Response to the comments

Comments to the Author:

Thank you for the revised manuscript. Please see the following comments from the reviewers recommending minor revisions:

The revised manuscript by Cao et al. is much improved, and these efforts made by the authors are much appreciated! The authors have addressed my previous comments. And the added section on the principle component analysis and the Pearson correlation really helped bridge the connections between the different measurement datasets! The revised manuscript should be accepted for publication with minor revisions (please refer to comments below) to further improve the final paper.

Re: We appreciated the reviewers and editor for the valuable comments, which are helpful for improving the quality of the manuscript. We have revised the manuscript based on the comments and provided a point-by-point response to all the comments.

Comments:

1. The conclusion that HULIS may be a major contributor of ROS production is very interesting and is consistent with previous studies as referenced by the author, as well as the review by Win et al., (J. Environmental Sciences, 71, 2018, 13, https://doi.org/10.1016/j.jes.2017.12.004). This fact that the current study have seen further evidence of this should be highlighted even more, as it is indeed an important finding!

Re: Thanks for your suggestion. As indicated in this study, the results again highlighted that HULIS may be a major contributor of ROS production. This is very interesting. According to your comments, we have added this information in the abstract and conclusion. Please refer to Lines 45-47 and line 702-704.

2. Please note that there have been many more studies that have quantified the oxidative

potential of biomass burning aerosols (in addition to the 4 journal articles mentioned by the authors, which gives the impression that much less work has been done in this area). Also, some of the papers that the authors have referenced later (comparison to literature values of DTTm, lines 561-563) were not referenced or mentioned in the introduction.

Some examples:

Hakimzadeh et al., The impact of biomass burning on the oxidative potential of PM2.5 in the metropolitan area of Milan, Atmospheric Environment, 224, 2020, 117328. https://doi.org/10.1016/j.atmosenv.2020.117328

Pietrogrande et al., Chemical composition and oxidative potential of atmospheric particles heavily impacted by residential wood burning in the alpine region of northern Italy, Atmospheric Environment, 253, 2021, 118360. https://doi.org/10.1016/j.atmosenv.2021.118360

Seo et al., Comparison of physical and chemical characteristics and oxidative potential of fine particles emitted from rice straw and pine stem burning. Environmental Pollution, 1267, 2020, 115599, https://doi.org/10.1016/j.envpol.2020.115599

Re: Thanks. According to your comments, we have added some references about oxidative potential of biomass burning aerosols in the revised manuscript. In addition, the literitures that referenced later (comparison to literature values of DTTm) were also added in the introduction. Please refer to Lines 101-105 and Lines 111-115.

3. Lines 580 -583: I am having trouble understanding how values of HULIS-C DTTm “are higher than the carbon portion of the HULIS-C/WSOC for the same sample”?

Re: We are sorry for the misleading sentences. In original manuscript, “these values” are the ratios of the DTT activity of HULIS-C to that of WSOC for the same sample. In order to avoid this misunderstand, we have revised that in the current manuscript. Please refer to Lines 587-589.

4. Line 626: What is the significance of the subscript “c” of DTTc? I think the authors are referring to DTT that is mass normalized by carbon mass, oppose to DTTm, which I gathered is DTT normalized by WSOC mass? Perhaps it might be more clear to refer to DTTm (that is normalized by WSOC, by DTTWSOC, or the alike?

Re: Yes. The subscript “c” of DTT_C is the organic carbon mass of BrC fractions (i.e., WSOC, HULIS-C and MSOC) in smoke samples. In this study, the DTT_m and DDT_C values were both calculated and presented in the paper. The DTT_m value was normalized by PM mass, which has been widely used in many previous studies. For better comparison with the data reported in other studies, the DTT_m value was presented in “Section 3.5 Oxidative potential”. However, it is noted that the optical and chemical properties of BrC fractions were all obtained based on organic carbon rather than PM, therefore, the DTT_C value was used to investigate the relationship between oxidative potential and chemical compositions of BrCs. To avoid misunderstanding, we have used DTT_{OC} instead of DTT_C in the revised manuscript. Please refer to Line 637-644.

5. Line 624: The methodology used for the principle component analysis is missing.

Re: We apologized for this missing. In the current manuscript, we have added the methodology used for principle component analysis in the supporting information (Section S6).