

Dear reviewer,

Thank you very much for the comments and suggestions, which contribute to improve the quality of our manuscript. We have replied all comments and suggestions in our point-by-point response attached below. In order to highlight the changes what we have done, the color of the revised text will become blue.

This study gives a very detailed measurement report on molecular composition, sources, and pollution evolution mechanisms of organic aerosols (OA) in PM<sub>2.5</sub> during winter in a big city Chengdu, Sichuan Basin, China. The methods and analysis are robust with results clearly presented. As the molecular-level OA research for this region is still very limited, this study is helpful understanding the OA formation mechanism in this region and providing insights for air pollution control measures. I recommend the publication of this study after a minor revision.

Minor comments:

1 Add one sentence in the end of the abstract clarifying the significance or implication of this study.

Response. According to the reviewer's suggestion, the necessary sentence has been added, i.e., *"These results are of great value for understanding the characteristics and formation mechanisms of OA, and its contribution to air pollution in the SCB."* (Line 25-27).

2 The use of GC/MS to quantify fatty acids, phthalate esters, and anhydrosugars etc, is well-established. However, for some more volatile organic compounds (e.g., isoprene SOA tracers), could the authors clarify potential losses of these relatively high volatility compounds during derivatization and describe correction measures?

Response. Thank you very much for this very important comment. Yes, we completely agree with that part of SOA<sub>I</sub> tracers like 3-MeTHF-3,4-diols, C5-alkene triols, and 2-methyltetrols would evaporate during thermal desorption and/or derivatization heating processes. However, the losses caused by this decomposition are difficult to determine (Lopez-Hilfiker et al., 2016). Therefore, as with previous study (Li et al., 2018), we have added necessary explanations in the section S1 of the supplementary materials, i.e., *"In addition, it is worth noting that part of detected SOA<sub>I</sub> tracers like 3-MeTHF-3,4-diols, C5-alkene triols, and 2-methyltetrols would evaporate during thermal desorption and/or derivatization heating processes. Thus these tracers in the current study were possibly somewhat underestimated."* (Line 34-37). This can enable readers to have a more accurate understanding of our study results. Meanwhile, the same measurement method has also been widely applied in the previous studies (Li et al., 2013; Cui et al., 2023; Fu et al., 2011; Fu et al., 2012; Wan et al., 2019). Therefore, we believe that with necessary explanations, this volatilization will not affect the credibility of our results.

## References

- Cui, L. L., Gao, Y., Chen, Y. B., Li, R., Bing, H. J., Wu, Y. H., and Wang, G. H.: Chemical characteristics and source apportionment of biogenic primary and secondary organic aerosols in an alpine ecosystem of Tibetan Plateau, J. Geophys. Res.-Atmos., 128, e2022JD037897, <https://doi.org/10.1029/2022jd037897>, 2023.
- Fu, P. Q., Kawamura, K., and Miura, K.: Molecular characterization of marine organic aerosols collected during a round-the-world cruise, J. Geophys. Res.-Atmos., 116, D13302,

<https://doi.org/10.1029/2011jd015604>, 2011.

Fu, P. Q., Kawamura, K., Chen, J., Li, J., Sun, Y. L., Liu, Y., Tachibana, E., Aggarwal, S. G., Okuzawa, K., Tanimoto, H., Kanaya, Y., and Wang, Z. F.: Diurnal variations of organic molecular tracers and stable carbon isotopic composition in atmospheric aerosols over Mt. Tai in the North China Plain: an influence of biomass burning, *Atmos. Chem. Phys.*, 12, 8359-8375, <https://doi.org/10.5194/acp-12-8359-2012>, 2012.

Li, J. J., Wang, G. H., Cao, J. J., Wang, X. M., and Zhang, R. J.: Observation of biogenic secondary organic aerosols in the atmosphere of a mountain site in central China: temperature and relative humidity effects, *Atmos. Chem. Phys.*, 13, 11535-11549, <https://doi.org/10.5194/acp-13-11535-2013>, 2013.

Li, J. J., Wang, G. H., Wu, C., Cao, C., Ren, Y. Q., Wang, J. Y., Li, J., Cao, J. J., Zeng, L. M., and Zhu, T.: Characterization of isoprene-derived secondary organic aerosols at a rural site in North China Plain with implications for anthropogenic pollution effects, *Sci. Rep.*, 8, 535, <https://doi.org/10.1038/s41598-017-18983-7>, 2018.

Lopez-Hilfiker, F. D., Mohr, C., D'Ambro, E. L., Lutz, A., Riedel, T. P., Gaston, C. J., Iyer, S., Zhang, Z., Gold, A., Surratt, J. D., Lee, B. H., Kurten, T., Hu, W. W., Jimenez, J., Hallquist, M., and Thornton, J. A.: Molecular composition and volatility of organic aerosol in the Southeastern U.S.: implications for IEPDX derived SOA, *Environ. Sci. Technol.*, 50, 2200-2209, <https://doi.org/10.1021/acs.est.5b04769>, 2016.

Wan, X., Kang, S. C., Rupakheti, M., Zhang, Q. G., Tripathi, L., Guo, J. M., Chen, P. F., Rupakheti, D., Panday, A. K., Lawrence, M. G., Kawamura, K., and Cong, Z. Y.: Molecular characterization of organic aerosols in the Kathmandu Valley, Nepal: insights into primary and secondary sources, *Atmos. Chem. Phys.*, 19, 2725-2747, <https://doi.org/10.5194/acp-19-2725-2019>, 2019.

3 For the PMF model, the factor of plastic-related sources is interesting. Could the authors include specific markers, e.g., styrene derivatives or tire-wear tracers, to strengthen the credibility of this factor? Response. Thank you very much for this important suggestion. We fully agree with the reviewer's comment that additional tracers are beneficial in strengthening the credibility of this factor. Unfortunately, this study did not measure these species. However, we believe that the current PMF results are still reliable. This is because: (1) The measurement method and results of phthalate esters in this study are reliable. Meanwhile, a large number of previous studies have indicated that phthalate esters are used as plasticizers in synthetic polymers and a softener in polyvinylchloride, and they have been used as the important tracers for plastic emission over the Arctic Ocean (Fu et al., 2013), in Mt. Tai (Fu et al., 2012), in Kathmandu Valley (Wan et al., 2019), and in fourteen Chinese cities (Wang et al., 2006). Correspondingly, in our PMF analysis results, these species appeared extensively in the source profiles of plastic-related sources. (2) Similar to our study, in some previous studies, such as Zhu et al. (2022), Liu et al. (2024), Kang et al. (2018), and Gadi et al. (2019), they also identified plastic-related sources mainly based on phthalate esters in PMF without measuring other tracers. Thanks again for this important comment, which have important guiding value for our future study.

#### References

Fu, P. Q., Kawamura, K., Chen, J., Charrière, B., and Sempéré, R.: Organic molecular composition of marine aerosols over the Arctic Ocean in summer: contributions of primary emission and secondary aerosol formation, *Biogeosciences*, 10, 653-667, <https://doi.org/10.5194/bg-10-653-2013>, 2013.

Fu, P. Q., Kawamura, K., Chen, J., Li, J., Sun, Y. L., Liu, Y., Tachibana, E., Aggarwal, S. G., Okuzawa, K., Tanimoto, H., Kanaya, Y., and Wang, Z. F.: Diurnal variations of organic molecular tracers and stable carbon isotopic composition in atmospheric aerosols over Mt. Tai in the North China Plain: an influence

of biomass burning, *Atmos. Chem. Phys.*, 12, 8359-8375, <https://doi.org/10.5194/acp-12-8359-2012>, 2012.

Gadi, R., Shivani, Sharma, S. K., and Mandal, T. K.: Source apportionment and health risk assessment of organic constituents in fine ambient aerosols (PM<sub>2.5</sub>): a complete year study over National Capital Region of India, *Chemosphere*, 221, 583-596, <https://doi.org/10.1016/j.chemosphere.2019.01.067>, 2019.

Kang, M. J., Fu, P. Q., Kawamura, K., Yang, F., Zhang, H. L., Zang, Z. C., Ren, H., Ren, L. J., Zhao, Y., Sun, Y. L., and Wang, Z. F.: Characterization of biogenic primary and secondary organic aerosols in the marine atmosphere over the East China Sea, *Atmos. Chem. Phys.*, 18, 13947-13967, <https://doi.org/10.5194/acp-18-13947-2018>, 2018.

Liu, Y. L., Shen, M. X., Liu, H. J., Dai, W. T., Qi, W. N., Zhang, Y. F., Li, L., Cao, Y., Wang, X., Guo, X., Jiang, Y. K., and Li, J. J.: Molecular compositions and sources of organic aerosols at a rural site on the Guanzhong Plain, Northwest China: the importance of biomass burning, *Particuology*, 89, 44-56, <https://doi.org/10.1016/j.partic.2023.10.014>, 2024.

Wan, X., Kang, S. C., Rupakheti, M., Zhang, Q. G., Tripathi, L., Guo, J. M., Chen, P. F., Rupakheti, D., Panday, A. K., Lawrence, M. G., Kawamura, K., and Cong, Z. Y.: Molecular characterization of organic aerosols in the Kathmandu Valley, Nepal: insights into primary and secondary sources, *Atmos. Chem. Phys.*, 19, 2725-2747, <https://doi.org/10.5194/acp-19-2725-2019>, 2019.

Wang, G. H., Kawamura, K., Lee, S., Ho, K. F., and Cao, J. J.: Molecular, seasonal, and spatial distributions of organic aerosols from fourteen Chinese cities, *Environ. Sci. Technol.*, 40, 4619-4625, <https://doi.org/10.1021/es060291x>, 2006.

Zhu, Y. H., Tilgner, A., Hans Hoffmann, E., Herrmann, H., Kawamura, K., Xue, L. K., Yang, L. X., and Wang, W. X.: Molecular distributions of dicarboxylic acids, oxocarboxylic acids, and  $\alpha$ -dicarbonyls in aerosols over Tuoji Island in the Bohai Sea: effects of East Asian continental outflow, *Atmos. Res.*, 272, 106154, <https://doi.org/10.1016/j.atmosres.2022.106154>, 2022.

4 Add references for Lines 357-360 about the formation mechanisms of OPAHs and its role in formation of ROS.

Response. Thank you very much for this important reminder, and the necessary references have been added, i.e.,

“Cassee, F. R., Heroux, M. E., Gerlofs-Nijland, M. E., and Kelly, F. J.: *Particulate matter beyond mass: recent health evidence on the role of fractions, chemical constituents and sources of emission*, *Inhal. Toxicol.*, 25, 802-812, <https://doi.org/10.3109/08958378.2013.850127>, 2013.

Chung, M. Y., Lazaro, R. A., Lim, D., Jackson, J., Lyon, J., Rendulic, D., and Hasson, A. S.: *Aerosol-borne quinones and reactive oxygen species generation by particulate matter extracts*, *Environ. Sci. Technol.*, 40, 4880-4886, <https://doi.org/10.1021/es0515957>, 2006.

Shin, S. M., Lee, J. Y., Shin, H. J., and Kim, Y. P.: *Seasonal variation and source apportionment of oxygenated polycyclic aromatic hydrocarbons (OPAHs) and polycyclic aromatic hydrocarbons (PAHs) in PM<sub>2.5</sub> in Seoul, Korea*, *Atmos. Environ.*, 272, 118937, <https://doi.org/10.1016/j.atmosenv.2022.118937>, 2022.”