

Response to CC1:

The authors conduct a study reporting a molecular-level characterization of primary and secondary constituents in PM_{2.5} during haze events, using high-time resolution data obtained from intensive sampling at approximately 2-hour intervals and comprehensive analytical methods. The findings are both informative and useful, as comparative analyses of PM_{2.5} components across different haze events are still lacking. With the growing global attention on the role of biomass burning, such as residential wood combustion, in atmospheric chemistry, and given the rarity of high-resolution molecular-level characterization, I recommend this work for publication in ACP, provided that the following issues are properly addressed.

We appreciate the reviewer's feedback on the manuscript, and we carefully reviewed the comments and addressed each individually below, highlighting changes made in the revised manuscript.

Specific comments:

In lines 26-28: The authors found the contribution of non-fossil fuels increased with increasing haze pollution. However, it was shown that fossil fuel sources made a dominant contribution to WSOC. More explanations should be given here.

The increased non-fossil contribution coincides with a significant intensification of biomass burning (BB) during that time, suggesting that BB could be a key driver of haze formation. This conclusion is also supported by recent research, which found that certain compounds emitted from BB, such as chlorine, can elevate oxidant levels, thereby enhancing secondary aerosol formation (Chang et al., 2024). Based on modeling work, Zhang et al. (2024) reported that the aqueous chemistry of biomass-burning phenols significantly contributes to secondary organic aerosol (SOA) formation, with this contribution increasing with relative humidity. All these findings support our conclusion that biomass burning plays a key role in WSOC formation and even the whole atmospheric chemistry.

In order to make our point clearer, we add some discussions in lines 298-304 as well: "This is further supported by previous reports that emphasized the contribution of aqueous-phase photochemical oxidation of BB organic gases to haze pollution (Zhang et al., 2024; Xiao et al., 2022). This aqueous-phase SOA formation could contribute more than the conventional semi-volatile SOA formation pathways, especially under polluted conditions with high relative humidity (Zhang et al., 2024). Additionally, BB-chlorine emissions could enhance oxidation capacity and further promote secondary aerosol formation (Chang et al., 2024)."

The references used are listed below:

Chang, D., Li, Q., Wang, Z., Dai, J., Fu, X., Guo, J., Zhu, L., Pu, D., Cuevas, C. A., Fernandez, R. P., Wang, W., Ge, M., Fung, J. C. H., Lau, A. K. H., Granier, C., Brasseur, G., Pozzer, A., Saiz-Lopez, A., Song, Y., and Wang, T.: Significant chlorine emissions from biomass burning affect the long-term atmospheric chemistry in Asia, *National Science Review*, nwae285, <https://doi.org/10.1093/nsr/nwae285>, 2024.

Xiao, Y., Hu, M., Li, X., Zong, T., Xu, N., Hu, S., Zeng, L., Chen, S., Song, Y., Guo, S., and Wu, Z.: Aqueous secondary organic aerosol formation attributed to phenols from biomass burning, *Science of The Total Environment*, 847, 157582, <https://doi.org/10.1016/j.scitotenv.2022.157582>, 2022.

Zhang, J., Shrivastava, M., Ma, L., Jiang, W., Anastasio, C., Zhang, Q., and Zelenyuk, A.: Modeling Novel Aqueous Particle and Cloud Chemistry Processes of Biomass Burning Phenols and Their Potential to Form Secondary Organic Aerosols, *Environ. Sci. Technol.*, 58, 3776–3786, <https://doi.org/10.1021/acs.est.3c07762>, 2024.

In lines 136-145: More experimental details need to be here about the quality control.

In the revised manuscript, we have added more description about the quality control. The whole paragraph looks like below:

“Sugar compounds, ... were measured using ion chromatography (Dionex ICS-5000+, ThermoFisher Scientific, USA) after being extracted with ultra-pure water (Milli-Q Reference, America). Standard curves establishment and blank correction were conducted during the analysis. Other organic compounds, including biogenic SOA tracers (isoprene, sesquiterpene, and monoterpene), diacids, and other main organic molecules appeared in the present study were determined by gas chromatography/mass spectrometry (Agilent Technologies; Santa Clara, CA). The average recoveries ranged from 70% to 110% and repeatability experiments showed that the deviation was less than 15%. All the data were corrected with field blanks. More details about measurements can be found in previous studies (Bao et al., 2023)...”

In lines 160-163: The $F^{14}\text{C}$ values were obtained by dividing by the reference isotopic ratio from 1950. Why is further correction needed by dividing by another reference value? This process is a bit confusing.

First, dividing by the reference isotopic ratio in 1950 (i.e., $(^{14}\text{C}/^{12}\text{C})_{1950}$) is to obtain $F^{14}\text{C}$ values. Then, correct these $F^{14}\text{C}$ values for the nuclear bomb effects of the 1950s and 1960s by dividing by the reference value ($f_{\text{nf,ref}}$) to determine the non-fossil carbon fractions (f_{nf}). Since modern carbon can originate from both BB and biogenic (bio) sources, $f_{\text{nf,ref}}$ should be divided into $f_{\text{bb,ref}}$ and $f_{\text{bio,ref}}$. In this study, $f_{\text{bb,ref}}$ (1.11) and $f_{\text{bio,ref}}$ (1.01) were used as the upper and lower limits of $f_{\text{nf,ref}}$, with their median value (1.06) representing the middle value of $f_{\text{nf,ref}}$.

In lines 418-420 and 499-500: The authors suggest that BB could enhance oxidation reactions and secondary formation of certain species. Could you provide additional evidence to support this claim?

In lines 451-452, we have added following evidence: “This is because BB is not only a significant source of air pollutants but also of oxidants (Chang et al., 2024), which enhances oxidation capacity and further promotes photochemistry and SOA formation” to provide a better explanation.”

In lines 533-535, we have included evidence such as: “Chlorine emissions from BB were found to increase oxidant levels, such as O_3 and OH radicals, largely impacting atmospheric chemistry and oxidation process (Chang et al., 2024).”

The reference cited is below:

Chang, D., Li, Q., Wang, Z., Dai, J., Fu, X., Guo, J., Zhu, L., Pu, D., Cuevas, C. A., Fernandez, R. P., Wang, W., Ge, M., Fung, J. C. H., Lau, A. K. H., Granier, C., Brasseur, G., Pozzer, A., Saiz-Lopez, A., Song, Y., and Wang, T.: Significant chlorine emissions from biomass burning affect the long-term atmospheric chemistry in Asia, *National Science Review*, nwae285, <https://doi.org/10.1093/nsr/nwae285>, 2024.