

Reply to Reviewer #1

This manuscript investigated the optical properties, sources and radiative impacts of water-soluble brown carbon in Tianjin, a representative megacity in the North China Plain. Daytime and nighttime samples were collected during winter and summer, and analyzed for aerosol compositions, light absorption spectra and fluorescence properties. Based on the measurement results, connections between the chemical compositions and optical characteristics of aerosols were explored; in addition, radiative impacts of brown carbon were estimated using different approaches. My overall assessment is that this manuscript could be considered for publication as a Measurement Report after a minor revision. My detailed comments are given below.

Reply: We appreciate for the positive comments and helpful suggestions. We have carefully revised our manuscript accordingly. We list our detailed reply in the following sections in blue text.

Line 16. Suggest change “aerosols” to “particles”.

Reply: Changed accordingly.

Lines 19-20. Check this sentence.

Reply: This sentence was modified as follows:

“In winter, BrC showed much stronger light absorbing ability, since mass absorption efficiency at 365 nm (MAE_{365}) in winter ($1.54 \pm 0.33 \text{ m}^2 \text{ g}^{-1}$) was 1.8 times larger than MAE_{365} in summer ($0.84 \pm 0.22 \text{ m}^2 \text{ g}^{-1}$).”

Line 21. I guess something was missing after “ $44.6 \pm 13.9 \%$ ”, e.g., were the two values for different seasons?

Reply: We have modified this sentence as follows:

“Direct radiative effects by BrC absorption relative to black carbon in the UV range were $54.3 \pm 16.9 \%$ and $44.6 \pm 13.9 \%$ in winter and summer, respectively.”

Line 45. It should be “On one hand”.

Reply: Sorry for the typo. It has been corrected.

Line 48. Secondary BrC could also be formed through gas-phase (i.e., photochemical) reactions.

Reply: We agree to the comment and have modified this sentences to “On the other hand, BrC can be formed through gas-phase, aqueous-phase or heterogeneous reactions from both biogenic and anthropogenic precursors.”

Section 2.1 The number of samples should be clarified. This information is important for the reliability of the PMF results.

Reply: Totally, 84 winter samples and 60 summer samples were collected during this campaign. Accordingly, this information has been added in the revised manuscript (Line 97).

Lines 107-108. Details on the EC-tracer method should be provided, e.g., determination of the OC/EC ratio representative of primary emissions.

Reply: The brief introduction of the method has been implemented accordingly as follows:

“Secondary organic carbon (SOC) was estimated with the EC tracer method, which assumed that in all samples the OC/EC ratio for the primary sources affecting the site remains constant (Castro et al., 1999):

$$SOC = OC - [EC \times (OC/EC)_{\min}]$$

where $(OC/EC)_{\min}$ is the minimum value of OC/EC ratios in each season.”

Line 133 and elsewhere in the manuscript. Check the unit of MAE (m^2/g or m^2/gC).

Reply: The unit of MAE should be $m^2 gC^{-1}$. We revised the units throughout the manuscript.

Equation (4). Check whether the WSOC mass has been converted to that of water-soluble organic matter.

Reply: In Eq.4, we used the mass concentration of WSOC rather than water-soluble organic matter according to previous studies (Liu et al., 2013; Shamjad et al., 2018).

References:

Liu, J., et al.: Size-resolved measurements of brown carbon in water and methanol extracts and estimates of their contribution to ambient fine-particle light absorption, *Atmos. Chem. Phys.*, 13, 12389–12404, 2013.

Shamjad, P. M., et al.: Absorbing Refractive Index and Direct Radiative Forcing of Atmospheric Brown Carbon over Gangetic Plain, *ACS Earth Space Chem.*, 2, 31–37, 2018.

Lines 197-199. Was the difference in AAE statistically significant?

Reply: Yes. The difference in AAE was statistically significant ($p < 0.01$) based on pair-sample *t*-test.

Lines 240-243. Were these values calculated by the same methodology?

Reply: Yes. The results were adopted from the limited available references using the same methodology.

Line 315 and elsewhere in the manuscript. Maybe it is better to use “*r*” (in italic) instead of “R”.

Reply: We have used *r* to replace R in the manuscript according to your suggestion.

Lines 403-404. Check the units of levoglucosan.

Reply: Sorry for the typo. We have modified the units to “ $ng m^{-3}$ ”.

Section 3.5. I would like to see the relationship between SOC estimated by the EC-tracer method and that derived from PMF analysis.

Reply: We calculate the contributions of different sources to BrC light absorption (Abs) rather than OC from PMF analysis. Therefore, according to the suggestion, we investigate the

relationship between Abs from SOC (Abs_{SOC}) derived from the PMF results and SOC estimated by the EC tracer method. As shown in the Figure, there is a strongly positive correlation between Abs_{SOC} and SOC with correlation coefficient r of 0.868.

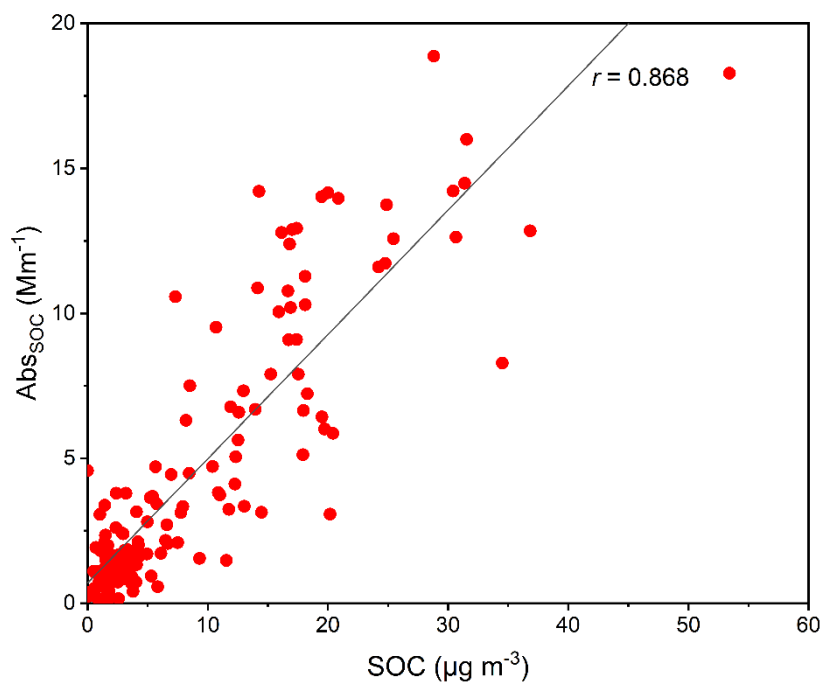


Fig. R1. Correlations between light absorption from SOC (Abs_{SOC}) derived from the PMF results and SOC concentration estimated by the EC tracer method.