Response to Reviewer #2

The manuscript by Cao et al. titled, "Chemical composition, optical properties and oxidative potential of water- and methanol-soluble organic compounds emitted from the combustion of biomass materials and coal" describes the characterization of various physicochemical properties of aerosols emitted from biomass and coal combustion. While the work here is important, as it provides necessary data that is currently missing in the literature, from BrC light absorptivity and oxidative potential values with corresponding chemical composition information, there is critical information missing that hinders the ability to review this manuscript at this time. As such, major revisions to the manuscript need to be done before it can be further reviewed for publication. Also, a more in-depth discussion of the author's observations is warranted to clearly highlight the novelty of this work.

Re: Thanks for the constructive and valuable comments, which is of great help to improve the quality of the manuscript. According to your comments, we have carefully and thoughtfully revised the manuscript, and responded to all comments point by point, and explained how the reviewers' comments and suggestions are handled in the current manuscript. The main revisions include:

(1) In the introduction, we start with the general importance of BrC and then described recent studies on BrC from biomass burning and coal combustion. As reviewed by previous studies, most of these studies only focused on the chemical compositions and optical properties of water-soluble BrC (i.e, HULIS) emitted from combustion process (Huo et al., 2018; Park et al., 2016; Fan et al., 2016). It is noted that water-insoluble BrC have been demonstrated exhibit a higher light absorption than water-soluble BrC in atmospheric aerosols in many studies (Chen et al., 2016, 2017, Bai et al., 2020, Huang et al., 2020, Li et al., 2018). But, knowledge on the chemical and optical properties of water-insoluble BrC from combustion sources are still lacking. Therefore, to gain more detailed information on BrC from combustion sources, a comprehensive characterization, including chemical, optical characteristics of the BrC fractions (include both water-soluble and water-insoluble BrC)

from the combustion of biomass materials and coals, is required. We have revised that in the manuscript. Please refer to Lines 80-92.

(2) We added some sentences to describe the recent studies on the oxidative potential of BrC from combustion sources and discuss the contributions and limitations of the previous research. It is obvious that knowledge on the OP of water-insoluble BB BrC and BrC fraction emitted from other combustion processes such as coal combustion are still lacking. In addition, the key components or functional groups that responsible for the ROS generation capacity of BrC are also unclear. Please refer to Lines 98-115.

(3) We also carefully revised the section of results and discussion to deeply analyze the data and highlight the novelty of this work (Please refer to Lines 286-292; 314-317; 386-396; 464-470; 560-575; 579-594 and section 3.6). Moreover, we have added a section to discuss the relationship between the DTT activities and their chemical and optical properties (e.g., fluorophores, proton functional groups) of BrC from combustion sources (see section 3.6). The results indicated that humic-like fluorophore (C4) mainly comprised with chemical species with a conjugated system and highly oxygenated species (e.g., quinones or aromatic acids) and may be the key components for the enhancement of the ability of BrC to produce ROS species. The detailed revision please refer to Lines 617-663 (Section 3.6).

References:

Bai, Z., Zhang, L., Cheng, Y., Zhang, W., Mao, J., Chen, H., Li, L., Wang, L., and Chen, J.: Water/Methanol-Insoluble Brown Carbon Can Dominate Aerosol-Enhanced Light Absorption in Port Cities, Environmental science & technology, 54, 14889-14898, 10.1021/acs.est.0c03844, 2020.

Chen, Q., Miyazaki, Y., Kawamura, K., Matsumoto, K., Coburn, S., Volkamer, R., Iwamoto, Y., Kagami, S., Deng, Y., Ogawa, S., Ramasamy, S., Kato, S., Ida, A., Kajii, Y., and Mochida, M.: Characterization of Chromophoric Water-Soluble Organic Matter in Urban, Forest, and Marine Aerosols by HR-ToF-AMS Analysis and

Excitation-Emission Matrix Spectroscopy, Environmental science & technology, 50, 10351-10360, 10.1021/acs.est.6b01643, 2016.

Chen, Q., Ikemori, F., Nakamura, Y., Vodicka, P., Kawamura, K., and Mochida, M.: Structural and Light-Absorption Characteristics of Complex Water-Insoluble Organic Mixtures in Urban Submicrometer Aerosols, Environmental science & technology, 51, 8293-8303, 10.1021/acs.est.7b01630, 2017.

Fan, X. J., Wei, S. Y., Zhu, M. B., Song, J. Z., and Peng, P. A.: Comprehensive characterization of humic-like substances in smoke PM2.5 emitted from the combustion of biomass materials and fossil fuels, Atmospheric Chemistry and Physics, 16, 13321-13340, 10.5194/acp-16-13321-2016, 2016.

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, Environmental science & technology, 54, 7836-7847, 10.1021/acs.est.0c01149, 2020.

Huo, Y. Q., Li, M., Jiang, M. H., and Qi, W. M.: Light absorption properties of HULIS in primary particulate matter produced by crop straw combustion under different moisture contents and stacking modes, Atmospheric Environment, 191, 490-499, 10.1016/j.atmosenv.2018.08.038, 2018.

Li, M., Fan, X., Zhu, M., Zou, C., Song, J., Wei, S., Jia, W., and Peng, P.: Abundances and light absorption properties of brown carbon emitted from residential coal combustion in China, Environmental science & technology, 10.1021/acs.est.8b05630, 2018.

Park, S. S., and Yu, J.: Chemical and light absorption properties of humic-like substances from biomass burning emissions under controlled combustion experiments, Atmospheric Environment, 136, 114-122, 10.1016/j.atmosenv.2016.04.022, 2016.

Specific Comment:

One of the major conclusions are: the DTT assay work is that the ROS activity is "weaker" compared to previous studies, likely due differences in chemical composition. This point should be further supported by comparing the chemical composition observation from this current study and other cited studies of ambient aerosol. This adds to the novelty of this study.

Re: Thanks for this comment. In this study, the DTT_m value of WSOC ranged from 0.5 pmol/min/μg (B-3) to 7.4 pmol/min/μg (CS), with a mean of 3.8 pmol/min/μg. These DTT_m values were lower than those in ambient aerosols in USA and China (Verma et al., 2012; Chen et al., 2019; Yu et al., 2019). These results suggested that the primary water-soluble organic fraction from BB and CC had a weaker ROS generation capacity than ambient aerosols, which was likely due to the differences in the chemical composition of WSOC in BB and CC smoke particles and ambient aerosols (Lin and Yu, 2011; Dou et al., 2015; Wong et al., 2019; Lin and Yu, 2019).

In the revised manuscript, we have conducted PCA and Pearson correlation coefficient analysis to explore how the chemical characteristic to explain the differences of DTT activities of brown carbon emitted from combustion sources (see section 3.6). The results indicated that the humic-like fluorophore (C4) component may mainly comprised with chemical species with a conjugated system and highly oxygenated species, such as quinones or aromatic acids and may be the key components for the enhancement of the ability of BrC to produce ROS species. These results also explained that the water-soluble BrC fractions in BB and CC smoke showed relatively lower DTT_m values than those in ambient aerosols, in which distinctly higher contents of fluorophore C4 were observed in the water-soluble fraction (Matos et al., 2015; Chen et al., 2016). The detailed revisions please refer to Lines 617-663.

References:

Chen, Q., Miyazaki, Y., Kawamura, K., Matsumoto, K., Coburn, S., Volkamer, R., Iwamoto, Y., Kagami, S., Deng, Y., Ogawa, S., Ramasamy, S., Kato, S., Ida, A., Kajii, Y., and Mochida, M.: Characterization of Chromophoric Water-Soluble Organic Matter in Urban, Forest, and Marine Aerosols by HR-ToF-AMS Analysis and Excitation-Emission Matrix Spectroscopy, Environmental science & technology, 50, 10351-10360, 10.1021/acs.est.6b01643, 2016.

Chen, Q., Wang, M., Wang, Y., Zhang, L., Li, Y., and Han, Y.: Oxidative Potential of Water-Soluble Matter Associated with Chromophoric Substances in PM2.5 over Xi'an, China, Environmental science & technology, 53, 8574-8584, 10.1021/acs.est.9b01976, 2019.

Dou, J., Lin, P., Kuang, B. Y., and Yu, J. Z.: Reactive Oxygen Species Production Mediated by Humic-like Substances in Atmospheric Aerosols: Enhancement Effects by Pyridine, Imidazole, and Their Derivatives, Environmental science & technology, 49, 6457-6465, 10.1021/es5059378, 2015.

Lin, M., and Yu, J. Z.: Dithiothreitol (DTT) concentration effect and its implications on the applicability of DTT assay to evaluate the oxidative potential of atmospheric aerosol samples, Environmental pollution, 251, 938-944, 10.1016/j.envpol.2019.05.074, 2019.

Lin, P., and Yu, J. Z.: Generation of reactive oxygen species mediated by humic-like substances in atmospheric aerosols, Environmental science & technology, 45, 10362-10368, 10.1021/es2028229, 2011.

Matos, J. T. V., Freire, S. M. S. C., Duarte, R. M. B. O., and Duarte, A. C.: Natural organic matter in urban aerosols: Comparison between water and alkaline soluble components using excitation-emission matrix fluorescence spectroscopy and multiway data analysis, Atmospheric Environment, 102, 1-10, 10.1016/j.atmosenv.2014.11.042, 2015.

Verma, V., Rico-Martinez, R., Kotra, N., King, L., Liu, J., Snell, T. W., and Weber, R. J.: Contribution of water-soluble and insoluble components and their hydrophobic/hydrophilic subfractions to the reactive oxygen species-generating potential of fine ambient aerosols, Environmental science & technology, 46, 11384-11392, 10.1021/es302484r, 2012.

Yu, S., Liu, W., Xu, Y., Yi, K., Zhou, M., Tao, S., and Liu, W.: Characteristics and oxidative potential of atmospheric PM2.5 in Beijing: Source apportionment and seasonal variation, The Science of the total environment, 650, 277-287, 10.1016/j.scitotenv.2018.09.021, 2019.

Detailed information about the methodology re: the TOC content analysis was supposed to be in the supplementary, but in the supplementary file, this "detailed measurement method is provided in the SI file" (line 133-134 of SI). This information needs to be presented, as some of the results presented are mass-normalized. However, it is unclear if "µg" is referring to the mass of carbon, or the mass of the WSOC, or the mass of the PM (e.g., the conversion of the mass of OC to organic PM concentration uses a conversion factor (Turpin and Lim, Aerosol Science and Technology, 2001, 35(1) 602-610). This lack of information makes it difficult to assess if the direct comparison of mass-normalized results as reported in the current manuscript (e.g., Figure 6) is applicable.

Re: Thanks. We are sorry for the mistake that "detailed measurement method is provided in the SI file". We have added the detailed information of TOC measurement method in revised supporting information file. The TOC content of WSOC and HULIS was determined by a high-temperature catalytic oxidation instrument (VCPH analyzer, Shimadzu, Kyoto, Japan) following the non-purgeable OC protocol. After the removal of inorganic carbon, the sample was oxidized at high temperature (680 °C) and the peak area of CO_2 was determined by a non-dispersive infrared detector. Please refer to Lines 136-138 in SI file.

In the original manuscript, the 'µg' is referring to the mass of carbon for the OC, EC, WSOC, HULIS, and MSOC and the mass of PM for smoke $PM_{2.5}$, respectively. We are sorry for this misleading weight unit "µg" used in the original manuscript. <u>In order to avoid these misunderstood.</u> We have revised "µg" to "µgC" when it referring to the mass of carbon for the OC, EC, WSOC, HULIS-C, and MSOC in the present manuscript. Please refer to Lines 142-143 in SI file.

L184: For the ^{1}H NMR work: What does it mean that the BrC fractions (e.g., dissolved in water, recovered from the SPE cartridges with solvent, and methanol extracted) were dissolved in D_2O? Effectively, this was a liquid-liquid extraction? I am not sure if the all the MSOC components dissolved in the D_2O (and which from my understanding is the portion of the sample that leads to the ^{1}H NMR signal), and as such, I am uncertain if the authors can equate the proton-NMR data are measurements of BrC in the MSOC BrC fraction. Why not use deuterated methanol instead?

Re: Thanks for the comments. We have double checked the experimental records and confirmed that we made a mistake. In fact, the water-soluble BrC fractions (WSOC and HULIS) were redissolved by D_2O, but MSOC were redissolved in deuterium methanol. We have revised that in the present manuscript. Please refer to Lines 220-222.

L224: How is HULIS a hydrophobic fraction of *water-soluble* organic carbon? Perhaps you mean it is less polar components of the WSOC?

Re: Thanks. We agreed with your comments that it is less polar components of the WSOC. In this study, the HULIS fraction was isolated with a HLB SPE cartridge. The lower molecular weight and high polar organic species and inorganic salt ions or metal ions was removed by the cartridge, whereas the higher molecular weight and less polar components was retained. Finally, the retained HULIS were eluted with methanol. According to previous studies, these less polar components (i.e., HULIS) of the WSOC were also refer to the relatively hydrophobic fraction of water-soluble organic carbon (Verma et al., 2012, Zheng et al., 2013, Katsumi et al., 2018). Therefore, HULIS was also described as a hydrophobic fraction of water-soluble organic carbon in this study. We have added some descriptions in the present manuscript. Please refer to Lines 120-122 in SI file.

References:

Katsumi, N., Miyake, S., Okochi, H., Minami, Y., Kobayashi, H., Kato, S., Wada, R., Takeuchi, M., Toda, K., and Miura, K.: Humic-like substances global levels and extraction methods in aerosols, Environmental Chemistry Letters, 17, 1023-1029, 10.1007/s10311-018-00820-6, 2018.

Verma, V., Rico-Martinez, R., Kotra, N., King, L., Liu, J., Snell, T. W., and Weber, R. J.: Contribution of water-soluble and insoluble components and their hydrophobic/hydrophilic subfractions to the reactive oxygen species-generating potential of fine ambient aerosols, Environmental science & technology, 46, 11384-11392, 10.1021/es302484r, 2012.

Zheng, G., He, K., Duan, F., Cheng, Y., and Ma, Y.: Measurement of humic-like substances in aerosols: a review, Environmental pollution, 181, 301-314, 10.1016/j.envpol.2013.05.055, 2013.

L125: It is not clear as to how these blank filters can be used to correct the mass of smoke, optical signal, and DTT consumption by BrC. Were these blank filters placed into the filter collection system behind the first filter that contains most of the aerosol (e.g., this is typically done for quartz filter for breakthrough, such as correction for semi-volatile organic carbon).

Re: Thanks for this comment. In this study, the field blank filters were collected followed the method for sampling smoke $PM_{2.5}$ samples, but without ignited fuel samples. The field blank filters were treated as the method for smoke samples. The average values of WSOC, HULIS and MSOC were 1.8±0.2 μgC/cm^2, 0.7±0.1 μgC/cm^2 and 5.3±0.9 μgC/cm^2, respectively. They were much less than the values of that in smoke particle.

In this study, all the results were blank-corrected by subtracting an average field blank value for each sample. The data were present as a mean ± standard deviation based on triplicate analysis of filter sample for each combustion experiment. We have added the detailed information in the supporting information (Section S6). Please refer to Lines 165-180 in SI file.

Minor comments:

L218: It is not clear which parameter was used to infer "average contribution of WSOC to wood smoke PM2.5" from Table 1 (is it WSOC-C/PM)?

Re: Yes, "WSOC-C/PM" was used to infer "average contribution of WSOC to wood smoke PM2.5" in the original manuscript. In order to avoid the misunderstood for the concepts of WSOC and WSOC-C, we have revised "water-soluble organic compounds (WSOC)" to "water-soluble organic carbon (WSOC)", "methanol-soluble organic compounds (MSOC)" to "methanol-soluble organic carbon (MSOC)" in the present manuscript. The weight unit was revised to "μgC" for carbon fractions (i.e., the OC, EC, WSOC, HULIS, and MSOC) and "μg" for $PM_{2.5}$, respectively. In addition, for the HULIS contribution, HULIS/$PM_{2.5}$ was corrected to HULIS-C/$PM_{2.5}$ in the present manuscript. The detailed revisions please refer to Lines 22-24, 68-70.

Figure 5: I recommend the authors abbreviate "typical biomass burning" as something else other than "WS" as it is confusing when "WS" is commonly used to refer to water-soluble.

Re: Thanks. We have used BB and CC to represent typical biomass burning and coal combustion, respectively, in Figure 5. Please refer to the revised Figure 5.

It is also useful to provide the DTT values that are normalized by volume of air, these values would be useful for the calculation of exposure. Can the authors provide these analogous values in the supplementary?

Re: Thanks. We agreed with your comments that the DTT values that are normalized by volume of air (DTT_v) would be useful for the calculation of exposure. However, due to the inherent weakness of the combustion and sampling system in the laboratory, the DTT_v values were not provided in the current manuscript. The main reasons are: (1) The combustion experiment was carried out in a relatively small combustion and sampling system, and the concentration of smoke $PM_{2.5}$ greatly higher than $PM_{2.5}$ aerosols in atmosphere despite it has been diluted. (2) The DTT_v values of smoke samples were mainly controlled by the dilution ratio and sampling time in this study, which is greatly differ from the actual exposure risk in atmosphere. Therefore, the DTT_m values that are normalized by the mass of PM were provided in this study.

The volumes and concentrations of reagents used in the current study are not as described by most of the papers cited by Bates et al., 2019 and Verma et al., 2012. The general approach is certainly identical, but the specific details are not. I highly recommend the authors to specify the differences, as this may be important for comparison to other literature value.

Re: Thanks. The DTT method applied in this study was mainly based on the methods of Fan et al (2018) and Gao et al (2020), and also with minor modifications. As comparison with those introduced by Bates et al. (2019) and Verma et al. (2012), the DTT assay used in this study only enlarge the volume of reagents, and the final concentration of each reagent was similar to the former ones. In this study, positive control of DTT assay was conducted using 1.4-phenanthraquinone as a standard. The resulting DTT consumption rate of control sample was 0.46 ± 0.03 μM DTT/min (n=10), which was comparable to those reported in previous studies (Fan et al., 2018; Lin and Yu, 2019). This suggested that the OP values determined by the DTT assay applied in this study were reliable. Please refer to line 252-253.

References:

Bates, J. T., Fang, T., Verma, V., Zeng, L., Weber, R. J., Tolbert, P. E., Abrams, J. Y., Sarnat, S. E., Klein, M., Mulholland, J. A., and Russell, A. G.: Review of Acellular Assays of Ambient Particulate Matter Oxidative Potential: Methods and Relationships with Composition, Sources, and Health Effects, Environmental science & technology, 53, 4003-4019, 10.1021/acs.est.8b03430, 2019.

Fan, X., Li, M., Cao, T., Cheng, C., Li, F., Xie, Y., Wei, S., Song, J., and Peng, P. a.: Optical properties and oxidative potential of water-and alkaline-soluble brown carbon in smoke particles emitted from laboratory simulated biomass burning, Atmospheric Environment, 194, 48-57,10.1016/j.atmosenv.2018.09.025, 2018.

Gao, D., Mulholland, J. A., Russell, A. G., and Weber, R. J.: Characterization of water-insoluble oxidative potential of PM2.5 using the dithiothreitol assay, Atmospheric Environment, 224, 117327, 10.1016/j.atmosenv.2020.117327, 2020.

Lin, M., and Yu, J. Z.: Dithiothreitol (DTT) concentration effect and its implications on the applicability of DTT assay to evaluate the oxidative potential of atmospheric aerosol samples, Environmental pollution, 251, 938-944, 10.1016/j.envpol.2019.05.074, 2019.

Verma, V., Rico-Martinez, R., Kotra, N., King, L., Liu, J., Snell, T. W., and Weber, R. J.: Contribution of water-soluble and insoluble components and their hydrophobic/hydrophilic subfractions to the reactive oxygen species-generating potential of fine ambient aerosols, Environmental science & technology, 46, 11384-11392, 10.1021/es302484r, 2012.

L56: The Lin and Yu, as well as Ma et al. papers, looked at the generation of ROS by HULIS - these paper did not actually report whether the HULIS is also BrC (e.g., it is still not known if all HULIS can be considered as BrC and vice versa). On a related note, the next sentence is written in a manner that suggests that all HULIS is BrC.

Re: Thanks. In general, HULIS is the strong light-absorbing organic fraction in WSOC, which have been demonstrated to be important component of BrC and were usually used as surrogates for water-soluble BrC in many previous studies (Li et al., 2019; Wu et al., 2018; Han and Kim, 2017). Therefore, the two references were used in this sentence. To avoid the misunderstanding, we have added a new reference. Please refer to Line 60-61

References:

Han, H., and Kim, G.: Significant Seasonal Change in Optical Properties by atmospheric humic-like substances (HULIS) in Water-Soluble Organic Carbon Aerosols, Atmospheric Chemistry and Physics Discussions, 1-19, 10.5194/acp-2017-554, 2017.

Li, X., Han, J., Hopke, P. K., Hu, J., Shu, Q., Chang, Q., and Ying, Q.: Quantifying primary and secondary humic-like substances in urban aerosol based on emission source characterization and a source-oriented air quality model, Atmospheric Chemistry and Physics, 19, 2327-2341, 10.5194/acp-19-2327-2019, 2019.

Wu, G., Wan, X., Gao, S., Fu, P., Yin, Y., Li, G., Zhang, G., Kang, S., Ram, K., and Cong, Z.: Humic-Like Substances (HULIS) in Aerosols of Central Tibetan Plateau (Nam Co, 4730 m asl): Abundance, Light Absorption Properties, and Sources, Environmental science & technology, 52, 7203-7211, 10.1021/acs.est.8b01251, 2018.

Technical Corrections:

The Ma et al. reference has some typos/weird characters.

Re: Thanks. We have revised that in the present manuscript. Please refer to Line 901-904.

Ma, Y., Cheng, Y., Qiu, X., Cao, G., Fang, Y., Wang, J., Zhu, T., Yu, J., and Hu, D.: Sources and oxidative potential of water-soluble humic-like substances ($HULIS_{WS}$) in fine particulate matter ($PM_{2.5}$) in Beijing, Atmospheric Chemistry and Physics, 18, 5607-5617, 10.5194/acp-18-5607-2018, 2018.

L77: I don't understand the following sentence: "However, these studies only focused on the BrC fractions emitted from BB or CC, and therefore the comprehensive characterization and full understanding of the BrC fractions from combustion processes are still required." Is the author referring to the fact that the cited studies only focused on certain fractions of BrC (e.g., water-soluble BrC?).

Re: I am sorry for this confusing sentence. In this manuscript, we want to said that the cited studies only focused on water-soluble BrC from combustion sources. This question was also point out by the other reviewer. Based on your comments, we have made a major revision and the detailed information were provided as follow:

"However, most of these studies only focused on the relative abundances, chemical composition, and optical properties of water-soluble BrC (e.g., HULIS) emitted from the combustion of various fuels and different combustion conditions (e.g., smoldering and flaming) (Huo et al., 2018; Park et al., 2016; Fan et al., 2016). It is noted that water-insoluble BrC even exhibits a higher light absorption than water-soluble BrC in ambient aerosols (Chen et al., 2016, 2017; Bai et al., 2020; Huang et al., 2020; Li et al., 2019). However, knowledge on the chemical and optical properties of water-insoluble BrC from combustion sources is still lacking. Moreover, the association of chemical compositions responsible for light absorption of BrC from combustion sources is still constrained. Therefore, to gain more detailed information on BrC from combustion sources, a comprehensive characterization, including the chemical and optical characteristics of the BrC fractions (including both water-soluble and water-insoluble BrC) from the combustion of biomass materials and coals, is required. We have revised that in the manuscript. Please refer to Lines 80-92.

References:

Bai, Z., Zhang, L., Cheng, Y., Zhang, W., Mao, J., Chen, H., Li, L., Wang, L., and Chen, J.: Water/Methanol-Insoluble Brown Carbon Can Dominate Aerosol-Enhanced Light Absorption in Port Cities, Environmental science & technology, 54, 14889-14898, 10.1021/acs.est.0c03844, 2020.

Fan, X., Li, M., Cao, T., Cheng, C., Li, F., Xie, Y., Wei, S., Song, J., and Peng, P. a.: Optical properties and oxidative potential of water-and alkaline-soluble brown carbon in smoke particles emitted from laboratory simulated biomass burning, Atmospheric Environment, 194, 48-57,10.1016/j.atmosenv.2018.09.025, 2018.

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, Environmental science & technology, 54, 7836-7847, 10.1021/acs.est.0c01149, 2020.

Huo, Y. Q., Li, M., Jiang, M. H., and Qi, W. M.: Light absorption properties of HULIS in primary particulate matter produced by crop straw combustion under different moisture contents and stacking modes, Atmospheric Environment, 191, 490-499, 10.1016/j.atmosenv.2018.08.038, 2018.

Park, S. S., and Yu, J.: Chemical and light absorption properties of humic-like substances from biomass burning emissions under controlled combustion experiments, Atmospheric Environment, 136, 114-122, 10.1016/j.atmosenv.2016.04.022, 2016.

Li, M., Fan, X., Zhu, M., Zou, C., Song, J., Wei, S., Jia, W., and Peng, P.: Abundances and light absorption properties of brown carbon emitted from residential coal combustion in China, Environmental science & technology, 10.1021/acs.est.8b05630, 2018.

L138: the Cheng et al. (2016) reference was included twice.

Re: Revised.

L206: It is not clear what the acronym DTPA stands for, it has not been explained prior.

Re: Thanks. DTPA is the abbreviation of diethylene triamine pentaacetic acid. As an efficient metal ion chelating agent, which can avoid the interferences of metal ions on the oxidation potential of BrC fraction and had been widely used in previous studies (Lin et al., 2011, Ma et al., 2018). We have added the specific name of DTPA in the present manuscript. Please refer to Line 244.

References:

Lin, P., and Yu, J. Z.: Generation of reactive oxygen species mediated by humic-like substances in atmospheric aerosols, Environmental science & technology, 45, 10362-10368, 10.1021/es2028229, 2011.

Ma, Y., Cheng, Y., Qiu, X., Cao, G., Fang, Y., Wang, J., Zhu, T., Yu, J., and Hu, D.: Sources and oxidative potential of water-soluble humic-like substances ($HULIS_{WS}$) in fine particulate matter ($PM_{2.5}$) in Beijing, Atmospheric Chemistry and Physics, 18, 5607-5617, 10.5194/acp-18-5607-2018, 2018.