

We are thankful to the two reviewers for their thoughtful and constructive comments and questions that help improve the manuscript substantially. We have revised the manuscript accordingly. Listed below is our point-to-point response each comment that was offered by the reviewers.

Response to Referee #1

General comments:

The manuscript is very well written, and the methods and results are presented with all necessary details. My main concern is related to the interpretation of the results, or rather to the lack of final conclusions (your conclusions at the moment are more like a summary) and implications. Now, the manuscript reads more like a measurement report, and for example, the abstract does not mention anything related to the question of “what do these results imply” in a larger context. ACP accepts different manuscript types (ACP - Manuscript types), and measurement report is one of them. At the present form, therefore, I would recommend adjusting the manuscript into a proper measurement report i.e. adding it also to the manuscript title. This would require sharing the data fully online via DOI, not just upon request. Another option would be to adjust the abstract and conclusions sections to include more discussion on the importance and significance of these results for future research and whether the observations made in these campaigns have larger implications (and reflect on whether these observations agree/disagree with other similar measurements and why).

For example, lines 90-91 in the introduction are very general and could be added to many other manuscripts too presenting such measurements. I think more practical insights are necessary here (how is the understanding advanced, how can these results be used to constrain models and so on) unless going with the Measurement Report. The manuscript title also reads very report-like, and does not reveal anything about the actual results or implications. We thank the reviewer for this insightful and constructive comment. We agree that the initial version of the manuscript placed more emphasis on presenting the measurements, while the broader implications and significance of the results were not sufficiently highlighted. Following the reviewer's suggestion, we have chosen to strengthen the interpretation of our results rather than reframe the manuscript as a measurement report. Specifically, we have revised the Abstract and Conclusions to more clearly emphasize the scientific implications of our findings. In particular, we now highlight (i) the role of cloud microphysical properties, especially droplet size and cloud lifetime, in regulating in-cloud organic aerosol processing, (ii) the importance of cloud-regime-dependent pathways for aerosol aging, and (iii) how these results provide process-level constraints for improving the representation of aqueous-phase SOA formation and aerosol–cloud interactions in atmospheric models. In addition, we have refined parts of the

Introduction to better articulate how this study advances current understanding beyond previous work and how the observations can be used to inform model development. We believe these revisions substantially enhance the interpretative depth of the manuscript and clarify its broader relevance beyond a measurement-focused study.

Specific comments:

Comment #1:

Line 42-43: The reference for the IPCC is from 2021, which was already 5 years ago. I would suggest deleting the word “recent” or adjust otherwise (i.e. change reference) to reflect this.

We thank the reviewer for this helpful suggestion. The word “recent” has been removed to ensure a more accurate description of the reference.

Comment #2:

Line 106: Where do these limits (3km, 95%) come from /are based on when defining cloud events? Absence of precipitation is also mentioned, so I assume only non-precipitating clouds are studied, however later in lines 195 and onwards, you do mention precipitating cloud events too? Can you reflect on any of the differences regarding size distribution or composition for activated or interstitial aerosols for precipitating clouds? Can we expect those to be different from those arising from non-precipitating clouds?

We thank the reviewer for this insightful comment regarding the definition of cloud events and the role of precipitation. The thresholds of visibility < 3 km and RH $> 95\%$ used to define cloud events are based on the operational principles and parameter settings of the GCVI, as well as previous field studies employing similar approaches (Gao et al., 2023; Guo et al., 2025). These criteria are commonly applied to ensure that the sampling corresponds to in-cloud conditions, where cloud droplets are present and can be efficiently separated from interstitial aerosols.

Regarding precipitation, the initial exclusion of precipitating periods is also related to the GCVI sampling principle. During precipitation, raindrops can be sampled by the GCVI inlet, which would significantly interfere with the selective sampling of cloud droplets and lead to biases in the measured cloud residual composition. Therefore, in the 2023 campaign, the GCVI system was automatically shut down during precipitation periods, and only interstitial aerosols were measured during these times. In contrast, during the spring 2024 campaign, the cloud droplet sampling setup was changed (ACSYS system), which effectively avoided the collection of raindrops.

Moreover, most cloud events during this campaign were associated with precipitation. As a result, precipitating cloud events were included in the analysis for 2024, which explains the difference in treatment between the two datasets.

Precipitation can have a substantial impact on aerosol chemical composition. The formation of precipitation generally indicates more efficient activation of cloud condensation nuclei (CCN), often associated with higher hygroscopicity of particles (e.g., enhanced inorganic fraction such as sulfate and nitrate) and stronger aqueous production (Isokääntä et al., 2022). This is consistent with our observations (Fig. 3), where a higher contribution of hygroscopic inorganic species was found in both INT and RES particles during the 2024 campaign compared to 2023. Therefore, the differences in aerosol composition between the two campaigns can be partly attributed to the presence of precipitating clouds in 2024. We agree that aerosols associated with precipitating clouds are expected to differ from those in non-precipitating clouds, due to stronger aqueous-phase processing and more efficient removal and redistribution processes.

We have clarified the treatment of precipitation in Section 2.1.1: **“In the 2023 campaign, periods with precipitation were excluded because raindrops can be sampled by the GCVI inlet, potentially biasing the chemical characterization of RES particles. During such periods, the GCVI system was shut down and only INT particles were measured. In contrast, during the spring 2024 campaign, ACSIS system was implemented, which effectively avoided the collection of raindrops. As most cloud events during this period were associated with precipitation, precipitating cloud events were included in the analysis.”** Plus, we added a discussion on the potential impacts of precipitation on aerosol composition in the third paragraph of Section 3.1: **“In addition, the inclusion of precipitating cloud events during the 2024 campaign may have further influenced aerosol chemical composition. Precipitating clouds are typically associated with more developed cloud systems and enhanced in-cloud aqueous-phase processing, which can promote the formation of secondary inorganic species (e.g., sulfate and nitrate) (Ervens, 2015; McNeill, 2015). This is consistent with the higher contributions of hygroscopic inorganic components observed in both INT and RES particles in 2024 compared to 2023, suggesting that differences in cloud conditions, including the occurrence of precipitation, partly contributed to the observed compositional variability.”**

Comment #3:

Line 127: I have difficulties understanding this sentence. What do you mean by that the size range for every bin varies for different sizes? Do you mean the width of the size bin varies?

We thank the reviewer for pointing out the lack of clarity in this sentence. Yes, we intended to indicate that the width of the size bins varies depending on the droplet size range. We have revised the sentence to make this clearer in Sect. 2.1.2: **“The instrument resolves particles into 30 size bins, with variable bin widths: 1 μm for droplets smaller than 14 μm and 2 μm for larger droplets.”**

Comment #4:

Line 130: How well does ERA5 correspond to the measured values at the site? I would assume large differences exist considering your site is located on a mountainous environment, thus having (potentially) large variation in the WD depending on where you measure. ERA5 does represent an average over larger gridbox, so might not be always representable.

We thank the reviewer for this important comment. We agree that, due to the complex mountainous terrain at our site, local wind conditions can vary substantially and may not always be well represented by ERA5 reanalysis data, which provides spatially averaged meteorological fields. During the autumn 2023 campaign, the in-situ wind direction and wind speed measurements were affected by an instrument malfunction, resulting in incomplete and unreliable data. Therefore, we used ERA5 data as a reference to provide general meteorological context. We acknowledge that ERA5 may not fully capture local-scale variability at the site, particularly for wind direction. However, wind-related parameters are not a central focus of this study, and our analysis and conclusions are not strongly dependent on wind direction or wind speed. Therefore, the use of ERA5 data does not significantly affect the main results presented in this work. We have added a clarification in Section 2.1.3 to state that ERA5 data were used as a reference due to missing in-situ measurements and to acknowledge the limitations of ERA5 in representing local meteorological conditions in complex terrain. We added: **“Due to an instrument malfunction, in-situ WD and WS measurements during the autumn 2023 campaign were incomplete and considered unreliable; therefore, ERA5 reanalysis data were used to provide general meteorological context only. Given the complex mountainous terrain of the site, ERA5 data may not fully capture local-scale variability, particularly for wind direction.”**

Comment #5:

Line 162: You mention orographic cloud events in few places. Consider explaining this out when mentioned first time for readers who are not familiar with this type of clouds / formation process. Later on (Line 165) you also bring up the persistent cloud events, please consider giving brief explanation on the differences and similarities of

these.

We thank the reviewer for this helpful suggestion. We agree that clearer definitions of orographic and long-persistent cloud events will improve readability for a broader audience. We have added brief explanations of both cloud types at the beginning of Sect.2.2.2, including their formation mechanisms as well as their key differences and similarities: **“In this study, two types of cloud events were identified, namely orographic and long-persistent cloud events. Orographic clouds form when air masses are forced to ascend over mountainous terrain, leading to rapid cooling and condensation; these clouds are typically short-lived and strongly influenced by local topography and wind conditions. In contrast, long-persistent cloud events are associated with more stable large-scale meteorological conditions, resulting in longer cloud lifetimes and more continuous cloud processing. Despite differences in formation mechanisms and duration, both cloud types provide environments for aqueous-phase processing of aerosols, although the extent and efficiency of such processing may vary.”**

Comment #6:

Line 170: Wet scavenging process is rather abstract term, especially when you discuss about wet scavenging efficiency. Could you be a bit more specific on what wet scavenging process do your numbers you calculate reflect on? Is this now in-cloud removal via activation, below cloud removal or what?

We thank the reviewer for this important comment. We agree that the term “wet scavenging” can be ambiguous and should be more clearly specified. In this study, the calculated scavenging efficiency primarily reflects in-cloud removal via activation of aerosol particles into cloud droplets during the cloud formation stage. The calculation is based on the comparison between INT particles and pre-cloud AMB particles, and therefore represents the fraction of particles that are incorporated into cloud droplets and removed from the interstitial phase. Since the analysis is restricted to the early cloud formation period (i.e., when visibility stabilizes below 100 m), the derived scavenging efficiency mainly captures the activation process rather than below-cloud scavenging or precipitation-driven removal. We have revised the text of Sect. 2.2.3 to clarify this point and to avoid potential ambiguity in the interpretation of the scavenging process: **“Here, the scavenging efficiency primarily represents in-cloud removal via activation of aerosol particles into cloud droplets, as inferred from the depletion of INT particles relative to pre-cloud AMB conditions.”**

Comment #7:

Figures 1-3 and others where you show bulk composition from AMS: I understand that the red-green-blue-orange-black colors are the ones traditionally used to present AMS data. However, the red-green combination is not very accommodating for the most common varieties of color vision deficiencies. ACP submission guidelines enforce this (ACP - Submission) and therefore I strongly recommend the authors to test their figures and select other color schemes or add perhaps line overlays to make the different colors distinguishable also for people with color vision deficiencies.

We thank the reviewer for this important suggestion and fully agree that the figures should be accessible to readers with color vision deficiencies. Following this recommendation and the ACP guidelines, we have revised the color scheme and added pattern overlays to improve the distinguishability of different components. Specifically, in Figures 1 and 2, we retained the original color scheme but added distinct hatch patterns to the red and green components (horizontal lines for red and diagonal lines for green) to enhance contrast. In Figure 3, we adjusted the presentation by separating the green component in the pie charts to better distinguish between organic and inorganic fractions. All modifications have been clearly indicated in the figure captions. These changes improve the accessibility of the figures while maintaining consistency with commonly used AMS color conventions.

Comment #8:

Line 228: What are the potential reasons for the more aged particles for 2024? Is this due to the different cloud events/types (orographic versus persistent)? You do mention the lifetime of the clouds later on in the paragraph, but could the reason be different origin of the aerosols between these two years?

We thank the reviewer for this insightful comment. We agree that the higher degree of aerosol aging observed in 2024 is likely influenced by multiple factors. While differences in cloud types (orographic versus persistent) and associated cloud lifetimes may contribute to enhanced in-cloud processing, they are unlikely to be the sole explanation. Differences in aerosol sources and background atmospheric conditions between the two campaigns may also play an important role. As shown in Fig. 4, even outside cloud processing periods (i.e., pre-cloud and post-cloud ambient particles), the fraction of MO-OOA in 2024 is consistently higher than in 2023. This suggests that the overall aging level of aerosols was already elevated in 2024 prior to cloud processing. Such differences may be related to variations in source regions and/or atmospheric oxidation capacity between the two periods, which can influence the baseline chemical state of the aerosols. Therefore, the enhanced aging observed in 2024 likely reflects a combined effect of different cloud processing conditions and differences in aerosol origin and atmospheric chemical environment. We have added a discussion in the last paragraph of Section 3.1 to clarify that

the higher aerosol aging observed in 2024 may be influenced by both cloud processing conditions and differences in aerosol sources and background atmospheric oxidation levels: **“This enhanced aging is likely influenced not only by differences in cloud processing but also by elevated background atmospheric oxidation conditions. Notably, even outside cloud processing periods, the fraction of MO-OOA in pre-cloud and post-cloud AMB particles was consistently higher in 2024 than in 2023, indicating a higher overall oxidation level prior to cloud interaction.”**

Comment #9:

Sect. 3.2: Ah I do see now that you have made some air mass comparisons here. So overall, just to confirm, your conclusion is that the air masses came from similar directions in the 2024 and 2023 campaign, and thus the differences I mentioned above are unlikely due to different air masses/source regions?

We thank the reviewer for this clarification. Based on the backward trajectory analysis presented in Sect. 3.2 (Fig. S8), the air masses during the 2023 and 2024 campaigns generally originated from similar directions. However, we note that there are still notable differences in the transport characteristics between the two periods. In particular, the trajectories in 2024 tend to be longer, suggesting that the air masses reaching the site may have originated from more distant source regions compared to those in 2023. This implies that, despite similar transport directions, differences in source regions and transport pathways may still exist. Therefore, the influence of air mass origin cannot be fully excluded and may still contribute to the observed differences in aerosol aging between the two campaigns. We have clarified this point in Sect 3.2 to avoid overinterpretation of the trajectory similarity: **“Although the air mass trajectories during the two campaigns were broadly similar in direction, the longer transport pathways observed in 2024 suggest a potential influence of more distant source regions.”**

Comment #10:

Lines 285-294: Could you give numeric values for the scavenging efficiencies also here, at least for the species you explicitly mention? Maybe just reporting the averages you later show in Fig. 7? This way the reader does not have to go back and forth between the figure and text.

We thank the reviewer for this helpful suggestion. We have added the corresponding numerical values of scavenging efficiencies for the key species discussed in this section, using the average values shown in Fig. 7. This allows the reader to directly access the quantitative information without referring back to the figure. We added: **“The scavenging efficiency of organics (65% in 2023 and 74% in 2024) was generally similar to that of nitrate**

(66% in 2023 and 72% in 2024). Among organic components, SOA were more efficiently scavenged than POA during both observation periods, likely due to the lower hygroscopicity of POA. This behaviour was also observed for BC, which showed limited removal efficiency (~60%). Among the four organic components identified, MO-OOA showed the highest wet scavenging efficiency (75-82%), followed by IEPOX-SOA (64-75%), consistent with their respective hygroscopic properties.”

Comment #11:

Figure 7 and Figure 8: You use different colors for 2023 and 2024, highlighting the comparison between those years. Is that your goal or is the main idea to compare the different cloud events (persistent versus orographic) which just happened to take place in different years? I would consider adding these cloud types to the figure alongside the observation years to mark this. After all, its not really the different years alone you are interested in but the very different clouds taking place those years. If it were the years, I would be expecting similar values which is not the case here.

We thank the reviewer for this important clarification. The primary objective of this study is indeed to compare different cloud types (persistent versus orographic), rather than the observation years themselves. The distinction between 2023 and 2024 mainly reflects the occurrence of different cloud types during the two campaigns. Following the reviewer's suggestion, we have revised the figure legends in Figures 7 and 8 to explicitly include the cloud types alongside the observation years.

Comment #12:

Line 351: Results partly align, a little ambiguous. Where do they agree and where do they not?

We thank the reviewer for pointing out this ambiguity. In Sect. 3.3.1, we showed that smaller cloud droplets tend to promote the formation of LO-OOA, likely due to enhanced aqueous-phase processing. At the same time, the limited LWC in smaller droplets may constrain further oxidation to highly oxygenated species. In Sect. 3.4, we observe that the mass fraction of LO-OOA increases as droplet size decreases. This is consistent with the idea that smaller droplets favor the production of LO-OOA. Therefore, the agreement lies in the consistent role of droplet size in modulating oxidation processes, while the difference is that Sect. 3.4 does not provide clear evidence for inhibited MO-OOA formation in smaller droplets, but instead suggests a more complex response of organic aerosol composition. We have clarified this statement in Sect. 3.4 to explicitly describe where the results are consistent: **“These results suggest that the formation of in-cloud SOA tends to occur preferentially in smaller droplets,**

which partly align consistent with the results in Sect. 3.3.1, where smaller droplets were found to promote the formation of LO-OOA. ... However, Sect. 3.4 does not provide clear evidence for suppressed MO-OOA formation in smaller droplets; in some cases (e.g., Case 6 in 2023), the MO-OOA fraction even increases with decreasing droplet size, indicating a more complex response of SOA composition.”

Comment #13:

Sect. 4: The conclusions are more like a summary of the results. I don't oppose a summary, however, if aiming for something else than a measurement report, this would need to be expanded. Please see my general comment.

We thank the reviewer for this helpful suggestion. We agree that the original Conclusions section was too focused on summarizing the results and did not sufficiently emphasize their broader implications. In the revised manuscript, we have expanded the Conclusions to go beyond a summary by explicitly discussing the significance of our findings. In particular, we now emphasize the key role of droplet-scale microphysical properties in controlling in-cloud organic aerosol processing, and we discuss how these mechanisms can influence aerosol aging, cloud chemistry, and aerosol–cloud–climate interactions more broadly. Furthermore, we have added discussion on how our results can help constrain atmospheric models, especially with regard to representing aqueous-phase SOA formation and cloud-regime-dependent processes. We also highlight the broader applicability of our findings to other cloud-influenced environments. These revisions align the Conclusions more closely with the expectations for a process-oriented study rather than a measurement report.

Response to Referee #2

General comments:

This manuscript presents two cloud campaigns at a high-altitude site in southeastern China, combining cloud droplet separation with real-time ACSM and microphysical measurements. The dataset is clearly valuable, particularly the INT/RES separation across contrasting cloud regimes. Such measurements remain relatively limited, especially in this region.

The measurements themselves are strong. The discussion of scavenging efficiency and seasonal contrasts in OA composition is interesting. I tend to agree with Reviewer 1 that the manuscript would be more fit “Measurement Report”. At the same time, two aspects of the analysis would benefit from further clarification and strengthening before publication in ACP.

We thank the reviewer for the positive evaluation of our dataset and for recognizing the value of the INT/RES separation under contrasting cloud regimes. We also appreciate the constructive suggestions for improving the analysis and interpretation. In line with the comments from both reviewers, we have chosen not to reframe the manuscript as a Measurement Report. Instead, we have strengthened the scientific interpretation and expanded the discussion of the results. Specifically, we have (i) clarified the role of cloud microphysical properties, particularly droplet size and cloud lifetime, in regulating in-cloud aerosol processing, (ii) improved the interpretation of scavenging efficiency and organic aerosol partitioning, and (iii) enhanced the discussion on the broader implications of our findings for aerosol–cloud interactions and atmospheric modeling. In addition, we have carefully revised the relevant sections to improve clarity and ensure that the conclusions are more clearly linked to the underlying mechanisms. We believe these revisions significantly strengthen the scientific depth of the manuscript and address the reviewer’s concerns.

Specific comments:

Comment #1:

The analysis compares pre-cloud, formation, in-cloud, and dissipation stages, but mostly through averaged periods. However, the analysis remains largely based on averaged “formation” and “dissipation” stages rather than resolving the continuous chemical–microphysical evolution throughout the cloud lifecycle.

Given the availability of high time-resolution ACSM and other measurements, a process-resolved analysis would substantially strengthen the interpretation. For example: (1) How do OA factors evolve progressively with increasing LWC and droplet size? (2) Is there evidence of gradual in-cloud oxidation within RES particles? (3) Do

chemical changes exhibit hysteresis between cloud formation and dissipation? (4) Is the reported enhancement of LO-OOA during dissipation progressive or abrupt?

Stage-averaged contrasts may obscure transient processes and potentially conflate chemical transformation with air mass variability. A time-resolved co-evolution analysis of LWC, droplet size, INT and RES composition, and OA factor dynamics for representative cases would considerably enhance the process-level insight of the study.

We thank the reviewer for this insightful and constructive comment. Following the suggestion, we have substantially strengthened the analysis by incorporating time-resolved investigations and additional discussion to better resolve the chemical–microphysical evolution during cloud events. Our detailed responses to the four specific questions are provided below.

(1) Evolution of OA factors with LWC and droplet size:

We have results (Fig. 10) to explicitly examine how OA factors evolve with cloud microphysical parameters. The results show that the mass fraction of LO-OOA in RES particles increases with decreasing LWC and D_e , whereas POA exhibits the opposite trend, with higher contributions under larger LWC and D_e conditions. In contrast, IEPOX-SOA shows relatively minor variation across different microphysical regimes. Additional discussion has been included in Sect. 3.4 to interpret these contrasting behaviors in relation to cloud processing and particle activation.

(2) Evidence of in-cloud oxidation:

We have incorporated new results and expanded the discussion in Sect. 3.4 to better characterize in-cloud oxidation processes. Specifically, we present time-resolved analyses of three representative cloud events (Fig. 11), showing a continuous increase in the mass fraction of MO-OOA in both RES and INT particles, accompanied by a gradual decrease in LO-OOA. The enhancement of MO-OOA is more pronounced in RES particles, indicating more efficient aqueous-phase oxidation in cloud-activated aerosols. We also added explanation and discussion about the new results in Sect. 3.4: **“To further examine the temporal evolution of RES and INT particle composition during cloud events, three representative cases (case 2, 3, and 7) were selected, in which anthropogenic influences were relatively limited and chemical composition data were simultaneously available for both particle types. The changes in SOA components over time are presented in Fig. 11a–c. As the cloud events progressed, the mass fraction of MO-OOA in both RES and INT particles showed a continuous increase, while that of LO-OOA gradually decreased. Notably, MO-OOA contributed more substantially in RES particles, suggesting that aqueous-phase secondary oxidation reactions proceeded more efficiently in cloud-activated aerosols than in those that remained unactivated. On average, the mass fraction of MO-OOA in**

RES particles was 44.8–62.2% higher than in INT particles. The contribution of aerosol liquid water (ALW) to the formation of aqueous-phase SOA within particulate matter has been previously documented. For instance, Rogers et al. (2025) reported that ALW played a crucial role in promoting the formation of oxygenated OA in PM_{2.5} during the summer over the United States. Similarly, field campaign conducted in the Whistler forest in the United States revealed that freshly formed biogenic SOA exhibited a lower oxygen content compared to cloud organics measured during periods dominated by non-biogenic sources (Lee et al., 2012). These findings underscore the critical role of particle-phase water in promoting the further aging of SOA and facilitating the formation of MO-OOA.”

In addition, we estimated the O/C ratio based on ACSM-derived f_{44} to further assess oxidation state. Distinct trends between RES and INT particles were observed as a function of OA/ Δ CO. In RES particles, O/C decreases with increasing OA/ Δ CO, suggesting a fragmentation-dominated oxidation pathway likely associated with nocturnal NO₃ radical chemistry. In contrast, INT particles exhibit increasing O/C with OA/ Δ CO, highlighting continued aging and SOA formation under high-RH conditions. These results provide additional evidence that oxidation pathways differ between activated and non-activated particles during cloud events. Based on the new results, we added: “Furthermore, in order to evaluate the oxidation state of organics in different particle types during cloud events, the oxygen-to-carbon (O/C) ratio was estimated based on f_{44} (i.e., the fraction of m/z 44 in the organic aerosol signal) measured by the ACSM. This estimation employed a site-specific linear regression relationship derived from aerosol mass spectrometer (AMS) measurements at the SH site, expressed as $O/C = 3.68 \times f_{44} - 0.37$. Detailed information regarding the derivation and application of this method can be found in Li et al. (2023). Figures 11d-f show the variations in O/C ratio as a function of OA/ Δ CO for RES and INT particles during nighttime orographic cloud events. Here, Δ CO represents the CO concentration after subtracting the regional background level (0.12 ppm), in order to minimize the influence of air mass transport and atmospheric dilution on OA concentrations (De Gouw and Jimenez, 2009; Decarlo et al., 2010). Notably, distinct trends are observed between the two particle types as OA/ Δ CO increases. In cloud-activated (RES) particles, the O/C ratio decreases with increasing OA/ Δ CO, suggesting that in-cloud aqueous-phase or heterogeneous reactions do not significantly enhance the mass of highly oxidized organic aerosol. Instead, the observed behavior is likely associated with SOA formed from biogenic precursors that undergo intensive nocturnal oxidation, predominantly driven by NO₃ radical chemistry. This process can increase the oxidation state of organic molecules while simultaneously reducing their mass concentration, indicating a fragmentation-dominated oxidation pathway (Lee et al., 2012). In contrast, INT particles exhibit

an increasing trend of O/C with OA/ Δ CO during the same cloud events. This behavior highlights the important role of aging processes under high RH conditions, which promote the formation and oxidation of SOA in non-activated particles (Zhang et al., 2024; Hu et al., 2017). These results suggest that, even without cloud activation, INT particles can continue to evolve through hygroscopic growth and multiphase reactions within the cloud environment.”

(3) Hysteresis between cloud formation and dissipation:

We have further investigated the temporal evolution of aerosol composition during cloud formation and dissipation stages, and added detailed discussion in Sect. 3.3.2. Based on three representative cases (Figs. S12–S14), we find that during cloud formation, decreases in aerosol chemical component concentrations lag slightly behind the reduction in visibility, indicating a delayed response to cloud activation. In contrast, during cloud dissipation, increases in particle mass concentrations occur nearly synchronously with visibility recovery. This asymmetric behavior demonstrates a clear hysteresis effect in aerosol–cloud interactions, reflecting differences in the timescales of activation and evaporation processes. We added in Sect. 3.3.2: **“To investigate whether chemical evolution exhibits hysteresis during the cloud lifecycle, we selected three representative cloud events (Figs. S12–S14) and examined the time-resolved co-evolution of visibility and particle composition. The results reveal an asymmetric response of aerosol chemical composition between the cloud formation and dissipation stages. During cloud formation, the decrease in the mass concentrations of chemical components lags slightly behind the reduction in visibility. Even after visibility stabilizes at low values, both INT and RES particle concentrations require additional time to reach steady-state levels, indicating a delayed adjustment of aerosol composition to cloud activation processes. In contrast, during cloud dissipation, the increase in fine particle mass concentrations occurs almost synchronously with the recovery of visibility. As the cloud dissipates, AMB particle concentrations rise concurrently, suggesting a rapid release of cloud-processed material back into the atmosphere. This asymmetric behavior between the formation and dissipation stages indicates the presence of hysteresis in aerosol-cloud interactions, reflecting differences in the timescales of cloud activation and droplet evaporation. Such hysteresis effects highlight the dynamic nature of aerosol chemical evolution during cloud cycles and suggest that stage-averaged analyses may overlook important transient processes.”**

(4) Progressive vs. abrupt enhancement of LO-OOA:

We further examined two representative cloud events and added new discussion in Sect. 3.3.2 (Fig. S15). The results show that LO-OOA continues to increase after cloud dissipation, rather than exhibiting an abrupt change at

the moment of cloud breakup. This increase persists under elevated RH conditions and is interrupted when RH decreases significantly. These findings indicate that the enhancement of LO-OOA is a progressive process, driven by continued aqueous-phase or multiphase oxidation in the post-cloud environment, and suggest that cloud processing effects can extend beyond the cloud lifetime. We added in Sect. 3.3.2: **“To further examine whether the enhancement of LO-OOA during cloud dissipation is progressive or abrupt, two representative cloud events were selected for detailed analysis (Fig. S15). The results show that LO-OOA continues to increase after the cloud has dissipated, rather than exhibiting an instantaneous enhancement at the point of cloud breakup. Notably, this increasing trend persists for a certain period following cloud dissipation and is interrupted when RH decreases significantly (Fig. S15b). This behavior suggests that the enhancement of LO-OOA is a progressive process, driven not only by in-cloud processing but also by continued aqueous-phase or multiphase oxidation under elevated RH conditions in the post-cloud environment. These findings indicate that cloud processing effects on OA can extend beyond the cloud lifetime, with high RH conditions after cloud dissipation still facilitating chemical aging and the formation of moderately oxidized SOA.”**

Overall, these additions provide a more process-resolved perspective on aerosol chemical evolution and directly address the reviewer's concern regarding the limitations of stage-averaged analysis.

Comment #2:

The manuscript attributes many differences between 2023 and 2024 to cloud-type contrasts. However, the two campaigns also differ in season, background aerosol composition, air mass patterns, and sampling systems. It therefore remains unclear whether the reported contrasts reflect intrinsic cloud-type controls or broader seasonal variability. A clearer separation between cloud-type effects and background differences would strengthen the attribution.

We thank the reviewer for raising this important point regarding the attribution of the observed differences between the 2023 and 2024 campaigns. We agree that, in addition to cloud-type contrasts, the two campaigns differ in several aspects, including season, background aerosol composition, air mass characteristics, and sampling conditions. As the reviewer correctly points out, fully disentangling the effects of cloud regime from broader seasonal variability would require a more extensive dataset (e.g., multiple campaigns covering different cloud types within the same season), which is beyond the scope of the present study. In this work, our primary objective is to contrast aerosol–cloud interactions under two distinct observational periods (autumn 2023 and spring 2024), during which different cloud regimes were predominantly observed. We acknowledge that the reported differences therefore reflect a

combination of cloud-regime effects and seasonal/background variability, rather than purely intrinsic cloud-type controls. To reduce the influence of air mass variability, we have (i) compared backward trajectories between the two campaigns (Sect. 3.2 and Fig. S8), which show broadly similar transport pathways, and (ii) applied normalization approaches (e.g., ΔCO) in the analysis to minimize the impact of dilution and regional background differences. In addition, we have focused part of our analysis on relatively clean and comparable cases to further reduce the influence of anthropogenic variability. Following the reviewer's suggestion, we have revised the manuscript to clarify this point and to adopt more cautious wording when interpreting the differences between the two campaigns. We added in Sect. 1: **“It should be noted that the two campaigns were conducted in different seasons (autumn 2023 and spring 2024), and thus differences in aerosol properties may reflect not only cloud-regime influences but also variations in background conditions, such as air mass characteristics and seasonal emission patterns. Therefore, the comparisons presented in this study are interpreted as the combined effects of cloud processes and seasonal variability, rather than purely intrinsic cloud-type differences.”** And we further stated in Sect. 3.1: **“While these differences are consistent with variations in cloud regimes, it should be noted that they may also be influenced by seasonal and background aerosol variability between the two campaigns.”** And we revised the conclusion part in Sect. 4: **“However, it should be emphasized that the observed contrasts between the two campaigns likely reflect a combination of cloud-regime effects and seasonal/background variability, and thus should not be attributed solely to intrinsic differences between cloud types.”**

In particular, we now explicitly state that the observed contrasts should be interpreted as reflecting combined effects of cloud regime and seasonal/background conditions, rather than attributing them solely to cloud-type differences. We believe this clarification improves the robustness of the interpretation while preserving the value of the dataset in illustrating how aerosol properties and processing differ under contrasting cloud-influenced environments.

References

- De Gouw, J. and Jimenez, J. L.: Organic Aerosols in the Earth's Atmosphere, *Environmental Science & Technology*, 43, 7614-7618, 10.1021/es9006004, 2009.
- DeCarlo, P. F., Ulbrich, I. M., Crouse, J., de Foy, B., Dunlea, E. J., Aiken, A. C., Knapp, D., Weinheimer, A. J.,

Campos, T., Wennberg, P. O., and Jimenez, J. L.: Investigation of the sources and processing of organic aerosol over the Central Mexican Plateau from aircraft measurements during MILAGRO, *Atmospheric Chemistry and Physics*, 10, 5257-5280, 10.5194/acp-10-5257-2010, 2010.

Gao, M., Zhou, S., He, Y., Zhang, G., Ma, N., Li, Y., Li, F., Yang, Y., Peng, L., Zhao, J., Bi, X., Hu, W., Sun, Y., Wang, B., and Wang, X.: In Situ Observation of Multiphase Oxidation-Driven Secondary Organic Aerosol Formation during Cloud Processing at a Mountain Site in Southern China, *Environmental Science & Technology Letters*, 10, 573-581, 10.1021/acs.estlett.3c00331, 2023.

Guo, Z., Zhang, G., Peng, X., Sun, W., Liu, F., Li, M., Pan, X., Du, X., Wang, J., Wang, Z., Wang, X., and Bi, X.: In situ measurement evidence of selective aqueous-phase formation of ammonium, nitrate, and sulfate in clouds, *Atmospheric Research*, 330, 108514, 10.1016/j.atmosres.2025.108514, 2025.

Hu, W., Hu, M., Hu, W. W., Zheng, J., Chen, C., Wu, Y., and Guo, S.: Seasonal variations in high time-resolved chemical compositions, sources, and evolution of atmospheric submicron aerosols in the megacity Beijing, *Atmospheric Chemistry and Physics*, 17, 9979-10000, 10.5194/acp-17-9979-2017, 2017.

Isokääntä, S., Kim, P., Mikkonen, S., Kühn, T., Kokkola, H., Yli-Juuti, T., Heikkinen, L., Luoma, K., Petäjä, T., Kipling, Z., Partridge, D., and Virtanen, A.: The effect of clouds and precipitation on the aerosol concentrations and composition in a boreal forest environment, *Atmos. Chem. Phys.*, 22, 11823-11843, 10.5194/acp-22-11823-2022, 2022.

Lee, A. K. Y., Hayden, K. L., Herckes, P., Leaitch, W. R., Liggió, J., Macdonald, A. M., and Abbatt, J. P. D.: Characterization of aerosol and cloud water at a mountain site during WACS 2010: secondary organic aerosol formation through oxidative cloud processing, *Atmospheric Chemistry and Physics*, 12, 7103-7116, 10.5194/acp-12-7103-2012, 2012.

Li, Z., Xu, W., Zhou, W., Lei, L., Sun, J., You, B., Wang, Z., and Sun, Y.: Insights into the compositional differences of PM₁ and PM_{2.5} from aerosol mass spectrometer measurements in Beijing, China, *Atmospheric Environment*, 301, 119709, 10.1016/j.atmosenv.2023.119709, 2023.

Rogers, M. J., Joo, T., Hass-Mitchell, T., Canagaratna, M. R., Campuzano-Jost, P., Sueper, D., Tran, M. N., Machesky, J. E., Roscioli, J. R., Jimenez, J. L., Krechmer, J. E., Lambe, A. T., Nault, B. A., and Gentner, D. R.: Humid Summers Promote Urban Aqueous-Phase Production of Oxygenated Organic Aerosol in the Northeastern United States, *Geophysical Research Letters*, 52, e2024GL112005, 10.1029/2024GL112005, 2025.

Zhang, Z., Xu, W., Zhang, Y., Zhou, W., Xu, X., Du, A., Zhang, Y., Qiao, H., Kuang, Y., Pan, X., Wang, Z., Cheng, X., Liu, L., Fu, Q., Worsnop, D. R., Li, J., and Sun, Y.: Measurement report: Impact of cloud processes on secondary

organic aerosols at a forested mountain site in southeastern China, *Atmospheric Chemistry and Physics*, 24, 8473-8488, 10.5194/acp-24-8473-2024, 2024.