Authors' Response to Reviews of

Model analysis of biases in satellite diagnosed aerosol effect on cloud liquid water path

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RC: ***Reviewers' Comment***, AR: Authors' Response, ☐ Manuscript Text

Reviewer #2

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Overview

RC: The joint relationship between LWP and cloud droplet number concentration (CDNC) is commonly used to infer the LWP adjustment to CDNC changes resulting from changes in CCN. This relationship has been assessed in satellite retrievals, but such retrievals depend on key assumptions in order to estimate CDNC and LWP. Biases associated with these assumptions are evaluated in LES simulations of the frequently analyzed DYCOMS RF02 case. Satellite retrievals of CDNC are high biased, and if constant values are used for adiabaticity and other inputs as commonly done, CDNC increases from cloud cell cores to edges, opposite of simulation output. Allowing those variables such as adiabaticity to vary based on model output produces the proper CDNC spatial pattern, though still with a high bias. The satellite retrieval LWP is unbiased overall if averaged across cells, though it tends to be overestimated in cell cores and underestimated on cell edges. For a set CCN concentration, LWP increases approximately linearly with CDNC on a log-log scale, and combining across different CCN simulations, the negative slope of the inverted "v" LWP-CDNC relationship is not produced. If adiabatic satellite retrievals of CDNC and LWP are used, inverted "v" shaped LWP-CDNC relationships are produced. Introducing 20% uncertainty to satellite retrievals of cloud optical depth and effective radius does not alter the overall LWP-CDNC relationship. The overestimate of CDNC is particularly biased at relatively low LWP. The authors thus argue that a good constraint on CCN is required because CDNC cannot be used as a proxy in such situations for representing LWP adjustments. Overall, the study presents compelling evidence that the negative slope portion of the inverted "v" LWP-CDNC relationship may not be caused by LWP adjustments to CDNC. This is a very informative study that is thorough in its methods. Its conclusions should aid improved interpretation of satellite retrieved cloud microphysical properties that are used to infer aerosol-cloud interactions. More studies like this are needed to sufficiently understand and design proper model-observation comparisons. I don't have any major concerns with the study but have some minor comments, mostly related to clarification, that I think could improve the study if addressed.

AR: We deeply appreciate your comments, questions and suggestions. We will proceed to resolve each one of them. In some cases, we have added subsections to address each item.

Comments

RC: **1.** Lines 22-24: This sentence would be more informative if it explicitly stated what the counteracting physical processes and satellite retrieval challenges were with respect to the studies cited.

AR: This is a very good comment and to explain this more clearly, we have modified this part to read as follows:

> These mixed results have been attributed to several counteracting physical processes, for example the effects of solar heating, cloud-top mixing, and variability in moisture on LWP (Feingold et al., (2022); Gryspeerdt et al., 2022; Glassmeier et al., 2021; Zhang et al., 2024). In addition, there are temporal and spatial averaging and variability and/or noise in satellite data which cause bias in satellite based estimates of aerosol-cloud interactions (Feingold et al., 2022; Arola et al., 2022).

RC: **2.** Line 38: I believe "higher boundary layer" should be "boundary layer depth change".

AR: The manuscript was changed accordingly.

> The cloud top pressure increases from 850 hPa to 890 hPa going from west to east indicating ~~higher boundary layer~~ boundary layer depth change which might affect boundary layer dynamics and thus cloud properties.

RC: **3.** Line 42: State what the increased CDNC at the cell boundaries is relative too, presumably real CDNC values?

AR: The manuscript was changed to clarify the sentence.

> However, calculation of CDNC based on the effective radius, and assuming constant sub-adiabaticity, would lead to overestimation in retrieved CDNC values compared to the real CDNC values at the cell boundaries (see Equation (2) in Section 2).

RC: **4.** Line 45: Is the estimate of the aerosol effect on LWP referenced here based on joint distributions of LWP and CDNC or how is the true effect quantified from which the bias is determined? Beyond the question of whether the LWP-CDNC joint distribution is accurately retrieved, there is the question of whether it can even be used to estimate LWP adjustment when derived from Eulerian statistics, e.g., considering that a process such as the entrainment-evaporation feedback can take several hours to days to alter the LWP in response to a change in CDNC along Lagrangian trajectories (e.g., Christensen et al. 2023, 2024).

AR: Yes, the estimate of the aerosol effect on LWP in Arola et al., (2022) is based on the analysis of joint distributions of CDNC and LWP. Variability in cloud fields and noise in retrievals will also affect the analysis of CDNC-LWP response when using Eulerian statistics.

RC: **5.** Line 108: Clarify that these parameters are not necessarily constant even though they can be and often are assumed to be constant.

AR: We have now clarified this in the revised manuscript as follows:

> The cloud parameters k, f_{ad}, c_{w}, and Q_{ext} vary with time along the cloud structure. However the actual values cannot be directly derived from MODIS observations and thus they are ~~can be~~ assumed to be constant and denoted by α for which an often used value for marine stratiform clouds is $1.37 \times 10^{-5}\ \mathrm{m}^{-\frac{1}{2}}$ (Quaas et al., 2006; Grosvenor

et al., 2018b; Gryspeerdt et al., 2022; Arola et al., 2022) Estimates of f_{ad} could possibly be improved combining MODIS/CALIOP observations.

RC: **6.** Line 143: I see low biases in LWP at the cloud cell boundaries and high LWP biases in the cell centers.

AR: We have modified the text to read:

> Biases in LWP also occur ~~at cloud cell boundaries~~differently across cloudy areas (Figure S5).
>
> Cloud cell boundaries tend to have low biased LWP values while cloud cell centers are biased high. In cloud cell boundaries processes such as entrainment and lateral mixing leads to sub-adiabaticity. Since these sources of variability are not considered in the formulation of satellite retrieval equations, there are important deviations from the assumptions of vertically constant values for droplet number concentration, droplet size distribution breadth and adiabaticity.

RC: **7.** Figure 3 and discussion about it: The positive slope of LWP with respect to CDNC is often assumed to be caused by precipitation suppression. That could be the case when combining multiple different CCN simulations, but for a single simulation, the positive slope is simply representing the horizontal structure of the cell where air moves from the high LWP, high CDNC core outward toward low LWP, low CDNC cell edges, correct? Is it worth clearly distinguishing between these 2 causes and interpretations?

AR: This is correct and a good point. We have added the following text to discuss this:

> For an individual simulation, the positive slope between CDNC and LWP reflects the horizontal structure of the cell, where air flows from the core, characterized by high LWP and high CDNC, outward toward the cell edges with lower LWP and CDNC.

RC: **8.** Line 161-162: Making adiabaticity the same in a model and satellite analyses also brings CDNC-LWP relationships into better alignment (e.g., Fig. 21 in Varble et al., 2023).

AR: We truly appreciate that you have pointed out this reference. The following changes have been made to the main manuscript.

> Previous studies have shown that selecting adiabatic pixels in a model and satellite analysis bring their results closer to each other (Dipu et al., 2022 ~~)~~ and Varble et al. [2023] showed that removing the differences between the adiabaticity in an Earth System Model and satellite retrievals brings the observed and satellite retrieved LWP adjustment closer to each other.)

RC: **9.** Lines 193-194: I don't completely follow the argument here regarding subadiabatic points not contributing to the LWP-CDNC inverted "v" shape since it is referencing the relationship of LWP with CCN rather than CDNC in Figure 5. Adiabaticity will impact the CDNC calculation, but CCN is not impacted, so what allows for the connection of Figure 5 to the LWP-CDNC correlation?

AR: Here we mean to say that even though in the high resolution of the LES model the sub-adiabatic points contribute to the CDNC-LWP "inverted-v" shape in data for individual simulations, the spatial averaging of the model data to a resolution that corresponds closer to the satellite data dilutes this effect. This results in very similar CCN-LWP correlation between the direct model diagnosed and satellite-equation diagnosed CCN and LWP. Using CDNC instead of CCN would result in an "inverted-v" shape if many different cloud cases are combined. This highlights the need for using observed CCN instead of using CDNC as a proxy for CCN.

This analysis indicates that when using domain averaged CCN and LWP values the non-adiabaticity of the cloud cell edges does not contribute significantly to the "inverted v" shaped correlation between CDNC and LWP seen in satellite data. Although there are issues in using Equations (2)-(3), coarse resolution of satellite data will reduce these issues significantly.

RC: **10.** Lines 203-204: This is true but if you combine Figures S12-14 like was done for Figures 4-5, could an inverted "v" LWP-CDNC correlation be produced?

AR: Yes, see Figure R1

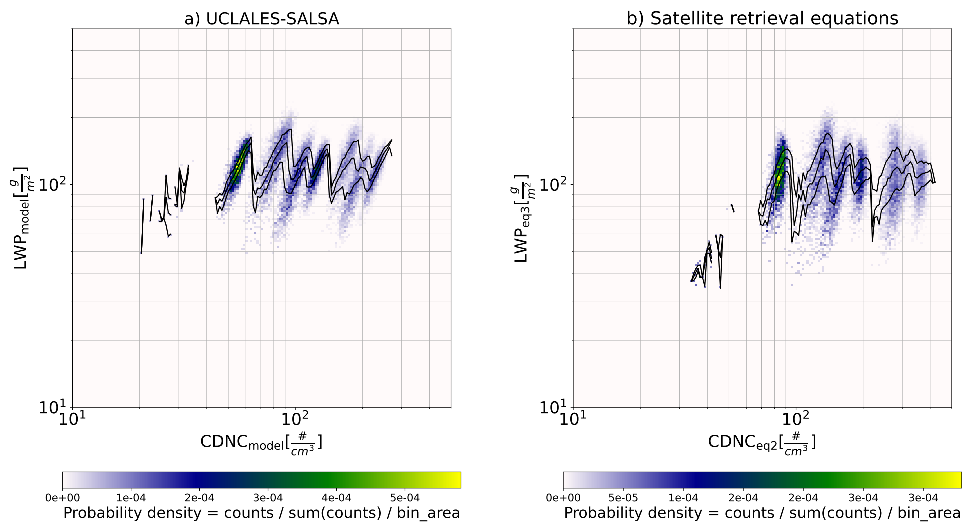

Figure R1: Joint histogram for spatially aggregated LWP and CDNC values a) from the direct output of UCLALES-SALSA, b) calculated using Equations (1) and (2) assuming a constant α. Black continuous lines indicate the 25th, 50th and 75th percentiles of LWP per bin. The color scale indicate the probability density calculated as counts/sum(counts)/bin area.

RC: **11.** In the supplemental material, it mentions that a negative LWP-CDNC slope is produced when combining different times (cloud types) and CCN concentrations, which seems important, but I didn't see it highlighted in the main manuscript (although maybe I missed it or misinterpreted the supplemental text).

AR: We have now highlighted this point in the first sentence of Conclusions

Our LES simulations show that variability in cloud properties when including different cloud types, CCN concentrations, and clouds in different phases of their cycle ~~dynamics~~will bias satellite derived correlation between CDNC and LWP similar to Arola et al. [2022].

RC: **12.** Supplemental text line 80: what is the correction factor introduced?

AR: The adiabatic factor defined by Brenguier et al. [2000] considers that the adiabatic value of the liquid water path increases linearly with increasing altitude from zero at the cloud base to its maximum value at the cloud top being equal to $\mathrm{LWP}_{\mathrm{adiab}} = 0.5 c_{w,\mathrm{model}} H^2$ where $c_{w,\mathrm{model}}$ is the water condensational lapse rate in the extended cloud top region, H is the cloud geometrical thickness. In our study, we defined the cloud base differently as the minimum altitude at which the liquid water content is equal or higher than 0.01 g m^{-3} instead of zero. To have comparable conditions at cloud base, we introduced the term $\mathrm{LWC_{model,CB}H}$ in Equation S.6

$$f_{\mathrm{ad}} = \frac{\mathrm{LWP}}{0.5c_{w,\mathrm{model}}H^2 + \mathrm{LWC_{model,CB}}H},$$

AR: The text in the supporting information was modified in accordance to the previous paragraph.

~~In this study, we introduced a correction factor to account for our definition of the cloud base (i.e. at the cloud base altitude, the liquid water content $\mathrm{LWC_{model,CB}}$ was assumed to be 0.01 gm^{-3}).~~

The adiabatic factor defined by Brenguier et al. (2000) considers that the adiabatic value of the liquid water path increases linearly with increasing altitude from zero at the cloud base to its maximum value at the cloud top being equal to $\mathrm{LWP_{adiab}} = 0.5c_{w,mathrmmodel}H^2$ where $c_{w,\mathrm{model}}$ is the water condensational lapse rate in the extended cloud top region, H is the cloud geometrical thickness. In our study, we defined the cloud base differently as the minimum altitude at which the liquid water content is equal or higher than 0.01 g m^{-3} instead of zero. To have comparable conditions at cloud base, we introduced the term $\mathrm{LWC_{model,CB}H}$ in Equation S.6

RC: **13.** Figure S5: The CER plot is saturated from the maximum value of 10 μm. Perhaps extend the CER range to show more structure. Also, the LWP Equation (3) panel uses a color scale that is not ideal for anomalies because it isn't clear where the zero crossover is. I suggest a diverging color scale or a contour of the 0 line (if not too noisy).

AR: Thanks. The color scale used to represent biases in LWP satellite retrievals was changed to a divergent one as it is shown in Figure R2. Changes are reflected in the new version of the supplement.

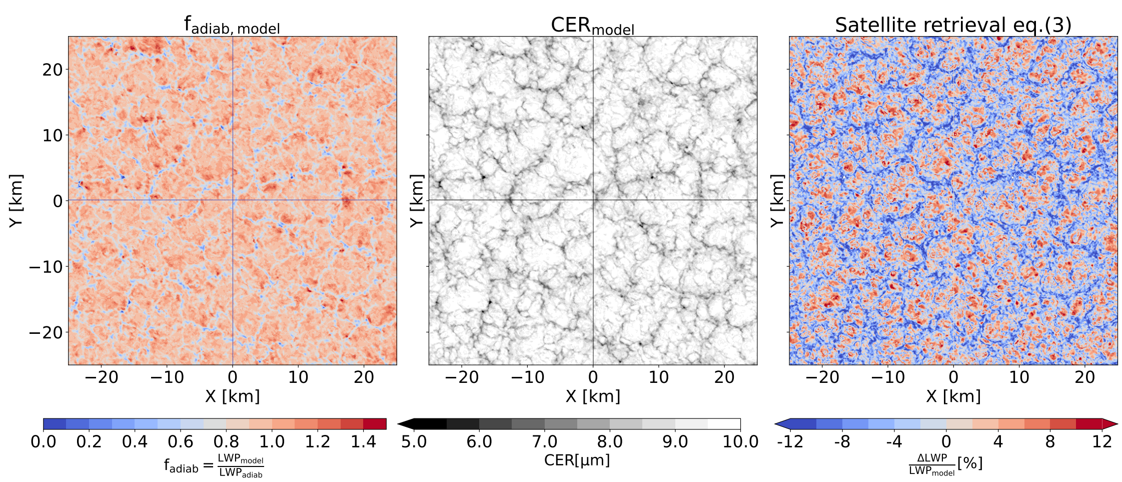

Figure R2: Biases between modelled and surrogate satellite-retrievals of LWP with Equation (3) in a simulation initialized with a CCN loading of 360 cm^{-3} at the time instant of 10 h

RC: **14.** Supplemental text lines 92-93: Larger biases are seen at cloud edges, but are the cores biased in the opposite direction too?

AR: This is correct, we have modified the text as follows:

> Nonetheless, biases in LWP between the model and Equation (3) are significantly lower remaining below ±20% with larger values at cloud edges and adiabatic cores as can be seen in Figure S5 (R3). All datasets at different simulation times show similar trends suggesting that biases are caused by processes at cloud edges related to stratocumulus dissipation (e.g. evaporative cooling during cloud top mixing or lateral mixing) which are not considered in the pseudo-adiabatic cloud model from which satellite equations are derived. Positive biases in satellite retrievals of LWP can be also expected when cloud top CER values do not reflect droplet growth fully driven by adiabatic cooling but instead correspond to super-adiabatic droplet growth after entrainment mixing (e.g., Yang et al., (2016), Zhu et al., (2019).

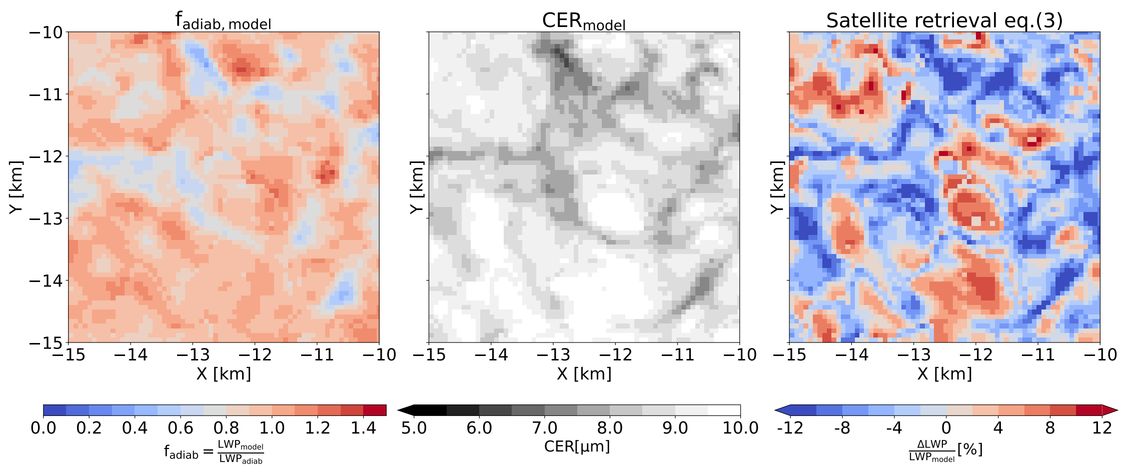

Figure R3: Biases between modelled and surrogate satellite-retrievals of LWP with Equation (3) in a simulation initialized with a CCN loading of 360 cm^{-3} at the time instant of 10 h in a section of the model domain shown in Figure 3.

RC: **15.** Figure S10: Why does the color bar have such a large range? It's difficult to see structure in the plots because of this.

AR: This is a good point. The color scale was improved to show better the cloud structure.

RC: **16.** Supplemental text lines 140-141: If using words like "extreme" and "very" here, I suggest adding numerical values to reference since interpretations of these words varies. Further, I suggest softening this sentence a bit to say that the satellite-retrieved inverted "v" can be caused by these biases rather than it is caused by them since this is based off of a single LES case.

AR: You are right. The following changes were made to the supplement:

This factor together with the fact that the signal from cloud edges is flatten out after spatial aggregation of cloud properties (Figure S9), support the hypothesis that the inverted-V shape in satellite-based studies ~~is caused by~~ is likely related to ~~extreme~~ positive biases in satellite retrievals of CDNC at ~~very~~small CER values. Additional cloud modelling studies reflecting a wider palette of meteorological conditions and background aerosol loadings would be needed to offer a definitive confirmation.

References

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