Responses to comments by Referee 2

General comments: This study presents the results of brown carbon measurements in cloud, including cloud droplet residuals, cloud interstitial particles, and cloud water. The authors attempted to demonstrate the role of cloud processing in the formation of brown carbon. The dataset covers both the collected cloud water and cloud residuals, and thus may offer new insight into cloud processing of brown carbon, which has been rarely investigated. The topic is appropriate for Atmospheric Chemistry and Physics, but there are some issues that need to be addressed before publication.

Reply: Thanks for the reviewer's positive comments.

Main comments:

Introduction: Generally, what are the major fractions contributing to the light-absorption of cloud water? The authors indicate that nitrophenols and aromatic carbonyls were the major fraction contributing to the light-absorption (~50%) of cloud water at Mt. Tai, but what about in other regions? Also, those related results for aerosol particles should be summarized herein.

Reply: Thanks for the reviewer's helpful comments. Although many light-absorption species such as nitrophenols, aromatic carbonyls, imidazole, and organosulfates have also been detected in cloud/fog water, there is only one research focusing on the optical properties of cloud water (Desyaterik et al., 2013), with the major light-absorption species detected as nitrophenols and aromatic carbonyls.

We agree with the comment, and the major light-absorption species of aerosol particles was also summarized in the revised manuscript: "*Many field studies focused on the optical properties of BrC in particulate matter. The light-absorption of BrC in $PM_{2.5}$ was well correlated with nitrophenols, polycyclic aromatic hydrocarbons, and oxygenated polycyclic aromatic (Wu et al., 2020). Nitrophenols and carbonyl oxygenated polycyclic aromatic hydrocarbons accounting 10-14% to the light-absorption at 365 nm in urban $PM_{2.5}$ (Huang et al., 2020). The contribution of nitrophenols and nitrated salicylic acids*

to the aqueous extract light-absorption of PM_{10} was 0.10-3.71% and 5 times higher than their mass contribution to WSOC (Teich et al., 2017)."

References

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Huang, R. J., Yang, L., Shen, J., Yuan, W., Gong, Y., Guo, J., Cao, W., Duan, J., Ni, H., Zhu, C., Dai, W., Li, Y., Chen, Y., Chen, Q., Wu, Y., Zhang, R., Dusek, U., O'Dowd, C. and Hoffmann, T.: Water-Insoluble Organics Dominate Brown Carbon in Wintertime Urban Aerosol of China: Chemical Characteristics and Optical Properties, Environ. Sci. Technol., 54(13), 7836–7847, doi:10.1021/acs.est.0c01149, 2020.

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Wu, C., Wang, G., Li, J., Li, J., Cao, C., Ge, S., Xie, Y., Chen, J., Li, X., Xue, G., Wang, X., Zhao, Z. and Cao, F.: The characteristics of atmospheric brown carbon in Xi'an, inland China: Sources, size distributions and optical properties, Atmos. Chem. Phys., 20(4), 2017–2030, doi:10.5194/acp-20-2017-2020, 2020.

Section 3.1 Line 172 The discussions related to the influence of aromaticity and molecular weight of WSOC in the light-absorption capacity should be improved. What is the real meaning for a medium negative correlation ($r > 0.43$, $p < 0.05$) with E250/E365? Is such evidence consistent with those obtained by the EEMs measurements in section 3.2?

Reply: Thanks for the comments. The E_{250}/E_{365} could be used as a qualitative measure of aromaticity and molecule weight, in which a lower E_{250}/E_{365} ratio means higher aromaticity and larger molecule weight (Peuravuori and Pihlaja, 1997; Kristensen et al., 2015). The medium negative correlation between MAE_{365} and E_{250}/E_{365} indicates the limited influence of aromatic and molecule weight on the MAE_{365}. For more accurate expression, this sentence has been rewritten as "*Both the MAE_{365} of WSOC in cloud water and $PM_{2.5}$ show a positive correlation ($r > 0.84$, $p < 0.01$) with $SUVA_{254/280}$, and a medium negative correlation ($r > 0.43$, $p < 0.05$) with E_{250}/E_{365}, which may indicate that higher MAE_{365} of WSOC has higher aromatic and molecule weight, the aromaticity and molecular weight of WSOC may influence the light-absorption capacity of cloud water and $PM_{2.5}$*".

No meaningful results could be obtained through correlation analysis between the E_{250}/E_{365} and fluorescent components (F_{max}). The E_{250}/E_{365} ratio was used to analyze the possible influencing factors of the light-absorption of WSOC, where EEMs was used to investigate the possible chemical components of WSOC, the purposes and results of the two analyses are different.

References

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Peuravuori, J. and Pihlaja, K.: Molecular size distribution and spectroscopic properties of aquatic humic substances, Anal. Chim. Acta, 337(2), 133–149, doi:10.1016/S0003-2670(96)00412-6, 1997.

Section 3.2: The authors came to the conclusion that NOx may enhance the formation of nitrogen-containing organics, based on the correlation analysis. I suggest including a discussion on the detailed mechanisms related to such a conclusion. Also, is there any evidence to exclude other pathways as indicated in the introduction?

Reply: Thanks for the reviewer's suggestion. As discussed in section 3.3, the humic-like substances may be formed through Maillard reaction involving carbonyls and ammonium/amines, however, we cannot exclude the other pathways such as photochemical oxidation mentioned in the introduction. The specific mechanism for the conclusion that NOx may enhance the formation of nitrogen-containing organics can be obtained from previous research, which was summarized in the revised manuscript: "*NO_2^- resulted from the dissolved NOx can react with benzene and finally formed nitrophenol in the presence of UV-A (Harrison et al., 2005; Vione et al., 2004). Various of reactive oxygen/nitrogen species generated from the photolysis of inorganic nitrate in aqueous-phase could also facilitate the photooxidation of organic compounds to form BrC (Seinfeld and Pandis, 2016; Yang et al., 2021)*"

References

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Seinfeld, J. H. and Pandis, S. N.: Atmospheric Chemistry and Physics: From Air Pollution to Climate Change, John Wiley & Sons., 2016.

Vione, D., Maurino, V., Minero, C., Lucchiari, M. and Pelizzetti, E.: Nitration and hydroxylation of benzene in the presence of nitrite/nitrous acid in aqueous solution, Chemosphere, 56(11), 1049–1059, doi:10.1016/j.chemosphere.2004.05.027, 2004.

Yang, J., Au, W. C., Law, H., Lam, C. H. and Nah, T.: Formation and evolution of brown carbon during aqueous-phase nitrate-mediated photooxidation of guaiacol and 5-nitroguaiacol, Atmos. Environ., doi:10.1016/j.atmosenv.2021.118401, 2021.

Section 3.2: PMF model indicates a possible influence of biomass burning on the formation of secondary brown carbon. It would be much better to include and compare with those found for aerosol particles. The paper needs to provide more discussion on this issue.

Reply: Thanks for the reviewer's comments. We agree with the reviewer's comments such an additional analysis would benefit the discussions. We tried to investigate the source apportionment of all particulate phase BrC (FREE-$PM_{2.5}$ and INT-$PM_{2.5}$, n=33) through PMF model with input parameters the same as the cloud water. Two to five factors were evaluated and the results are summarized in Table.1. No meaningful results could be obtained for such a limited sample number, and thus such results were not included in the discussion.

Table 1 Q values for PMF analysis with different number of factors.

Num. of factors	$R^{2\#}$ for all input species	R^2 for WSOC	R^2 for Abs_{365}	Q_{robust}*	Q_{robust}/Q_{theory}	Bootstrap (100 runs)
2	0.03-0.93	0.55	0.46	2674.1	0.73	>77
3	0.02-0.99	0.59	0.38	1770.8	0.72	>68
4	0.29-0.99	0.66	0.33	1156.9	0.91	>93
5	0.26-0.99	0.67	0.90	822.6	0.88	>47

#R^2 between the observed and predicted species