Dear Reviewers,

We appreciate the comments of reviewers and have revised the manuscript to address the review comments from all reviewers. We have provided a point-to-point response to the concerns, suggestions, and questions. 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.

This study reported the direct ambient observation of SOA formation and absorption properties in the aqueous phase from the Sichuan Basin, China. The results showed considerable aqSOA was originated from the aged biomass-burning emissions via aqueous-phase reactions instead of photo-chemical reactions under high aerosol liquid water content (ALWC) conditions, especially during the polluted period. This study revealed the aqSOA formation and brownness from aged biomass-burning emissions and highlighted the importance of aqueous-phase reactions on air quality and climate. This is a manuscript with potential. I recommend the manuscript be revised before being accepted for publication.

1. Lines 83-87. The authors mention the geographic variability of the North China Plain amount the Sichuan Basin, but there is a study on aqSOA in these two regions is not described in detail, it is recommended to add a comparative analysis to better highlight the regional uniqueness of this study.

Response: Thanks for the reviewer's comment. It has been revised accordingly in the revised manuscript, which reads "However, observations on aqSOA formation and optical properties in China are limited and most research concentrate on the North China Plain (NCP). Similar to NCP, the Sichuan Basin (SCB) characterized by high humidity and frequent biomass burning is also the main region with severe aerosol pollution in China (Tian et al., 2019; Wang et al., 2018; Yang et al., 2011). Previous research indicated that aqSOA from different regions exhibited distinct formation mechanisms and optical properties, due to the diverse sources and ambient conditions (Bao et al., 2023; Bao et al., 2024; Wang et al., 2021; Xu et al., 2017). For instance, Wang et al. (2021) revealed fast aqueous-phase conversion of fossil-fuel primary organic aerosol (FF-POA) to aqSOA under high-humidity conditions during a Beijing winter haze event, and found that aqSOA exhibited much lower light absorption than its primary precursor due to decreased aromaticity. Similarly, Huang et al. (2023) illustrated that the aqueous-phase oxidation of fossil fuel combustion emissions played a critical role in SOA formation under high RH conditions. Unlike these studies in NCP, the effect of aqueous-phase reactions on oxygenated OA (OOA) formation was significant when aerosol liquid water content (ALWC) was below 200 μg m^{-3}, but was insignificant when ALWC > 200 μg m^{-3} in SCB. Additionally, the aqueous-phase oxidation process probably did not play a role in the decay of BrC during summer in Chengdu (Bao et al., 2024). Currently, few studies explored the dynamic evolution and optical properties of aqSOA, and the knowledge of ambient aqSOA processing is still limited in SCB. Therefore, a more detailed characterization of aqSOA formation and optical properties is of great importance to reveal the key factors contributing to haze formation." **(Lines 82-104).**

2. Section 2.1. As a field measurement report, this paper lacks a map of the geographic location of the observatory sites, which does not allow us to visualize the

geographic information about the Sichuan Basin, so we suggest that a map of the observatory sites and the surrounding terrain be added.

Response: A map of the observatory site and the surrounding terrain has be added in the revised SI:

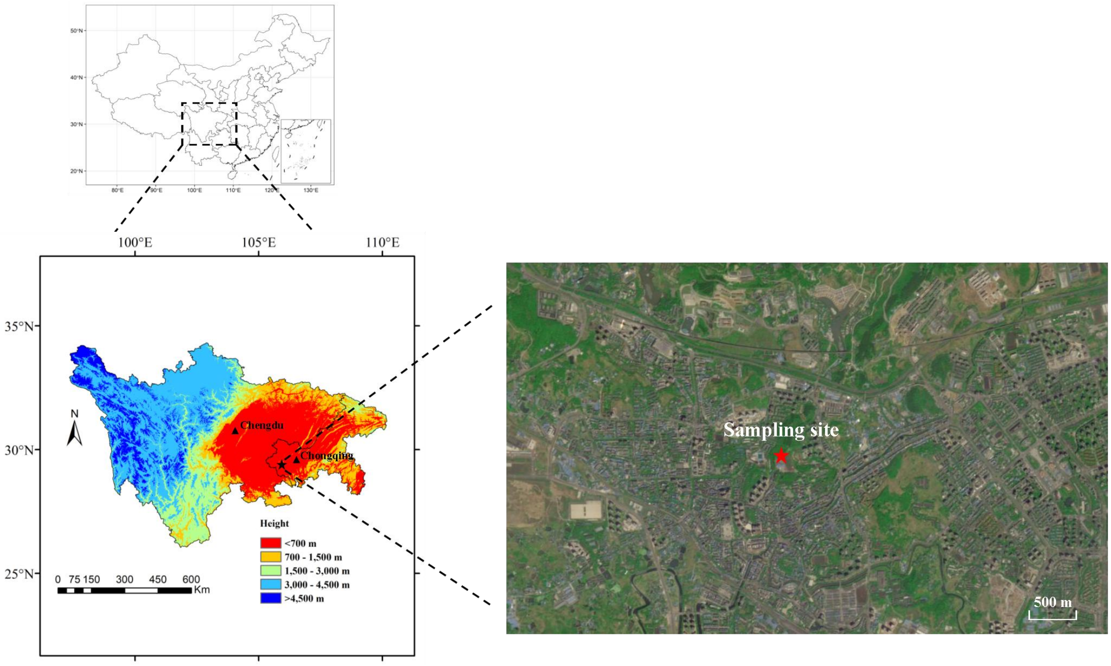

Figure S1. Location of the observation site in the Sichuan Basin (from Baidu Maps, © 2025 Baidu – GS(2023)3206).

3. Line 151. Further details are required on the basis for setting the CE value of ToF-ACSM to 0.5.

Response: Thanks for the reviewer's comment. It has been revised accordingly in the revised manuscript, which reads "Meanwhile, a particle collection efficiency (CE) was introduced to compensate for the particle loss, as the acidity, the contribution of ammonium nitrate (ANMF) and phase state changed the particle bounce effects at the vaporiser (Matthew et al., 2008). Middlebrook et al. (2012) developed a CE algorithm for ToF-ACSM to quantify the aerosol species. Their results indicated that a constant CE value of 0.45 should be used when: (1) the ANMF is below 40%, or (2) particles are partially or fully neutralized. In this study, aerosol particles were dried by Nafion dryer (RH < 30%) before sampling by ToF-ACSM, and the ANMF was always below 40%. As shown in Fig. S2, the average ratio of the measured NH_4 to the predicted NH_4 needed to fully neutralize the SO_4, NO_3 and Chl was approximately 1. All of these conditions did not affect the CE value that had usually been used at this site. The typical default CE value (0.5) was applied during the whole sampling period, which was consistent with previous research (Bao et al., 2025; Peng et al., 2025; Sun et al., 2016a; Sun et al., 2016b; Zhao et al., 2019). While the typical default CE is 10% higher than 0.45, the difference is small considering the 30% uncertainty determined for CE (Bahreini et al., 2009). Additionally, the strong correlation between NR-$PM_{2.5}$ and $PM_{2.5}$ mass concentrations supported that the CE value was reasonable (Fig. S3)." **(Lines 165-182).**

4. Section 2.3.2. The ISORROPIA-II model estimates ALWC from these inorganic constituents only, but will the organic constituents affect the estimates, especially at higher OA conditions.

Response: Thanks for the reviewer's comment. In this study, the organic contribution for ALWC was calculated by Zdanovskii−Stokes−Robinson (ZSR) mixing rule as discussed in SI Text S2 (Guo et al., 2015; Huang et al., 2020; Nguyen et al., 2016; Xu et al., 2022). The average ALWC with organic species only took up 3.7 ± 2.2% of total ALWC, thus the ALWC was determined only considering inorganic species in this study. It has been revised accordingly in the revised manuscript, which reads

"**2.3.2 Aerosol liquid water content**

The ALWC is controlled by meteorological conditions (T and RH) and also by inorganic and organic components. During the campaign, the ALWC with inorganic species was estimated by the ISORROPIA-II model based on the ammonium, nitrates, sulfates, and chlorides mass concentrations from ToF-ACSM and the meteorological parameters (T and RH) from National Environmental Monitoring Station (Fountoukis and Nenes, 2007). Here, the forward type and metastable mode were used in the ISORROPIA-II model (Hennigan et al., 2015). The thermodynamic equilibrium of the $NH_4^+-SO_4^{2-}-NO_3^--Cl^--H_2O$ system was modeled and ALWC was then calculated. Consistent with previous research, the organic contribution for ALWC was calculated by Zdanovskii−Stokes−Robinson (ZSR) mixing rule as discussed in SI Text S2 (Guo et al., 2015; Huang et al., 2020; Nguyen et al., 2016; Xu et al., 2022). In this study, the ALWC with organic species ranged from 0.1 to 35.2 μg m^{-3}, with an average of 1.9 ± 3.0 μg m^{-3}, taking up 3.7 ± 2.2% of total ALWC. As organic species had minor effects on total ALWC (< 5%), the ALWC was determined only considering inorganic species (Chen et al., 2021; Guo et al., 2015; Liu et al., 2017)." **(Lines 197-212).**

"**Text S2.** Estimation of Aerosol Liquid Water Content (ALWC)

The effect of inorganic ions on ALWC was estimated using a thermodynamic equilibrium model for the $NH_4^+-SO_4^{2-}-NO_3^--Cl^--H_2O$ aerosol system, ISORROPIA-II (Fountoukis and Nenes, 2007). Here we ran the ISORROPIA II in forward mode, and the particles were assumed to be deliquescent, i.e., in metastable mode (Hennigan et al., 2015). The ammonium, nitrates, sulfates, and chlorides mass concentrations were measured by ToF-ACSM and the meteorological parameters (T and RH) were obtained from National Environmental Monitoring Station.

Meanwhile, the contribution of organics to ALWC was calculated by Zdanovskii−Stokes−Robinson (ZSR) mixing rule using the following equation (Guo et al., 2015; Huang et al., 2020; Nguyen et al., 2016; Xu et al., 2022):

$$W_{OA}=\frac{OA}{\rho_{OA}}\rho_{w}\frac{k_{OA}}{(100\%/a_{w}\text{-}1)} \tag{S4}$$

where OA was the mass concentration of organics, ρ_w was the density of water (1.0 g cm^{-3}) and ρ_{OA} was the density of organics (1.4 g cm^{-3}) (Nguyen et al., 2016; Xu et al., 2022). k_{OA} was the hygroscopicity parameter of organic aerosol composition. We applied k_{OA} value of 0.08 for organic aerosols at urban site (Nguyen et al., 2016). a_w was the water activity (dimensionless). We assumed a_w was equivalent to RH for the sake of simplicity because of a lack of particle diameter data (Huang et al., 2020; Xu et al., 2022)."

5. Lines 220-222. There is a need for further clarification on the judgment of clean-up and contamination periods, the standards are different in each country and it is recommended to add comparisons with other standards.

Response: Thanks for the reviewer's comment. It has been revised accordingly in the revised manuscript, which reads "According to the Chinese National Ambient Air Quality Standard (NAAQS) (GB 3095-2012) (MEP, 2012), the Grade I annd Grade II levels for daily $PM_{2.5}$ mass concentration are 35 μg m^{-3} and 75 μg m^{-3}, respectively. The Chinese NAAQS Grade II level, based on WHO's Phase-1 interim target (IT-1), is higher than the WHO Air Quality Guideline (AQG) value (15 μg m^{-3}), the EU daily limit (25 μg m^{-3}), and U.S. 24-hour standard (35 μg m^{-3}). During the campaign, the average of $PM_{2.5}$ mass concentration was 1.4 times NAAQS Grade I level (35 μg m^{-3}). Therefore, the pollution periods (PP) were defined by the daily $PM_{2.5}$ mass concentration exceeding NAAQS Grade II level of 75 μg m^{-3}. Similarly, the days with $PM_{2.5}$ mass concentration below 75 μg m^{-3} were characterized as clean periods (CP). " **(Lines 261-270).**

6. Lines 294-296. The authors suggest that aqSOA may also be produced by aqueous phase reactions of fossil fuel-related OA components, but this is only speculative and a more convincing basis is needed.

Response: We agree that the suggestion regarding aqSOA production from fossil fuel-related OA components is speculative. Our current study does not provide direct experimental evidence to definitively support this hypothesis and can not specify the physical or chemical mechanisms behind the fossil fuel-related OA to aqSOA transformation. However, the observed correlation between fossil-fuel-related OA components and ALWC is consistent with recent research (Wang et al., 2021). Moreover, previous studies indicated that aqueous-phase processes could play a role in the formation of SOA from fossil fuel emissions (Ervens et al., 2011; Huang et al., 2023; Wang et al., 2021; Xu et al., 2022; Yan et al., 2017). For example, Wang et al. (2021) and Xu et al. (2022) have highlighted the potential for aqueous-phase reactions to contribute to SOA formation, particularly in regions with high levels of anthropogenic emissions. Based on these findings, we suggest that while our data do not conclusively prove this mechanism, further investigation is necessary to strengthen this hypothesis and explore its relevance in different atmospheric environments.

It has been revised accordingly in the revised manuscript, which reads "Additionally, previous studies indicated that aqueous-phase processes could play a role in the formation of SOA from fossil fuel emissions (Ervens et al., 2011; Huang et al., 2023;

Wang et al., 2021; Xu et al., 2022; Yan et al., 2017). For example, Wang et al. (2021) and Xu et al. (2022) have highlighted the potential for aqueous-phase reactions to contribute to SOA formation, particularly in regions with high levels of anthropogenic emissions. In this study, a strong anticorrelation between the mass fraction of fossil-fuel related OA components (sum of CCOA, HOA and OOA) and ALWC at the high f_{29} values was also observed ($r^2 = 0.48$, $p < 0.001$) (not shown), consistent with recent research (Wang et al., 2021). This indicated that aqSOA might also be produced by aqueous-phase reactions of fossil-fuel related OA components." **(Lines 365-375).**

7. Lines 350-352. These results indicated that considerable aqSOA might be formed from BBOA, which was more intensive than OOA at high ALWC levels during PP. What are the authors' judgments about the magnitude of their intensity, and what are the differences in "intensive"?

Response: Thanks for the reviewer's comment. We have revised this sentence and provided a detailed discussion on the correlations between ALWC and both aqSOA and OOA under high ALWC conditions during PP and CP to clarify the differences in "intensive". It has been revised accordingly in the revised manuscript, which reads "Moreover, the mass fraction of aqSOA showed a stable increasing trend and remained high levels (from 18% to 22%) at ALWC > 80 μg m^{-3}, which was associated with a corresponding decrease in OOA (from 15% to 10%) (Fig. 4b). Compared with OOA ($p > 0.1$), the aqSOA concentrations showed strong positive correlation with ALWC ($r^2 = 0.73$, $p < 0.001$) when ALWC > 80 μg m^{-3} during PP. In contrast, ALWC showed the weak correlations with aqSOA and OOA concentrations during CP ($p > 0.1$). It should be noted that a strong anticorrelation between aqSOA and OOA concentrations was observed during PP at ALWC > 80 μg m^{-3} when $f_{29} > 0.15$ ($r^2 = 0.76$, $p < 0.001$), but not during CP ($p > 0.1$) (Fig. 5b and d). These results indicated that the aqSOA formation was more intensive than OOA at high ALWC levels during PP." **(Lines 431-441).**

8. Lines 372-373. These results indicated that considerable aqSOA might be formed from BBOA, which was more intensive than OOA at high ALWC levels during PP. Any details on what the mechanism is for this result?

Response: Thanks for the reviewer's comment. It has been revised accordingly in the revised manuscript, which reads "Moreover, the mass fraction of aqSOA showed a stable increasing trend and remained high levels (from 18% to 22%) at ALWC > 80 μg m^{-3}, which was associated with a corresponding decrease in OOA (from 15% to 10%) (Fig. 4b). Compared with OOA ($p > 0.1$), the aqSOA concentrations showed strong positive correlation with ALWC ($r^2 = 0.73$, $p < 0.001$) when ALWC > 80 μg m^{-3} during PP. In contrast, ALWC showed the weak correlations with aqSOA and OOA concentrations during CP ($p > 0.1$). It should be noted that a strong anticorrelation between aqSOA and OOA concentrations was observed during PP at ALWC > 80 μg m^{-3} when $f_{29} > 0.15$ ($r^2 = 0.76$, $p < 0.001$), but not during CP ($p > 0.1$) (Fig. 5b and d). These results indicated that the aqSOA formation was more intensive than OOA at high ALWC levels during PP." **(Lines 431-441).**

Additionally, we have shown the details on the mechanism for this result: "The relative abundance of m/z 45 (mainly HCO_2^+), a tracer ion for carboxylic acids, was higher in the aqSOA spectra than in the OOA spectrum (Fig. S9). It was consist with previous research which found that aqueous-phase reactions were important sources of oxygenated organic compounds, including organic acids (Ervens et al., 2011; Kim et al., 2019; McNeill, 2015; Sun et al., 2010; Yu et al., 2014). Fig. 6b shows BBOA and aqSOA with higher f_{60} values (0.019 and 0.011) than CCOA (0.009) and HOA

(0.008). The f_{60} value of OOA was 0.002, lower than the typical background value (0.003) in the atmospheric without biomass burning influence (Cubison et al., 2011). The mass spectrometry feature of aqSOA showed large f_{44} and f_{60} values, laying in a schematic space of aged BBOA based on mass spectrometry features in previous research (Cubison et al., 2011; Ortega et al., 2013). Additionally, BBOA contains abundant water-soluble organic compounds (WSOC) with (i.e., sugars, phenols, and organic acids), that can form aqSOA via efficient 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) (i.e., OH radicals), which has low concentrations in BBOA (Akagi et al., 2011; Jimenez et al., 2009; Shrivastava et al., 2017; Yokelson et al., 2007). This suggested that BBOA could be the important precursors for aqSOA instead of OOA via aqueous-phase reactions. These results were consistent with previous research and most of the observation data were within the triangle space (Bao et al., 2023; Kim et al., 2019; Paglione et al., 2020)." **(Lines 456-478).**

9. Lines 406-407. The authors emphasize the importance of aqueous-phase reactions, but how can the contribution of photochemical pathways be judged?

Response: In the revised manuscript, we have used O_x observational data to analyze the contribution of photochemical pathways to SOA formation in this study, which reads "...In contrast, the odd oxygen ($O_x = O_3 + NO_2$) showed weak correlations with both OOA and aqSOA concentrations during the campaign ($p > 0.1$) (not shown). Although the average O_x concentration was higher during PP (51.1 ± 19.6 ppb) than

CP (36.9 ± 14.0 ppb), no significant correlations were observed in either period (not shown). These results suggested that photochemical reactions might played a limited role in SOA formation in this study." **(Lines 322-328).**

10. Lines 497-500. How was this result obtained and please provide further clarification on the mechanism?

Response: Thanks for the reviewer's comment and suggestion. In this study, the effect of boundary layer height was minimized by $Abs_{370,BrC,sec}/\Delta CO$ to characterize the evolution of secondary BrC absorption. During the campaign, the values of $Abs_{370,BrC,sec}/\Delta CO$ increased with the increases of aqSOA and ALWC concentrations from 17:00 to 03:00 LT (r^2 = 0.63, 0.57, $p < 0.001$), when the average ALWC concentration (45 μg m^{-3}) was higher than that from 03:00 to 17:00 LT (39 μg m^{-3}) (Fig. 8). Previous research found that the SOA with strong absorption across UV to Vis range (i.e., guaiacol dimer and nitroguaiacol) could be formed via aqueous-phase reactions under high ALWC conditions (Gilardoni et al., 2016; Kroflic et al., 2015; Yang et al., 2021). This suggested secondary BrC chromophores with strong absorption at 370 nm were formed under the high ALWC from 17:00 to 03:00 LT. Additionally, we have provided further clarification on the mechanism of aqSOA with strong absorption formation from the aged BBOA via aqueous-phase reactions.

It has been revised accordingly in the revised manuscript, which reads "During the campaign, the relationship between $Abs_{370,BrC,sec}$ and SOA factors mass concentrations was analyzed to understand the correlation between secondary BrC absorption and its chromophores. As shown in Fig. S16, $Abs_{370,BrC,sec}$ significantly increased with the increase of aqSOA concentrations ($r^2 = 0.44$, $p < 0.001$) and high $Abs_{370,BrC,sec}$ values were consistent with the high ALWC values, this was not the case for OOA ($p > 0.1$).

The slope of the linear regression (3.50) between aqSOA mass concentrations and $Abs_{370,BrC,sec}$ was higher than OOA (Fig. S16), so was the MAC values of aqSOA across UV to Vis range (Fig. S14). To further characterize the evolution of secondary BrC absorption, $Abs_{370,BrC,sec}$ was normalized by ΔCO (the background-corrected CO mixing ratios) to minimize the effect of boundary layer height (Fig. 8) (DeCarlo et al., 2010). Here, the background CO value (400 ppb) was defined as the lowest 1.25th percentile of the CO values during the campaign (Kondo et al., 2006). Fig. 8 shows that the values of $Abs_{370,BrC,sec}/\Delta CO$ increased with the increases of aqSOA and ALWC concentrations from 17:00 to 03:00 LT ($r^2 = 0.63$, 0.57, $p < 0.001$), while $Abs_{370,BrC,pri}/\Delta CO$ slightly decreased with the increases of BBOA and m/z 60 concentrations ($r^2 = 0.35$, 0.33, $p < 0.001$). Additionally, the mass concentrations of NO_3, NH_4, and NO_2 from 17:00 to 03:00 LT were 1.2, 1.2, and 1.3 times that from 04:00 to 16:00 LT during the campaign. These results were similar to those observed in SCB during winter (Peng et al., 2025; Wu et al., 2024). As described in section 3.2, the SOA with hydroxyl groups (i.e., glyoxal and methylglyoxal) could be formed from the aged BBOA via aqueous-phase reactions under high ALWC during the campaign. Previous research have shown that oligomers (involving two glyoxal molecules) formed via aqueous reactions of glyoxal with NH_3 contain C=C or C=N bonds, exhibiting strong absorption at near-UV (Laskin et al., 2015; Lee et al., 2013; Nozière et al., 2009; Powelsonet al., 2014). This suggested secondary BrC chromophores with strong absorption at 370 nm were formed under the high ALWC from 17:00 to 03:00 LT, which might be related to the aqSOA from the aged BBOA via aqueous-phase reactions. The low values of $Abs_{370,BrC,sec}/\Delta CO$ at 12:00−14:00 LT could be related to the photolysis and/or photooxidation causing BrC photobleaching (Sareen et al., 2013; Zhao et al., 2015). Overall, we suggested that aqSOA formed from biomass-burning emissions might be important for BrC absorption, especially at night."**(Lines 584-615).**

11. The conclusion section needs to be further condensed; it seems that parts of the conclusion have been emphasized several times, and there is a need for the author to reorganize the structure of the conclusion to give a complete and clear conclusion.

Response: It has been revised accordingly in the revised manuscript, which reads

"4 Conclusions

Field observations indicated that secondary organic aerosol (SOA) accounts for most of organic aerosol (OA) worldwide and aqueous-phase oxidation is an important pathway for the SOA formation. Our results demonstrated the fact that aqSOA was originated from the aged biomass-burning emissions via aqueous-phase reactions under high ALWC in the ambient atmosphere. 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 the polluted period. This study also indicated that the impact on secondary BrC absorption should not be ignored, although primary BrC dominated the BrC absorption across ultraviolet to visible range. The aqSOA formed from aged biomass-burning emissions significantly contributed to the BrC budget and showed stronger absorption across ultraviolet to visible range than other OA components (except BBOA). The similarity between ambient data and 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. Higher values of $AAE_{370–880}$ and $MAC_{\lambda,aqSOA}$ reinforced that aqSOA formation from aged biomass-burning emissions via aqueous-phase reactions had stronger absorption than that via photo-chemically reactions.

In conclusion, our results revealed the aqSOA formation and brownness from aged BBOA via aqueous-phase reactions and highlighted the importance of aqSOA on aerosol pollution and absorption in the Sichuan Basin, China. Brown aqSOA originating from biomass-burning emissions was an important player in air quality budget and climate forcing balance worldwide. And it should be taken into account in air quality and climate models for a correct description of the global OA budget and its climate-relevant optical properties. This study was helpful in understanding the formation, light properties, and impacts of aqSOA in the ambient atmosphere. Future research should focus on the molecular-level characterization, transportation, and reactivities of gas and particle-phase aqSOA precursors to improve understanding of aqSOA formation processes and absorption properties." **(Lines 652-680).**

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