Reply to Reviewer #2

We appreciate the scientific insight of the reviewer's comments and suggestions. His/her report helped clarify sections of the manuscript and put our analysis in the context of other relevant studies in the field of aerosol-cloud interactions and radiative forcing. His/her specific comments are addressed below (highlighted in blue).

I feel many important references were not mentioned in this study. Most points discussed in the Introduction have been already well-documented in previous review papers, e.g., https://doi.org/10.5194/acp-20-15079-2020 and more recent https://doi.org/10.1029/2022RG000799 and the references therein. Would be nice to acknowledge previous work.

We thank the reviewer for suggesting these relevant studies. Both review articles, Rosenfeld et al. and Quaas et al., are now properly cited in the introduction. Quaas et al. is of particular interest, as the article nicely summarizes the challenges in the quantification of ACI and the importance of applying observational constraints to improve current estimates of radiative forcing.

For the effect of retrieval bias, I don't think (Varnai and Marshak, 2009) really touched the effect on ACI, instead more about AOD error. A detailed investigation can refer to https://doi.org/10.5194/acp-22-7353-2022, which may be more relevant here. Regarding the aerosol retrieval issue in low aerosol conditions, a reference should be provided (see the discussion about this in above two review papers).

Correct, Varnai and Marshak did not specifically address the effect of aerosol biases on ACI. We appreciate the reference suggested by Referee 2. In the revised version, we will be adding the following sentence. "Analysis of passive satellite aerosol and cloud retrievals reveal that biases in AOD can yield underestimations of the N_d-AOD regression of at least 3% (Jia et al., 2022)".

We agree with the reviewer about limitations for regions with low aerosol loading. We will provide additional discussion in section 4.

Figure. 2: Similar plots but showing sample size would be help here. Also, it's interesting to see negative signals in some regions, particularly in (b) this seems to be more visible in regions with strong precipitation. Any explanation about this? As I can understand these plots were for all clouds; would be interesting to look at non-precipitating clouds only. Similarly, for L233-237: The easiest way to investigate the effect of precipitation on ACI is making the similar plots as Fig. 4 but distinguishing non-precipitating and precipitating clouds

Samples size figures are now included in the manuscript (Fig. R1). In addition, we follow the reviewer's suggestion of separating the analysis into precipitating and non-precipitating samples (also recommended by Reviewer1). To that end, we used the probability of precipitation (POP) defined as the fraction of precipitation CloudSat pixels (reflectivity>-15dBZ) relative to the total cloudy pixels along the 25 km segment. Because precipitation is somewhat patchy in boundary layer clouds, we select a POP>0.3 for defining precipitating scenes. Conversely, non-precipitating segments are defined as having POP <0.05 (Fig. R2). Generally speaking, ACI for precipitating samples decreases, primarily encompassing regions over the open ocean in the subtropics and midlatitudes. We also note that ACI substantially decrease south of Australia, where

ACI is statistically zero. In contrast the same coastal region exhibits ACI up to 0.25 for non-precipitating samples.

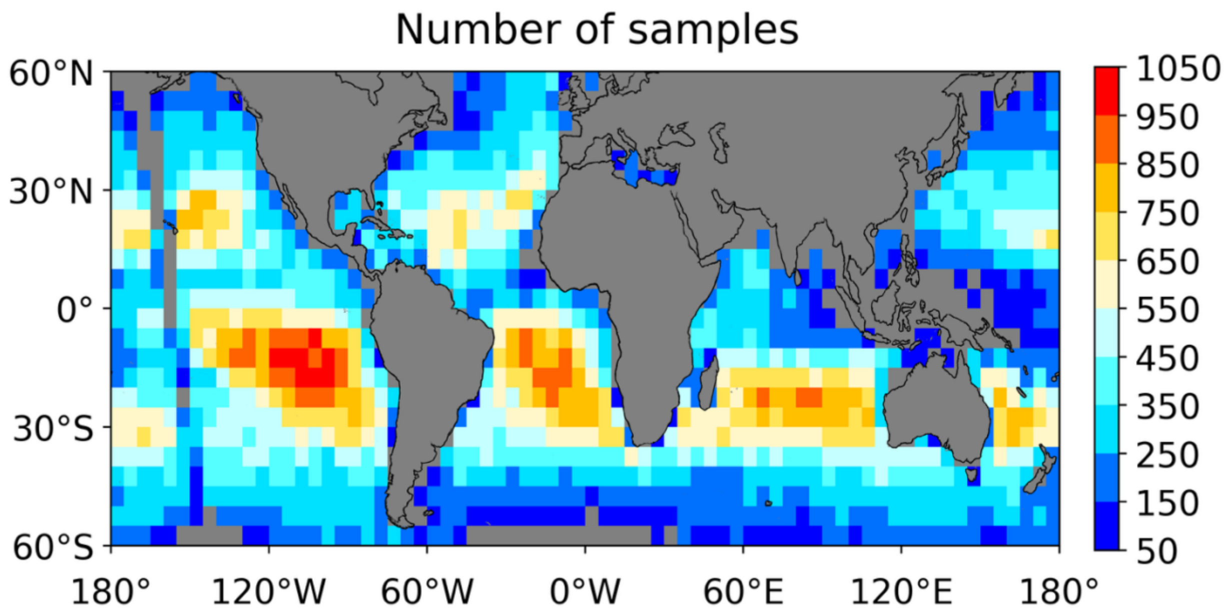


Figure R1: Number of 25-km segments used for the derivation of the ACI index.

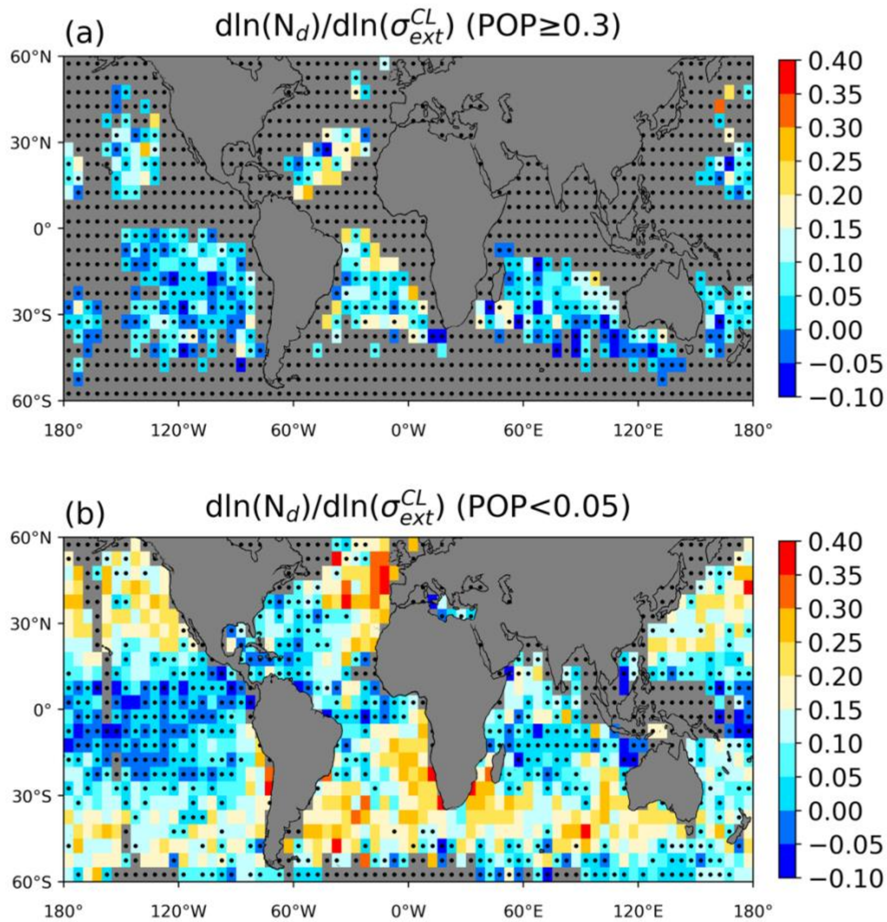


Figure R2: ACI index map. Black dots indicate grids that are undistinguishable from zero according to a student's t test at 95% confidence level. a) Grids with probability of precipitation (POP) > 0.3, b) Grids with POP<0.05.

L213: maybe an explanation on Spearman correlation would help. It's confusing that the data in Fig. 3a apparently are not concentrated around the regression line, but r_s are mostly larger than 0.95 and even being 1. Please clarify.

The Spearman correlation is less affected by outliers and primarily captures the monotonic increase of the relationship. In the revised manuscript, we will report both Spearman and Pearson correlations.

L216-219 (also the argument on 'S-shape' in abstract): I'm not sure how much I can be convinced by this statement.

- I feel the reason why we didn't see a clear 'flat curve' in high σ is the insufficient samples there; binning data into same sample-size bins induces a weak representativity where data are sparse. Even with sparse data, we still see the saturated Nd when σ starts going beyond 0.2-0.3. Thus, the analysis here is not sufficient to demonstrate the S-shape is non-physical.

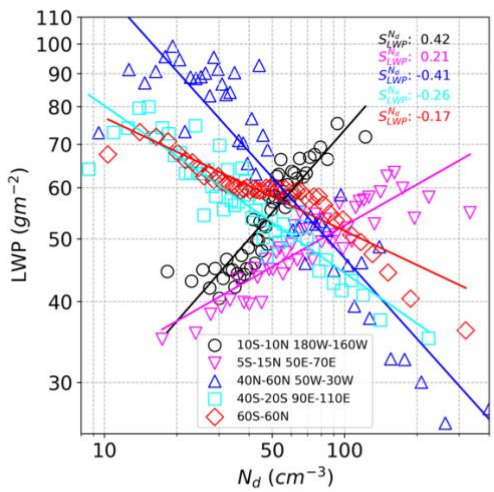
-Even using boundary-layer SO4 (closer to σ here), the sigmoid shape is still quite clear (Fig. 1b in https://doi.org/10.5194/acp-23-4115-2023). A recent study further provided the observational evidence for this sigmoid curve based on long-term trends (https://doi.org/10.1038/s41558-023-01775-5). These should be discussed.

- I’d suggest formulating it in a way that the non-lineaer behavior reported in earlier studies tends to be less pronounce when using cloud-base extinction than column AOD, instead of saying it’s non-physical as the results presented cannot justify this strong statement.

The way the binning is conducted could certainly change the shape of the curve. We did try multiple bin sizes and the results remained unchanged. With the available dataset, the binning in n-tiles is the appropriate approach to faithfully represent the sampling distribution and reduce the effect of outliers. As suggested by the reviewer, it is pertinent to cite Jia and Quaas (2023) in the discussion section, however, we do not have a hypothesis for why S-shape appears to be present when applied to sulfate. In general, relationships between Nd and other aerosol proxies do not necessarily capture the same physical information, processes, or biases; and, therefore, our conclusions are only valid for optical aerosol properties derived from satellites. However, the contrast between the sigmoid curve for AOD and a semi-linear relationship for aerosol extinction is quite evident, and the saturation of the curve for high AOD is not observed for aerosol extinction coefficient. However, we will revise the text to convey the idea that the non-linearity is less pronounced when vertically-resolved aerosol extinction coefficient is used in the analysis.

L260-261: This is an interesting point. The authors could demonstrate this even clearer by making a panel (b) for Fig 7 but lumping all global data together.

The updated figure is included below, with red diamond representing global data, and will be discussed in the revised version.



Minor comment:

Since only the vertical co-location of cloud and aerosol layers is studied, the term ‘Progress’ in the title seems too broad and gives the impression of a review-like paper. I suggest removing it.

Following the reviewer’s suggestion, we modify the title to: “Advancing the quantification of aerosol-cloud interactions with the use of the CALIPSO-CloudSat-Aqua/MODIS record”

L26: Estimates -> Observational estimates

Corrected, thanks

This work largely follows Painemal et al. (2020). The importance of using vertically collocated aerosol has been already well-justified. Would be good to explain what new message one could get beyond the existing literature.

Unlike Painemal et al. (2020), the global quantification of ACI and susceptibilities is a key contribution of our manuscript. Also, the integrated study of cloud susceptibilities and ACI using MODIS-CALIOP-and CloudSat is also another contribution. We will be highlighting these points in the revised manuscript.

L68: 'shortcomings': Since the text so far only highlights the benefits, it might be helpful to flag what shortcomings the readers can expect next.

Correct, we are going to discuss some outstanding issues, primarily associated with the fact that optical properties differ from aerosol concentration.

' metrics of cloud susceptibilities of ACI' and 'cloud susceptibility' are the same, aren't they?
Correct, we modified the sentence to read: "metrics of ACI and cloud susceptibility"

L80-82: this sentence is hard to read. Please explain what 'This choice of CALIPSO-based dataset responds to limitations of the standard CALIPSO product' means

In short, our research version of the CALIPSO retrievals are estimated by solving the lidar equation, using an iterative process that finds both the extinction coefficient and lidar ratio that matches an independent AOD, derived from the SODA algorithm. This eliminates the need of classifying aerosols into specific aerosol types and assuming a constant lidar ratio. Because the lidar ratio is highly variable, assuming a constant lidar ratio introduces significant uncertainties in the CALIPSO aerosol product. Painemal et al. (2019) show that that this new aerosol extinction coefficient better compares with airborne observations from a high spectral resolution lidar. In the revised version, we are going to explicitly explain the advantages of our dataset over the standard CALIPSO products.

Reference:

Painemal, D., Clayton, M., Ferrare, R., Burton, S., Josset, D., and Vaughan, M.: Novel aerosol extinction coefficients and lidar ratios over the ocean from CALIPSO–CloudSat: evaluation and global statistics, Atmos. Meas. Tech., 12, 2201–2217, https://doi.org/10.5194/amt-12-2201-2019, 2019.

L93: I personally think CTH is a better term than ZT for cloud top height, which has been widely used. Would be easier for readers

We generally use a simple notation instead acronymous because the variables are easier to express and understand in equations.

L99: 'height': is it cloud top height?
Correct, all the variables in the sentence are for clouds.

Eq 1: Though the authors referred to (Albertcht et., 1990), it's good to provide the full formulation here along with all parameter values need in the calculation so that people can easily follow.

The formulation is a bit more complicated than a single formula solely depending on temperature and pressure. Because the expression has been utilized in a number of studies, we refer the reader to Albrecht et al.

112-113: How it can categorize the low-cloud precipitation rate is not clear. I guess the authors put a 'raining' flag if Zmax>-15, otherwise 'non-raining', right?

We appreciate the reviewer's comment. Yes, we identify precipitation at the CLoudSat pixel level as Zmax>-15dBZ. The categorization is not discussed in the later sections, thus we removed the sentence "The impact of additional precipitation categorization (drizzle: -15< Zmax<=-7), light rain: -7< Zmax<=0, and rain: Zmax>0) is discussed in Section 4."

L124: Could you clarify what you mean by "the closest CloudSat CPR pixels to the 25-km line"? What exactly does the 25 km line refer to here? L134-136: it's a bit unclear if the cloud-top height is from MODIS or CALIOP as stated earlier? To match and 1x1 modis pixels I'd assume it's from MODIS right?

It refers to the closest pixels (and ground-track) to the CALIPSO ground-track, represented by the 25 km segment in Figure 1 (in blue). The derivation of cloud layer height for computing aerosol extinction coefficient is from CALIOP. Filtering of MODIS pixels are based on MODIS cloud top height. We are going to clarify this in the revised manuscript.

L140: the threshold is generally set to 4; could you explain why 2 is used here? Does it mean more optically thin clouds are included in this study?

The rationale is based on the Painemal et al. (2025). Using airborne polarimetric retrievals, Painemal et al (2025) show that retrievals tend to be more robust for optical depth greater than 2, especially if the satellite data correspond to a cloudy scene (high cloud coverage).

Reference:
Painemal, D., Smith, W. L. Jr., Gupta, S., Moore, R., Cairns, B., McFarquhar, G. M., & O'Brien, J. (2025). Can we rely on satellite visible/infrared microphysical retrievals of boundary layer clouds in partially cloudy scenes? implications for climate research. *Geophysical Research Letters*, 52, e2024GL113825. **https://doi.org/10.1029/2024GL113825**

Eq3: Only data with CF>80% are analyzed. In this case, S_CF cannot reflect the real effect. would be nice to mention this limitation here though it appears quite later in the paper

Correct. We will emphasize the fact that the changes in cloud coverage due to aerosols cannot be investigated because the different filters applied in the analysis, by design, remove a substantial part of the aerosol-cloud fraction relationship.

L207: 'is constrained using AOD' what does this mean? Is σ vertically integrated into the value of AOD?

Correct, the vertically integrated extinction coefficient is AOD. We modified the sentence to read: It is noteworthy to mention that because the vertically integrated σ_{ext} in the CALIOP-S data product is AOD…"

Fig.7: It would be easier to follow if the authors marked these 4 regions in Fig 6.
Good suggestions. This will be implemented in the revised version.

l261-262: I think the story in Arola et al. (2022) is quite different to the argument here. They attributed the invert-V to retrieval errors. Citing this paper here seems a bit confusing unless the authors make this clear.

The reviewer is correct in the sense that Arola et al. (2022) address a number of potential biases, including retrieval uncertainties. However, the article also explores the effect of cloud natural heterogeneity and discusses how spatial changes can yield a relationship that are not necessarily the manifestation of the aerosol indirect effect. This concept is partially encapsulated in the article title ("Aerosol effects on clouds are concealed by natural cloud heterogeneity and satellite retrieval errors").

L266: It's Intuitive that the product of S_nd_lwp (Iig. 6a) and ACI (Fig. 4a) should be negative as their signs are opposite, especially in Tropics; so it's kinda surprising that it turns to be negligible. Could the authors explain this a bit more?

From a point of view of the statistical estimation, this is the result of having a negligible ACI index for regions with positive susceptibility of LWP to Nd. The physical interpretation of these results is, nevertheless, challenging. Cognizant that these estimates need to be validated with other datasets and methods, we interpret this negligible susceptibility as the modest effect of aerosols to modify the relationship between Nd and LWP for those specific regions. This will be discussed further in the article, including potential sources of uncertainties that can also challenge our interpretation.

L298: I'd avoid words like "novel" or "new," or anything implying the study is the first to show a particular conclusion. A more neutral phrasing would work better. It would be easier for readers to follow the results if Fig. 4 were placed as a separate panel within Fig. 9; so that readers would not need to scroll back and forth. And why does the ACI(based on AOD) index appear to be negative here? It's overall positive in previous studies. Would be good to discuss.

We agree on the use of "novel" or "new" and the inclusion of Fig.4 as a subpanel of Fig. 9. We note that regions with seemingly negative slopes are statistically indistinguishable from zero and, therefore, the slope sign is not discussed in the article.

L301-303: The authors stressed a lot on the difference in ACI between σ and AOD; but it's very important to mention here that in the end we care about anthropogenic perturbation of Nd and forcing which also relies on PI-PD change in the utilized proxy, not the simple slope (https://doi.org/10.1029/2022RG000799).

This is a fair criticism. While assessing PD-PI changes is important for understanding the anthropogenic forcing, these satellite-based metrics are also useful for evaluating climate models (e.g. Zheng et al., 2025). Also, anthropogenic forcing estimates benefit from quantifying these slopes (e.g. Bellouin et al., 2021).

Reference:

Zheng, X., Feng, Y., Painemal, D., Zhang, M., Xie, S., Li, Z., Jacob, R., and Lusch, B.: Regime-based aerosol–cloud interactions from CALIPSO-MODIS and the Energy Exascale Earth System

Model version 2 (E3SMv2) over the Eastern North Atlantic, Atmos. Chem. Phys., 25, 17473–17499, https://doi.org/10.5194/acp-25-17473-2025, 2025.

L311-312: the use of AOD doesn't misguide the modelers as long as they are looking at AOD as well. I think this sentence can be dropped.

It could still misguide modelers in the sense that the relationship between AOD and Nd could point to processes and covariations that are not necessarily related to aerosol-cloud interactions. We will slightly modify the sentence to reflect our response.

L311: I find the phrase "unlike previous assessments, but similar to Gryspeerdt et al. (2016)" a bit confusing. Gryspeerdt et al. (2016) is also a 'previous' study, so it might help to clarify what you mean here. For example, do you refer to a specific group of 'previous assessments' using a different methodology?

We agree with the reviewer. Because we are talking about methodological differences rather than differences in conclusions, we are going to omit the "unlike previous assessments" phrase.