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.

Thank you for submitting your revised manuscript “Measurement report: Formation and brownness of aqueous secondary organic aerosol from the aged biomass-burning emissions in the Sichuan Basin, China” and for addressing the reviewers' comments. The paper has improved very much since the initial submission, and I appreciate the authors' efforts to incorporate the feedback provided.

It is now much clearer that the manuscript presents valuable insights into aqSOA formation from aged biomass-burning emissions and can make significant contributions to our understanding of aqueous-phase atmospheric chemistry. It now has the potential to be published as a manuscript, not as a measurement report, in ACP. What is particularly unique about this work is the direct ambient observation of aqSOA formation from aged biomass-burning emissions under high aerosol liquid water content conditions, with a comprehensive discussion of both the formation mechanisms and the optical properties in a region with frequent biomass burning activity over a long time period.

However, before the manuscript can be sent to a further round of reviews and can be considered for publication in Atmospheric Chemistry and Physics, major revisions are required to align with the journal's guidelines and strengthen the scientific contribution.

Response: Thanks for your positive feedback and constructive comments on our revised manuscript. We greatly appreciate your acknowledgment of our efforts in improving the manuscript and the potential you see for its publication in ACP. We are pleased to hear that the revisions have significantly enhanced the clarity and scientific contribution of the manuscript. Your suggestion to consider publishing it as a full manuscript rather than a measurement report is particularly encouraging. In

accordance with your recommendations, we have undertaken major revisions to further align the manuscript with ACP's guidelines and strengthen its scientific impact. Below, we outline the key changes made in response to your comments.

1. Structural Requirements per ACP Guidelines

According to the ACP guidelines for authors (https://www.atmospheric-chemistry-and-physics.net/policies/guidelines_for_authors.html), the current manuscript structure needs significant revision of the Concluding Section: The journal requires that "every article must have a final section where the overall advances are concisely summarized and put in context." Your current "Conclusions" section lacks the comprehensive structure expected by ACP. The concluding section should include:

• Summary: Main results related to objectives with quantitative results

• Synthesis/interpretation: Concise explanation enabling readers to understand results as a whole

• Comparison and context: How results compare with previous studies, explaining consistencies, inconsistencies, and advances

• Caveats and limitations: How these affect confidence in results and where future work is needed

• Implications: What results mean for understanding atmospheric and climate behavior

Discussion Integration: Much of your valuable discussion is currently embedded in the Results section (Section 3). Per ACP guidelines, this synthesis and interpretation should be moved to or significantly expanded in the concluding section.

Response: We sincerely thank the editor for this constructive feedback regarding the structure of the manuscript, particularly the Conclusions Section. We have thoroughly revised the "Conclusions" section in accordance with the ACP guidelines to provide a more comprehensive and contextualized summary of the study. The revised section now includes all specified components: a quantitative summary, synthesis/interpretation, comparison with previous studies, caveats and limitations, and broader implications. The following elements have been incorporated:

• Summary:

We have concisely summarized the main quantitative findings of the study, directly relating them to the original research objectives.

• Synthesis/Interpretation:

The revised conclusion now provides an integrated interpretation of the results, explaining how the pieces of evidence collectively support the identified formation mechanisms of aqSOA and its optical properties.

• Comparison and Context:

We have expanded the context by comparing our results with those of previous studies, noting both consistencies (e.g., similarities in reaction pathways observed in laboratory studies) and inconsistencies (e.g., different with that observed in fresh and photo-chemically aged biomass-burning emissions), thereby clarifying the advancements made by this work.

• Caveats and Limitations:

The seasonal specificity of the campaign and relative uncertainty of Abs_{BrC} have been discussed in this study, and how these limitations affect the interpretation of the results. We also indicate directions for future work.

• Implications:

The broader implications of our findings for atmospheric chemistry and climate modeling, especially in regions impacted by biomass burning, are now clearly stated.

Additionally, in accordance with the comment regarding Discussion Integration, the synthesis and interpretation in Results section (Section 3) have been significantly expanded in the concluding section. This restructuring ensures that the "Conclusions" now serve as a stand-alone section that synthesizes and contextualizes the findings, as required by ACP. We believe these revisions greatly enhance the clarity, depth, and scholarly rigor of the manuscript′s conclusion.

"This study conducted comprehensive real-time measurements of the light absorption properties and chemical composition of carbonaceous aerosols during autumn in the Sichuan Basin, China. The findings provided direct ambient evidence demonstrating that aqueous secondary organic aerosol (aqSOA) formed from the aged biomass-burning emissions under high aerosol liquid water content (ALWC > 60 μg m^{-3}) conditions significantly contributes to both aerosol pollution and light absorption.

Organic aerosol (OA) was identified as the dominant component of $PM_{2.5}$ (46.6 ± 10.7%) and exhibited strong absorption properties at UV wavelengths ($Abs_{370,BrC}$ = 42.4 ± 28.5 Mm^{-1}). During pollution periods (PP), the average contribution and concentration of aqSOA to OA were 14.1% and 7.6 μg m^{-3}, exhibiting enhanced oxidation (f_{29} = 0.141 ± 0.062, f_{44} = 0.080 ± 0.035) and substantial light absorption characteristics of OA ($Abs_{370,BrC}$ = 91.6 Mm^{-1}, $AAE_{370–880}$ = 2.1). Additionally, the less oxidized aqSOA formation via aqueous-phase reactions instead of photo-chemical reactions played a key role in the haze pollution dynamic evolution during PP. Furthermore, backward trajectory analysis revealed regional transport of BBOA primarily originated from northeastern Chongqing, and aged to aqSOA within approximately 12 to 48 hours. These results underscore that aqueous-phase reactions of BBOA—particularly during the transport of biomass-burning emissions—converts primary emissions into strong light-absorbing aqSOA, substantially influencing regional haze formation and radiative effects.

Our findings align with previous laboratory studies on biomass-burning BrC formation (Lu et al., 2015; Powelsonet al., 2014), while providing novel ambient quantification of these processes under realistic atmospheric conditions. The parameterized curve of $AAE_{370–880}$ versus BC-to-OA ratios in this study was consistent with the previous laboratory research on biomass-burning emissions. The $AAE_{370–880}$ values observed in this study (average = 1.95) were higher than those in the laboratory experiments of fresh and photo-chemically aged biomass-burning emissions (Saleh et al., 2013), and significantly increased with the increase of f_{aqSOA} (r^2 = 0.49, p < 0.001). Additionally, the elevated $Abs_{370,BrC,sec}$ values were consistent with the high ALWC, NO_3, and NH_4 values, and significantly increased with the increase of aqSOA concentrations (r^2 = 0.44, p < 0.001). These results suggest that aqueous-phase reactions of BBOA in high-NO_x and high-NH_4 conditions produce secondary BrC with particularly strong light absorption. The campaign was conducted during autumn, which features intensive biomass-burning activity, and thus may not fully represent aerosol processes in other seasons. The relative uncertainty range of Abs_{BrC} was [−112%, +42%] at 370 nm due to the choice of AAE_{BC}. Our results underscore the importance of aqueous-phase processing in transforming biomass-burning emissions, with important implications for climate and air quality modeling. The significant contribution of aqSOA to both aerosol mass and absorption highlights

the need for improved representation of aqueous processes in models. The linkages between aging timescales, transport pathways, and aqSOA formation in this study provide a transferable framework for understanding aqSOA processing in other humid regions influenced by biomass burning. 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 705-753).**

2. 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.

Response: We sincerely appreciate the editor′s insightful comments regarding the temporal scope of our study and its implications for the generalizability of our findings. We fully acknowledge that the concentration of our sampling campaign during the autumn harvest period represents a significant limitation. In response, we have substantially revised the manuscript to explicitly address these points and to better situate our conclusions within an annual perspective. Below, we outline the specific revisions made in response to each sub-point:

• Annual Relevance:

We have explicitly stated that the autumn harvest period (October-November 2022) is indeed a period of intensified biomass burning in the Sichuan Basin. It has

been revised accordingly in the revised manuscript, which reads "Previous research indicated that the autumn harvest period − specifically October and November − is a period of intensified biomass burning in SCB, primarily due to post-harvest crop residue burning (Chen et al., 2017; Tao et al., 2014)." **(Lines 336-339).**

We clarify that while the processes of aqSOA formation identified herein (e.g., glycolysis, oligomerization) are chemically robust and likely occur year-round, their intensity and relative importance are certainly heightened during autumn due to elevated precursor concentrations. We also note that meteorological conditions typical of autumn (e.g., high humidity) favor the conditions for aqueous aerosol chemistry. It has been revised accordingly in the revised manuscript, which reads "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 they are chemically robust and likely occur year-round." **(Lines 485-488).** Thus, autumn represents a period of peak production for biomass-burning-influenced aqSOA, but the mechanisms remain relevant across seasons.

• Seasonal Variability:

To address how aqSOA formation and BrC properties might differ in other seasons, we have revised accordingly in the revised manuscript based on the literature and known seasonal characteristics of the basin: "Previous studies also provided evidence that spring may experience similar but less intense biomass-burning activity (Chen et al., 2014; Chen et al., 2017; Tao et al., 2014), with comparable aqueous chemistry though potentially increased photochemical bleaching of BrC (Wang et al., 2019a). Though winter features lower biomass-burning emissions, secondary BrC likely formed from BBOA through aqueous-phase reactions under high NO_x and NH_4 concentrations and stagnant conditions after sunset during winter in SCB (Peng et al., 2025; Wu et al., 2024). Elevated temperature and O_x (O_x = NO_2 +O_3) levels could enhance photochemical oxidation for secondary BrC formation during summer, while also promoting BrC photobleaching (Wu et al., 2024). It should be noted that the seasonal biomass-burning emissions and associated chemical processing of carbonaceous aerosols must be accounted for in climate and air quality models. This is critical to avoid underestimating aerosol impacts in autumn and overestimating them in other seasons. In general, the research of BrC chromophores is still in the early stage, and more studies of the quantitative link between the chemical

composition and light absorption properties of BrC chromophores and biomass-burning emissions during different season are necessary to better understand." **(Lines 651-667).** This qualitative discussion helps contextualize our autumn-specific findings within the broader seasonal cycle.

• Quantitative Significance:

Although our study did not measure other seasons, we have strengthened the narrative by incorporating quantitative seasonal biomass-burning emissions estimates from published regional studies. We reference recent work (e.g., [Wang et al., 2019]) indicating that biomass-burning emissions in the Sichuan Basin were significantly higher in autumn than the annual average. Thus, we argue that the aqSOA formation processes we observe are likely dominant in autumn but less influential in other seasons.

We clearly state the implications for regional and climate models, which reads "It should be noted that the seasonal biomass-burning emissions and associated chemical processing of carbonaceous aerosols must be accounted for in climate and air quality models. This is critical to avoid underestimating aerosol impacts in autumn and overestimating them in other seasons." **(Lines 660-664).** We explicitly note the need for future multi-seasonal campaigns to quantitatively constrain these dynamics, which reads "In general, the research of BrC chromophores is still in the early stage, and more studies of the quantitative link between the chemical composition and light absorption properties of BrC chromophores and biomass-burning emissions during different season are necessary to better understand." **(Lines 664-667).**

These revisions have been integrated throughout the manuscript to ensure that the temporal limitation is transparently addressed and that the contributions and context of our work are clearly understood.

3. Missing Temporal and Spatial Context

• How long has the aerosol been aged before reaching your measurement site? The paper discusses “aged” biomass-burning emissions but provides no quantitative assessment of aging timescales.

• Where did the air masses originate (local, regional, long-range transport)? Back-trajectory analysis or similar approaches would strengthen the manuscript significantly. Please consider that "Articles with a local focus must clearly explain how the results extend and compare with current knowledge".

• Formation Timescales: As noted in previous reviews, the timescales of aqSOA formation processes are not well discussed. What are the typical transport times and pathways affecting your site? This represents a significant gap given that you identify aqueous-phase reactions as the dominant pathway.

Response: We thank the editor for this critical comment, which has helped us significantly improve the contextual rigor of our manuscript. We agree that a more quantitative treatment of air mass history and aging timescales is essential to support our conclusions regarding aqueous-phase processing of aged biomass-burning plumes. In response, we have conducted new analyses and substantially revised the manuscript to provide the missing temporal and spatial context.

• Quantitative Assessment of Aging Timescales & Air Mass Origin:

As suggested, we have performed a 48-hour back-trajectory analysis using the HYSPLIT model to determine the origin and transport pathways of air masses arriving at our sampling site. This new analysis is presented in Section 3.2 of the revised manuscript and visualized in Figure S11.

The results indicated that during the campaign, the majority of air masses originated from northeastern Chongqing, regions known for widespread agricultural burning during the autumn harvest. Higher contributions and concentrations of BBOA and aqSOA, values of ALWC, f_{44}, and the NO_3/SO_4 ratio were observed in the cluster originating from northeastern Chongqing than other clusters during PP. The typical transport time to the sampling site was estimated to be between 12 to 48 hours. We have revised the manuscript to include quantitative assessment of aging timescales.

• Local Focus and Broader Relevance:

We have explicitly stated how our findings, while focused on a specific region and season, extend the current knowledge. We emphasize that the Sichuan Basin acts as a natural reactor for aqueous-phase chemistry due to its unique humid and stagnant meteorological conditions. We establish linkages between aging timescales, transport pathways, and aqSOA formation, providing a transferable framework for

understanding aqSOA processing in other humid regions influenced by biomass burning.

• Formation Timescales:

We acknowledge that the timescales of aqSOA formation were not sufficiently discussed. In Section 3.2, we have added a new paragraph dedicated to this issue. We synthesize the available literature on aqueous-phase reaction rates and juxtapose them with our estimated transport times. We argue that the 12-48 hour aging time frame is highly congruent with the known timescales for efficient aqueous-phase processing of BBOA, which can occur on the order of several hours to several days. This alignment strongly supports our hypothesis that aqueous-phase process of BBOA was an important pathway for the observed SOA formation and BrC evolution under the high ALWC conditions of the Sichuan Basin autumn.

"Regional transport significantly influences the aging of BBOA in the SCB. Previous studies have demonstrated that the northeast winds prevail during autumn in the SCB and facilitate the transport of pollutants along the Dazhou → Guang'an → Hechuan pathway, and this northeast-southwest transport pathway had a significant impact on Chongqing (Peng et al., 2019; Wang et al., 2018). As shown in Fig. S11, air masses predominantly originated from northeastern Chongqing, which is an area with widespread agricultural burning activities (He et al., 2015; Luo et al., 2020), and were transported over short ranges during the campaign. Compared to other air mass clusters, the highest contribution and concentration of BBOA in total $PM_{2.5}$ were observed when air masses passed through northeastern Chongqing. The percentage of air mass trajectories that passed through biomass-burning-influenced regions was higher during PP (~57%) than CP (~35%). Unlike cluster 3 during CP, air masses originating from northeastern Chongqing (cluster 2) during PP showed significantly higher contributions and concentrations of BBOA (24.7%, 27.8 μg m^{-3}) and aqSOA (9.4%, 10.6 μg m^{-3}) than other clusters ($p < 0.001$). In addition, cluster 2 exhibited notably higher values of ALWC (93.7 μg m^{-3}), $Abs_{370,BrC,sec}$ (105.1 Mm^{-1}), f_{44} (0.113), and the NO_3/SO_4 ratio (2.1, a tracer for BBOA aging (Liu et al., 2024; Zhang et al., 2025)) than other clusters during PP ($p < 0.001$). During PP, approximately 68% of the trajectories in cluster 2 passed through biomass-burning-influenced regions, with transport times to the sampling site ranging between 12 and 48 hours. This time frame

is consistent with previous research on the aqueous-phase aging process of biomass burning emissions (Cubison et al., 2011; Hennigan et al., 2010; Ortega et al., 2013; Zhu et al., 2023). These results suggested that regional transport of BBOA primarily originated from northeastern Chongqing, and aged to aqSOA within approximately 12 to 48 hours." **(Lines 512-531).**

4. Strengthen Scientific Significance

While the manuscript presents interesting observations and discusses the atmospheric implications throughout the text, the broader atmospheric implications need clearer articulation: What are the implications for atmospheric chemistry models in regions with and without significant biomass burning?

Response: We thank the editor for this valuable comment, which has helped us to better articulate the broader scientific significance of our work. We have substantially expanded the implications for atmospheric chemistry models in the Conclusions section of the revised manuscript. "Our results underscore the importance of aqueous-phase processing in transforming biomass-burning emissions, with important implications for climate and air quality modeling. The significant contribution of aqSOA to both aerosol mass and absorption highlights the need for improved representation of aqueous processes in models. The linkages between aging timescales, transport pathways, and aqSOA formation in this study provide a transferable framework for understanding aqSOA processing in other humid regions influenced by biomass burning. 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." **(Lines 740-750).**

Our results provide a time frame for aqSOA formation and BrC evolution from aged biomass-burning emissions. Models often underestimate SOA mass and radiative forcing in the biomass-burning-influenced regions (i.e., Southeast Asia, Amazonia, Sub-Saharan Africa). We explicitly state that incorporating the aqueous-phase pathways identified here—triggered by high ALWC, high-NO_x and high-NH_4 conditions—will improve the predictive accuracy of chemical transport models for aerosol mass and light absorption during and after burning seasons. While the primary

precursor in our study is biomass burning, the aqueous-phase reaction pathways are also highly relevant for the oxidation of anthropogenic volatile organic compounds from fossil fuel combustion and industrial sources. Our findings suggest that in any polluted, humid environment with sufficient ALWC, these aqueous-phase processes could be a major source of SOA and BrC. Therefore, we recommend that models for the regions without significant biomass burning (i.e., urban areas in Europe and North America) also consider implementing more detailed aqueous-phase chemical mechanisms.

The manuscript has clear potential for publication in ACP after these major revisions. The observations are valuable, and the comprehensive approach to characterizing both formation mechanisms and optical properties is commendable. What are the uncertainties in discussing the brown carbon absorption estimates? I have significant uncertainties in aethalometer-based BrC measurements in mind.

I look forward to receiving your revised manuscript. If you prefer to submit as a measurement report, the discussion can be somewhat shorter. Nevertheless, it should be in the Conclusion section.

Response: We thank the editor for this insightful comment and for acknowledging the value of our comprehensive approach. We agree that critically discussing the uncertainties associated with aethalometer-based BrC measurements is crucial for a robust interpretation of our optical data. In response, we have added new paragraphs dedicated to this issue and have expanded the discussion of these limitations in the Text S3:

"Previous research have demonstrated that AAE_{BC} is sensitive to the refractive index, size distribution, and coating of carbonaceous aerosols (Gyawali et al., 2009; Lack and Langridge, 2013; Li et al., 2019). In this study, the relative uncertainties of Abs_{BrC} and Abs_{BC} were estimated following the methodology described by Tian et al. (2019):

$$U_{Abs_{BC}}(\lambda)=\sqrt{(U_{Abs}(880\ nm))^2+(AU_{AAE_{BC}}\times \ln(\frac{880}{\lambda}))^2} \quad (S11)$$

$$U_{Abs_{BrC}}(\lambda)=\frac{\sqrt{(U_{Abs}(\lambda)\times Abs(\lambda))^2+(U_{Abs_{BC}}(\lambda)\times Abs_{BC}(\lambda))^2}}{Abs(\lambda)-Abs_{BC}(\lambda)}=\frac{\sqrt{(U_{Abs}(\lambda))^2+(U_{Abs_{BC}}(\lambda)\times(1-C_{BrC}(\lambda))^2}}{C_{BrC}(\lambda)} \quad (S12)$$

$$U_{C_{BrC}}(\lambda)=\sqrt{(U_{Abs}(\lambda))^2+(U_{Abs_{BC}}(\lambda))^2} \quad (S13)$$

where $U_{Abs}(\lambda)$, $U_{Abs_{BC}}(\lambda)$, and $U_{Abs_{BrC}}(\lambda)$ represent the relative uncertainties of Abs, Abs_{BC}, and Abs_{BrC}, respectively. $AU_{AAE_{BC}}$ represents the absolute uncertainty of the $AAE_{BC}=-\log_{10}(Abs_{880}/Abs_{950})\div\log_{10}(880/950)$, $\ln(880/\lambda)$ is used to adjust the $AU_{AAE_{BC}}$ to the relative uncertainty. Qin et al. (2018) fould AAE_{BC} ranges from 0.67 to 1.03 in core-shell scenarios with different refractive indexes, cited in this study. $U_{Abs}(\lambda)$ is specified as a constant value (± 5%) to represent uncertainty in the absorption measurements of the model AE33 at all the wavelengths (Tian et al., 2019; Titos et al., 2015). $C_{BrC}(\lambda)$ is the absorbance fraction of BrC relative to Abs.

The relative uncertainties of Abs_{BC} were estimated to be [−46%, +21%] at 370 nm, [−34%, +16%] at 470 nm, [−28%, +13%] at 520 nm, [−22%, +11%] at 590 nm, [−16%, +8%] at 660 nm, and [−5%, +5%] at 880 nm. The relative uncertainty of Abs_{BrC}, as well as the relative uncertainties of C_{BrC}, can be calculated using equations (S12) and (S13), with a resulting uncertainty range of [−112%, +42%] at 370 nm." **(Lines 161-181).**

Meanwhile, we have revised accordingly in the revised manuscript, which reads "Previous research have demonstrated that AAE_{BC} is sensitive to the refractive index, size distribution, and coating of carbonaceous aerosols (Gyawali et al., 2009; Lack and Langridge, 2013; Li et al., 2019). In this study, the uncertainty associated with the estimation of Abs_{BC} and Abs_{BrC} was analyzed (Text S3). The relative uncertainty ranges of Abs_{BC} and Abs_{BrC} were [−46%, +21%] and [−112%, +42%] at 370 nm, respectively." **(Lines 226-232).**

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