Notes on 'Assessing Nonlinear Responses of Low-Level Warm Clouds Under the Impacts of Absorbing Aerosols Using the Cloud-Aerosol Mixing Ratio' by Lin et al.

This study investigates interactions between clouds and biomass burning aerosols using comprehensive in-situ measurements in a high-frequency (one-minute). They proposed a very interesting metric – cloud-aerosol mixing ratio – to separate signals from the Twomey effect and the semi-direct effect. I think they have nicely demonstrated how the evaporation due to aerosol absorption modulates the relative importance of two effects.

Overall, this is an interesting piece of work, and the proposed metric provides new insights. However, the presentation quality could be improved, and the manuscript feels somewhat longer than necessary. The writing could be more concise, and some interpretations of the key results need clarification (see my detailed comments). As such, I would recommend major revision to address the following concerns.

## **Specific comments:**

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.

## Abstract:

- 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.
- 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 (<a href="https://doi.org/10.1029/2008JD011006">https://doi.org/10.1029/2008JD011006</a>).
- I think it's nice to also mention ACI index under low MCr.

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).

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., <a href="https://doi.org/10.5194/acp-20-15079-2020">https://doi.org/10.5194/acp-20-7353-2022</a>).

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

L184:  $\alpha$  refers to aerosol parameters such as AOD, PM2.5, or aerosol number concentration (Na)  $\rightarrow$   $\alpha$  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.

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.

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.

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.

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

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.

Figure 6: it's interesting to see the different ACI behaviours 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.

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

L332-333: This sentence is a bit confusing. could you clarify from what to what the proportion increases from 13% to 30%?

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.

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.

## **Technical corrections:**

L16: better → better

L32: 'cloud optical thickness' -> cloud optical thickness and thus cloud albedo

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

L57: suppress supersaturation - > reduce ambient supersaturation

L194: cloud number concentration → cloud droplet number concentration

L195: is less constrained by LWC → does not rely on the fixed-LWC assumption L233-234: Not to mention that RH measurements cannot capture supersaturation conditions: I didn't get this sentence.