

Dear Professor Irena Grgić,

Appended is our revised manuscript entitled "Technical note: Identification of chemical composition and source of fluorescent components in atmospheric water-soluble brown carbon by excitation-emission matrix with parallel factor analysis: Potential limitation and application". My coauthors and I have made all necessary revisions in the current version of the manuscript according to the comments and suggestions provided by the editor and the two reviewers. We feel that the revised manuscript has much improved quality and more convincing evidence than the prior version.

Thank you and the two reviewers for your comments which we found greatly improve the quality of manuscript. In the reply file, we have given a point-by-point response to all the comments and explained how the comments and suggestions by the editor and reviewer were addressed in the current version of the manuscript.

Please let me know if you have any question about our revised manuscript and thank you again for your assistance.

Looking forward to hearing from you.

Best regards,

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## Response to Additional comments on acp-2022-676

This is a well-prepared manuscript; there is also some discussion, but not much on atmospheric implications of authors' results. However, their findings provide new insights for the analysis of chemical properties and sources of atmospheric fluorophores using the excitation-emission matrix fluorescence spectroscopy.

Besides the comments of the two experts in the field, I have few additional suggestions (lines in MS version2):

Line 49: Some newer references on BrC organic compounds could be added (e.g., Frka et al., *Chemosphere* 2022, 299(11), 134381).

Re: Thanks for your suggestion. We have added some newer references on BrC organic compounds in the present manuscript. Please refer to Lines 49-54.

### References:

- Frka, S., Šala, M., Brodnik, H., Štefane, B., Kroflič, A., and Grgić, I.: Seasonal variability of nitroaromatic compounds in ambient aerosols: Mass size distribution, possible sources and contribution to water-soluble brown carbon light absorption, *Chemosphere*, 299, 134381, <https://doi.org/10.1016/j.chemosphere.2022.134381>, 2022.
- Huang, S., Luo, Y., Wang, X., Zhang, T., Lei, Y., Zeng, Y., Sun, J., Che, H., Xu, H., Cao, J., and Shen, Z.: Optical properties, chemical functional group, and oxidative activity of different polarity levels of water-soluble organic matter in PM<sub>2.5</sub> from biomass and coal combustion in rural areas in Northwest China, *Atmos. Environ.*, 283, 119179, <https://doi.org/10.1016/j.atmosenv.2022.119179>, 2022.
- Ma, L., Li, B., Yabo, S. D., Li, Z., and Qi, H.: Fluorescence fingerprinting characteristics of water-soluble organic carbon from size-resolved particles

during pollution event, *Chemosphere*, 307, 135748, <https://doi.org/10.1016/j.chemosphere.2022.135748>, 2022.

Zhang, T., Shen, Z., Huang, S., Lei, Y., Zeng, Y., Sun, J., Zhang, Q., Ho, S. S. H., Xu, H., and Cao, J.: Optical properties, molecular characterizations, and oxidative potentials of different polarity levels of water-soluble organic matters in winter PM<sub>2.5</sub> in six China's megacities, *Sci. Total Environ.*, 853, 158600, <https://doi.org/10.1016/j.scitotenv.2022.158600>, 2022.

Line 67: For fluorescent components in aerosol particles, reference of Ma et al. (*Chemosphere*, 2022) should be added.

Re: Thanks for your suggestion. This is an important reference for the fluorescent components in ambient particles. We have added it in the present manuscript. Please refer to Line 71.

References:

Ma, L., Li, B., Yabo, S. D., Li, Z., and Qi, H.: Fluorescence fingerprinting characteristics of water-soluble organic carbon from size-resolved particles during pollution event, *Chemosphere*, 307, 135748, <https://doi.org/10.1016/j.chemosphere.2022.135748>, 2022.

Line 127 & lines 210-213: some references on typical atmospheric BrC compounds could be added (see e.g., Frka et al, 2022, *Chemosphere*).

Re: Thanks. According to your comments, we have added some references on typical atmospheric BrC compounds in the present manuscript. Please refer to Lines 134-135 and 223.

References:

Frka, S., Šala, M., Brodnik, H., Štefane, B., Kroflič, A., and Grgić, I.: Seasonal

variability of nitroaromatic compounds in ambient aerosols: Mass size distribution, possible sources and contribution to water-soluble brown carbon light absorption, *Chemosphere*, 299, 134381, <https://doi.org/10.1016/j.chemosphere.2022.134381>, 2022.

Huang, R.-J., Yang, L., Shen, J., Yuan, W., Gong, Y., Ni, H., Duan, J., Yan, J., Huang, H., You, Q., and Li, Y. J.: Chromophoric Fingerprinting of Brown Carbon from Residential Biomass Burning, *Environ. Sci. Tech. Lett.*, 9, <https://doi.org/10.1021/acs.estlett.1c00837>, 2021.

Wang, X., Gu, R., Wang, L., Xu, W., Zhang, Y., Chen, B., Li, W., Xue, L., Chen, J., and Wang, W.: Emissions of fine particulate nitrated phenols from the burning of five common types of biomass, *Environ. Pollut.*, 230, 405-412, <https://doi.org/10.1016/j.envpol.2017.06.072>, 2017.

Line 154: Error. Unit for the resistivity is  $M\Omega$  cm, so for Milli-Q water (ultra-pure) is 18.2  $M\Omega$  cm.

Re: We are sorry for this mistake. In the present manuscript, we have corrected that to 18.2  $M\Omega$  cm. In addition, we also double check the full text to avoid the similar errors. Please refer to Line 164.

Page 14 (end of paragraph): Please, check the references on formation of new secondary organic compounds (second generation) absorbing light at longer wavelengths (for example: Vidović et al., *Environ. Sci. Technol.* 2019, 53, 11195-11203; Vidović et al, *Atmosphere* 2020,11, 131).

Re: Thanks for your suggestion. We have checked the references citation on the formation of new secondary organic compounds absorbing light at longer wavelengths and added some new references in the present manuscript. Please refer to Lines 312-313.

## References:

- Powelson, M. H., Espelien, B. M., Hawkins, L. N., Galloway, M. M., and De Haan, D. O.: Brown carbon formation by aqueous-phase carbonyl compound reactions with amines and ammonium sulfate, *Environ. Sci. Tech.*, 48, 985-993, <https://doi.org/10.1021/es4038325>, 2014.
- Vidović, K., Kroflič, A., Jovanovič, P., Šala, M., and Grgić, I.: Electrochemistry as a Tool for Studies of Complex Reaction Mechanisms: The Case of the Atmospheric Aqueous-Phase Aging of Catechols, *Environ. Sci. Tech.*, 53, 11195-11203, <https://doi.org/10.1021/acs.est.9b02456>, 2019.
- Vidović, K., Kroflič, A., Šala, M., and Grgić, I.: Aqueous-Phase Brown Carbon Formation from Aromatic Precursors under Sunlight Conditions, *Atmosphere*, 11, 131, <https://doi.org/10.3390/atmos11020131>, 2020.
- Vione, D., Albinet, A., Barsotti, F., Mekic, M., Jiang, B., Minero, C., Brigante, M., and Gligorovski, S.: Formation of substances with humic-like fluorescence properties, upon photoinduced oligomerization of typical phenolic compounds emitted by biomass burning, *Atmos. Environ.*, 206, 197-207, <https://doi.org/10.1016/j.atmosenv.2019.03.005>, 2019.
- Yu, L., Smith, J., Laskin, A., George, K. M., Anastasio, C., Laskin, J., Dillner, A. M., and Zhang, Q.: Molecular transformations of phenolic SOA during photochemical aging in the aqueous phase: competition among oligomerization, functionalization, and fragmentation, *Atmos. Chem. Phys.*, 16, 4511-4527, <https://doi.org/10.5194/acp-16-4511-2016>, 2016.