Review of “Assessing Nonlinear Responses of Low-Level Warm Clouds Under the Impacts of Absorbing Aerosols Using the Cloud-Aerosol Mixing Ratio” (Manuscript ID: egusphere-2025-3777)

Summary

The research aims to quantify cloud microphysical responses to different mixing ratios of aerosol mass/number to cloud liquid water content using data collected in Taiwan during episodes characterized by absorbing aerosol. The authors address the question: How do cloud microphysical properties respond to a range of mass/number ratios between cloud water and aerosols (MCr/NCr)? The science question is indeed important to address uncertainties in the relationships between aerosols and clouds under different cloud water-aerosol mixing states. The authors introduce a novel quantity (MCr, NCr) and demonstrate its applicability to diagnosing aerosol-cloud relationships, specifically the Twomey and semi-direct effects. The data are relevant for addressing their science question. The figures and tables support the key points of the manuscript but require revision for improved clarity/communication. Some parts of the analysis and writing should be improved as described below. I recommend major revisions prior to consideration of publication.

Major Comments

- MCr = 4.5 as a transition point for Twomey and semi-direct effects: Is this valid only for absorbing aerosols? I think there needs to be a control case where the ACI-MCr analysis is applied to a period characterized by scattering/non-absorbing aerosols (e.g., background, non-biomass burning (BB) air) to demonstrate that the semi-direct effect the authors observed here is strongly linked to the presence of absorbing aerosols. Since identifying the effect of absorbing aerosols is a central goal of the study, I believe a control case would be a clear demonstration that the study’s findings are indeed related to the semi-direct effect.

- Regarding the possibility of “different air mass properties” (L349), the authors must demonstrate that the observed cloud responses are mainly due to the aerosol-cloud mass ratio (MCr) and not shifting air mass sources/aerosol properties over the sampling period (e.g., shift between aerosols from local vs regional, urban vs BB). As an example, Fig 6 shows the observed response of cloud microphysics grouped by PM2.5 loading. But for a causal relationship to be established, Fig 6 implies consistent aerosol source/composition/properties across different PM2.5 loadings, which the authors must demonstrate. The authors must constrain this confounding variable (aerosol composition/properties) to draw more robust conclusions about the relationship between cloud microphysics and PM2.5 loading. Alternatively, the authors can demonstrate that aerosol composition/properties are roughly consistent across PM2.5 loadings. To do this, I suggest plotting PM10 aerosol optical properties from the Lulin Aerosol System grouped by 11-D PM2.5 loading. Although this is not a one-to-one comparison, this may at least suggest that there is some degree of consistency in PM properties vs. loading.

- Washout of BB aerosol in frontal system: Won’t precipitation in frontal system scavenge the biomass burning aerosols? How do the authors know that the aerosols they’re sampling aren’t local (e.g., Kaohsiung)? While Fig 2 show trajectories reaching the Southeast Asian Peninsula, these trajectories do not preclude the possibility of local influence and do not imply that the BB aerosols were unscavenged during the 2-3 days of transport in the warm front. The authors

must either demonstrate that the source of the absorbing aerosol is Southeast Asian biomass burning or rephrase the text on the origin of the absorbing aerosol.

- I strongly recommend adding a conceptual aerosol-cloud figure to visually explain the key findings of the paper, which can be used as a graphical abstract as well.

Minor Comments

- L11-12/L81-86/L106-108: The trajectories from Fig. 2c appear to mainly pass through southeast China and the surrounding waters around Taiwan. The trajectories that do reach the Southeast Asian Peninsula are quite few (only two). The specific source of the absorbing aerosol should not affect the conclusions of the paper, but since Southeast Asian biomass burning was mentioned several times, more evidence that the aerosols originated from biomass burning in that region rather than southeast China is required. For example, the authors could add hotspots over the period of the backtrajectories in Fig. 2c to reinforce claim over the source of absorbing aerosols.

- L80-81: The sentence "Observational studies have…" should be moved to the paragraph before this one for better organization.
- L102: Please mention if the aerosol measurements at LABS have a drying tube for in-cloud periods. Please also specify if the optical properties in Fig 4 refer to dried or ambient aerosols.
- L129: Please specify if the 11-D aerosol measurement during cloud events refers to interstitial aerosol and/or the in-droplet aerosol.
- L237: Please justify the use of PM2.5 over PM10 in the formula for MCr.
- L238: Please clarify what aerosol size range N_a refers to.
- Sect 3.1: It would be helpful to the reader if the authors explicitly stated in this section that the aerosol optical properties refer to PM10 while the rest of the paper refers to PM2.5.
- L260/Sect 3.1: The purpose of Sect 3.1 (PM10 optical properties) needs to be explicitly/more clearly explained in the context of the rest of the paper which uses PM2.5 and focuses only on cloud events. I suggest reframing Sect 3.1 to introduce the cloud events (Table 2) and then describe PM10 aerosol properties before, during, and after each cloud event (Fig 4). This would then flow more smoothly into Sect 3.2. If the LABS optical data is valid in-cloud (e.g., if a dryer was placed upstream of the instruments), it would be informative to include this information as additional columns in Table 2 and added to the discussion in this section.
- L260/Sect 3.1: I recommend adding a meteorological time series to add context to Fig 4 and Sect 3.1 (with similar markings/shadings to show when cloud events occurred). This would help establish a stronger link between the key findings and the physical environment during the sampling period. This meteorological time series could be in the supplementary information.

- L285/Section 3.2: This section is discussed in terms of Group Number. To keep the discussion physically tied to the atmosphere, I suggest that the discussion be based on the PM2.5 concentration ranges (physically based) rather than group number (statistically based). Please rephrase this section so that Group Numbers are replaced by PM2.5 concentration ranges. This will also make the section more succinct. Please apply this change to other sections that mention the PM groups too (e.g., L463).

- L296: Please define ED here or sooner if it's mentioned.

- L318/Sect 3.3: This section requires major revision for clarity, specifically more explicitly connecting the conclusions in the text to the figures so it will be easier for the reader to understand how the data supports the authors' conclusions. See my comments below.
- L336-340: it is not clear in any figure that the PM2.5 > LWC when MCr < 2.5 (since MCr is a ratio, a drop in MCr could be due to changes in LWC and/or PM2.5 and MCr < 2.5 does not necessarily imply that PM2.5 > LWC). It is also not evident from Fig 7 that ACI shifts from negative to positive as aerosol concentrations increase, please provide some x/y values to orient the reader to which part of Fig 7 to refer to. The panels in Fig. 7 are not clearly referenced in the text (i.e., only Fig. 7 is mentioned so it is unclear which panel to refer to while reading the text).
- L344-345: It is not evident from the figures that Event 3 occurred under high aerosol concentrations and low LWC. To add physical context to each cloud event, please add new columns to Table 2 with statistics on cloud microphysical and aerosol properties (e.g., mean+STD PM2.5) for each event. This would help with contextualizing the differences discussed in Sect 3.3 and explaining differences in how each event fills the 2D space in Fig. 7.
- L357: "sufficient liquid water content" please clarify: sufficient for what? For the MCr ratio to be applicable? Or for the Twomey effect to be dominant?
- L358: "division" please clarify what this division is for. If this division/threshold of MCr refers to the point where Twomey or semi-direct effects dominate, please specify this explicitly to make the purpose of Sect 3.4 clearer.
- L375-383: Please state the implication/importance behind why ED does not strongly correlate with ln(PM2.5). Please also justify the use of ln(PM2.5) rather than PM2.5.
- L386-388: Can this conclusion about the droplet size distribution be seen in Fig. 11-12? If so, please mention that to support this conclusion.
- L454/Sect 4: Please describe some suggestions for future analysis based on the main conclusions of the manuscript.
- Fig 4: Please mark each cloud event from Table 2 (e.g., grey areas spanning the start until end of each event). Also mention in caption that daily averages and STDs are also shown.
- Fig 5: please mention the PM size range for these measurements in the caption.
- Fig 6: please mention in the caption how many samples are in each group.
- Fig 6: Because of the uneven width of each group (a consequence of the equal-number groupings), I suggest the x-axis should be PM2.5 concentration rather than group number. This will show the true scale of the x-axis to make it clear that the observed trends are most conclusive for PM2.5 < 10 micrograms m-3 (where most data are). For clarity, one variable can take up one panel (so 6 panels).
- Fig 6: Instead of infinity, Group 10 should list the max PM2.5 concentration recorded during the period.

- Fig 8: Please include a panel to show the number of data points included in the calculation of the y-axis per threshold value (x-axis) to demonstrate that the converging trends mentioned are robust.
- Fig 8b: Please explicitly state in the caption what two quantities the correlation is being calculated for (e.g., ACI and MCr for data where MCr > threshold?). Please apply this to Fig. 9 too perhaps by changing the notation of r(ACI) (e.g., r(ACI, MCr)) for clarity.
- Fig 9: For visual clarity, please use different colors (rather than shades of blue) for different LWC groupings.
- Fig 11: Please double check the coloring of the aerosol size distributions in (a). The observation mean size distribution is not between the low and high aerosol loadings. If my understanding is right (that low/high aerosol loadings are relative to the median aerosol loading), then the low and high aerosol loading groups should be below and above the total observation mean, respectively (similar to Fig. 11b). Please correct me if I am mistaken and add this clarification to the text.
- Table 1: Please list the relevant PM size range measured by the Lulin Aerosol System (PM10 based on Fig 4).
- Table 3: Please add standard deviations. Please also perform a statistical significance test for each meteorological parameter between high and low aerosol loadings. Values that are statistically significant (between low vs. high loadings) can be highlighted in bold in Table 3. This would add support for the conclusions of Sect 3.5.
- Table 3: To demonstrate the semi-direct effect, RH is often mentioned in the paper; however, the instrument has diminishing sensitivity when RH approaches supersaturation (L229-231). The authors must show some evidence that the RH data for RH=[94-98%] (Table 3) is a robust enough measurement so that RH could be used to support their conclusions on the semi-direct effect. Details on the operating range from the RH sensor's manufacturer would be sufficient.