Authors' response to reviewer' comments

We would like to thank the reviewer for the thoughtful comments and suggestions to improve the manuscript. We address each comment individually below, with the reviewer's comment **in black** and our responses **in blue** and the revised text **in green**.

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The manuscript by Qin et al. discusses the possible source of brown carbon at Guanzhou, China. The major finding includes 1) biomass burning is the most important source of brown carbon in the region, and 2) importance of nitrogen containing compounds on brown carbon. The topic will attract the interest of the readers of the journal. The manuscript is clearly written, and easy to understand. I suggest publication of the manuscript after addressing the following comments.
We thank the reviewer for this overall very positive assessment of our manuscript.

1. P3L6 'BC is major contributor to light absorption that increases the atmospheric energy budget,' I am not sure what 'increases' means in this context. Is it possible to make the description to be more specific?

Reply:
We thank the reviewer for pointing out the ambiguous sentence. We were trying to say a positive radiative forcing. We clarified the sentence in the revised text.

Revised text:
BC is major contributor to light absorption that leads to positive radiative forcing, increasing the average temperature of the atmosphere.

2. P5 'Measurements and data analysis': I wonder how the relative humidity of the instruments was controlled, especially for the CRD and nephelometer. As water contents influence both extinction and scattering, it is ideal to have clear descriptions on it. In addition, it is ideal to have comprehensive descriptions on how the instruments were calibrated.

Reply:
The reviewer raised an important point. For controlling relative humidity (RH), a diffusion drier was used to dry the sampled air stream, which reduced the RH of the air to below 30 %. The nephelometer was calibrated by CO_2 weekly during the field campaign. Particle-free air was checked once a day. The CRD was calibrated using polystyrene spheres with known indices of refraction before the campaign. We have added the above sentences in the Measurements to make this point clear.

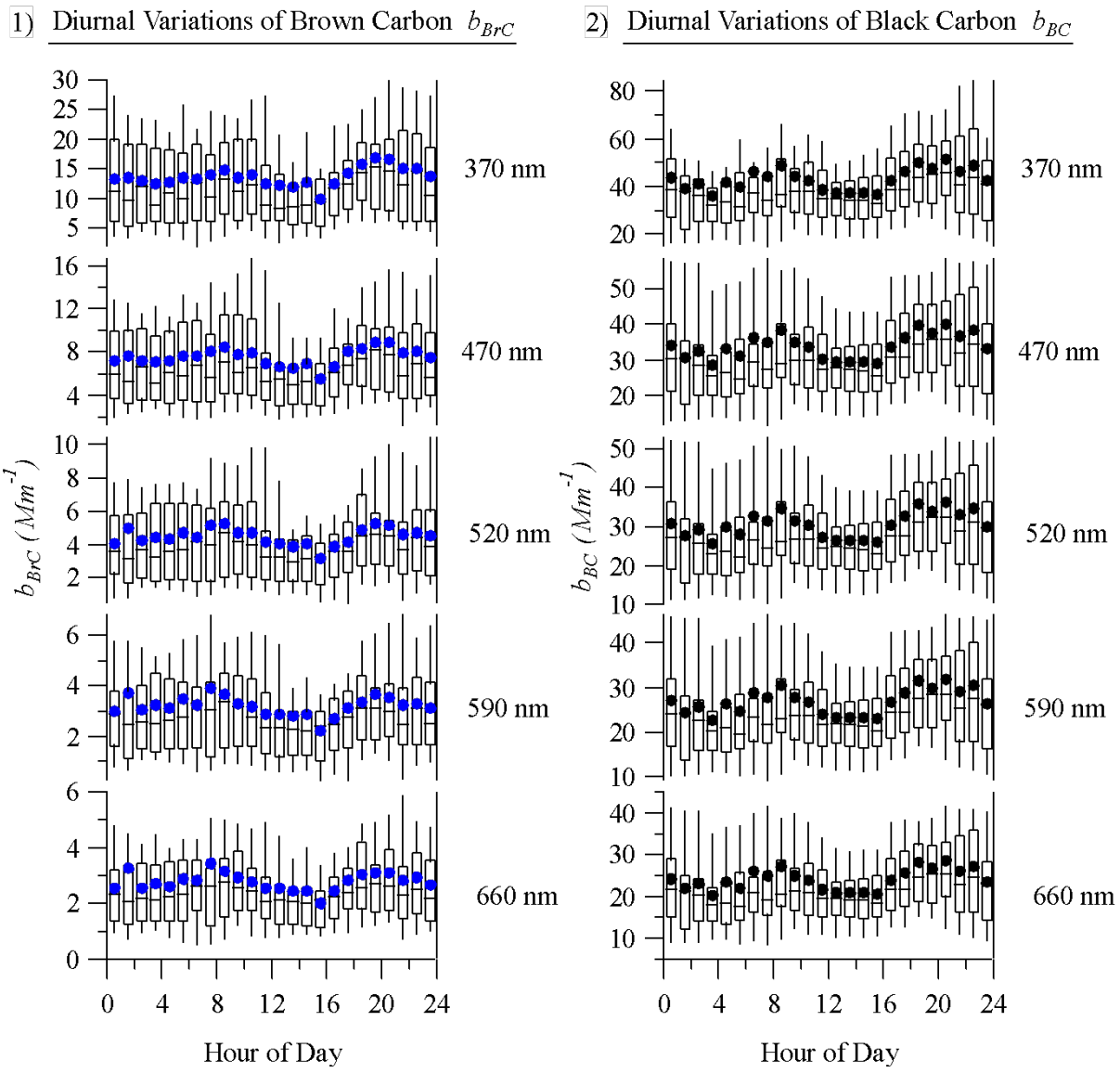
3. P7L126 'As shown in Figure 2b, the AAE values, which average at 1.43, are almost always higher than 1,' The histogram has a variation. It would be interesting to discuss how the temporal variation of AAE was controlled.

Reply:
Thanks for the comment. Total absorption Ångström exponent (AAE) values were calculated by a power-law fitting of the absorption coefficient over all available wavelengths at each time point. Because a plot is generated from the power law fitting at each time point and we have many timepoints, we did not present all the plots in the manuscript. An example of the power law fitting is added in Figure 7 in conjunction with reviewer #2's comment. In terms of the temporal variation, there may be differences in the sources and the relative contribution of each source. We discussed that in section 2 and section 3.

4. P8L154 'The diurnal variations of the different wavelengths were not significantly different, although short wavelengths exhibited more obvious diurnal variations.' There are some interesting patterns in the diurnal variation. For example, the peak at 1AM is clearer for longer wavelength. The morning peak occurred before 8AM for longer wavelength, while it happens after 8AM for shorter wavelength. It would be ideal to have further detailed descriptions on the pattern of the diurnal variation. P9L186 'our results suggest that the absorption coefficient of nascent BBOA' Would you be able to show evidence to consider it as nascent?

Reply:
We agree with the reviewer that the morning peak occurred before 8AM for longer wavelength, while it happened after 8AM for shorter wavelength. However, the peak at 1AM for the long wavelength may be due to some episodic events as the median data is relatively flat. A previous study showed that these changes may be attributed to diurnal changes in BrC sources, which most likely originated from crop residue burning in fall and winter in nearby regions (Wang et al., 2017). We added the discussion as follow in together with Reviewer #2's comment.

Revised text:
Figure 4 shows the diurnal variations of both b_{BrC} and b_{BC} at 370, 470, 520, 590, and 660 m respectively. In general, the diurnal cycles of b_{BrC} and b_{BC} share similar patterns, indicating that they may have similar sources. However, it should be noted that some OA factors, such as BBOA and HOA, also share similar pattern(Qin et al., 2017) .Overall, there were two peaks at each wavelength. The first peak appeared in the morning at around 8:00 LT, with a peak before 8:00 LT for longer wavelength and after 8:00 LT for shorter wavelength. The second peak appeared at 21:00 LT and its intensity decreased untill 24:00 LT. These changes may be attributed to diurnal changes in sources, which most likely originated from crop residue burning in fall and winter in nearby regions(Wang et al., 2017).



5. P9L202 'light absorption depends on the extent of sp 2 hybridization, in which electrons are usually found. I wonder what the 'extent of sp2 hybridization' means. Is it the number of sp2 bonding, or length of sp2 conjugated system?

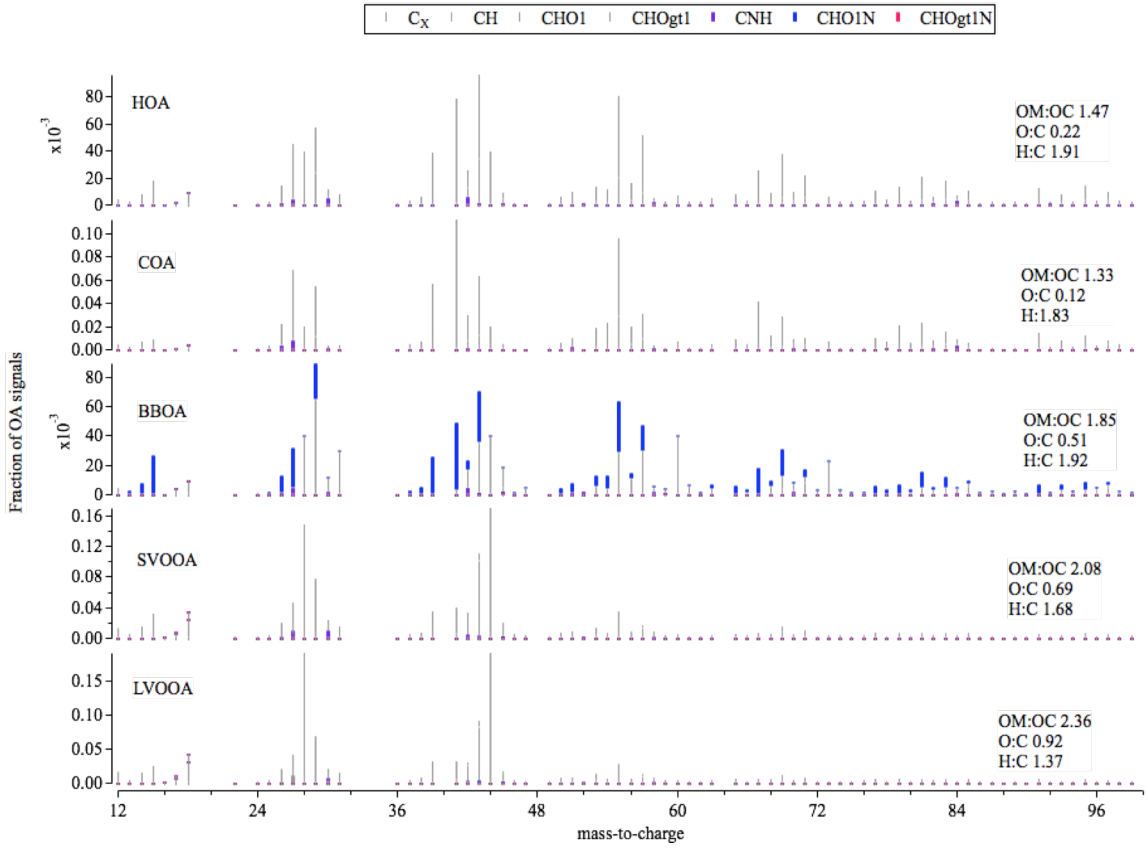
Reply:

Thanks for the comments. We have classified this point in the revised text. By 'extent of sp2 hybridization' we mean the length of the conjugated system. As the conjugation gets larger, the energy difference between the excited state and the ground state goes down, which makes the absorption band shift to longer wavelengths.

6. P9L209 ‘CxHyN + and CxH yOzN ++’Do they predominantly exist in BBOA, or in other types of OA?

Reply:

The reviewer raised an important point. As shown in the following figure, the N-containing ion fragments are distributed in all the OA factors, although the relative contribution is higher in BBOA than that in other OA factors. However, as the signal intensities are already normalized in the PMF analysis, the distribution of these fragments among the OA factors also depend on the mass concentration of each OA factor. We have added this discussion in the supporting information.



7. P13L339 ‘Laskin, J., Laskin, A., Nizkorodov, S. A., Roach, P., Eckert, P., Gilles, M. K., Wang, B., Ji, H., Lee, J. and Hu, Q.: Molecular Selectivity of Brown Carbon Chromophores, 2014.’Journal name is missing.

Response:

Added as suggested.

Reference:

Qin, Y. M., Tan, H. B., Li, Y. J., Schurman, M. I., Li, F., Canonaco, F., Prévôt, A. S. H. and Chan, C. K.: Impacts of traffic emissions on atmospheric particulate nitrate and organics at a downwind site on the periphery of Guangzhou, China, Atmos. Chem. Phys., 2017(x), 1–31, doi:10.5194/acp-2017-116, 2017.

Wang, Y., Hu, M., Lin, P., Guo, Q., Wu, Z., Li, M., Zeng, L., Song, Y., Zeng, L., Wu, Y., Guo, S., Huang, X. and He, L.: Molecular Characterization of Nitrogen-Containing Organic Compounds in Humic-like Substances Emitted from Straw Residue Burning, Environ. Sci. Technol., 51(11), 5951–5961, doi:10.1021/acs.est.7b00248, 2017.