Response to Review Comments by Anonymous Referee #1 on "Short-term Source Apportionment of Fine Particulate Matter with Time-dependent Profiles Using SoFi: Exploring the Reliability of Rolling Positive Matrix Factorization (PMF) Applied to Bihourly Molecular and Elemental Tracer Data" by Q. Wang et al.

General Comments by Anonymous Referee #1:

I thoroughly reviewed this manuscript. The manuscript titled "Short-term Source Apportionment of Fine Particulate Matter with Time-dependent Profiles Using SoFi: Exploring the Reliability of Rolling Positive Matrix Factorization (PMF) Applied to Bihourly Molecular and Elemental Tracer Data" suggests that short-term PMF analysis with the *a*-value constraints in SoFi can be utilized to apportion primary sources accurately. This may have implications for short-term pollution episodes management. The topic is interesting and meaningful. However, there are some problems in the article. I would recommend authors concerns on the following comments. Minor revision is recommended.

Response to General Comments: We thank the reviewer for the comments and acknowledgment of the importance of our work. Below is our point-by-point response to each comment, marked in blue. The related text in the manuscript is copied here for reference, with newly added/revised text underlined.
Changes made to the main text will also be marked in blue in the revised manuscript file.

Line 35, "The implementation of stringent control measures since 2013 has led to declining concentrations of PM5 in many megacities in China", it is recommended to provide specific data on the decrease in PM2.5.

Response: Suggestion taken; the following statement will be added:
"The implementation of stringent control measures since 2013 has led to declining concentrations of $PM_{2.5}$ in many megacities in China, with annual-average decreased from 72.3 μg m^{-3} in 2013 to 47.4 μg m^{-3} in 2017, as calculated from measurements in 74 cities across China (Chow et al., 2022a; Wang et al., 2020)."

Line 55, "This limitation explains the common observation that PMF with robust mode tends to underestimate the high concentration data while overestimating the low concentration data.", it is recommended to modify this sentence to make it coherent with the preceding and following sentences.

Response: The reviewer may have misunderstood the following sentences. The sentence "Sporadic sources, such as firework emission…" mainly describe the pollution characteristics of the sporadic sources such as firework contributions, which can contribute significantly to PM pollution and offset the pollution controls of other anthropogenic sources. While the sentence "This limitation explains …"

describe the model performance of the PMF on the high conc. data, i.e., underestimation of the data, thus leading to the lower estimation of the corresponding sources. To improve the clarity, we will rephrase the sentences in the revised manuscript as following:

"Source profile changes over extended observation period are often expected for certain sources. For instance, biomass burning exhibits variations in dominant biomass materials across different seasons; the implementation of catalytic converter replacement program alters the source profiles of vehicular emissions (Lee et al., 2017). Sporadic sources, such as firework emission during holidays or wildfires during dry seasons, can significantly contribute to PM pollution episodes that persist for hours to days, often overshadowing the effects of reductions in anthropogenic emissions (Song et al., 2021; Kong et al., 2015). The PMF analysis using the long-term data sets could not properly reflect source profile changes experienced during the longtime span, which is commonly expected. In other words, long-term PMF is inherently unsuitable for apportioning sporadic sources or ephemeral pollution events. This limitation explains the common observation that PMF with robust mode tends to underestimate the high concentration data while overestimating the low concentration data (Henry and Christensen, 2010). Contribution estimates of these sources would be biased when apportioned alongside other regular sources using long-term observational data."

Line 99, please define SOA when it first appears in the text.

Response: We will revise it as suggested.

"…and α-pinene secondary organic aerosol (SOA) tracers …"

Line 121, it is recommended to explain what the meaning of k and j in Eq 4.

Response: We will revise it as suggested.

"…Here, the index *j*, which varies between 0 and the number of species-*m*, represents the species of the k^{th} factor. The index *i*, which varies between 0 and the number of samples-*n*, is the sample of the k^{th} factor. f_{kj} and g_{ik} are the anchoring profiles and anchoring contributions…"

Line 170, Do you think these "18 days" must be continuous? Does discontinuity have an impact on the results?

Response: No, the continuity is not a prerequisite. Hypothetically, there could be data gaps, as the individual sets of hourly observations are used as inputs and there is not a requirement that the individual observations should be continuous. Previous studies with higher time resolution measurement suggested a window length of 14 d, we did a preliminary test with our bihourly data sets and found that a longer time period would provide a more robust solution with cleaner source profiles.

We will revise the wording to improve the clarity in the revised manuscript:

"The short-term source apportionment analysis was conducted using data from the first sampling period, spanning 18 days from 29 Dec. 2019 to 15 Jan. 2020. The selection of the window length may vary depending on the specific data sets under study. The determination of the window length for our observational data set is shown in Text S2, where 4 d, 7 d, 10 d, 14 d and 18 d were initially evaluated."

Line 121, please define Q/Qexp.

Response: Suggestion taken; the following statement will be added:

"The ratio Q/Q_{exp}, where $Q_{exp} \approx n \times m - p \times (n + m)$, indicates the overall fitting of all input species and is reciprocally associated with the fitting (Norris et al., 2014)."

Reference:

Norris, G. A., Duvall, R., Brown, S. G., and Bai, S.: EPA Positive Matrix Factorization (PMF) 5.0 Fundamentals and User Guide, Environ. Prot. Agency Off. Researc Dev. Publushing House Whashington, DC 20460, 136, 2014.

Line 221, please explain what Nfirework_data

Response: Suggestion taken; the following statement will be added:

"…was excluded due to the limited availability of data points influenced by firework emissions ($N_{firework\_data}$=2, representing the number of data points under the influence of firework emissions)."

Line 240, please define

Response: Suggestion taken; the following statement will be added:

"Among the four secondary sources, secondary nitrate showed good agreement with the reference result (slope of 1.0 and Pearson correlation coefficient-R_p of 1.0)."

Line 273, "Secondary nitrate, SOA_I and SOA_II showed large variations, with an average relative difference of 173%, 162%, and 75%, respectively.", it is recommended to provide a more detailed explanation of the reasons for the significant differences.

Response: We offer the following potential explanations for the significant differences. However, accurately attributing the contribution of each individual secondary source is challenging and requires additional measurement datasets to test and tune the methods.

The underlined text is newly added.

"Secondary nitrate, SOA_I and SOA_II showed large variations, with an average relative difference of 173%, 162%, and 75%, respectively. In the case of secondary nitrate factor, although the apportioned $PM_{2.5}$ contributions from individual source factors were comparable to the reference results, the resolved source profiles exhibited high time-dependent variabilities. We postulate this may be attributed to the sensitivity of nitrate formation to the reduction of NO_x and VOC precursors during the lockdown restriction (Yang et al., 2022). Previous laboratory studies indicated that reducing anthropogenic pollutants such as SO_2 and NO_x can also reduce the biogenic SOA formation via the anthropogenic–biogenic interaction (Zhang et al., 2019; Xu et al., 2015). This, to some extent, explains the high variabilities in source profiles of the two SOA factors. Additionally, the high variabilities may also arise from the uncertainties in the PMF analysis due to the limited data points available from the short-term

time span (Wang et al., 2018). Therefore, in future studies, alternative approaches are needed to independently assess the contribution of secondary sources. Also, we recommend deploying higher time resolution measurement of the organic tracers. This will help ensure accurate source apportionment results for individual secondary sources, especially within the confines of a short-term time span."

References:

Wang, Q., Qiao, L., Zhou, M., Zhu, S., Griffith, S., Li, L., and Yu, J. Z.: Source Apportionment of PM2.5 Using Hourly Measurements of Elemental Tracers and Major Constituents in an Urban Environment: Investigation of Time-Resolution Influence, J. Geophys. Res. Atmos., 123, 5284–5300, https://doi.org/10.1029/2017JD027877, 2018.

Xu, L., Guo, H., Boyd, C. M., Klein, M., Bougiatioti, A., Cerully, K. M., Hite, J. R., Isaacman-VanWertz, G., Kreisberg, N. M., Knote, C., Olson, K., Koss, A., Goldstein, A. H., Hering, S. V., De Gouw, J., Baumann, K., Lee, S. H., Nenes, A., Weber, R. J., and Ng, N. L.: Effects of anthropogenic emissions on aerosol formation from isoprene and monoterpenes in the southeastern United States, Proc. Natl. Acad. Sci. U. S. A., 112, 37–42, https://doi.org/10.1073/pnas.1417609112, 2015.

Zhang, Y. Q., Chen, D. H., Ding, X., Li, J., Zhang, T., Wang, J. Q., Cheng, Q., Jiang, H., Song, W., Ou, Y. B., Ye, P. L., Zhang, G., and Wang, X. M.: Impact of anthropogenic emissions on biogenic secondary organic aerosol: Observation in the Pearl River Delta, southern China, Atmos. Chem. Phys., 19, 14403–14415, https://doi.org/10.5194/acp-19-14403-2019, 2019.

Abstract and conclusion of the work can be improved as per the methods applied.

Response: We will revise the abstract and the conclusion to improve their readability and clarity in conveying the main findings in the revised manuscript.