kosetic the polish and german contributions are significant even during the cold period.

Author's response:

The high cross-border contributions in Melpitz refer only to the 'Other Combustion' sector. The public power sector also exhibits higher levels of cross-border pollution in Košetice and Frýdlant. The relevant sentence has been revised for clarity.

Author's changes in manuscript:

Line 405 ff:

'Contributions to fine OM from the 'other Combustion' sector are highest in the Czech Republic and in urban agglomerations in Poland and around Berlin, Germany (see Fig. 8, right panel). The main contributors to the concentrations observed at the stations are emissions originating within the country where the station is located.'

Reviewer Comment:

15.) Figure 7: I would suggest removing the wind barbs since it doesn't add any information's. or did the authors miss the y axis with the degrees?

Author's response:

We consider the correlation between fluctuations in wind direction and alterations in country contributions to be a subject of interest. Therefore, the windbarbs provide information on the prevailing wind at that time. To provide further clarification, the windbarbs have been included in the legend. The orientation of the barbs indicates the wind direction. We therefore assume that an additional y-axis is not necessary.

Author's changes in manuscript:

We have included a description of the wind barbs in the legend for Figure 7.

Reviewer Comment:

16.) Table 3: why is alpha 1 same for S1 and S3? Shouldn't it be same for S2 and S3? Some explanation is needed in the main body or the table caption.

Author's response:

S1 only includes the SOA yield changes, which is done by adjusting alpha 1. While S2 only includes the additional emissions with SOA yields as in the base run. S3 then includes both. We added more detailed explanation in the table description

Author's changes in manuscript:

The table referred to here is now Table 4. We have improved the description:
'Table 4. Overview of the sensitivity simulations. Shown are changes to α_1 to adjust the SOA yield parameter for aromatic precursors and scaling of CSL emissions based on CO emissions from GNFR C to account for phenol contributions.'

Reviewer Comment:

17.) L496-497: it would be interesting to see if the increase in OM holds true during the cold period of stagnant airmass at these sites.

Author's response:

The underestimation during this period is substantial and cannot be attributed solely to the underprediction of SOA. The increased emissions used in our sensitivity simulation do not fully explain the significant discrepancy. Therefore, it would be highly beneficial to incorporate temperature-dependent combustion emissions in future simulations to better capture increased heating activity during cold periods. This is planned as the next step to further improve model performance.

Author's changes in manuscript:

None

Reviewer Comment:

18.) L 537-538: I would suggest saying that the diurnal profiles are reproduced, not the total OM magnitude.

Author's response:

Yes! This is true since the AMS data is represented well but Sunset data is underestimated at all stations. We changed the sentence accordingly.

Author's changes in manuscript:

Line 597 ff:

‘Although the present study reproduced diurnal OM profiles well at two monitoring sites, measurements at Košetice are underestimated, partly due to an inadequate representation of SOA formation from residential heating (wood combustion), a major source of anthropogenic VOCs.’