Point-by-point response to review comments

The authors presented very comprehensive results on the sources of brown carbon (BrC) in urban Shanghai in spring and summer, 2020. The mass concentrations of BrC were resolved from a Bayesian inference (BI) approach, and the comprehensive chemical characterization results were subjected to positive matrix factorization (PMF) analysis together with BrC results. The authors showed that among the 15 identified sources, the 7 secondary sources contributed to over 40% of BrC in these two seasons. The authors further analyzed multiple cases with incremental secondary BrC growth, and showed that there were different pathways leading to secondary BrC formation at this urban site. The study was well designed and conducted, and the manuscript clearly written. I recommend Minor Revision, with comments as below.

Response: We thank the reviewer for appreciating the importance of our work. We also greatly appreciate the insightful comments, which have helped us improve the manuscript. Each of the questions raised has been addressed in detail in our point-by-point response below. Our response text is marked in blue in this document and references cited are placed at the end. Corresponding changes in main text are also highlighted in blue.

- *PMF results: 1) the individual species measured by TAG-GC-MS contain useful information on the dynamics of their emission and formation. But it seems that some tracers are grouped together to run PMF, e.g., as aromatic SOA tracers, isoprene SOA tracers etc.). Please justify on why they are not used as individual species in PMF runs. 2) The first resolved factor of secondary nitrate/sulfate formation processes contains quite a bit of metal signals. Why are they associated with secondary formation?*

  Response: 1) individual organic tracer from the same category showed good correlations with each other, suggesting common sources. Therefore, they were grouped together to reduce the number of input species and avoid collinearity issues. Following text has been added in the revised ms to improve the clarity.

  Lines 199-203: "A complete list of input species is provided in Table S1, and the naming and grouping of the organic tracers are shown in Table S2. As organic tracers from the same category showed good correlations with each other, they were grouped together to reduce the number of input variables and avoid collinearity issues (Wang et al., 2017)."

  Reference:

  Wang, Q., He, X., Huang, X. H. H., Gri, S. M., Feng, Y., Zhang, T., Zhang, Q., Wu, D., and Yu, J. Z.: Impact of Secondary Organic Aerosol Tracers on Tracer-Based Source Apportionment of Organic Carbon and PM 2.5: A Case Study in the Pearl River Delta, China, ACS Earth Sp. Chem., 1, 562−571, https://doi.org/10.1021/acsearthspacechem.7b00088, 2017.

  2) Certain amount of elements such as Se and Pb presented in the secondary nitrate/sulfate formation processes factor. The two elements were primarily emitted from coal-fired industrial sources, which also release SO_2 and NO_x, key precursors of sulfate and nitrate formation. Their presence in this factor may suggest the importance

of industrial sources in sulfate and nitrate aerosol formation. Following text has been added in the revised ms to improve the clarity.

Lines 288-291: "This factor was enriched in Se and Pb, elements primarily emitted from coal-fired industrial sources that also release SO_2 and NO_x, key precursors of sulfate and nitrate formation. Their presence in this factor may suggest the importance of industrial sources in sulfate and nitrate aerosol formation."

- *The discussion on SOA contribution to secondary BrC (e.g., p10) should be backed by some evidence suggesting that these SOA types are really light-absorbing. For example, pinenes such as limonene might have been shown to generate SOA that can absorb near UV light, but does isoprene SOA really can contribute light absorption significantly? Or is it related to heavy nitration during its formation?*

  Response: suggestion taken, we've added the supportive evidence of the SOA factors which can absorb UV light from laboratory studies in the revised ms. For isoprene SOA, previous study has demonstrated the formation of light-absorbing oligomers during reactive uptake of isoprene epoxydiol onto acidified sulfate aerosols. On the other hand, isoprene SOA is a minor contributor to secondary BrC under the urban atmospheric conditions (~4% of BrC), except under the episodic period. Following text has been added in the revised ms to improve the clarity.

  Lines 294-301: "Among SOA factors, we resolved aromatic SOA factor, α-pinene SOA, isoprene SOA and β-caryphyllene SOA based on their respective tracers, which trace back to specific volatile organic compound (VOC) precursors. Laboratory studies have demonstrated the oxidation of toluene, naphthalene, terpenes and sesquiterpene in the presence of NO_x or NH_3 can generate light-absorbing SOA (Lambe et al., 2013; Li et al., 2021; Updyke et al., 2012). Formation of light-absorbing oligomers during reactive uptake of isoprene epoxydiol onto acidified sulfate aerosols were also observed in laboratory studies (Lin et al., 2014)."

- *I understand that the main focus of this study is on BrC mass concentration. But the light absorption capability of BrC is really the key to its climate effect. I suggest that some data with quantified absorption capability (e.g., b_abs, MAE, AAE etc.) should be mentioned during the discussion.*

  Response: the reviewer is correct that the light absorption of BrC is of great importance to its climate effect. We've added their values in the Table S1 and the discussion in the revised ms. Considering the scope of this work, we will explore the relationship of BrC light absorption properties with the key chemical components and the sources in an upcoming paper.

  Lines 251-259: "BrC optical properties showed larger sample-by-sample variabilities. Both MAE_{370} and Abs_{370} displayed higher values in spring compared to summer (Table S1). The average values of Abs_{370} and MAE_{370} were 1.21 ± 2.14 Mm^{-1} and 0.72 ± 1.15 m^2/gC in spring and 0.60 ± 1.23 Mm^{-1} and 0.39 ± 0.80 m^2/gC in summer, respectively. On the other hand, the AAE of BrC, reflecting the spectral absorption dependence,

averaged at 2.40 ± 0.32 and 2.56 ± 0.33 in spring and summer, respectively. The obtained AAE values in this study are comparable to previous observations in other locations (Li et al., 2026; Liao et al., 2024). Given the scope of the present work, a detailed exploration of BrC light absorption properties and their relationship with key chemical components and sources will be presented in an upcoming paper."

Minor editorial comments:

- *L158: a "." is missing after the citation that ends the sentence.*

  Response: revised accordingly.

- *In a number of places where alpha-pinene and beta-caryophyllene were used as the first word of the sentence (e.g., L254-255), their first English letter should be capitalized, as "alpha-Pinene" etc.*

  Response: revised accordingly.

- *L326: why high wind speed cases should be considered as local air masses? Should it be transported instead?*

  Response: we thank the reviewer for pointing out this typo mistake, we've corrected it the in revised ms.

  Lines 384-385: "Wind speed lower than 3 m/s were taken to indicate the impact from local air masses, thereby representing local secondary BrC formation."

- *L341: the phrase "which shared NOx" reads odd here. Please rephrase.*

  Response: suggestion taken. We've rewritten the sentence to improve the clarity.

  Lines 398-401: "Notably, secondary nitrate and sulfate formation processes were the main sources of secondary BrC during this event. These processes share NO_x as a common precursor with NACs formation, suggesting a concurrent formation regime."

- *L360: does this "high RH" condition hint on possible aqueous-phase processes?*

  Response: Based on the atmospheric conditions of Type 4 event, it is possible that aqueous-phase reactions may contribute to the SOA formation. For example, previous laboratory studies have shown that the enhanced SOA formation from toluene-NO_2 irradiations under humid conditions is attributed primarily to aqueous reactions of water-soluble products such as glyoxal (Jia and Xu, 2018). We've added the description in the revised ms.

  Lines 424-426: "For example, laboratory studies have shown that the enhanced SOA formation from toluene-NO_2 irradiations under humid conditions was attributed

primarily to aqueous reactions of water-soluble products such as glyoxal (Jia and Xu, 2018)."

Reference:

Jia, L., & Xu, Y. (2018). Different roles of water in secondary organic aerosol formation from toluene and isoprene. Atmospheric Chemistry and Physics, 18(11), 8137-8154.