Authors' responses to referee comments on **egusphere-2025-4972 titled "Emitted yesterday, polluting today: temporal source apportionment of fine particulate matter pollution over Central Europe"** by Huszar et al.

Referee #3

Dear Anonymous Referee #3,

thank you for your time and effort to review our paper and for all your comments. Please find our point-by-point responses to the points of your revision below (in bold italic) .

General Comments

This study focuses on the critical scientific gap in the temporal source apportionment of fine particulate matter ($PM_{2.5}$) pollution in Central Europe. It innovatively introduces the Temporal Source Apportionment (TSA) method, combined with the CAMx chemical transport model, to investigate the contribution of emissions from the past 14 days to the $PM_{2.5}$ and its components concentrations on a given day. This method fills the gap in existing studies that have insufficient attention to the temporal dimension of pollution, quantifying the impact of historical emissions accumulation on pollution events. The study offers new insights into regional air pollution with strong scientific and practical significance. The results show that current-day emissions dominate winter $PM_{2.5}$ pollution, while past emissions also play an important role in pollution formation, especially under adverse meteorological conditions. These findings provide valuable insights for pollution control policies, emphasizing the importance of emission reductions several days in advance.

However, the study also has some limitations, particularly the significant underestimation of pollutant concentrations by the model, which may affect the quantitative assessment of past emission contributions. Additionally, the limitations of the emission inventory may also influence the results. Some of the model assumptions and the credibility of the findings need further clarification to strengthen the reliability of the conclusions. I recommend minor revision.

Specific Comments

1.Data validation

The model validation shows a systematic underestimation of $PM_{2.5}$ and NO_2 concentrations. This issue should be discussed more explicitly in the Discussion section. In particular, the authors should clarify how such underestimation may influence both absolute and relative contributions of past

emissions, and whether the relative temporal patterns are expected to remain robust despite these biases.

Author response:

Indeed, we encountered a systematic and strong underestimation of both PM and their precursors (SO_2 and NO_2), which is probably caused by the combined effect of underestimated emissions and too strong vertical mixing resulting in fast upward transport and overall dispersion of the emitted pollutants (see our previous validation study Prieto Perez et al.(2025)). We can expect that due to this negative bias the absolute contributions of previous days emissions are underestimated. However, the relative contributions are probably not affected too much as these represent the ratio of the absolute contribution and total concentration. We clarified this in the discussion section.

2.Methodology

The 14-day emission tagging strategy is effective in revealing temporal dynamics. However, further clarification is needed regarding how overlapping contributions within the 14-day cycle are treated (e.g., interactions between day-0 and day-1 emissions). In addition, a more detailed discussion of how meteorological conditions modulate the estimated emission contributions would improve the methodological transparency.

Author response: The source apportionment technology used here provides PM attribution for a given emission matrix - which here means dividing emissions into different days in a cyclic 14 day period defining 14 temporal "sources" - but does not provide quantitative information as to how PM contributions would change as emissions are altered. This means that the interaction of these 14 "sources" is not accounted for. If one is interested in the chemical interactions of emissions from different days, the sensitivities/impacts have to be calculated which can be achieved by removing a day from the 14 day period and running CAMx without the emissions for this day. This would require to run 14 additional 10yr simulations which is computationally extremely demanding so we decided to analyze only the contributions determined by the PSAT technology rather than the impacts.

(Regarding the contributions in PSAT, e.g. for sulfur emissions this means that S can contribute only to sulfates in PSAT but it is clear, that adding SO_2 to the chemical system alters also ammonium concentrations via ammonium sulfates and this in turn impact nitrates via removing ammonium making it less available for nitrates – see

Bartik et al., 2024) . We made these points clear in the revised manuscript.

We also added a paragraph about the potential impact of different meteorological conditions to the lifetime of PM pollution and thus the theoretical contribution of previous days emissions. In particulate, we considered vertical mixing (and stratification), wind speed, temperature and precipitation and their impact.

3.Results analysis

The finding that current-day emissions dominate winter $PM_{2.5}$ concentrations, while past emissions remain important under stagnant conditions, is compelling. Additional discussion on the generality and variability of this behavior under different urban meteorological regimes would further strengthen the results, especially for low wind speed and stable boundary layer conditions.

Author response: We agree that the study misses some further evidence about the role of stagnant conditions on the absolute and relative impact of past anthropogenic emissions on fine PM concentrations, therefor we made two additions to the study that further support our interpretation of the modelled impacts: i) we added a new figure to the revised text, Fig.18 that shows the distribution of the average DJF and JJA ventilation coefficient for the whole domain. We have chosen this metric as it combines both the vertical mixing (in terms of the PBL height) and wind-speed, knowing that under stagnant conditions ventilation is very low. This figure helps to identify regions with potential for accumulation of PM, and indeed, such areas is e.g. Northern Italy, were our results indicated the highest absolute and relative impact of past emissions. We added a paragraph to the discussion of the results to discuss the modelled PM contributions in the light of these average ventilation conditions.

Moreover, ii) we added to the supplement the scatter plots between the daily average impact of past emissions and selected meteorological variables (windspeed, pbl height, ventilation coefficient and 2m temperature) for all other cities as so far only Milan was presented (which, on the other hand, has been moved to the main text as these figures are important for the interpretation of the results). We have chosen the impact of day-7 emissions to demonstrate the potential importance of such old emissions and their connection to meteorological conditions. These new figures in the Supplement now add important information on the intercity variability of the association

of emissions impact and meteorological conditions and at the same time they show, that high relative contribution of past emissions (1 week old) is almost exclusively associated with very low ventilation (in each city).

4.Lines 1–15 (Abstract)

The description of the novelty of the Temporal Source Apportionment approach is relatively general. It would be helpful to more clearly distinguish TSA from existing PSAT-based or age-distribution approaches and explicitly state the added value of this method.

Author response: We added a few sentences to point out the real novelty of the study, which lies in the continuous long-term adoption of TSA. However, we had to be brief due to limitation on the length of the abstract (250 characters).

5.Lines 25–40 (Introduction)

While the review of regional and long-range transport studies is comprehensive, the distinction between previous short-term event-based temporal studies and the lack of long-term statistical analyses remains somewhat unclear. Clarifying this distinction would better position the contribution of the present work.

Author response: Indeed, we agree that this distinction deserves a paragraph. Therefor we added a few references in the Introduction to highlight the importance of "long term" studies that examine the long term (statistical) impact of regional and global emissions (acting via long-range transport) and what is their added value compared to short-term impact studies.

6.Lines 105–115

The use of a two-mode aerosol scheme in CAMx may limit the representation of aerosol aging processes. A brief discussion of this limitation and its potential implications would be appropriate.

Author response: Indeed, in the two-mode CF (coarse-fine) aerosol scheme, PM is treated either of size 2.5 μm in diameter or larger than this threshold (coarse). There is no mass transfer between these size bins. All the secondary aerosol components are considered as fine and their size evolution is not treated, i.e. they are not aged, which is the case also of the secondary organic aerosol within the SOAP module. We understand this as a limitation as aging has direct impact on the aerosol lifetime, e.g. it can increase their hygroscopicity (Rudich et al., 2007., Georgopoulou et al., 2025) which enhances wet removal. We added these notes to the description of the limitation of the CF method

(also noted in the Conclusion section under the summary of the limitations of the study)

7.Lines 180–190

Given the systematic underestimation of PM concentrations, the magnitude of model bias across different seasons and between urban and rural stations should be quantified more clearly.

Author response: We included more quantitative information on the bias while attempting to point the differences in model biases between rural and urban stations and between different seasons (DJF vs JJA).

8.Lines 270–275

The rapid decay of ammonium contributions is closely linked to its chemical formation pathways. A comparative discussion with sulfate and nitrate behavior would help contextualize this result.

Author response: Ammonium ($NH4^+$, denoted PNH4 in our study) is not directly emitted by instead, it forms from ammonia (NH_3) together with nitrogen oxides and sulfur oxides forming ammonium nitrates and ammonium sulfates. While the lifetime of NH_3 is short (a few hours), the lifetime of ammonium is much longer. Ie the impact of previous days emissions on PNH4 is limited by the quick transformation of ammonia to ammonium resulting in older emissions contain almost no NH_3. But this decay is also in line with the contribution of previous days emissions to sulfates and nitrates, which show that day-2 emissions in both cases contribute to total PSO4 and PNO3 only by around 8-12%. Limited contribution to these consequently means also limited contribution to ammonium formation which requires PSO4 and/or PNO3 to form. We added these notes in the revised manuscript, however, we did so in the Discussion section to separate the presentation of the result from their detailed discussion.

9.Lines 325–330 (Figure 12)

The substantial inter-city differences in day-0 contributions would benefit from quantitative support using meteorological indicators such as wind speed or boundary layer height.

Author response: Indeed, the day-0 relative contribution exhibit a large spread, so we added a paragraph to the Discussion section to try to explain at least partly the causes for such differences. We supported our argument with a new table (Tab 1) where we indicate the average winter and summer values of the relevant meteorological variables which could explain those differences. Mainly the wind speed and

ventilation coefficient serves as strong evidence by noting that windier cities remove the emissions more rapidly making their local contribution to the actual day's concentrations low, and vice versa.

10.Lines 425–430

The attribution of inter-city differences in day-0 contributions to ventilation conditions is plausible but not directly demonstrated. Additional quantitative evidence or references are recommended.

Author response: We would like to refer here also to Tab 1. which demonstrates substantial differences in the ventilation coefficient among cities (which is further seen also from the Fig. 18).

11.Figures 4–11 and 14–18

Many figures exhibit highly similar spatial patterns and temporal decay structures, which may give an impression of redundancy. The authors are encouraged to assess whether all figures are necessary in the main text, or whether some results could be summarized schematically or moved to the Supplement.

Author response: We agree that figures often show similar results, however we considered also this as a valuable information for the reader. It is not straightforward that all primary/secondary aerosol components will exhibit similar decay structures. However, we tried to at least remove some parts of the figures which show little information: e.g. the JJA PM2.5 contributions (Fig. 5) show very rapid decay, therefore we removed the older day emissions contributions and kept only the day-0 to day-3 ones (along with the BVOC contributions). The "full" (original) figure has moved to the Supplement. We did the same with the JJA SOA contributions, the new Fig. 10 is now smaller, while the full contribution of older emissions is presented in the supplement. Furthermore, we also moved Fig. 18 (POA daily variability) to the supplement as it shows very similar results to PM2.5.

Technical Corrections

1.The term $PM_{2.5}$ is sometimes written without proper subscript formatting (e.g., Lines 5, 405). Consistent use of $PM_{2.5}$ is recommended throughout the manuscript. In Figure 12, "PM25" should be corrected to "$PM_{2.5}$".

Author response: We understand the sometimes, PM2.5 is used in subscripted version as $PM_{2.5}$.or PM_{10}) however this is considered only as an alternative to PM2.5/PM10 notation, which we prefer, as subscript are standarly used for chemical substances (e.g. NO_2, SO_2) to

characterize the number of atoms while here the number stands for the size. Also, in our previous studies we have used PM2.5 instead of subscript so we would prefer to keep this consistency. Fig. 12 was corrected.

2.Figures 1 and 2 Axis labels should be made clearer to improve readability.

Author response: Figure 1 (the concept of temporal source apportionment) has been enlarged to increase the readability. We did the same for Fig.2 and Fig 3, which we believe are now much easier to comprehend (including the smallest fonts used for Axis labels).

3.Units and symbols

Concentration units are inconsistently written (e.g., μg/m³, μgm⁻³, ugm-3). Please standardize unit notation across the text and all figures.

Author response: We unified the unit notation and now in the whole text and all figures, units of concentrations are in μgm⁻³.

4.Line 150

The full name of NMVOC should be defined at its first occurrence.

Author response: Defined.

5.Line 215

Typographical error: “dye to” should be corrected to “due to”.

Author response: Corrected.

6.Line 291

“majority of SOA” should be revised to “the majority of SOA”.

Author response: Corrected.

7.Lines 412–414

Typographical error: “therefor” should be corrected to “therefore”.

Author response: Corrected.

8.Reference formatting

Instances of “Karlický et **a**., 2020” should be corrected to “Karlický et **al.,** 2020”.

Author response: Corrected.

9.Lines 81–83

Subject–verb agreement error: “Xie et al. (2023) has chosen …” should be revised to “Xie et al. (2023) have chosen …”.

Author response: Corrected.

References:

Bartík, L., Huszár, P., Karlický, J., Vlček, O., and Eben, K.: Modeling the drivers of fine PM pollution over Central Europe: impacts and contributions of emissions from different sources, Atmos. Chem. Phys., 24, 4347–4387, https://doi.org/10.5194/acp-24-4347-2024, 2024.

Georgopoulou, M. P., Florou, K., Matrali, A., Starida, G., Kaltsonoudis, C., Nenes, A., and Pandis, S. N.: Diurnal aging of biomass burning emissions: impacts on secondary organic aerosol formation and oxidative potential, Atmos. Chem. Phys., 25, 15835–15855, https://doi.org/10.5194/acp-25-15835-2025, 2025.

Rudich, Y., Neil M. Donahue, Thomas F. Mentel: Aging of Organic Aerosol: Bridging the Gap Between Laboratory and Field Studies. Annual Review Physical Chemistry. 58:321-352. https://doi.org/10.1146/annurev.physchem.58.032806.104432, 2007.

Referee #2

Dear Anonymous Referee #2,

thank you for your time and effort to review our paper and for all your comments. Please find our point-by-point answers to the points of your revision below (in bold italic).

The present manuscript by Peter Huszár and co-workers described a novel Temporal Source Apportionment approach within the CAMx chemical transport model to address the absolute and relative role of emissions from previous days on PM2.5 pollution. The research results showed that the contribution of previous emissions to PM2.5 concentrations gradually decreases by days. These conclusions drawn are similar to our common sense. The results of this study not only enrich the basic knowledge in the field of atmospheric chemistry models, but also provide scientific support for the development of refined PM2.5 control strategies. The following issues still need to be clarified by the authors.

1. As shown in Fig 1, the author “split emissions into 14 artificial "temporal sectors" corresponding to one day within 14 day period and this means that

emissions from each of these 14 "sectors" occur once in 14 days." In my opinion, such "emissions" is a continuous behavior. Why it occured once in 14 days. So, the approach is an important issue that requires clarification.

Author response: Emissions are continually introduced to the system as if no temporal source apportionment was used, the only difference is the tagging of emissions according to the day they have been emitted and there are 14 different tags which cycle throughout the whole 10 yr period. This is why we noted in our manuscript that the results – in terms of absolute concentrations – are exactly the same as in Prieto Perez et al. (2025), where these emissions where applied in a standard continuous manner. Here, what we did is that we "labelled" the emissions according to which day they belong in the cyclic 14 period. We clarified this in the revision.

2. What is the mean of "DJF" and "JJA"?

Author response: These abbreviations have been defined at their first occurrence.

3. The resuls showed in Figure 13-18, two noticeable sudden changes in May and October from the relative contribution of emissions from day-X can be observed, why?

Author response: As written in the manuscript, there is indeed a sudden drop of values seen in the daily variability of the relative contributions for PM and basically all of its components (both primary and secondary) occurring in the beginning of April. This is explained by the monthly temporal factors that are used to dissaggregate the annual totals. They expected a sudden decrease (to zero) of emissions from domestic combustion due to heating on 1 April. As this is the primary source of PM in urban areas, once it is switched off, the contribution of anthropogenic emissions show a large drop and long range transport becomes more important (which manifests itself in the contributions from the boundary conditions - not shown in this paper). The Nov 1 increase has the same explanation – this is the date when our emissions temporal factors expect the domestic heating is turned on and the local emissions sources become dominant.

4. why the contributions of previous days emissions to PM2.5 and aerosol components were disscussed only during summer and winter?

Author response: it is clear that analyzing only the annual means would hinder information about the seasonal differences, so we decided to look at seasonal means. In this regard we had to make decision between showing all four seasons or only DJF/JJA, but due to

the large number of outputs we preferred to present only the two "extreme" seasons, so the hottest and the coldest one (for the region of interest). Moreover, the emissions temporal patterns show very clear distinction between these two seasons supporting our decision.

References:

Prieto Perez, A.P., Huszár, P. and Karlický, J.: Validation of multi-model decadal simulations of present-day central European air-quality, Atmos. Environ., 349, 121077, 1352-2310, https://doi.org/10.1016/j.atmosenv.2025.121077., 2025.