Response to the Reviewer's Comments

May 2, 2022

Comments by Reviewer #2

I would like to thank the authors for the changes made in the article and the answers to my comments. Reading the new version of the article, I am confident that the changes improved the results, the readability, and the understanding for the readers. I confirm that the study provided a deep analysis on the aerosol-cloud interactions, the results are very interesting and they are suitable for ACP. Nevertheless, I still have some concerns from the analysis and the answer to the comments, therefore I still recommend major revisions before publication in ACP. I detailed the reasons below. I encourage the authors to consider my comments because the results are very interesting and would be useful for the community.

We thank the reviewer for this thoughtful and constructive statement. We have revised the manuscript carefully according to the reviewer's comments. Please see the following detailed point-by-point responses.

Major revisions

1. I still disagree on the fact that what is inferred from the study is the Twomey effect (even considering the microphysical part of it). I still believe that calculating the sensitivity of N Vs CCN is the aerosol-cloud interaction but not the Twomey effect (same comment would go for studies looking at re Vs CCN constraining for LWP). Therefore, I find the wording confusing and particularly the title. I advise the authors to keep reference to Twomey as a motivation only. McComiskey and Feingold (2012) made explicit that : "... the terminology for this calculation was changed to ACI (aerosol-cloud interaction) to clarify that the result represents not the indirect effect, which is a response of cloud albedo to aerosol, but instead the microphysical response of the albedo"..

   Thanks for the comment. According to the reviewer's suggestion, we change the 'the Twomey effect' to 'droplet number response to aerosol'. The title is now rephrased as 'Addressing the difficulties in quantifying droplet number response to aerosol from satellite observations'

2. l.192 : The all-data method has one more degree of averaging compared to the pre-binned. Also the 95% confidence interval is also lower with the all data-method. I understand the point about comparing the methods but I do not understand why the pre-binned is preferred in the rest of the study.

   Thanks for the comment. Given that both methods were employed extensively by previous studies and the choice is somewhat ambiguous, here we would not really place a preference. Thus, we show both results (one in the main text and the other in the supplement information) in the revised manuscript, which lead to similar conclusions. The reason why we put the all-data method in the supplement information is that the statistical significance tests become rather meaningless when using the very large amounts of data points.

3. l. 300 : About the aerosol measurements next to clouds, the authors mentioned that the uncertainties potentially come from 3D effects, swelling etc. They did a nice analysis with the DeltaL parameter. Nevertheless I think the study could have gone a bit further with concrete quantifications of the uncertainties. It might be difficult for some parameters but for example RH might be possible to infer from reanalysis. Did the authors look at the uncertainty related to DeltaL constraining for RH or directly related to RH ?

   The reviewer has a great point. We agree that relative humidity (RH) is an important factor to be considered since it can affect Nd via entrainment mixing and also AOD/AI via aerosol swelling. We also thank the suggestion of using RH from reanalysis; actually we have done such analysis in a previous work (Jia et al., 2019) but for CER-to-AI sensitivity, and found a more negative CER-AI slope (equivalent to larger S here) for higher RH condition, which was attributed to the weaker entrainment mixing. Thus, the evidently lower S in the first DeltaL bin, where data are tightly linked to large CF (Fig. 6), in turn, associated with relatively high RH (Engström and Ekman, 2010), is unlikely to be caused by the effect of RH on clouds.

Considering enormous subgrid-scale variability of RH and un-avoidable co-location issues between reanalysis and satellite observations, it is difficult to obtain precise and coincident RH from reanalysis to match the ΔL observations. Instead, we choose to utilize the published in-situ aircraft measurements to roughly isolate the contribution of aerosol swelling to the greatly high AOD in the fisrt ΔL bin from retrieval issues (i.e., 3D radiative effects and cloud contamination). During the Indian Ocean Experiment (INDOEX), Twohy et al. (2009) measured a rise in RH from about 70% at more than 20 km from cloud to 90% with 1–4 km of cloud edge (equivalent to the distances of the third and first ΔL bins in Fig. 5a), which in turn results in about a 69% increase in aerosol scattering cross section (Twohy et al., 2009). Considering the aerosol humidification only occurs near cloud level, i.e., one quarter to one third of the aerosol column could be affected according to Lidar observations (Twohy et al., 2009), the increase in AOD by aerosol swelling is estimated to be 17–23%. This is up to about a third of relative increase in AOD from the third to first ΔL bins (64%) in Fig. 5a, implying that the retrieval errors in aerosol contribute the majority of the S reduction in the first ΔL bin. Note that the estimated AOD rise due to humidification relies on observed RH variability surrounding cloud and also the vertical profile and chemical composition of aerosol. The associated discussion is now added on lines 313-323.

Added/Modified text:

Lines 313-323: 'Based on the published in-situ aircraft measurements, we roughly isolate the contribution of aerosol swelling from retrieval issues (i.e., 3D radiative effects and cloud contamination). During the Indian Ocean Experiment (INDOEX), Twohy et al. (2009) measured a rise in relative humidity (RH) from about 70% at more than 20 km from cloud to 90% with 1–4 km of cloud edge (equivalent to the distances of the third and first ΔL bins in Fig. 5a), which in turn results in about a 69% increase in aerosol scattering cross section (Twohy et al., 2009). Considering the aerosol humidification only occurs near cloud level, i.e., one quarter to one third of the aerosol column could be affected according to Lidar observations (Twohy et al., 2009), the increase in AOD by aerosol swelling is estimated to be 17–23%. This is up to about a third of relative increase in AOD from the third to first ΔL bins (64%) in Fig. 5a, implying that the retrieval errors in aerosol could contribute the majority of the S reduction in the first ΔL bin. It should be noted that the estimated AOD rise due to humidification relies on observed RH variability surrounding cloud and also the vertical profile and chemical composition of aerosol, which could vary with geographic location.

Minor revisions

1. l. 137-140 : The study from Anderson et al. (2003) considers clear-sky pixels but they do not conclude anything when it is cloudy (I think). I still do not know if considering aerosols next to clouds represent aerosols within the clouds. There might be a reason why cloudy pixels are cloudy and adjacent clear-sky pixels, clear sky. Not everything can be uniform, maybe it comes from the aerosols. I do not expect the authors to redo the analysis with another proxy (but I would recommend it for their future analysis) but I would like the text to highlight this concern clearly.

   Thanks for the suggestion. We agree with the reviewer that aerosol is not always as homogeneous as assumed, especially when precipitation occurs. We now clarify this on lines 135-136.

   Added text:

   Lines 135-136: ' Note that this assumption would be questionable especially when aerosol is scavenged by precipitation (Gryspeerdt et al., 2015).'

2. About the uncertainties, I was referring to the ones derived from the algorithms directly, there is no quantification about the uncertainty from Nd or CER for example. The authors constrained on the latitudes but the uncertainties on the retrieved parameters can still be high..

   Thanks for the comment. The discussion on retrieval uncertainties is now added on lines 155-158.

   Added/Modified text:

   Lines 155-158: 'With the above sampling strategy, the random uncertainty in N_d was reported at 78% on a pixel level and this dropped substantially when averaged to a 1° by 1° region (Grosvenor et al., 2018).

However, as stated in Gryspeerdt et al. (2021), the systematic bias in the N_d retrievals to in situ measurements is low, with determination coefficients of 0.48 for all cloud types and 0.5-0.8 for stratocumulus clouds. '

3. l. 149-151 : "Nd id the independent variable in the S calculation", I am not sure to understand what is meant here, can the author rephrase?.

   Here we meant that the adiabaticity doesn't affect the sensitivity calculation if it is constant or doesn't significantly vary with AOD (AI). Thanks for the reminder. We find this sentence is not necessary, and remove it now.

4. Table 3 is interesting but I am wondering how the authors determined the magnitude of the biases for each issue. Also would it be possible to retrieve a percentage of the bias (even a rough estimate) for each issue to estimate the uncertainties on S? If a future study cannot account for an effect, it would be very useful to use this information..

   We thanks the reviewer for this thoughtful suggestion. This is a great point! We now update Table 3 with the percentage of each bias as suggested.

   Added/Modified text:

   Lines 486-492: ' Aerosol retrieval biases (3D radiative effects and cloud contamination), aerosol swelling, and cloud retrieval bias (heterogeneity effect) tend to lead to an underestimation of S. Although S_{AI} (S_{AOD}) for the first ΔL bin, where evident AI(AOD) enhancement exists, is about 29% (50%) less than other unaffected bins, the overall underestimation is only ~3% because of the small data volume in the first bin (Fig. 5a). Nevertheless, for low-ΔL dominated regions (e.g., stratocumulus regions), the underestimation can be more pronounced. By comparing S_{AI} (S_{AOD}) calculated by N_{dAll} and N_d, the underestimation by cloud retrieval issues is roughly estimated to be ~8% (~17%). '

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