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 #1

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General comment

RC: The manuscript investigates the biases in satellite retrieval of liquid water path (LWP) and cloud condensation nuclei (CDNC) using LES simulations configured to DYCOMS-II RF02. The authors compare the correlations of the two variables from three sources: direct model outputs, retrievals from equations using LES simulated values for the parameters, and retrievals from equations assuming constant parameters. The authors find that CDNC retrievals assuming constant parameters tend to increase CDNC at the cloud boundaries. They conclude that satellite-derived CDNC shows a significant positive bias, but the correlation between LWP and CDNC was very similar between the methods. The authors also find that the instrumental noise in satellite retrievals do not affect the correlation between CDNC and LWP. The topic of this manuscript is suitable for publishing in ACP. However, the manuscript is poorly articulated, and the results are not effectively presented. The confusing variable labels in the figures further contribute to the overall lack of clarity. The current version isn't ready for publication but may be considered after some revisions. I would like the authors to respond to and address my comments. Details of my comments are as follows: Recommendation: Major revisions

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.

Major comments

RC: **1.** I don't find the figures to provide sufficient support for the authors' arguments in the text. I suggest adding more figures to better substantiate the arguments. Please see the detailed comments below.

AR: We have addressed your comments and modified the figures accordingly. A more detailed explanations of modifications will be provided later in this reply.

RC: **2.** The axis variables are not clearly labeled. The authors need to clearly label the LWP and CDNC in Figs. 3-6, indicating whether they are direct model outputs or computed from equations 1-3, and whether they are pixel-level data or domain-average data. An easy way to address this is by using subscripts, e.g., LWPtrue, LWPeq3, CDNCeq2, etc. Use overhead bars if the variables are domain average values. The titles of Figs. 3b, 4b, and 6b are confusing because the LWP in these panels is computed from equation 3, not equation 2. I suggest using a title like "Computed LWP and CDNC" or something similar.

AR: This is indeed a very good suggestion. We have changed the variable names according to it.

Detailed comments

RC: **1.** caption: "the calculated CDNC". Is it calculated from eq. 1 or 2? "the retrieved LWP". Is it calculated from eq. 3? Please also specify the sizes of the squares on the right panels.

AR: CDNC is calculated from Eq. (2) and we have modified the text to read CDNC calculated according to **?**. The small squares are approximately 30 km in size, while the width of the larger rectangle is approximately 500 km. We have added this information also in the figure caption.

> Cloud properties of a stratocumulus cloud deck west of Peru and Chile over South Pacific on Aug 30th, 2003. The upper left panel shows the ~~calculated CDNC~~ CDNC calculated according to Quaas et al. (2006) and lower left panel shows the retrieved LWP from Moderate Resolution Imaging Spectroradiometer (MODIS) Level-2 (L2) Collection 6.1 **?**. Right panels show a magnification of the structure of a cloud cell within the cloud field denoting the cloud effective radius and the cloud reflectance for the corresponding cloud cell. Small squares are approximately 3 km × 3 km and large rectangles are approx. 500 m × 6 km.

RC: **2.** Lines 35-38. The authors argue that the cloud top effective radius and liquid water path change with cloud top pressure. However, cloud top pressure is not shown in the paper or supplementary material. Please provide a figure of cloud top pressure to support this argument.

AR: Since this is introductory text and not results of this study, we have not included the figure in the manuscript or in a supplement. Below is the figure of MODIS AQUA cloud top pressure for Aug 30th, 2003.

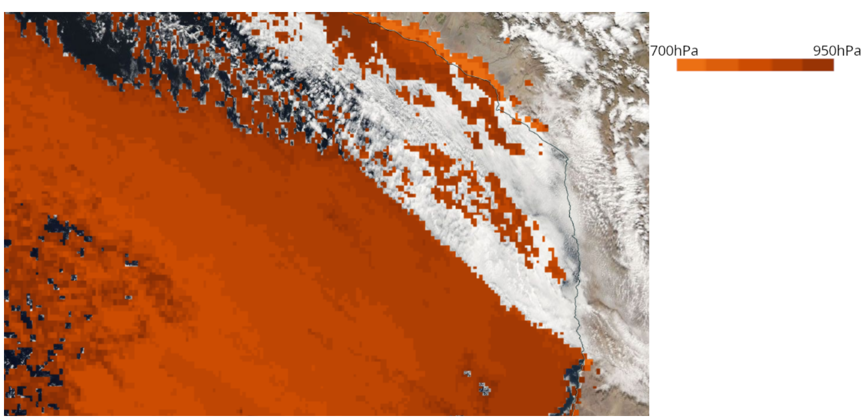

Figure R1: Cloud top pressure for MODIS AQUA

RC: **3.** Line 41. Replace "liquid water content" with "liquid water path" as the former is not shown in Fig. 1.

AR: Thank you. The manuscript reflects the replacement.

> The decrease in retrieved cloud effective radius results from the entrainment mixing at the cloud top and downdrafts in the cloud cell boundaries which both reduce the ~~liquid water content~~liquid water path.

RC: **4.** Lines 41-43. I don't see how this sentence connects to the previous one. Please revise the paragraph for better flow.

AR: Paragraphs were rephrased as follows

> The decrease in retrieved cloud effective radius results from the entrainment mixing at the cloud top and downdrafts in the cloud cell boundaries which both reduce the ~~liquid water content~~liquid water path. ~~However~~At the cloud cell edges this is in conflict with the assumptions made in the calculation of CDNC.Calculation of CDNC based on the effective radius, and assuming constant sub-adiabaticity, would lead to increased CDNC at the cell boundaries (see Equation (2) in Section 2). In addition to actual variability in physical properties of clouds, satellite retrievals include uncertainties and instrument noise causing another potential source of bias in the satellite-derived correlation between CDNC and LWP.

RC: **5.** Lines 126-132. Since the section predominantly centers on Fig. 2, the brief reference to Fig. 3 interrupts the flow. To improve continuity, I suggest removing the discussion of Fig. 3.

AR: We have moved the discussion of Figure 3 towards the end of this subsection where Figure 3 is also otherwise discussed.

RC: **6.** Line 133. "The leftmost panel shows a closed cell type structure in the cloud". It's difficult to discern cloud structure in Fig. 2. Please include a snapshot of LWP to clarify.

AR: A snapshot of LWP is shown in Supplementary Material, Figure S3.

RC: **7.** In Figure 3 caption: "Simulations are colour coded according to CCN concentrations used in the model initialization". Please provide a legend in the figure to reflect this.

AR: Legend of Figure 3 was improved to help identifying simulation scenarios with different CCN background conditions.

RC: **8.** Lines 136-143. I think the paper would benefit from a separate figure comparing CDNC from model output and equations 1 and 2 to support the arguments in the text. I suggest adding a scatter plot with the true CDNC from the LES on the x-axis and the computed CDNC on the y-axis. Mark the domain-average CDNCs in the plot. It would be helpful if the authors could overlay the results of aggregation so that readers do not need to refer to the Supporting Information for details. The authors might consider coloring the scatter plot with LWP values to illustrate the bias of computed CDNC in relation to cloud structure. Similar plots can be made for LWP from model output and equation 3.

AR: The comparison of modeled and satellite-retrievals CDNC distributions was shown using marginal histograms in Figure 3 and Figures S6-S8 and S12-S14. We acknowledge that the size ratio between the joint and marginal histogram was not optimal and it is difficult to compare CDNC distributions. We have the correspondent figures in the main manuscript and supplement. Your suggestion is very good indeed. However, each time instance for a single simulation scenario comprises millions of points and it is very difficult to visualize trends due to overlapping layers caused by data variability.Nonetheless, we have addressed your comment and here we show a graphical comparison of CDNC distributions. Values for probability, overlapping index and correlation coefficient correspond to the entire dataset for a single time and initial aerosol loading. Scatter plots, instead, reflect random samples equivalent to 1% of the total data, approximately. CDNC distributions of model outputs and satellite-retrieval equations in a simulation initialized with a CCN loading of 360 cm^{-3} at the time instant of 10 h are compared in Figure R2 for the high resolution case (75 m $\times$ 75 m) and in Figure R3 for the low resolution case (1425 m $\times$ 1425 m) obtained from spatial aggregation.

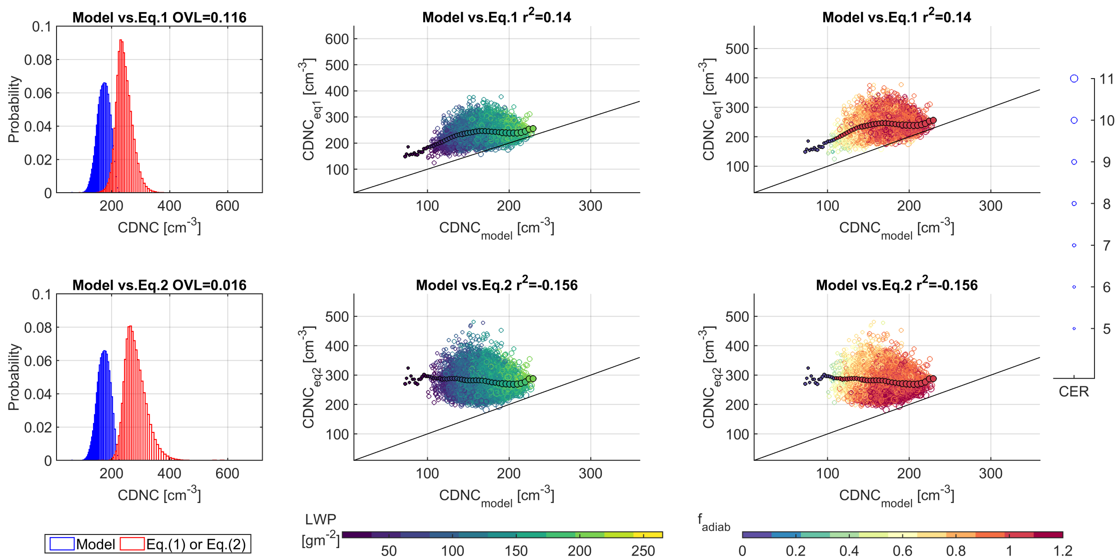

Figure R2: Comparison of distributions for cloud droplet number concentration (CDNC) obtained at high resolution (75 m $\times$ 75 m) from model outputs and satellite-retrieval equations in a simulation initialized with a CCN loading of 360 cm^{-3} at the time instant of 10 h a) Histograms of CDNC distribution indicating the overlapping index value (OVL) (i.e. If OVL=1 distributions are equivalent to each other) b) Scatter plot using LWP in the color scale and CER in μm for marker size c) Scatter plot using the adiabatic factor in the color scale and CER in μm for marker size. In the scatter plots, we have indicated linear correlation coefficient values ($p<0.05$) and added continuous black lines of perfect correlation as a visual guide. Mean values are highlighted with black edges keeping the variable color scale. For both satellite equations, larger biases correspond to thinner and subadiabatic columns with smaller droplet effective radius, conditions that are likely to happen in cloud edges. Histograms for CDNC-satellite values from Equation (2) show lower overlapping index as well as more frequent and higher positive deviations. Despite having a more robust approach that considers deviations from the adiabatic liquid water path as well as changes in the droplet distribution breadth, CDNC-satellite values from Equation (1) are still much higher than those from the model.

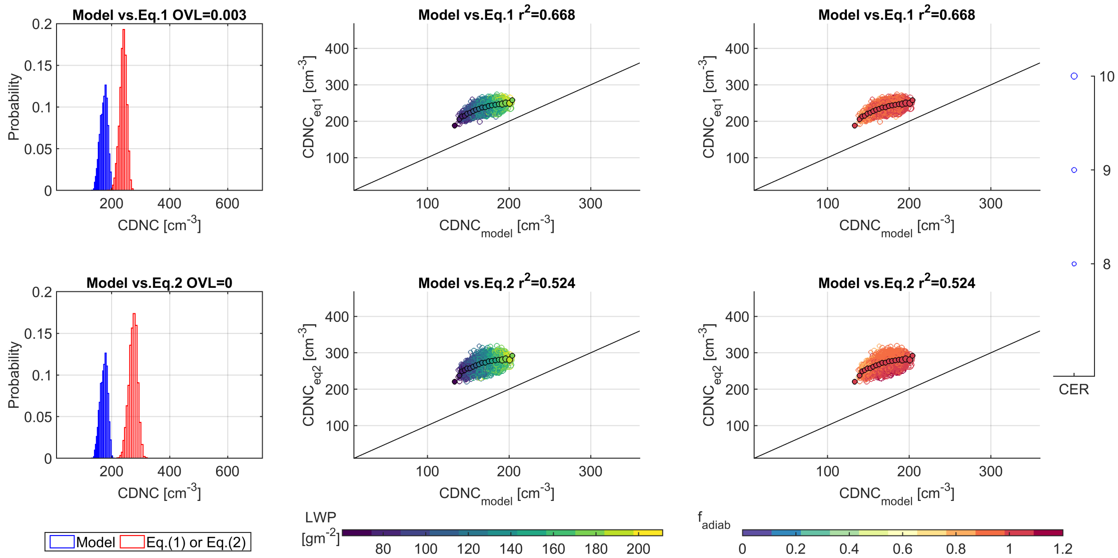

Figure R3: Comparison of distributions for cloud droplet number concentration (CDNC) obtained at low resolution (1425 m $\times$ 1425 m) from model outputs and satellite-retrieval equations in a simulation initialized with a CCN loading of 360 cm^{-3} at the time instant of 10 h. a) Histograms of CDNC distribution indicating the overlapping index value (OVL) (i.e. If OVL=1 distributions are equivalent to each other) b) Scatter plot using LWP in the color scale and CER in μm for marker size c) Scatter plot using the adiabatic factor in the color scale and CER in μm for marker size. In scatter plots, we have indicated linear correlation coefficient values ($p<0.05$) and added continuous black lines of perfect correlation as a visual guide. Mean values are highlighted with black edges keeping the variable color scale. After spatial aggregation using COT as a weighting factor, CDNC distributions become more symmetric and less spread out around the mean which in turn results in a reduction of the overlapping index between modeled and satellite-retrieval distributions. Although the aggregated dataset have a much lower influence of model columns with thinner sub-adiabatic clouds with smaller CER values, CDNC satellite-retrievals are still higher and linearly proportional to modeled ones (i.e. correlation coefficients in Figure R3 are larger than 0.5) confirming the systematic deviation.

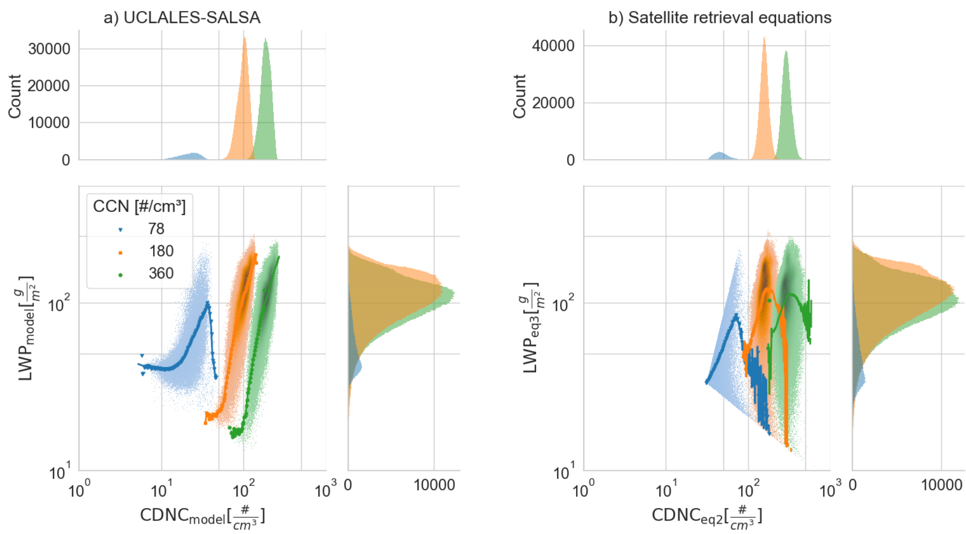

Figure R4: Joint and marginal histograms for LWP and CDNC values using a) UCLALES-SALSA and b) Equation (2) at a time instance of 6 hours. Simulations are colour coded according to CCN concentrations used in the model initialization. The intensity of colour in joint histograms increases when the probability increases. The probability is represented as a density function calculated as counts/sum(counts)/bin area. Continuous lines indicate the arithmetic mean.

RC: **9.** Line 142. Please list all possible assumption biases here instead of using "e.g".

AR: The main manuscript was modified to include possible biases.

> 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: **10.** Line 152. "Satellite derived CDNC values are at least two times higher than the direct LES values". Do the authors have any idea what might be causing this?

AR: Satellite derived CDNC values are calculated using direct LES values of cloud effective radius (CER) and cloud optical thickness (COT) that correspond to the expected optical penetration depth. However, they are positively biased due to the non-fulfilment of the underlying assumptions in the pseudo-adiabatic cloud model (i.e. vertically constant values for droplet number concentration, droplet size distribution breadth, adiabaticity). These criteria are hardly satisfied in thin cloud layers such as those observed in cloud edges.

> In addition, CDNC values have a clear high bias. In this case, satellite derived CDNC values are at least two times higher than the direct LES values. Values are positively biased due to the assumption of vertically uniform cloud

columns which is not valid in thin cloud layers such as those observed in cloud edges.

RC: **11.** Line 157. “Within individual ensemble members, the cloud internal variability contributes to the CDNC-LWP correlation and cannot be considered to be an aerosol effect on clouds”. Zhou and Feingold, (2023) has reached a similar conclusion. (Zhou, X., & Feingold, G. (2023). Impacts of mesoscale cloud organization on aerosol-induced cloud water adjustment and cloud brightness. Geophysical Research Letters, 50(13), e2023GL103417.)

AR: We have added the reference to Zhou and Feingold in the revised manuscript.

> Within individual ensemble members, the cloud internal variability contributes to the CDNC-LWP correlation and cannot be considered to be an aerosol effect on clouds, also shown by **?**.

RC: **12.** Line 177. To improve the connection with the subsequent discussion, please make it clear that Fig. 5 represents a proxy for satellite aggregation.

AR: The text in the manuscript has been modified as follows:

> Figure 5 represents a proxy for satellite aggregation. It shows the LES domain mean LWP at three different time instances into the simulation for three different runs as a function of the initial CCN concentration. Solid lines denote the mean LWP in the domain and the shading indicates the standard deviation in the data.

RC: **13.** Fig. 6a is identical to Fig. 4a. Replotting it is unnecessary.

AR: This is correct and we have removed Figure 4a as suggested.