

Dear Editor,

The authors gratefully thank you for your comments and suggestions. We have revised our manuscript to address these suggestions and comments. All the changes and responses to the comments are listed below point-by-point. We hope our work could satisfy both the reviewers and the editor. Our answers to each question have been marked in blue text. The annotated line numbers refer to the revised copy of the manuscript.

#### 1) Language

Before further consideration, the manuscript has to undergo language editing. Please note that the publisher does not provide this service. Therefore, please seek external services by a proficient English speaker or a professional language editing service.

Particular attention should be paid to the correct use of tenses, articles, plural vs singular and grammar.

**Response:** Thanks for the comments and suggestions. The manuscript has been thoroughly revised by a native English-speaking colleague in our field.

#### 2) Your responses to the previous editor comments are too brief and general:

- Critical Limitations and Temporal Context

The study was conducted during the autumn harvest period (October-November 2022) when biomass burning is particularly frequent ( is it?) in the Sichuan Basin. This represents a significant limitation that **must be thoroughly addressed:**

- Annual Relevance: What is the relevance of these findings on an annual basis? How representative are autumn conditions of year-round aqSOA formation processes?
- Seasonal Variability: How might aqSOA formation and brown carbon properties differ during other seasons when biomass burning is less prevalent?
- Quantitative Significance: Provide quantitative estimates or at least a qualitative discussion of how seasonal variations in biomass burning might affect the broader implications of your findings.

The first two points are only addressed by one sentence each. The response to the last point seems largely off-topic. The editor asked how seasonal variations would affect your conclusions. I acknowledge that you added new text in l. 651 – 667. However, L. 651 – 660 are a very general, at most a qualitative comparison. Can you quantify how much lower emissions are in other seasons?

l. 660 – 667 is text that seems out of place here as it does not add to the discussion of how your results compare to different seasons. Such considerations are more fitting for a conclusion section, if substantiated by the findings of the current study.

Please improve this discussion and add some quantitative comparison, e.g. of OA loading or emissions.

**Response:** We sincerely appreciate the editor's insightful comments regarding the temporal scope of our study. We sincerely appreciate the editor's insightful comments regarding the temporal scope of our study. Biomass burning is indeed frequent in China during the autumn harvest period (October to November 2022), particularly in the Sichuan Basin. It has been revised accordingly in "1 Introduction" in the revised manuscript, highlighting the importance of conducting aqSOA research in autumn in the Sichuan Basin: ".....Xu et al. (2022) indicated that biomass burning serves as a significant non-fossil source of aqSOA under high RH and high aerosol liquid water content (ALWC) conditions, especially during fall-to-winter period when open burning of post-harvest agricultural crop residues is widespread in China. While aqSOA formation has been extensively studied during winter, autumn—despite featuring both high ALWC and strong biomass burning emissions—has received considerably less attention (Feng et al., 2022; Qiu et al., 2016; Zhao et al., 2019). In contrast to these studies in NCP, research in the SCB has shown that the effect of aqueous-phase reactions on oxygenated OA (OOA) formation was significant when ALWC was below  $200 \mu\text{g m}^{-3}$ , but became insignificant at  $\text{ALWC} > 200 \mu\text{g m}^{-3}$ . Additionally, aqueous-phase oxidation probably did not contribute to the decay of BrC during summer in the SCB (Bao et al., 2024). It should be pointed out that autumn is the typical biomass burning season following the harvest of rape and rice in the SCB, and the contribution of

biomass burning to OA is significantly higher in autumn than in other seasons (Chen et al., 2017; Tao et al., 2014; Yang et al., 2019). In summary, the intensive biomass burning emissions and high ALWC during autumn in the SCB likely result in aqSOA formation pathways that differ from those in other seasons. To date, few studies have explored the dynamic evolution and optical properties of aqSOA in the SCB, especially during autumn, leaving ambient aqSOA processing poorly understood. Therefore, a more detailed characterization of aqSOA formation and optical properties is of great importance to reveal the key factors contributing to haze formation in this region." (Lines 99–120).

In the revised manuscript, we first quantified the concentrations of BBOA, OOA, CCOA, HOA, and aqSOA during the observation period, along with their contributions to OA. We then compared the observed OA and BBOA concentrations, as well as the OA/PM<sub>2.5</sub> and BBOA/OA ratios obtained under the high-ALWC and strong biomass-burning conditions characteristic of autumn and winter, with those reported for other regions of China: "The OA fractions of PM<sub>2.5</sub> in this study are as high as those reported in wintertime studies in the SCB, and significantly higher than those observed in winter in other regions of China (Table S1)." (Lines 281–284) and "Overall, the average concentration of BBOA was  $8.6 \pm 7.7 \mu\text{g m}^{-3}$  and accounted for the highest proportion of OA ( $34.8 \pm 11.2\%$ ), followed by OOA ( $5.5 \pm 3.5 \mu\text{g m}^{-3}$ ,  $21.7 \pm 11.4\%$ ), CCOA ( $4.0 \pm 3.3 \mu\text{g m}^{-3}$ ,  $15.7 \pm 7.1\%$ ), HOA ( $3.5 \pm 2.8 \mu\text{g m}^{-3}$ ,  $14.6 \pm 8.1\%$ ), and aqSOA ( $3.3 \pm 2.9 \mu\text{g m}^{-3}$ ,  $13.2 \pm 5.9\%$ ) (Fig. 1). These results demonstrated BBOA as the dominant component of OA in autumn in the SCB. It is worth noting that fractions of BBOA to OA in this study were much higher than those reported in wintertime studies in China (Table S1)." (Lines 322–328). We pointed that autumn represents a period of peak production for biomass-burning-influenced aqSOA, but the mechanisms remain relevant across seasons: "Additionally, BBOA contains abundant water-soluble organic compounds (WSOC) (i.e., sugars, phenols, and organic acids) that can efficiently form aqSOA via aqueous-phase reactions (i.e., oxidation and oligomerization reactions) (Ervens et al., 2011; Gilardoni et al., 2016; Lee et al., 2013; Lei et al., 2024; Li et al., 2020; Powelson et al., 2014). In contrast, OOA

formation primarily relies on gas-phase oxidation of VOCs with high-reactivity (i.e., aromatics and long-chain alkanes by OH radicals, which have low concentrations in BBOA plumes (Akagi et al., 2011; Jimenez et al., 2009; Shrivastava et al., 2017; Yokelson et al., 2007). This suggested that BBOA acted as the important precursors for aqSOA instead of OOA via aqueous-phase reactions. These results were consistent with previous studies and most of the observed data fall within the triangle space (Bao et al., 2023; Kim et al., 2019; Paglione et al., 2020). It should be noted that these processes of aqSOA formation could be more intense and important during autumn due to elevated precursor concentrations (i.e., BBOA), ALWC, and RH values, though the underlying chemical pathways are robust and can occur year-round (Bao et al., 2023; Tang et al., 2025; Zeng et al., 2025)." (Lines 494-509).

We argue that the aqSOA formation processes and secondary BrC formation from BBOA through aqueous-phase reactions we observe are likely dominant in autumn but less influential in other seasons. To address how aqSOA formation and BrC absorption properties might differ in other seasons, we have revised accordingly and add some quantitative comparison in the revised manuscript based on the literature and known seasonal characteristics of the basin: "Previous studies indicated that biomass-burning activity is negligible in summer, and although it may also experience in spring and winter, its intensity is typically less intense than in autumn (Chen et al., 2014; Chen et al., 2017; Tao et al., 2014; Yang et al., 2019). For example, Tao et al. (2014) found that the contribution of biomass burning to PM<sub>2.5</sub> during autumn ( $19 \pm 11\%$ ) was significantly higher than in other seasons. Meanwhile, the highest fraction of organic matter (OM) in PM<sub>2.5</sub> was also observed during autumn (33.4%), with biomass burning as the dominant contributor. Additionally, the concentration of BBOA and its fraction in OA during autumn in this study ( $8.6 \pm 7.7 \mu\text{g m}^{-3}$  and  $34.8 \pm 11.2\%$ , respectively) were respectively higher than those observed during summer ( $0.41 \mu\text{g m}^{-3}$  and 5.7%, respectively) (Zeng et al., 2025) and winter in the SCB (Tang et al., 2025; Zhang et al., 2023) (Table S1). It should be noted that the ALWC during autumn in this study ( $41.6 \pm 24.9 \mu\text{g m}^{-3}$ ) was substantially higher than that reported for summer

( $18.6 \pm 35.3 \mu\text{g m}^{-3}$ ) (Zeng et al., 2025) and winter ( $27.4 \pm 9.7 \mu\text{g m}^{-3}$ ) (Tang et al., 2025) in the SCB. Wang et al. (2018) observed that RH values at Chengdu and Chongqing in autumn were also higher than those in other seasons. Previous research indicated that while aqueous-chemistry pathways in spring were comparable to those in autumn, photochemical bleaching of BrC was potentially stronger in spring (Wang et al., 2019a). Although winter features lower biomass-burning emissions, secondary BrC could still form from BBOA through aqueous-phase reactions under high  $\text{NO}_x$  and  $\text{NH}_4$  concentrations and stagnant nighttime conditions, as observed during winter in the SCB (Peng et al., 2025; Wu et al., 2024). In summer, elevated temperature and  $\text{O}_x$  ( $\text{O}_x = \text{NO}_2 + \text{O}_3$ ) levels could enhance photochemical oxidation, promoting secondary BrC formation while also intensifying BrC photobleaching (Wu et al., 2024). In summary, while secondary BrC can form from BBOA through aqueous-phase reactions in all seasons in the SCB (with the possible exception of spring), the elevated ALWC and BBOA concentrations during autumn are particularly favorable for its aqueous-phase formation." (Lines 675-701).

In the revised manuscript, based on the results described above, we have revised the conclusions section as follows: "Our findings align with previous laboratory studies on biomass-burning BrC formation (Lu et al., 2015; Powelson et al., 2014), while providing novel ambient quantification of these processes under realistic atmospheric conditions. The parameterized curve of  $\text{AAE}_{370-880}$  versus BC-to-OA ratios in this study was consistent with the previous laboratory research on biomass-burning emissions. The mean  $\text{AAE}_{370-880}$  observed in this study (1.95) were higher than values reported for fresh and photochemically aged biomass-burning emissions in laboratory experiments (Saleh et al., 2013), and increased significantly with rising  $f_{\text{aqSOA}}$  ( $r^2 = 0.49$ ,  $p < 0.001$ ). Additionally, elevated  $\text{Abs}_{370, \text{BrC}, \text{sec}}$  values coincided with high ALWC,  $\text{NO}_3$ , and  $\text{NH}_4$  levels and correlated strongly with aqSOA concentration ( $r^2 = 0.44$ ,  $p < 0.001$ ). These results suggest that aqueous-phase reactions of BBOA under high- $\text{NO}_x$  and high- $\text{NH}_4$  conditions produce secondary BrC with particularly strong light absorption. It should be noted that seasonal variations in biomass-burning emissions and the

associated chemical processing of carbonaceous aerosols must be adequately represented in climate and air quality models. This is critical to avoid underestimating aerosol impacts in autumn and overestimating them in other seasons. The campaign was conducted during autumn, when biomass-burning activity is intense, and thus may not fully represent aerosol processes in other seasons. The relative uncertainty of  $Abs_{BrC}$  at 370 nm, resulting from the choice of  $AAE_{BrC}$ , ranged from  $-112\%$  to  $42\%$ . Nevertheless, our results underscore the importance of aqueous-phase processing in transforming biomass-burning emissions, with important implications for climate and air quality modeling. The substantial contribution of aqSOA to both aerosol mass and light absorption highlights the need for improved representation of aqueous processes in models. The linkages established here among aging timescales, transport pathways, and aqSOA formation provide a transferable framework for understanding aqSOA processing in other humid regions influenced by biomass burning. In general, research on BrC chromophores is still at an early stage, and more studies that quantitatively link the chemical composition and light absorption properties of BrC chromophores to biomass-burning emissions across different seasons are necessary to improve our understanding of their climatic and environmental effects. Future research should prioritize molecular-level characterization of aqSOA precursors and products, quantification of aqueous reaction rates under ambient conditions, and multi-scale modeling to assess regional climate impacts. This study highlights that aqueous processes play an important role in the evolution of biomass-burning emissions and should be adequately considered in both air quality budgets and climate forcing balance on a global scale." (Lines 758-793).

3) Additional comments:

- a) Section 3.1: Use a different, more descriptive Section title that tells the reader what to expect in this section.
- b) l. 717: "...aqSOA formation via aqueous-phase reactions instead of photo-chemical reactions"

Are you implying that aqueous phase reactions are not photochemical? I do not think that this is correct, since many aqSOA products are formed by OH and other photochemically driven reactions.

c) Data availability: I checked the data set deposited on Zenodo. I could only find a table with 8 columns, including date, the six major aerosol constituents without any unit (is it % or  $\mu\text{g}/\text{m}^3$  or...?) and Abs 370. This data set is certainly not complete. Please provide a full data set so that your analysis can be reconciled. Note that the full data availability is a requirement for Measurement Reports.

**Response:** Thanks for the careful reading and constructive comments. We have addressed all points raised, and our detailed point-by-point responses are provided below. All changes have been incorporated into the revised manuscript.

a) We agree with that the original title was not sufficiently informative. We have changed it to a more descriptive one that clearly states the section's content: "3.1 Enhanced OA formation from BBOA and aqSOA during pollution periods".

b) Thanks for this important clarification. Our phrasing was indeed imprecise and could be misinterpreted. We intended to state that less-oxidized aqSOA was formed predominantly through aqueous-phase reactions, instead of via gas-phase photochemical oxidations of the precursors during the studied pollution episodes. We have revised the sentence to clarify this point: "Additionally, less-oxidized aqSOA, formed predominantly via aqueous-phase reactions, instead of gas-phase photochemical oxidations of their precursors, played a key role in the dynamic evolution of haze pollution during PP." **(Lines 749-751)**

c) We sincerely apologize for this oversight and the inconvenience it caused. We have now uploaded the complete dataset to the same Zenodo repository: "<https://doi.org/10.5281/zenodo.18635386>". The new data file contains all key parameters used in the analysis, including: RH, T, the mass concentrations of  $\text{NH}_4$ ,  $\text{NO}_3$ ,  $\text{SO}_4$ , Chl, Org, BC, OOA, BBOA, CCOA, aqSOA, and HOA in  $\text{PM}_{2.5}$ , the values of ALWC,  $f_{29}$ ,  $f_{43}$ ,  $f_{44}$ ,  $f_{60}$ ,  $\text{Abs}_{370,\text{BrC}}$ ,  $\text{Abs}_{370,\text{brc,pri}}$ ,  $\text{Abs}_{370,\text{brc,sec}}$ ,  $\text{Abs}_{370,\text{OOA}}$ ,  $\text{Abs}_{370,\text{BBOA}}$ ,  $\text{Abs}_{370,\text{CCOA}}$ ,  $\text{Abs}_{370,\text{aqSOA}}$ ,  $\text{Abs}_{370,\text{HOA}}$ ,  $\text{MAC}_{370,\text{BrC}}$ , and  $\text{AAE}_{370-880}$ .

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