Anonymous Referee #2

We appreciate the suggestion from the referee. Reviewer reports are marked in black font, our responses are marked in green bold font, and the changes to the revised manuscript are marked in blue bold font.

- The title is a bit hard to read. I'd suggest revising it to "Separating the Twomey effect and the semi-direct effect through the cloud–aerosol mixing ratio," which more clearly conveys the main points of the paper.

We appreciate the reviewer's constructive suggestion. The original title was indeed not sufficiently concise. However, distinguishing the aerosol-cloud interactions specifically under absorbing-aerosol conditions is a central theme of this study. Although absorbing aerosols are implicitly related to the semi-direct effect, we believe that explicitly stating this context is important to avoid misunderstanding the cloud–aerosol mixing ratio as a metric for differentiating absorbing versus non-absorbing aerosols. Clarifying that both the Twomey effect and the semi-direct effect are examined within absorbing-aerosol environments therefore strengthens the accuracy of the title. If title modification is permitted, we are willing to revise it to:
"Separating the Twomey Effect and the Semi-Direct Effect in Absorbing Aerosol Environments through the Cloud-Aerosol Mixing Ratio."

- Abstract:

1 The term "nonlinear" should be clearly defined, especially since it is emphasized in the title. After reading the abstract, it remains unclear whether it refers to the nonlinear response to aerosol amount or to MCr.

2 Including simple equations for the key metrics (the cloud–aerosol ratio and the ACI index) would be helpful. In particular, the interpretation of the ACI index depends on whether it is defined in terms of droplet number concentration (Nd) or droplet size (https://doi.org/10.1029/2008JD011006).

3 I think it's nice to also mention ACI index under low MCr.

- L16: better -> ~~better~~

Below are the revisions we made to the abstract.

L15-20: …This study applies the cloud-aerosol mixing ratio (e.g., mass concentration mixing ratio (MCr), "$log\left(\frac{LWC\times10^6}{PM_{2.5}}\right)$") in conjunction with the ACI

index "($ACI_{N_d} = \frac{1}{3}\left(\frac{\partial \ln N_d}{\partial \ln \alpha}\right)$)" to describe the behavior of aerosol-cloud interactions. Results identify … (2) under low MCr conditions (MCr = 3-4.5), Not only is the ACI_{N_d} less than -0.06, but the high aerosol loading … This study provides a comprehensive explanation of how "absorbing aerosols" influence cloud systems over East Asia and highlights "the critical role of the cloud-aerosol mixing ratio in characterizing the microphysical responses associated with the Twomey effect and the semi-direct effect."

- L45-50: It would be good to note that such nonlinear behavior has been widely observed from space recently, and understanding it is crucial for improving climate predictions, especially in the context of continuously decreasing aerosol emissions (https://doi.org/10.1038/s41558-023-01775-5).

We have added this study to the reference list and moved L80-81 to follow the discussion of that paper, thereby improving the logical flow of the manuscript.

L49: …effect on cloud droplet size and number "(Feingold et al., 2001; Saponaro et al., 2017; Chen et al., 2021; Jia and Quaas, 2023)". "Observational studies have also shown that the water content in the atmosphere plays a critical role in determining whether aerosols ultimately enhance or suppress cloud formation (Zhang and Zuidema, 2019, 2021). Furthermore," some cloud…

L80-81: ~~Observational studies have also shown that the water content in the atmosphere plays a critical role in determining whether aerosols ultimately enhance or suppress cloud formation (Zhang and Zuidema, 2019, 2021).~~

- L62-75: This section discusses why the quantification of ACI remains so uncertain. I think it would benefit from referencing some more timely review papers that summarize the main sources of uncertainty in the Twomey effect (e.g., https://doi.org/10.5194/acp-20-15079-2020, https://doi.org/10.5194/acp-22-7353-2022).

We have rewritten the relevant section and incorporated a discussion on the uncertainties associated with the Twomey effect.

L62: To quantify the impact of aerosols on cloud properties, previous studies have utilized aerosol-cloud interaction (ACI) index as a measure (Kaufman and

Fraser, 1997; Feingold et al., 2001; McComiskey et al., 2009; Lihavainen et al., 2010; Zheng et al., 2020; Chen et al., 2021). "In particular, positive ACI index values (ranging from 0 to 0.33) are generally interpreted as an indication of the strength of the Twomey effect (McComiskey et al., 2009; Lihavainen et al., 2010)." However, … to specific liquid water regimes (McComiskey et al., 2009). "In addition, studies based primarily on satellite data are often constrained by retrieval limitations, as simultaneous estimation of cloud and aerosol properties within the same spatial domain remains challenging (Jia et al., 2021). Moreover, variations in cloud-base and cloud-top CCN concentrations, uncertainties in N_d, and the low vertical resolution of aerosol optical depth (AOD) introduce substantial uncertainty in estimating in-cloud aerosol concentrations (Quaas et al., 2020; Jia et al., 2022). Furthermore, variations in aerosol composition, number concentration, hygroscopicity, and optical properties (Wang et al., 2010; McMeeking et al., 2011; Ohata et al., 2016; Pöhlker et al., 2023) make the conversion from aerosols to CCN difficult to quantify (Quaas et al., 2020), significantly increasing the complexity of retrieving and interpreting aerosol-cloud interaction signals. These factors contribute to the persistent difficulty in robustly quantifying aerosol effects on clouds. Consequently, observational ACI estimates frequently demonstrate significant spatial and temporal constraints, and may not generalise effectively across varied cloud regimes or environmental conditions (McComiskey et al., 2009; Lihavainen et al., 2010; Quaas et al., 2020; Jia et al., 2022)." To present, …

- L92: a definition of cloud water-aerosol mixing ratio should be given here'

L92: Furthermore, "this study introduces the use of the cloud-aerosol mixing ratio to represent how much liquid water is present per unit of aerosol concentration, thereby providing a more realistic indication of aerosol loading within cloud systems." This framework allows for a systematic examination of the transition between Twomey effect-dominated …

- L184: α refers to aerosol parameters such as AOD, PM2.5, or aerosol number concentration (Na) à α refers to proxies for CCN number concentration, such as AOD, aerosol index, sulphate mass concentration, PM2.5, or aerosol number concentration (Na)… many widely used CCN proxies should be metioned.
- L194: cloud number concentration -> cloud droplet number concentration
- L195: is less constrained by LWC -> does not rely on the fixed-LWC assumption

We have integrated this discussion into L194 to improve the logical flow of the manuscript, and we have incorporated the CCN proxies suggested by the reviewer.

~~**L184: Here, Nd is the cloud droplet number concentration, COT is the cloud optical thickness, and …**~~

L194: "Here, N_d is the cloud droplet number concentration, COT is the cloud optical thickness, and LWP is the liquid water path, while α refers to proxies for CCN number concentration, such as AOD, aerosol index (AI), sulphate mass concentration, $PM_{2.5}$, or aerosol number concentration (N_a)." In contrast, McComiskey et al. (2009) indicated that the "cloud droplet number concentration" emphasizes the activation process from aerosol to cloud droplet. This metric, particularly when direct measurements of droplet number are available, "does not rely on the fixed-LWC assumption". While Eq. (3) …

- Section 2.4: The Nd-to-aerosol susceptibility has become more commonly used than the ACI index in recent years (e.g., https://doi.org/10.1038/s41467-018-05028-4, https://doi.org/10.5194/acp-20-15079-2020). I wouldn't suggest removing this part, but perhaps linking ACINd to the Nd-to-aerosol susceptibility would be a good compromise.

Thank you for the reviewer's insightful suggestion. We present the reorganized discussion below.

L199: …into cloud droplets. "In recent years, the calculation formula derived from the ACI index has been increasingly regarded as one of the approaches to quantify the sensitivity of cloud systems to aerosol perturbations (Ma et al., 2018; Liu et al., 2024). In particular, the N_d-to-aerosol susceptibility provides a straightforward representation of the relationship between aerosol variations and changes in cloud droplet number (Quaas et al., 2020), thereby giving the index a clear physical meaning."

- Figure 3(b): what does the 'differential results' mean? Is it the derivative of curve in fig3a? Would be good to have an equation for this.

This is indeed the derivative of the curve shown in Figure 3(a). Our intention is to more clearly illustrate the differences that arise from selecting different time

intervals for the calculation. To achieve this, we computed the values in a discrete manner, and the corresponding equation has been added in the updated Figure 3 (b).

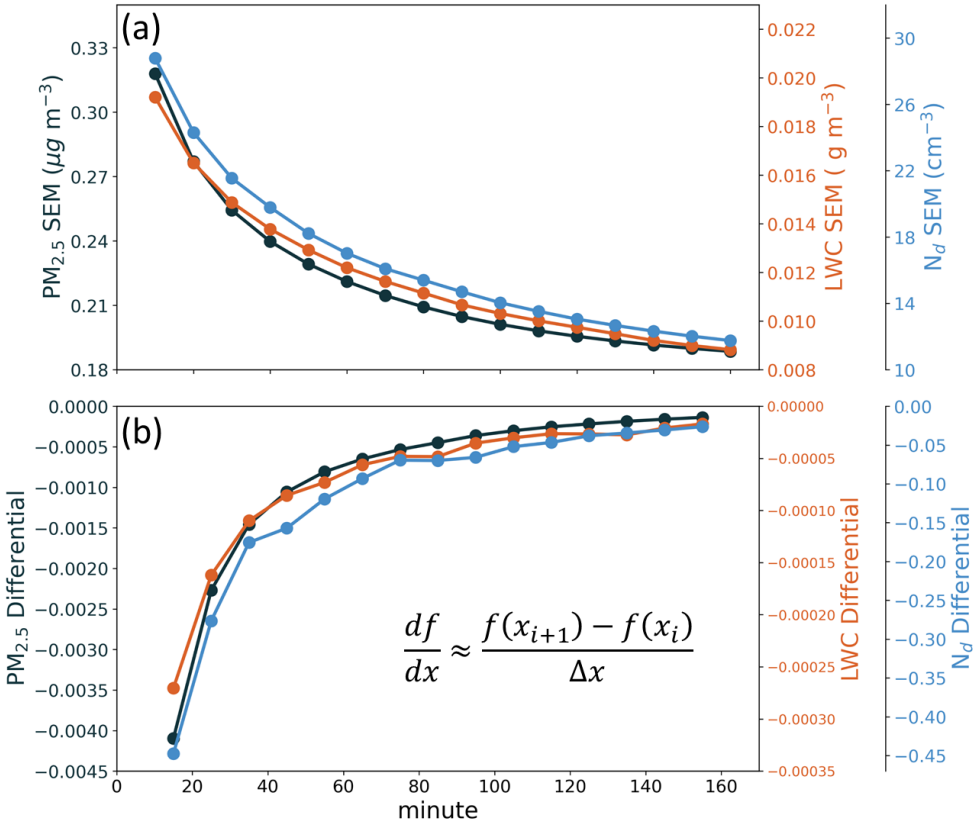

Figure 3: (a) the standard error of the mean for $PM_{2.5}$ (black), LWC (orange), and N_d (blue) at different time resolutions; (b) the differential results of the standard error of the mean.

- Section 2.6: SEM sounds a smart idea to remove the high-frequency signal. Could the authors explain a bit more about its physical meaning? And, the use of 60 minute is nicely justified, but I wonder to what extent the time interval can change the ACI results in section 3.

When the selected time interval is too short, the calculations may be dominated by excessive fluctuations, and the number of data points becomes insufficient to ensure that the derived ACI index is statistically meaningful. Conversely, if the interval is too long, variations in meteorological conditions may overshadow the aerosol-related signals. Therefore, when examining the temporal evolution of aerosol–cloud interactions, our core objective is to identify the shortest reasonable time interval that preserves the physical linkage between aerosols and clouds while ensuring statistical robustness.

The results in Figure A1 clearly demonstrate this behavior. When the averaging interval exceeds 120 minutes (Fig. A1(d)–(f)), most ACI index values cluster around zero, indicating a loss of sensitivity to aerosol–cloud interaction signals. In addition, for some cloud events (Events 3 and 4), a single long averaging window can span nearly the entire event. In such cases, the resulting distributions may reflect differences in meteorological conditions between cloud events rather than aerosol-induced perturbations within an event.

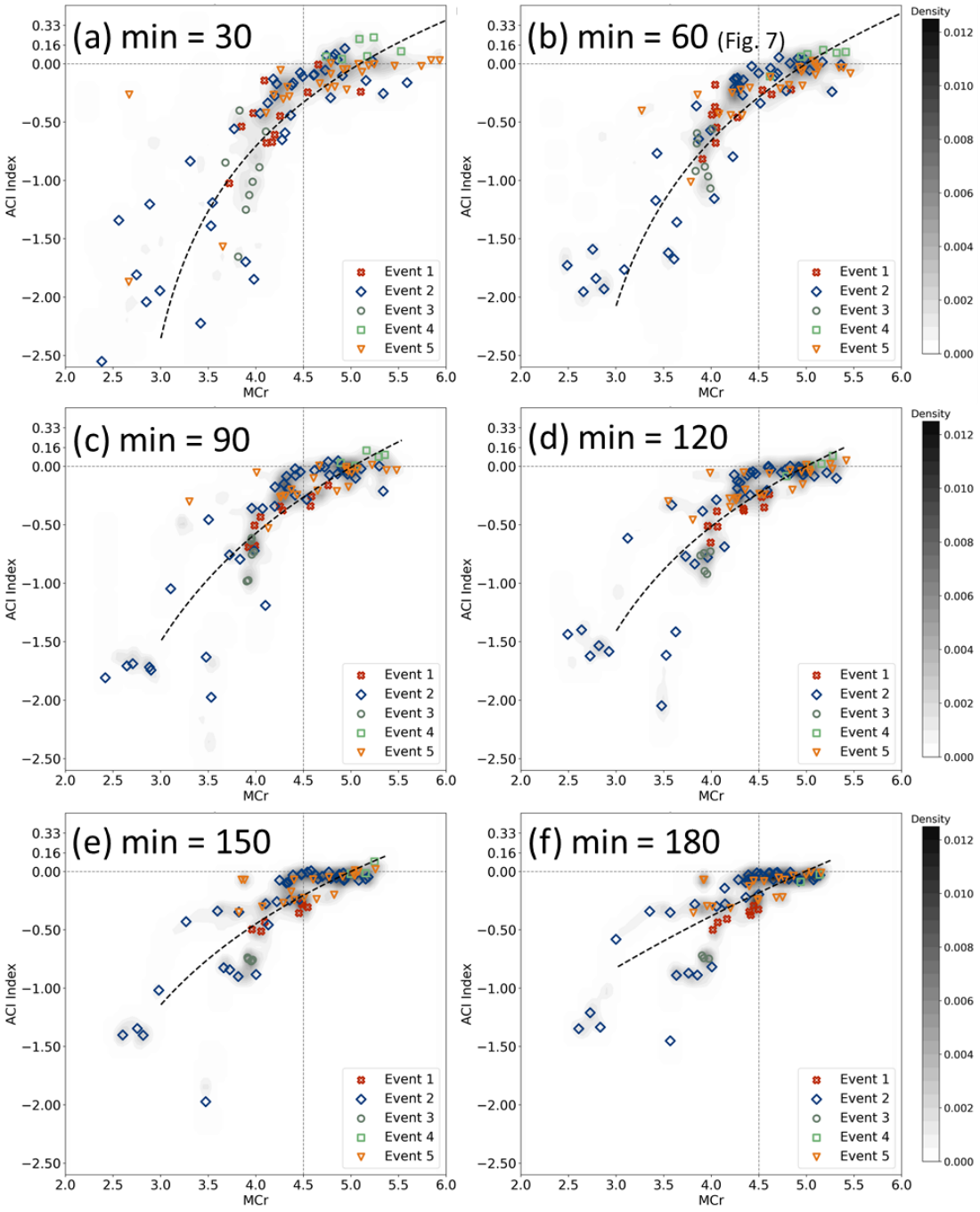

Figure A1: Scatter and density plots of the MCr-ACI index for the continuous

cloud events. The density plot illustrates the overall distribution of all calculated results, while the scatter plot shows the distribution of individual cloud events using different colors and symbols (one point shown for every 30 calculations). (a)-(f) represent different sizes of the sampling time intervals.

- Figure 4 & Table2: I suggest to plot the duration of each event (shown in Table 2) in Fig. 4, making it easier to read L279-284

We thank the reviewer for the suggestion. We have reorganized the discussion and included the corresponding meteorological information at the time of observation in the supplementary (Fig. S2) material to aid readability. In addition, we have rewritten the section to frame the analysis around the evolution of cloud events, allowing a more focused comparison of whether aerosol optical properties are consistent across different cloud events.

In Figure S2(c), it can be seen that not all periods with low visibility and relative humidity near 100% were classified as continuous cloud events. This is because some of these periods were caused solely by precipitation, and sometimes the CDP laser windows were wet or contaminated by rain, resulting in poor-quality observations that were excluded prior to analysis.

L261:

Figure 4 presents the aerosol observations from the LABS aerosol system, with the corresponding meteorological information shown in Fig. S2. In this section, the aerosol optical properties of aerosols are analyzed using PM10 measurements from the aerosol system, whereas PM2.5 observations from the 11-D instrument are primarily used in the other sections. The gray-shaded intervals denote continuous cloud events lasting more than three hours and detected by CDP (Table 2). Based on the absorption and scattering coefficients as well as aerosol mass concentrations (Fig. 4 (a), (b) and Fig. S2 (d)), it is evident that during cloud events accompanied by significant precipitation (Fig. S2 (d)), aerosol concentrations decreased due to washout effect. However, in periods without precipitation signals prior to 10 March, aerosol concentrations clearly increased, indicating continuous long-range transport of aerosols to the LABS region. Overall, higher pollutant levels were observed during the early stage of the cloud event on March 1 (Event 1), both the early and late stages of the event on March 7 (Event 2), and throughout the event on March 9 (Event 3). In contrast, lower concentrations occurred during the precipitation periods on March 1 and March 7, and during the cloud events on March 10 and 11 (Event 4

and Event 5).

Further analysis was conducted using AAE and SAE to evaluate the light absorption and scattering abilities of aerosols at blue and red wavelengths (Fig. 4 (c), (d)). Except for the Event 1, when the mean AAE value was significantly lower than 1 (0.61 ± 0.12), the average AAE during other cloud events was close to 1, suggesting minimal wavelength dependence of aerosol absorption. The SAE remained relatively stable throughout the observation period but showed a slight decrease on March 9 (1.47 ± 0.01), possibly indicating the presence of coarse-mode aerosols related to local sources. The single-scattering albedo (SSA), used to evaluate the overall aerosol absorption capacity (Fig. 4(e)), was generally below 0.9 during most of the observation period except on March 12. Notably, no significant correlation was found between SSA values and aerosol concentrations before and after cloud events, suggesting that despite effects of wet deposition, the proportion of light-absorbing aerosols within the total aerosol population remained relatively consistent. In some cloud events (Events 2 and 5), SSA even decreased during precipitation (Fig. S2 (d)), indicating an increased fraction of absorbing aerosols, likely due to the relatively hydrophobic nature of black carbon (McMeeking et al., 2011; Ohata et al., 2016; Pöhlker et al., 2023), which reduces its susceptibility to wet removal compared to more hygroscopic components.

The overall…

Table 2: the start, end local times and the mean of $PM_{2.5}$, LWC of continuous cloud events (UTC +8).

Cloud Event	Start Time (LT)	End Time (LT)	Average $PM_{2.5}$	Average LWC
Event 1	3/1 09:37	3/1 15:40	7.8 ± 3.1	0.130 ± 0.130
Event 2	3/7 09:16	3/8 07:28	11.4 ± 9.3	0.138 ± 0.126
Event 3	3/9 11:07	3/9 15:27	20.3 ± 4.5	0.163 ± 0.164
Event 4	3/10 02:13	3/10 06:00	2.3 ± 1.6	0.229 ± 0.085
Event 5	3/11 09:47	3/12 05:21	2.5 ± 3.2	0.147 ± 0.121

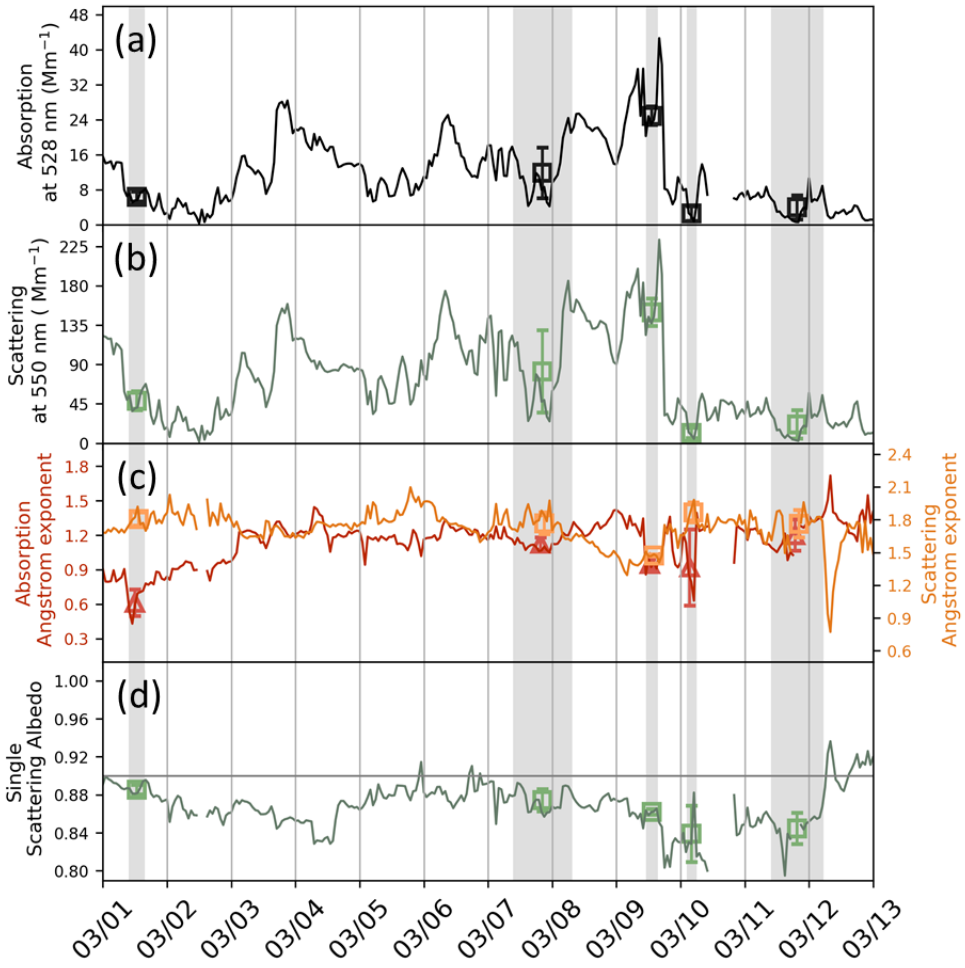

Figure 4: PM_{10} aerosol optical properties observed by the aerosol system at LABS, the gray-shaded intervals represent continuous cloud events. (a) absorption coefficient; (b) scattering coefficient; (c) Absorption Ångström Exponent (AAE, red) and Scattering Ångström Exponent (SAE, orange); (d) Single Scattering Albedo (SSA), and gray dash line means SSA = 0.9.

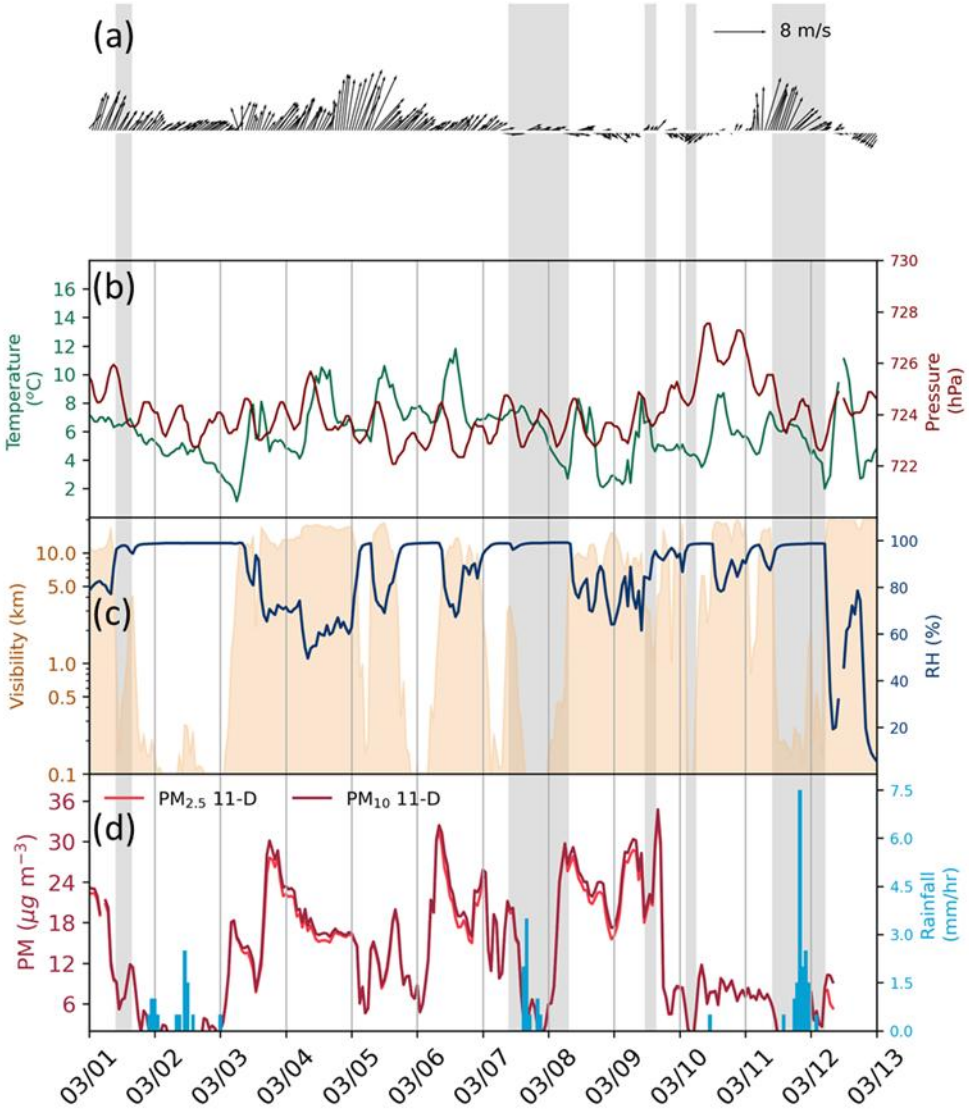

Figure S2: Hourly averages of meteorological parameters and aerosol mass concentrations observed at LABS from March 1 to 12, 2024: (a) wind speed and direction; (b) temperature (green) and pressure (red); (c) visibility (orange) and relative humidity (dark blue); (d) PM_{10} (dark red) and $PM_{2.5}$ (red) measured by the 11-D instrument, along with hourly precipitation (light blue bars).

- L287-289: 'Due to significant…between high and low aerosol loads.' I don't quite see how this statement is relevant here. As I understand it, all aerosol and cloud measurements were made at the same ground level at this mountain site, so vertical co-location shouldn't be an issue in this study.

At LABS, a high-altitude background station, the baseline concentrations of pollutants are substantially lower than those typically observed in urban or suburban environments influenced by anthropogenic emissions, provided that no

significant long-range transport or strong vertical transport mechanisms (e.g., subsidence or valley winds) occur. Moreover, the diurnal variability of pollutants at LABS differs markedly from that within the boundary layer. Therefore, the conventional definition of a "pollution event" is not directly applicable to this site. We agree with the reviewer that the original statement may have been difficult to interpret, and the revised version is presented below for clarity.

L288: … served as the distinguishing criterion. "Because LABS is situated at a high altitude within the free atmosphere, the variability in pollutant concentrations is markedly different from that in the boundary layer dominated by anthropogenic emissions. As a result, conventional air quality indices may not adequately capture the magnitude of aerosol loading observed at the site." Therefore, …

- Figure 6: it's interesting to see the different ACI behaviors in low and high aerosol groups. I wonder if there is significant difference in SSA between two groups, which will help to understand the shift.

Following the reviewer's suggestion, we analyzed the minute-resolution data from the aerosol system at LABS and plotted the results in Figure A2. As shown in Figures A2(a) and (b), the mean differences in both AAE and SAE across different aerosol concentration ranges are minimal. Most data points can be categorized as BC-type aerosols based on the optical characteristics (Schmeisser et al., 2017). It is worth noting that the standard deviation of AAE is larger in the low-concentration range ($PM_{2.5}$ < 11.1 μg m^{-3}), which can be partly attributed to the increased uncertainty of the CLAP instrument under low absorption coefficients and high temporal resolution conditions (Ogren et al., 2017). For the comparison of SSA (Fig. A1(c)), the SSA values remain consistently below 0.9 regardless of aerosol concentration, indicating that the proportion of absorbing aerosols in the environment is relatively stable. In particular, the fraction of absorbing aerosols does not increase under low-concentration conditions, confirming that the consistency of aerosol optical properties does not affect the validity of the analysis in this study. Since the aerosol system at LABS is designed for long-term monitoring, we prefer to use hourly averaged data for better QA/QC reliability. Additionally, the system alternately measures PM_{10} and PM_1 every 30 minutes, meaning that only up to 30 minutes of data are available for each size fraction per hour. This characteristic is also reflected in the sample counts of each group shown in Figure A2.

According to the reviewer's recommendation, we further analyzed the aerosol characteristics inside and outside of cloud events in Sect. 3.1 of the revised manuscript (see precious responses (Fig. 4)). The conclusions are consistent with the results in Figure A2. To keep the subsequent analysis more focused on the variations in cloud microphysical properties, we prefer to retain the original set of variables in Figure 6 of the manuscript.

For clarity, we have revised Figure 6 into six subpanels and modified the x-axis to display $PM_{2.5}$ concentrations. This adjustment more clearly highlights the concentration differences among the groups and improves overall readability.

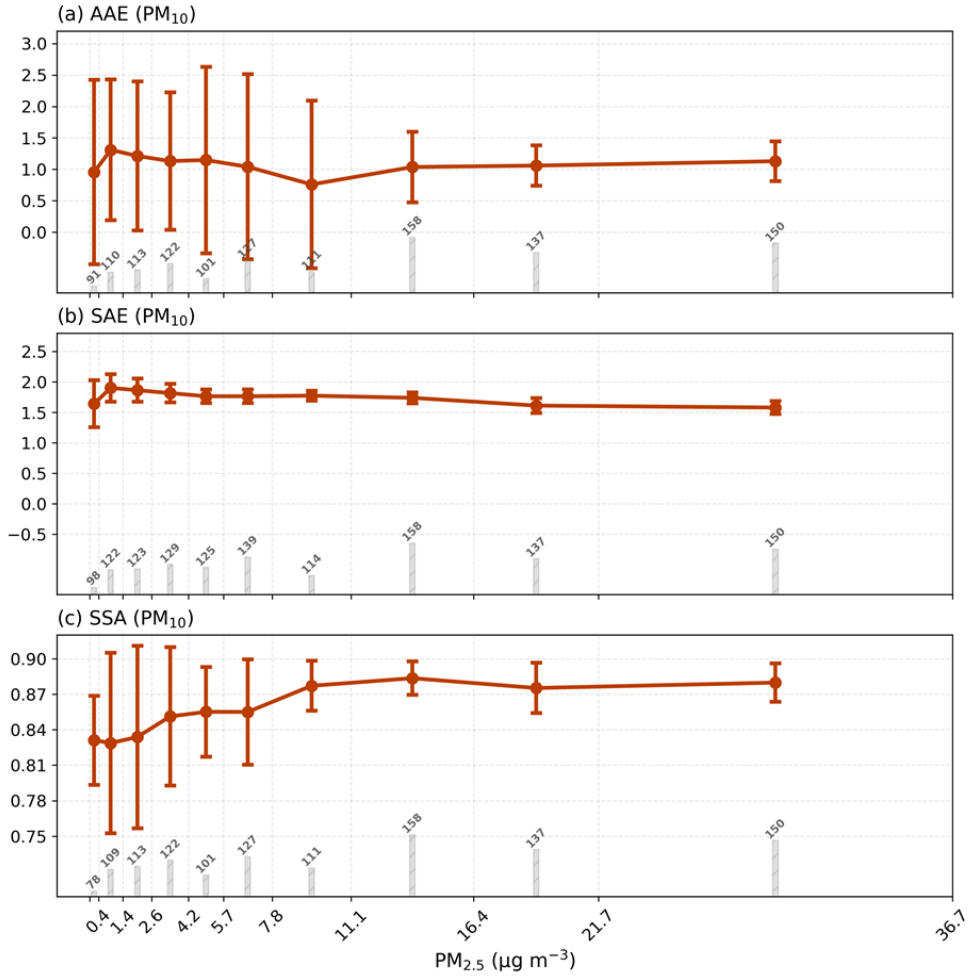

Figure A2: Aerosol optical properties categorized into 10 groups of equal sample size based on $PM_{2.5}$ obtained from 11-D (the data number in each group is shown below as gray bars and values). (a) AAE, (b) SAE, (c) SSA.

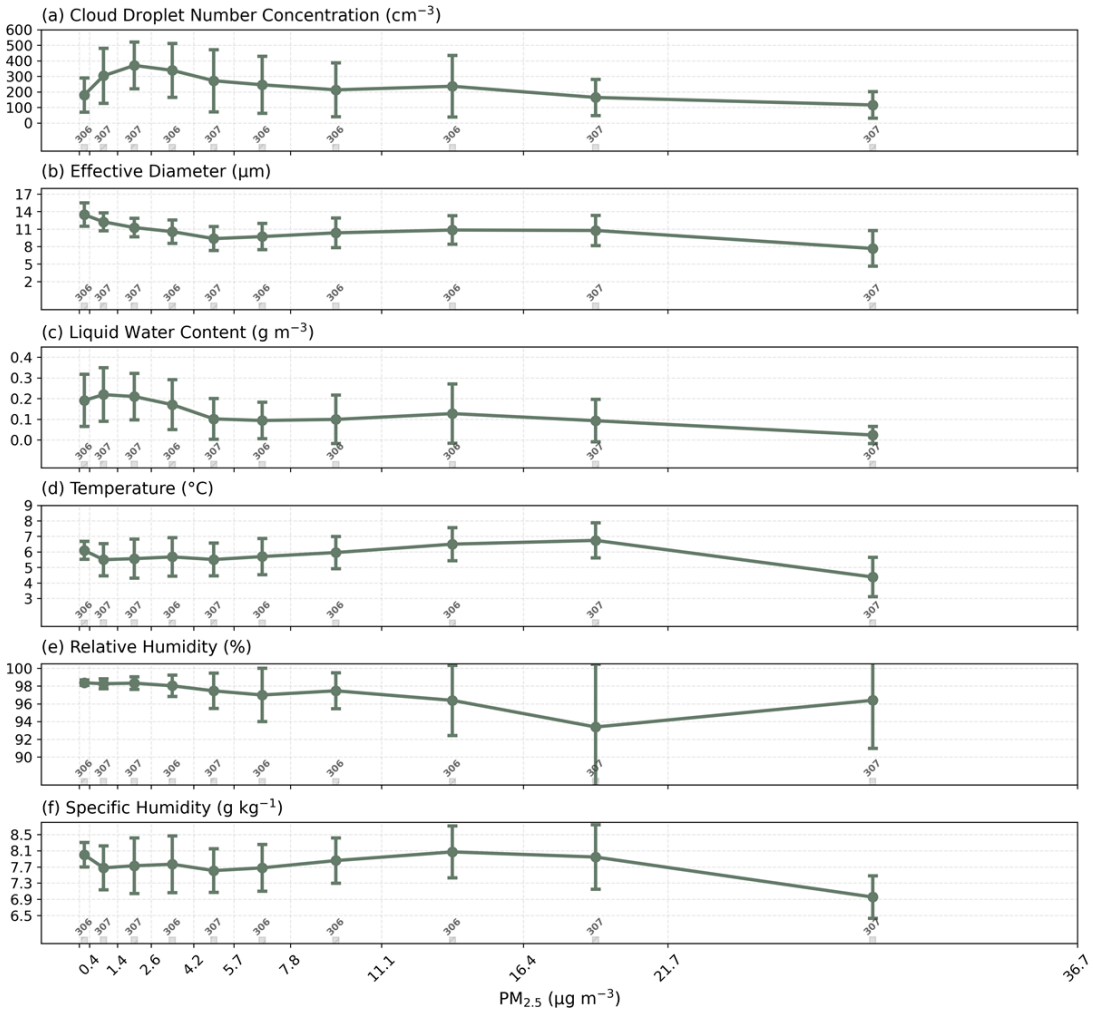

Figure 6: Cloud microphysical and weather data categorized into 10 groups of equal sample size based on $PM_{2.5}$ obtained from 11-D (the data number in each group is shown below as gray bars and values). (a) N_d, (b) ED, (c) LWC from CDP observation results; (d) temperature, (e) relative humidity and (f) specific humidity are weather results measured at LABS.

- L332: why 4.5 not 4.64 - the minimal MCr with ACI index>0?

From our perspective, 4.64 indeed represents the minimum value derived from the analysis. However, from the standpoint of real-world observations, we believe that additional measurements, across different regions, meteorological conditions, and aerosol types, are necessary before the MCr threshold can be more firmly constrained. Defining the value too rigidly at this stage would not strengthen the main message of this study. Moreover, variations in instruments, sampling strategies, and site characteristics across global observatories may

introduce uncertainties, and an overly precise value (with many decimal places) would likely reflect these uncertainties rather than provide meaningful physical insight, thereby distracting from the key findings.

Nevertheless, to demonstrate the robustness of using 4.5 as a representative threshold in our study, we further evaluate and validate this choice through analyzes conducted across different MCr ranges in Sect. 3.3.

- L332-333: This sentence is a bit confusing. could you clarify from what to what the proportion increases from 13% to 30%?
- L337-338: A very confusing part again… 'Although the ACI shifted from negative to positive as aerosol concentrations increased': however, from the figure, it seems that the negative-to-positive shift occurs as MCr increases, not aerosol concentration. Also, when MCr < 2.5, the proportion of aerosol should be lower than that of cloud water, shouldn't it? Please clarify these.

We agree that the descriptions in these two parts were not sufficiently precise in the original manuscript.

1. **The comparison of 13% and 30% refers to the fraction of cases in which the ACI index is positive. Specifically, 13% represents the proportion of all calculated ACI values that are positive, whereas 30% represents the proportion of positive ACI values when only cases with** MCr > 4.5 **are considered. This distinction highlights that positive ACI_{Nd} are predominantly concentrated in the higher MCr regime.**
2. **In this study, MCr inherently accounts for both aerosol loading and liquid water content within the cloud system. Physically, it represents how much liquid water corresponds to each unit of aerosol concentration. Therefore, MCr is not simply a direct comparison between $PM_{2.5}$ and LWC, as the earlier phrasing may have implied.**

To improve clarity, we revised and streamlined the discussion in Section 3.3 and reorganized the structure. We also added Table 2**, which provides the mean and standard deviation of $PM_{2.5}$ and LWC for different cloud events. In the latter part of Section 3.2, we now focus on how variations in $PM_{2.5}$, LWC, and MCr across cloud events correspond to changes in the ACI values, thereby emphasizing the central role of MCr. For a more focused presentation,** Figure 7 **now shows only the MCr-based results, while the NCr results have been moved to the supplementary material (Fig. S3).**

L319-331:

L323: … display a positive ACI index value. "Of these, over 95% (334 out of 351) occurred in regions where MCr exceeded 4.5 (with the minimum MCr value within the reasonable range for ACI index > 0 being 4.64), especially when positive ACI values within the reasonable range (0-0.33, Fig. 7). This finding suggests that under the influence of high aerosol concentrations, cloud systems tend to adjust by reducing the cloud droplet number concentrations most of the time. As shown in Figure S3, the distribution of NCr-ACI reveals a similar tendency to that observed in the MCr-ACI relationship. After constraining aerosol properties and meteorological conditions, negative ACI index can be meaningfully interpreted (as described in Sect. 2.4). However, most of the previous studies have focused only on positive region. It is important to note that negative values do not have a defined "reasonable range", they simply indicate that an increase in aerosol concentration is accompanied by a decrease in cloud droplets number." The physical mechanism …

L332-354:

Comparative analysis among individual cloud events reveals several notable trends (Fig. 7). All events show a consistent relationship between MCr and ACI, where smaller ACI values (more negative) correspond to lower liquid water content per unit aerosol abundance (i.e., smaller MCr). A closer examination of cases with MCr > 4.5 shows that 31% of ACI values (135 data points) are positive in Event 2, and 93% (148 data points) are positive in Event 4. In contrast, although ACI values in Events 1 and 5 tend to increase with MCr, most of them remain negative. These findings suggest that short-term variations in cloud systems are more sensitive to environmental changes induced by absorbing aerosols, where increased aerosol loading can lead to the dissipation of cloud droplets. It is noteworthy that although Event 3 exhibited relatively high mean LWC (the second highest among the five consecutive cloud events), it occurred under high aerosol loading conditions (Table 2). Consequently, the liquid water content per unit aerosol abundance was relatively low (MCr ≈ 4; Fig. 7), and the calculated ACI values remained negative throughout the event (Fig. 7). This result suggests that aerosol-cloud interactions under the influence of absorbing aerosols cannot be interpreted solely based on aerosol concentration. In particular, since the liquid water content varies substantially among real world cloud events, MCr provides a more representative measure of the actual aerosol loading within clouds.

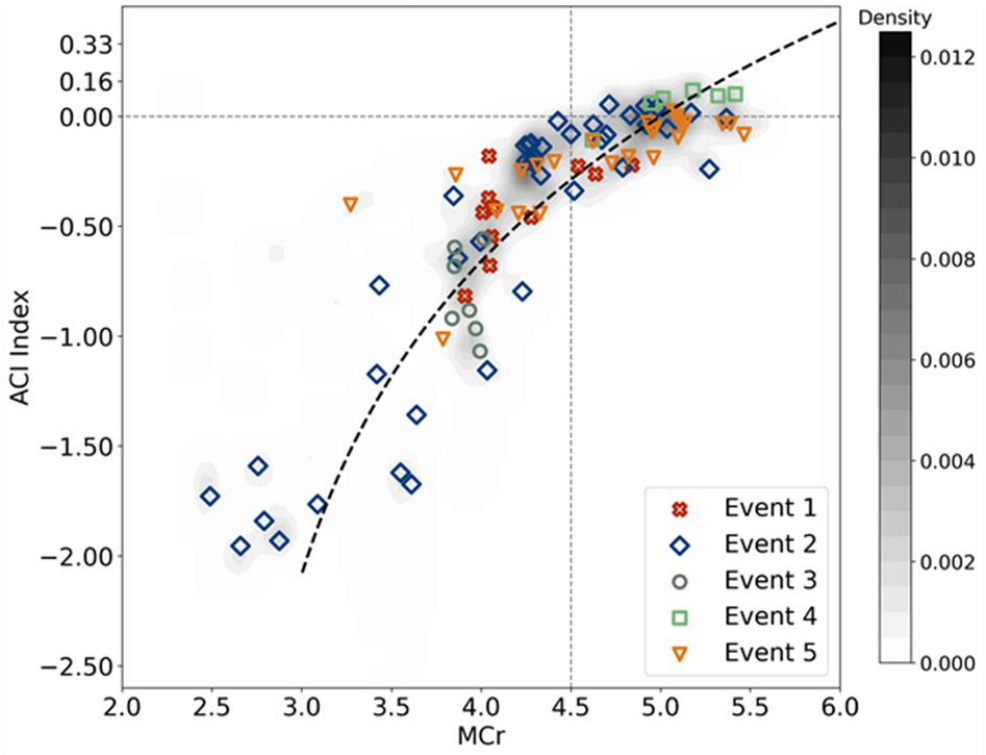

Figure 7: Scatter and density plots of the MCr-ACI index for the continuous cloud events. The density plot illustrates the overall distribution of all calculated results, while the scatter plot shows the distribution of individual cloud events using different colors and symbols (one point shown for every 30 calculations).

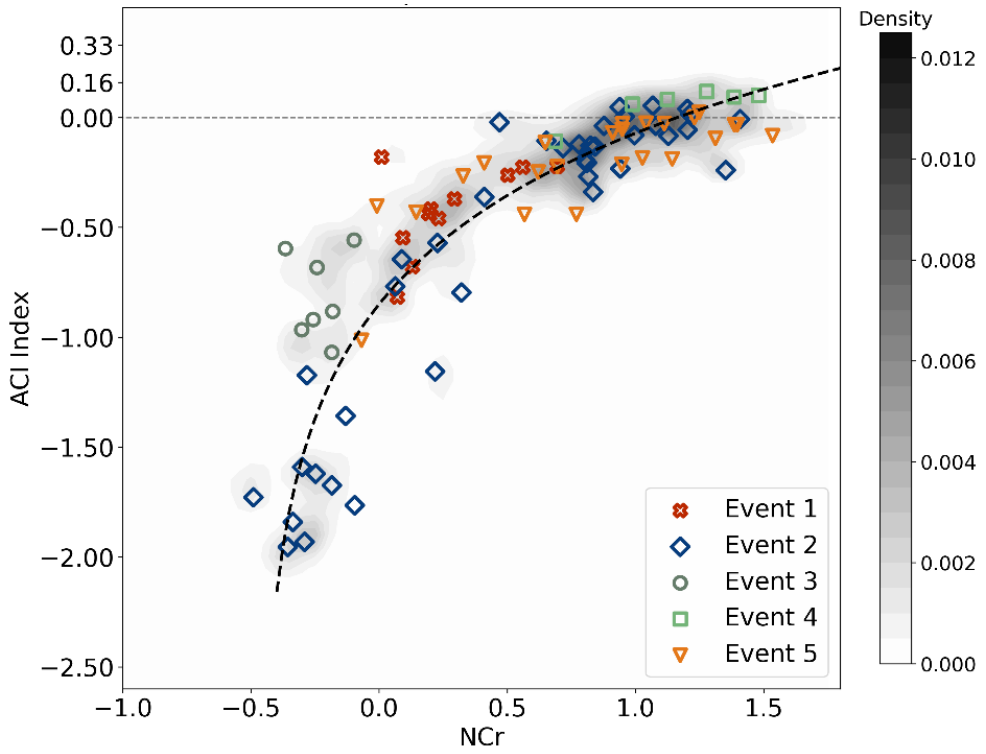

Figure S3: Scatter and density plots of the NCr-ACI index for the continuous cloud events. The settings are identical to those used in Figure 7.

- Figure 7: Including SSA could help explain the role of absorption in evaporation. It might be interesting to make a plot similar to Fig. 7, but with each point colored by its corresponding SSA value.

The single-scattering albedo (SSA) represents the average absorptivity of the aerosol population and therefore provides a good indication of the fraction of purely absorbing aerosols within the total aerosol mixture. However, for evaluating the environmental relevance of the semi-direct effect, we consider that the absolute amount of absorbing aerosols is more important than their relative proportion.

Figure A3 presents the absorption coefficient derived from the CLAP PM_{10} measurements (Fig. A3 (a)) together with the minute-resolution SSA retrieved from the aerosol system (Fig. A3 (b)). Although SSA tends to be lower in the MCr > 4.5 regime (discussion in Fig. A2) the corresponding absorption coefficients remain very small. This is because aerosol concentrations in this regime are intrinsically low during our field campaign. In contrast, for MCr < 4.5, the absorption coefficient is noticeably higher despite the larger SSA values.

Nevertheless, the interpretation of the ACI index cannot rely solely on aerosol loading. The relevant discussion, including how these differences reflect aerosol-cloud interactions under different mixing ratios, has been clarified in the revised Section 3.3.

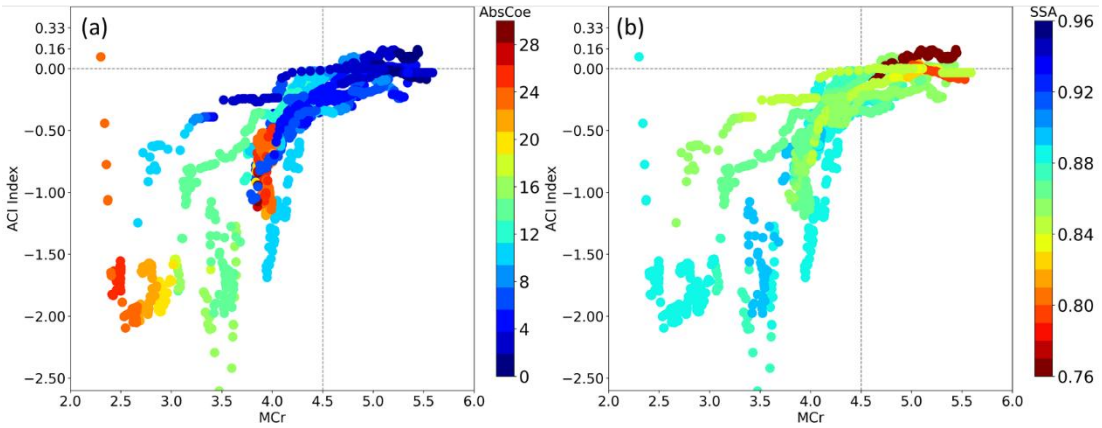

Figure A3: Scatter plots of the MCr-ACI index for the continuous cloud events. The color scale represents (a) the mean CLAP absorption coefficient at 528 nm and (b) the mean single-scattering albedo (SSA).

- L32: ‘cloud optical thickness’ -> cloud optical thickness and thus cloud albedo

L32: **…a greater number of cloud droplets with smaller droplet sizes will form, leading to an increase in cloud optical thickness “and thus cloud albedo”, which …**

- L40: comparing to cloud amount, LWP is even more uncertain and should be mentioned here

L40: …instead enhance evaporation (Chen et al., 2012; Fan et al., 2016; Toll et al., 2019). “This not only indicates large uncertainties in the estimation of LWP but may also lead to misinterpretations in models regarding aerosol-induced variations in LWP and cloud fraction (CF) (Toll et al., 2019; Chen et al., 2022).” It is worth noting that…

- L57: suppress supersaturation - > reduce ambient supersaturation

L57: …absorbing aerosols may “reduce ambient supersaturation levels” …

- L233-234: Not to mention that RH measurements cannot capture supersaturation conditions: I didn’t get this sentence.

The purpose of this statement is to clarify the measurement limitations of the RH sensor. Specifically, the RH instrument cannot detect supersaturated conditions (RH > 100%), and thus the variations that occur beyond saturation cannot be captured by the sensor. Below are the revisions of the sentence.

L233: “Furthermore, conventional relative humidity instruments cannot capture variations under supersaturated conditions (RH > 100%).” While LWC also does not directly quantify supersaturation …

In addition, we refined the textual descriptions in Sections 3.2, 3.4and 3.5 to improve clarity and readability. In Section 3.2, the descriptions of the groups were revised to directly reference $PM_{2.5}$ concentration, making the interpretation more intuitive. In Section 3.4 and 3.5, we reorganized and streamlined theoretical discussions that had been mentioned multiple times to enhance the logical flow and avoid redundancy.

Sect. 3.2:

L292: Figure 6 shows the aerosol loading in the environment, ranging from low to high. "The maximum mean value of N_d (370 cm^{-3}) was observed when the $PM_{2.5}$ concentration ranged between 1.4 and 2.6 μg m^{-3} (Fig. 6 (a)), whereas LWC reached the maximum mean value (0.219 g m^{-3}) within the lower $PM2_{2.5}$ concentration range of 0.4-1.4 μg m^{-3}(Fig. 6 (c)). Notably, when $PM_{2.5} \geq 2.6$ μg m^{-3}, both N_d and LWC exhibited a decreasing trend as environmental aerosol concentrations increased. The variation in effective diameter (ED) can be characterized by a threshold at $PM_{2.5} = 5.7$ μg m^{-3} (Fig. 6 (b)). When $PM_{2.5}$ concentrations were below this value, ED decreased with increasing aerosol loading. However, when $PM_{2.5} \geq 5.7$ μg m^{-3}, ED maintained an average size or slightly increased despite the higher aerosol concentration. A further comparison of weather data across different $PM_{2.5}$ concentration ranges revealed that when $PM_{2.5} \geq 21.7$ μg m^{-3}, the average temperature (4.3°C, Fig. 6 (d)) and specific humidity (7.0 g kg^{-1}, Fig. 6 (f)) reached the lowest values, possibly constrained by specific time and weather conditions. Additionally, relative humidity (Fig. 6 (e)) displayed a decreasing trend with increasing environmental aerosol loading, and the standard deviation of relative humidity increased, reaching the lowest value (93.4%) and the highest standard deviation (7.1%) between $PM_{2.5}$ concentrations of 16.4 and 21.7 μg m^{-3}."

L305: … below 4.2 μg m^{-3}. "Compared with environments where $PM_{2.5} \geq 5.7$ μg m^{-3}, those with extremely low aerosol concentrations ($PM_{2.5} < 2.6$ μg m^{-3}) exhibited noticeably higher LWC, highlighting the substantial differences in cloud liquid water among different aerosol loading conditions. The range of $PM_{2.5}$ between 2.6 and 5.7 μg m^{-3} appeared to represent a transitional phase. In environments with relatively low average aerosol concentrations, higher LWC coexisted with higher N_d and lower ED values, indicating that sufficient water availability allowed more aerosol particles to be activated." It is important to note that … In the high aerosol loading environment "($PM_{2.5} \geq 5.7$ μg m^{-3})," increased aerosol concentrations result in a decrease in N_d and a slight increase in ED …

L460: "In a comparative analysis involving 10 data groups stratified by both $PM_{2.5}$ concentration levels and sample size considerations, a distinct trend emerged: in the low-concentration ($PM_{2.5} < 2.6$ μg m^{-3}), an increase in $PM_{2.5}$ was associated with an increase in N_d and a significant decrease in ED. However, in the high-concentration when $PM_{2.5} \geq 5.7$ μg m^{-3}, further increases in $PM_{2.5}$ corresponded to a decrease in N_d and a slight increase in ED. These results

suggest a transition in cloud microphysical responses under elevated aerosol loading conditions." …

Sect. 3.4:

L389: ~~As observed in Fig. 6, an increase in temperature is also noted in Groups 6-9. The reduction in environmental supersaturation primarily affects smaller cloud droplets, which may fail to sustain growth or even be unable to surpass the Köhler critical point before developing into clouds. Therefore, as aerosol loading transitions from low to high, the microphysical effects of increasing absorbing aerosols on clouds shift from the Twomey effect to semi-direct effect.~~

Sect. 3.5:

L397: …MCr grouping. "For each group, data were further separated into high and low aerosol loading based on the median PM2.5 concentration. In MCr [4.5, 6], high aerosol loading yields a markedly higher frequency of N_d between 300-600 cm^{-3} compared with both the overall observations and the low-aerosol subset (Fig. 10 (c)). In the frequency distribution of ED (Fig. 10 (b)), it was also found that frequencies below 12 μm are significantly higher, which is consistent with the results in Sect. 3.4." In the MCr [3, 4.5] range …

L409: …across these regimes. "However, restricting the LWC range is necessary to reduce uncertainties when assessing aerosol effects on droplet size." Considering weather conditions, Table 3 shows the mean value of temperature, relative humidity, specific humidity in different constraint conditions. "In the high aerosol loading of MCr [3, 4.5]," the temperature…

L420: …20 μm are observed under high aerosol load. "When LWC is restricted (Fig. 12 (a)), the aerosol size distribution remains similar except for fewer particles larger than 2.5 μm." Under high aerosol loading, an increase in cloud droplets smaller than 12 μm is observed, while the larger droplets (> 12 μm) markedly decrease. ~~To further minimize systematic interference and better align with the assumptions of the Twomey effect, applying an LWC constraint is both necessary and appropriate (Fig. 12 (a)).~~ **"Constraint LWC (Fig.12 (a)) more clearly characterizes the Twomey effect, illustrating how aerosol particle concentrations influence the droplet size distribution, "in particular, there are minimal differences between high and low aerosol pollution in terms of weather**

conditions (Table 3).”

L439: …~~dividing point. Combined with the observation of larger coarse-mode aerosol particles in high aerosol loading conditions, this suggests that coarse-mode aerosols can activate and grow into larger droplets under lower supersaturation conditions, but small droplets are more prone to dissipation, consistent with their position on the Köhler curve (Klemm and Lin, 2016). In terms of weather conditions, under constrained LWC, the high aerosol pollution interval in MCr [3, 4.5] has a mean temperature of 6.8°C, relative humidity of 94%, and specific humidity of 8.02 g kg^{-1}. Under low aerosol pollution, the mean temperature is 5.8°C, relative humidity is 98%, and specific humidity is 7.80 g kg^{-1}. Differences in these results agree with the previous discussion (Table 3) in this section.~~

L448: …the interpretation becomes more complex. “Cloud droplet size distributions can be explained by Köhler curve, coarse-mode aerosols under high aerosol loading can more easily activate and grow into larger droplets under lower supersaturation conditions, while smaller droplets are more prone to evaporation (Klemm and Lin, 2016).” Although …

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