

Li et al. combine multiple spaceborne A-Train sensors to investigate aerosol–cloud interactions (ACI). They assess the susceptibility of cloud droplet number concentration (Nd) and liquid water path (LWP), derived from MODIS, to aerosol extinction coefficients retrieved at cloud level from CALIPSO. They also examine the relationship between aerosol extinction coefficients and the occurrence of precipitation using CloudSat observations. The effort to relate aerosol properties at cloud level with cloud characteristics on a global scale is scientifically significant. However, the implementation of this study contains several methodological shortcomings that have been extensively documented in previous literature and should be carefully considered when correlating optical aerosol properties with cloud parameters.

In the introduction, the authors appropriately highlight several known limitations of using AOD or AI in ACI studies:

- **Lines 41–43:** This is because AOD (or aerosol index) does not uniquely represent aerosol concentration or CCN concentration, as variations in aerosol composition, particle size distribution, and optical properties can yield the same AOD for different aerosol concentrations.
- **Lines 43–44:** A second limitation is the inability to disentangle the contributions of different aerosol layers to the total AOD, which prevents any meaningful vertical collocation between aerosol and cloud layer.
- **Lines 58–60:** Moreover, enhanced aerosol swelling, cloud contamination, and three-dimensional radiative effects can affect the collocated satellite AOD pixels near cloud edges (Varnai and Marshak, 2009).

Given these acknowledgments and the manuscript's title, one would expect substantive progress in addressing these limitations. However, only the issue of vertical collocation has been considered, following their previous work (Paneimal et al., 2020). Other important sources of uncertainty, such as variations in aerosol type and size distribution and the influence of aerosol hygroscopic growth, are equally relevant for CALIPSO-derived extinction profiles. These factors are either neglected, deemed insignificant without sufficient justification, or, surprisingly, suggested to be not important for future ACI studies in the discussion section.

I have several major concerns, primarily regarding the aerosol and cloud sampling criteria employed in this analysis. These include the inappropriate inclusion of precipitating clouds in the computation of Nd susceptibility, the use of aerosol properties from highly humid regions adjacent to clouds, the restriction to broken-cloud 25 km X 25 km scenes for estimating LWP susceptibility, and the fine spatial aggregation applied in the analysis. Each

of these issues could significantly affect the derived sensitivities and should be carefully revisited. Addressing these points is essential for the manuscript to substantiate its claim of advancing the assessment of aerosol–cloud interactions.

Major Comments:

1. **Line 145:** The authors limit the 25 by 25 km cloud fraction (CF) to 90% to exclude cases where aerosols are fully embedded within cloudy regions, on the premise that such situations are affected by aerosol swelling due to hygroscopic growth at high relative humidity (RH). However, this filtering does not adequately ensure that hygroscopic growth is properly accounted for. Aerosol retrievals in direct contact with cloudy pixels (likely cloud-contaminated pixels) can still be significantly influenced by hygroscopic growth effects, irrespective of CF. As demonstrated in Christensen et al. (2017), this can lead to artificially enhanced correlations between Nd and AOD or AI. Since the cloud-level aerosol extinction coefficients are considered in the present manuscript, where the RH effect is likely significant, the derived susceptibilities may be biased.

I recommend redoing the calculations after omitting aerosol retrievals in pixels directly adjacent to cloudy columns irrespective of total CF. This will also address another issue in computing $d\ln LWP/d\ln Nd$ (see next paragraph). This approach has been adopted in several recent ACI studies using satellite-derived AI to estimate Nd susceptibility (e.g., Jia et al., 2022). Alternatively, aerosol retrievals can be filtered using an RH threshold (e.g., only including retrievals where $RH < 70\text{--}80\%$), within which hygroscopic growth is limited for both continental and marine aerosol types. RH values can be obtained from the operational CALIPSO product (which includes interpolated meteorological parameters) or directly from reanalysis datasets such as ERA5 or MERRA-2. This is a fundamental consideration in satellite-based ACI studies and should not be overlooked, particularly in a study aiming to advance current estimates of Nd susceptibility.

Furthermore, the decision to omit cloud retrievals with $CF > 90\%$ (within 25×25 km scenes) when computing $d\ln LWP/d\ln Nd$ is not justified. Both LWP and Nd are derived from MODIS cloud retrievals, which tend to be more reliable in overcast cloud fields due to their higher spatial homogeneity. Such conditions better satisfy the plane-parallel cloud approximation, and consequently, three-dimensional radiative effects are minimized (Zhang and Platnick, 2011). I recommend removing the CF filtering from Nd-LWP susceptibility calculations.

2. **Lines 236–237:** The authors state, “Indeed, global ACI for non-precipitating ($Z_{\max} < -15$ dBZ) and precipitating ($Z_{\max} > -15$ dBZ) segments is 0.13 and 0.08, respectively.” It is unclear how this information can be inferred from Fig. 5. I assume that the authors averaged the ACI indices over grid points with the minimum or maximum probability of precipitation (POP). If this interpretation is correct, further clarification is necessary on how this separation was implemented and statistically represented in the figure. Based on this assumption, I have an additional related comment below.
3. Another fundamental issue not addressed in this study is the inclusion of precipitating clouds in the calculation of the ACI index or Nd susceptibility, which leads to two key issues. First, precipitating clouds introduce significant uncertainty in Nd retrievals, as the assumption of adiabaticity no longer holds. Second, collision-coalescence reduces Nd independent of aerosol loading, thereby distorting the aerosol–cloud relationship. The inclusion of precipitating scenes can lead to a non-causal positive bias in Nd susceptibility of approximately 21% (Jia et al., 2022). Since the authors already utilize CloudSat observations to identify precipitating clouds, it would be straightforward to exclude precipitating clouds from the analysis and recompute Nd susceptibility accordingly.
4. Since the authors use LWP and Nd from MODIS following a similar approach to previous studies (e.g., Gryspeerdt et al., 2019), the primary differences between their results and those in the literature appear to stem from the finer aggregation scale ($25\text{ km} \times 25\text{ km}$ instead of $100\text{ km} \times 100\text{ km}$) and the exclusion of pixels with $\text{CF} > 90\%$. One concern here is the use of such a fine grid size. A $25\text{ km} \times 25\text{ km}$ domain may not be sufficiently large to capture the structural or morphological variability within cloud systems over oceans. While cloud-top Nd tends to be relatively homogeneous in non-precipitating clouds, as it is primarily governed by the initially activated CCN population, the situation is different for LWP. Within a cloud, LWP typically peaks in the core regions and decreases toward the periphery, leading to substantial intra-cloud heterogeneity. This variability becomes even more pronounced in precipitating clouds. So, for similar Nd, we can have two different LWP, because of the cloud morphology, not directly because of aerosols. It is unclear how these in-cloud variations are accounted for in the current analysis, and clarification on this point is necessary to assess the robustness of the derived susceptibilities.

5. **Line 319:** The authors state that “future analyses should be framed in terms of the ambient aerosol extinction coefficient.” It is unclear how this recommendation is justified, given that aerosol hygroscopic growth is known to bias Nd susceptibility estimates. Numerous previous studies have recognized and explicitly accounted for this effect (e.g., Christensen et al., 2017; Hasekamp et al., 2019; Jia et al., 2022; Quaas et al., 2020). The authors should clarify the rationale behind this suggestion.

Minor comments:

6. Line 26: “Observational estimates ...” instead of “Estimates”?
7. Lines 48-49: Do you mean the “updraft limited regime” (Reutter et al., 2009)?
8. Line 64: Citing the authors: “Regrettably, the application of spaceborne lidar observations to the ACI computation is still surprisingly lacking.” This is not entirely true. Alexandri et al. (2024) combined CALIPSO-derived CCN concentrations with Nd from geostationary observations in a sophisticated cloud-by-cloud framework using an advanced cloud tracking and matching algorithm.
9. Line 106: Which wavelength was used for the effective radius and why? Did the authors apply the condensation rate temperature correction based on Gryspeerdt et al. (2019) when calculating Nd?
10. Which correlation coefficient is shown in Figures 2 and 3? Please mention it in the caption. I recommend the Pearson’s correlation coefficient. If the authors prefer Spearman, please provide the figures with Pearson’s correlation coefficient in the supplementary.
11. Figure 4: How do the authors interpret negative $d\ln Nd/d\ln EXT$?
12. Line 262: $d\ln LWP/d\ln Nd$ is also affected by sampling bias due to missing cloud properties in MODIS as a result of retrieval failure, particularly the positive $d\ln LWP/d\ln Nd$ response (Choudhury and Goren, 2025).
13. I suggest the authors provide a supplementary figure showing $d\ln Nd/d\ln(EXT_{surface})$ and $d\ln Nd/d\ln(AOD)$?
14. A general observation from Figures 4 and 9 is low or negative ACI index over pristine oceans. Can the authors comment on why this could happen in both CALIPSO and MODIS retrievals?

References:

Alexandri, F., Müller, F., Choudhury, G., Achtert, P., Seelig, T., and Tesche, M.: A cloud-by-cloud approach for studying aerosol–cloud interaction in satellite observations, *Atmos. Meas. Tech.*, 17, 1739–1757, <https://doi.org/10.5194/amt-17-1739-2024>, 2024.

Christensen, M. W., Neubauer, D., Poulsen, C. A., Thomas, G. E., McGarragh, G. R., Povey, A. C., Proud, S. R., and Grainger, R. G.: Unveiling aerosol–cloud interactions – Part 1: Cloud contamination in satellite products enhances the aerosol indirect forcing estimate, *Atmos. Chem. Phys.*, 17, 13151–13164, <https://doi.org/10.5194/acp-17-13151-2017>, 2017.

Choudhury, G., & Goren, T. (2025). Sampling bias from satellite retrieval failures of cloud properties and its implications for aerosol–cloud interactions. *Geophysical Research Letters*, 52, e2025GL115429. <https://doi.org/10.1029/2025GL115429>

Gryspeerdt, E., Goren, T., Sourdeval, O., Quaas, J., Mülmenstädt, J., Dipu, S., Unglaub, C., Gettelman, A., and Christensen, M.: Constraining the aerosol influence on cloud liquid water path, *Atmos. Chem. Phys.*, 19, 5331–5347, <https://doi.org/10.5194/acp-19-5331-2019>, 2019.

Painemal, D., Chang, F.-L., Ferrare, R., Burton, S., Li, Z., Smith Jr., W. L., Minnis, P., Feng, Y., and Clayton, M.: Reducing uncertainties in satellite estimates of aerosol–cloud interactions over the subtropical ocean by integrating vertically resolved aerosol observations, *Atmos. Chem. Phys.*, 20, 7167–7177, doi:10.5194/acp-20-7167-2020, 2020.

Hasekamp, O.P., Gryspeerdt, E. & Quaas, J. Analysis of polarimetric satellite measurements suggests stronger cooling due to aerosol–cloud interactions. *Nat Commun* 10, 5405 (2019). <https://doi.org/10.1038/s41467-019-13372-2>

Jia, H., Quaas, J., Gryspeerdt, E., Böhm, C., and Sourdeval, O.: Addressing the difficulties in quantifying droplet number response to aerosol from satellite observations, *Atmos. Chem. Phys.*, 22, 7353–7372, <https://doi.org/10.5194/acp-22-7353-2022>, 2022.

Quaas, J., Arola, A., Cairns, B., Christensen, M., Deneke, H., Ekman, A. M. L., Feingold, G., Fridlind, A., Gryspeerdt, E., Hasekamp, O., Li, Z., Lipponen, A., Ma, P.-L., Mülmenstädt, J., Nenes, A., Penner, J. E., Rosenfeld, D., Schrödner, R., Sinclair, K., Sourdeval, O., Stier, P., Tesche, M., van Dierenhoven, B., and Wendisch, M.: Constraining the Twomey effect from satellite observations: issues and perspectives, *Atmos. Chem. Phys.*, 20, 15079–15099, <https://doi.org/10.5194/acp-20-15079-2020>, 2020.

Reutter, P., Su, H., Trentmann, J., Simmel, M., Rose, D., Gunthe, S. S., Wernli, H., Andreae, M. O., and Pöschl, U.: Aerosol- and updraft-limited regimes of cloud droplet formation: influence of particle number, size and hygroscopicity on the activation of cloud condensation nuclei (CCN), *Atmos. Chem. Phys.*, 9, 7067–7080, <https://doi.org/10.5194/acp-9-7067-2009>, 2009.

Zhang, Z., and S. Platnick (2011), An assessment of differences between cloud effective particle radius retrievals for marine water clouds from three MODIS spectral bands, *J. Geophys. Res.*, 116, D20215, doi:10.1029/2011JD016216.