Dear editor and reviewers,

We appreciate all your detail and valuable suggestions on our manuscript (acp-2020-676). We have carefully considered the comments and revised the manuscript accordingly. Please see the point-by-point response below and changes are marked blue in the revised manuscript.

Thanks for your kind help.

Best regards,

Min Hu

Point-by-point response to review comments

Referee #1

This study investigated the emission factors (EFs) and light absorption of carbonaceous components, including water-soluble organic carbon (WSOC), humic-like substances (HULIS), and water-insoluble organic carbon (WISOC), from burning crop residues (wheat and corn). Also, the influences of biofuel moisture and burning conditions on EFs and brown carbon (BrC) absorption were analyzed by using data of modified combustion efficiency (MCE). Although a clear dependence of EFs on MCE values was illustrated, the influence of burning conditions on biomass burning (BB) BrC absorption can hardly be observed or validated. This might be due to the limit in sample number, smoldering combustion conditions (MCE = 0.68 – 0.88), or uncertainties in the calculation of mass absorption efficiency (MAE_λ). Before the acceptance to publication, the following issues should be addressed.

Response: Thanks for your valuable comments on our manuscript and pointing out the deficiency in discussing the influence of burning conditions on biomass burning BrC absorption. We have now carefully revised the manuscript and addressed the following comments.

1. Page 5, line 137. Why total OC was used to represent the concentration of extracted OC?

In Page 127, it was stated that "total OC was analyzed by a thermal/optical carbon analyzer (Subset Laboratory)".

These two "total OC" should be different. The first is used to calculate EFs of OC, while the second is derived for MAE_λ calculation.

Furthermore, the authors can perform better estimation on extracted OC (or WISOC) mass by measuring residue OC on filter samples after solvent extractions. Then the calculation of MAE_λ of WISOC will be less uncertain.

Typically, the residue OC would account for ~10% of the total, and WSOC contributed more than 50% of total OC in this work. Then the inter-sample variability of residue OC will lead to substantial uncertainty on the estimation of WISOC mass and absorption.

Response: Thanks for pointing out the uncertainties in estimating "extracted OC" concentrations. Yes, we used the total OC analyzed by the thermal/optical carbon analyzer to calculate the EFs and also to represent the "total extracted OC" to derive the MAE_λ. We agree with the referee that this may lead to an underestimation of MAE of WISOC, which we mentioned in lines 143-144.

The OC concentration measured by the thermal/optical carbon analyzer has been widely employed to estimate the absorption capability of OC or WISOC extracted by methanol in previous studies (Liu, 2014; Zhu et al., 2018). Chen and Bond (2010) suggested that more than 92% of OC emitted from biomass pyrolysis could be extracted by methanol. Xie et al. (2019) suggested that 93.6%-99.7% of biomass burning-generated OC could be extracted by methanol (added in lines 144-146). Thus, the residue OC was relatively small compared with the methanol-extracted OC, and the difference between the total WISOC (estimated by the difference between thermal/optical carbon analyzer measured OC and WSOC) and methanol-extracted WISOC were relatively small and not taken into consideration in this study.

We agree with the referee that it is very important to estimate the residue OC and make an accurate calculation of WISOC absorption. However, during the extraction procedures in this study, we first cut up the filter samples and then extracted them by water and then methanol. The thermal/optical carbon analyzer quantified OC on certain size filter by converting the carbon species to methane, and measuring by a flame ionization detector. Thus, it's difficult for us to measure the residue OC due to the organic solvent absorbed on the extracted filters and the fragmentized filters after extraction. We will design experiments, such as those conducted in Chen and Bond (2010) and Xie et al. (2019), to estimate the residue OC concentrations and make a more accurate calculation of the MAE of WISOC in our future studies.

Lines 143-146:

"It is noted that the total OC was used to represent the concentration of total extracted OC, which may lead to an underestimation of MAE of WISOC. Previous studies suggested that 92%-99.7% of BBOA could be extracted by methanol (Chen and Bond, 2010; Xie et al., 2019), thus the residue OC un-extracted by methanol was relatively small compared with the extracted fraction."

References:

Chen, Y. and Bond, T. C.: Light absorption by organic carbon from wood combustion, *Atmos. Chem. Phys.*, 10, 1773-1787, 10.5194/acp-10-1773-2010, 2010.

Liu, J., Scheuer, E., Dibb, J., Ziemba, L. D., Thornhill, K. L., Anderson, B. E., Wisthaler, A., Mikoviny, T., Devi, J. J., Bergin, M., and Weber, R. J.: Brown carbon in the continental troposphere, *Geophys. Res. Lett.*, 41, 2191-2195, 10.1002/2013gl058976, 2014.

Xie, M., Chen, X., Hays, M. D., and Holder, A. L.: Composition and light absorption of N-containing aromatic compounds in organic aerosols from laboratory biomass burning, *Atmos Chem Phys*, 19, 2899-2915, 10.5194/acp-19-2899-2019, 2019.

Zhu, C. S., Cao, J. J., Huang, R. J., et al.: Light absorption properties of brown carbon over the southeastern Tibetan Plateau, *Sci. Total Environ.*, 625, 246-251, 10.1016/j.scitotenv.2017.12.183, 2018.

2. The dependence of EFs on burn conditions was well illustrated in Figures 3 and 4. But Figure 7d-f did not show any influence of burn conditions on light absorption. Figure 7a-c and Figure 3b-d tell the same thing——smoldering combustion has higher EFs of carbonaceous aerosols.

Page 11, lines 296-297, "Furthermore, the MAE365 of WSOC and HULIS emitted from straw burning were slightly higher under less efficient burning conditions (Figures 7d, 7e)"

In previous studies, MAE values tend to be greater under more flaming conditions or higher burning temperatures. The observation results reported here seems not reasonable.

Due to the sample number limit and small variability in MAE365 for most observations, the light *absorption of BB BrC did not show any dependence on burn conditions.*

Response: We deleted the statement "*Furthermore, the MAE_{365} of WSOC and HULIS emitted from straw burning were slightly higher under less efficient burning conditions*." in the revised version and revised the sentence as in lines 320-321. We carefully checked the correlation between MAE_{365} and MCE again, and found their correlations were not significant at the 0.01 level (2-tailed) for either WSOC or HULIS. The slight increasing trends of MAE_{365} for WSOC and HULIS as the decreasing of MCE were really due to the two outliers in wheat burning experiments.

We agree with the referee that limited sample number conducted light absorption measurements and small variability in MAE_{365} for most observations may be the reasons for lack of dependence of MAE on burning conditions. We added this reasons in the revised manuscript (lines 310-312). In this study, we intended to simulate the real combustion conditions of agriculture residue burning in the field. Smoldering-dominated conditions, with expected MCE<0.9 or even lower, have been widely observed during the burns of agricultural residues in the agricultural area in China (Figure R1) and India (Figure R2). Thus, the burning conditions were controlled to be dominated by smoldering (as shown in Figure 1, with MCE=0.68-0.88) in our experiments, and the small variation in burning conditions could be the reasons for small variability in MAE_{365} for most observations. More lab-controlled burning experiments, involving larger numbers of experiments and more variable burning conditions, are required in our future studies to address the influence of combustion conditions on the BrC absorption (lines 327-331).

Figure R1 Intense straw burning in agriculture area (Anhui province, China) in China. (Wang, et al., 2017)

Figure R2 Post-harvest crop residue burning in northwest India. (IARI, 2012)

Lines 320-321:

"We did not observe obvious dependence of MAE_{365} on the combustion efficiency for either water-soluble fractions or WISOC (Figure 7d-f)."

Lines 327-331:

"It is noted that limited sample population was selected to conduct the light absorption measurements and smoldering dominated the burning conditions in this study, which could be the reasons that we did not observe an obvious dependence of MAE on combustion conditions. More lab experiments, involving larger numbers of experiments and more variable burning conditions, are required to address the influence of combustion efficiency on light absorption capability of biomass burning-emitted carbonaceous aerosols in future studies."

References:

Crop Residues Management with Conservation Agriculture: Potential, Constraints and Policy Needs, edited by: Institute, I. A. R., India, 2012.

Wang, Y., Hu, M., Lin, P., et al.: Molecular characterization of nitrogen-containing organic compounds in humic-like substances emitted from straw residue burning, *Environ. Sci. Technol.*, 51, 5951-5961, 10.1021/acs.est.7b00248, 2017.

Page 12, lines 339-342, the final conclusion "Our results suggested that the influence of varied combustion efficiency on the emission levels and light absorption of BBOA could surpass the differences between biofuel types. Thus, the burning efficiency or combustion conditions should be taken into consideration when estimate the influence of biomass burning." was not fully supported by the experiments results.

Response: We agree with the referee that the influence of burning conditions on MAE of BBOA was not obvious in this study, as mentioned above. We revised this sentence as follows: "Our results suggested that the influence of varied combustion efficiency on the emission levels of BBOA could surpass the differences between biofuel types. (lines 361-362)". The emission factors of $PM_{2.5}$ or BBOA (OC, WSOC, HULIS) from more smoldering conditions were 2.8-4.3 times of those from more flaming conditions in the present study. While the differences between wheat burning and corn burning under similar combustion conditions (or MCE) were not such obvious, as shown in Figures 3.