Response to Reviewer 2

Summary:

In this paper, the author's examine the relationship between liquid water path (L) and cloud droplet number concentration (N) in the DYCOMS-II nocturnal stratocumulus case using an entrainment parameterization from Deardorff (1976), a partitioning of the buoyancy flux profile into contributions from surface fluxes, entrainment fluxes, long-wave radiation, and precipitation, and a slope parameter (m = dlog(L)/dlog(N)). The mixed-layer model assumes well-mixed boundary layers; therefore, the analysis is restricted to non-precipitating or weakly precipitating profiles. The mixed-layer analysis finds that at low N, the major driver of negative L adjustments is precipitation (virga). At high N, decreased sedimentation velocities increase droplet residence times near cloud top and result in greater entrainment and negative L. At low L (less than 30 g/m^2), an increase in droplet size can cause negative L through more efficient long-wave cooling (which enhances entrainment). Overall, L adjustments tend to be strongly negative for N = 10-100 cm^{-3} and tail off to smaller values at very high N (1000 cm^{-3}), which differs from the steady-state m in Glassmeier et al. (2021).

We thank the reviewer for the careful reading of our manuscript and the insightful comments made below. Our answers are stated in green font, while excerpts from the (revised) manuscript are printed in blue font with italic characters. All references not explicitly stated in this letter can be obtained from the revised manuscript.

Main comments:

The writing is thorough and technical, but at times hard to digest. I would suggest a title change, as one of the main results of the paper pertains to the effects of precipitating condensate on the mixed-layer state.

We changed the title to *"On the Processes Determining the Slope of Cloud-Water Adjustments in Weakly and Non-Precipitating Stratocumulus"*, with subsequent changes throughout the document.

I find reduced-order approaches such as the MLM outlined in this paper to be attractive and helpful in teasing out causality, but I question just how many "real" environments the results may apply to. If I am not mistaken, all of the L-N phase space is explored from a single research flight with fixed surface fluxes, moisture/energy jumps, and boundary layer depth. Some of these imposed environments may be very unlikely to occur in nature.

The reviewer is right that we present a very large phase space in Figs. 1 and 2, with combinations that are probably not realistic. However, the core of our study is Fig. 3 and the associated discussion in Section 3.3. Here, liquid water paths between 10 and 100 g m^{-2} and droplet concentrations between 100 cm^{-3} and 1000 cm^{-3} are tested, i.e., values typically observed in nature, as we clarified in the revised manuscript: *"Figure 3 shows $m \equiv d \ln (L)/d \ln (N)$ as a function of N for two different L values: L = 100 g m^{-2} (continuous lines) is representative for most stratocumulus (e.g., Wood, 2012). L=10 g m^{-2} (dashed lines) is representative for optically thin stratocumulus, which reflect markedly less solar radiation, but cover substantial regions of the globe (e.g., Leahy et al., 2012)."* As we agree that liquid water paths substantially higher than 100 g m^{-2} exceed the typical range of observed stratocumulus, we added the following statement: *"In fact, all L>150gm−2 are rather untypical for stratocumulus (Wood, 2012), and results in this part of the phase space should be analyzed with care."*

It would be beneficial to the reader to see some connection between the DYCOMS-II case (perhaps even a few LES cases probing the L-N phase space) and MLM results. For example, how do entrainment rates in the MLM compare to the field campaign (or buoyancy flux profiles from the LES)? It would be nice to see some exercise to constrain the behavior of the MLM and describe differences between the idealized and observed profiles.

We have added a black rectangle to Figs. 1 and 2 indicating the L-N range observed during the second research flight of the DYCOMS-II campaign, as reported by Ackerman et al. (2009). Our entrainment rate in this region (6 to 8 mm s^{-1}) agrees very well with the observed range of values (Fig. 1a). Moreover, the integrated buoyancy flux (Fig. 1d) or the surface precipitation rate also agree well with the large-eddy simulation intercomparison values reported in Ackerman et al. (2009). We added the following statement to Sec. 2.4: *"Figures 1 and 2 contain black rectangles that indicate the L-N range observed during the second research flight of the DYCOMS-II campaign (Ackerman et al. 2009). The w_e determined from (8) agrees very well with the observed range of 6 to 8 mm s^{-1} (cf. Ackerman et al., 2009). Additionally, $\langle B \rangle$ (Fig. 1d) and the surface precipitation rate $P_p(0)$ (dashed lines in Figs. 1 and 2) are in general agreement with the values reported from the large-eddy simulation intercomparison by Ackerman et al. (2009). This indicates that the framework applied in this study reproduces the behavior of observed stratocumulus at least in some parts of the analyzed phase space."*

It is stated within the conclusions that "This study was built upon the assumption that an increase in entrainment rate we due to an increase in N is exactly offset by a commensurate decrease in L, resulting in the same we irrespective of N." This statement is at odds with Figure 1a, with the entrainment rate contours sloping with changing N.

Entrainment increases with N. At several places, we state that *"In the absence of precipitation, further increases in N are found to increase the mixing of clouds with their surroundings (entrainment), leading to a decrease in L (Wang et al., 2003; Ackerman et al., 2004; Bretherton et al., 2007)."* and *"The large-eddy simulations in our companion paper (Chen et al., 2024a) show that a positive perturbation of N , δN > 0, results in an increase in we in response to an aerosol perturbation (see their Fig. 4a)."* However, this increase is transient because entrainment introduces warm and dry free-tropospheric air into the boundary layer, which evaporates the cloud. As a result, L decreases. And since entrainment is also a function of L, an initial increase in the entrainment rate due to a perturbation in N is offset by a (negative) adjustment in L: *"After sufficient time (18 h), this increase in w_e is diminished, resulting in negligible differences in w_e among the perturbed and unperturbed simulations. This decrease in w_e is enabled by a commensurate decrease in L in the perturbed simulations, δL < 0, resulting in increasingly stronger negative m with time (see their Fig. 2c)."* While the assumption (35) is an idealization of this behavior, it is based on accepted theories. For instance, this idealization can be considered a corollary of the feedback mechanism between entrainment rate and liquid water path originally suggested by Zhu et al. (2005) (see their Fig. 7), which has been also highlighted by Wood (2012) as a major mechanism responsible for the observed stability of stratocumulus (see their Fig. 26). We state this as: *"This idea is based on large-eddy simulations presented in our companion paper (Chen et al., 2024a) (see their Fig.4a), and can be considered a corollary of the w_e-L feedback mechanism suggested by Zhu et al. (2005) (see their Fig. 7)."*

Additionally, it is not clear from Chen et al. (2024a) that this assumption is justified. In Chen et al. (2024a), the L-N phase space (L ranges from 65-95 g/m^2; N ranges from 100-600 cm^{-3}) is much smaller than the phase space examined in this paper. Given how critical this assumption remains to the results of the paper, justification for this simplification needs to be provided within the manuscript.

While we show a much greater phase in Figs. 1 and 2, these results do not rely on the assumption (35). These results are based on the established entrainment parameterization discussed in Sections 2.2. There is nothing new about the entrainment parameterization, besides showing how the entrainment parameterization and its components behave in an L-N phase space, which is necessary to better understand Fig. 3.

Only for Fig. 3 and the associated discussions in Section 3.3, has the assumption (35) been applied to liquid water paths between 10 and 100 g m^{-2} and droplet concentrations between 100 cm^{-3} and 1000 cm^{-3}: *"Figure 3 shows $m \equiv d \ln(L)/d \ln(N)$ as a function of N for L = 10 and 100 g m^{-2} (dashed and continuous lines, respectively), representing cases with unsaturated and saturated longwave radiative cooling. [...] For clarity, we will focus our discussion on how m changes between N = 100 and 1000 cm^{-3}, typical values that circumscribe non-precipitating stratocumulus."* While these values slightly exceed the range of values tested in our companion paper (Chen et al. 2024a), in which liquid water paths in the range of 40 to 100 g m^{-2} and droplet concentrations between 100 and 400 cm^{-3} are analyzed (see their Fig. 1), the assumption (35) is probably valid for the largest part of the phase space shown in Fig. 3.

It's also unclear why this assumption is necessary. In Section 2.3.2, the timescale of 18 hours to reach an equilibrium entrainment rate is mentioned, but this would require a longer time period than a single night to equilibrate.

We agree that 18 hours exceed the nighttime in the subtropics where most stratocumulus are found. However, for this study and many previous studies on aerosol-cloud interactions (e.g., Glassmeier et al. 2021) or our companion paper (Chen et al. 2024a), we assume perpetual night to better understand the interactions of cloud microphysics and dynamics. As soon as solar radiation is considered, the interpretation of results is much harder. We clarify that we do not consider solar radiation: *"As for our companion paper (Chen et al., 2024a), we neglect interactions with solar radiation for simplicity."* and *"Additionally, (35) does not consider adjustments in surface fluxes (Bretherton and Wyant, 1997), as well as the impact of solar radiation (Chen et al., 2024b), for which more complex models would be required."*

Please explain why entrainment cannot vary as a function of N.

See our previous answer to "It is stated within the conclusions that "This study was built upon the assumption that an increase in entrainment rate we due to an increase in N is exactly offset by a commensurate decrease in L, resulting in the same we irrespective of N." This statement is at odds with Figure 1a, with the entrainment rate contours sloping with changing N.".

In addition to the entrainment assumption, it seems odd to fix the boundary layer depth in the MLM. Is there a reason the moisture and energy equations are decoupled from the mass budget?

There are no coupled or decoupled budgets used in this study. The only equation evaluated in this study is (8): *"In this study, we utilize a simple entrainment parameterization used in mixed-layer models to determine entrainment- mediated cloud-water adjustments in non-precipitating stratocumulus."* In 2.3, we further clarify that *"Using the mixed-layer model entrainment parameterization w_e outlined above, (35) is solved iteratively for δL using prescribed values of L, N , and δN , while keeping all other parameters constant."* One could address changes in boundary layer depth in (35). However, this would require knowledge on how the boundary layer depth changes with N. While this is interesting, this study aims to provide deeper understanding of cloud water adjustments, i.e., the change in L with N. While the boundary layer depth certainly has an impact on the cloud water adjustments shown in Fig. 3c, the change in boundary layer depth over the course of 18 h, as implicitly assumed for (35), is probably small in contrast to the more direct impact of entrainment on L. Still, investigating these nuances is important, and we amended our last to paragraph accordingly: *"All in all, this study showed that even comparably simple models, such as the one used here, can be applied to increase our fundamental understanding of aerosol-cloud interactions. In fact, the simplicity of the applied model allowed us to directly link cause and effect of cloud-water adjustments, which can be difficult in more complex models such as global circulation or even large-eddy simulation models due to confounding factors (Mülmenstädt et al., 2024).. That being said, the nuance provided by these models should not be disregarded as they help to quantify the effects that have been neglected here (e.g., interactive surface fluxes, changes in boundary layer depth, or the diurnal cycle of solar radiation). Thus, we advocate that the assessment of aerosol-cloud interactions should balance the use of complex and simple approaches by substantiating quantitative understanding with qualitative insight."*

Minor comments:

1. Line 6: "At higher N, the cessation"… here and on line 370, I would recommend not using "cessation", as sedimentation continues to exist, it is just smaller.

   Changed to *"decrease in"*.

2. Line 23: It may be useful to mention the diurnal cycle of LWP (Wood et al., 2002). Recent LES of polluted environments have shown larger diurnal variability of LWP (at nearly constant N). It's important to consider how the diurnal cycle of entrainment rate/buoyancy flux may influence the interpretation of m. Additionally, it may be worth noting that the free troposphere can act as a source of N, complicating a clean relationship between the entrainment rate and the liquid water response (Chun et al., 2023).

   Those processes confound the understanding that can be gained from observations. Thus, disregarding them in our simplified framework helps to reveal the underlying physics more clearly. We have added the following statement to the manuscript's introduction: *"Additionally, observational approaches to estimate m need to consider the natural* *variability in L due to, e.g., the diurnal cycle of solar radiation (e.g., Wood et al., 2002) or changes in the large-scale meteorological conditions (e.g., Chun et al., 2023; Goren et al., 2024). While models can be used to eliminate the influence of some of these confounding factors, the variability of m in weakly or non-precipitating clouds is likely associated with a complex network of interactions and dependencies that comprise entrainment (Mellado, 2017; Igel, 2024), making it hard to obtain direct process understanding from models that represent the underlying dynamics explicitly, e.g., three-dimensional large-eddy simulations. Thus, to understand m for weakly and non-precipitating stratocumulus better, we will base our work on a simple, zero-dimensional mixed-layer model (Lilly, 1968; Schubert et al., 1979; Bretherton and Wyant, 1997; Stevens, 2002). We will focus on the representation of the entrainment rate in such models and how it depends on L and N , which allows us to directly assess the individual aspects of the entrainment process (Nicholls and Turton, 1986; Turton and Nicholls, 1987; Bretherton et al., 2007)."*

3. Line 55: The boundary-layer depth mass budget can also depend on horizontal advection of ht (Caldwell et al., 2005).

   This probably depends on whether one considers the simulated volume of air in a Lagrangian framework or at a predefined location. We added the following statement: *"Lastly, s, qt, and ht can change due to large-scale advection."*

4. Line 217: This may need to be quantified. What value of negative cloud-base buoyancy flux was used to determine decoupling?

   We have added the following statement: *"These regions have been determined using the approach by Turton and Nicholls (1987), which diagnoses decoupling if the ratio of the integrated sub-cloud-layer buoyancy flux to the integrated cloud-layer buoyancy flux is smaller than −0.4."*

5. Line 226: Why is a cloud-base precipitation rate necessary to omit cases if we only care about decoupling? If a precipitating case remains fairly well-mixed (no significant negative cloud-base buoyancy flux) there is no need to get rid of that profile.

   We have clarified this sentence as: *"Since our approach does not consider losses in L due to surface precipitation, we neglect regions of the L-N phase space that are substantially affected by this process."*

6. Line 232: A decrease in the buoyancy jump does not necessarily guarantee an increase in entrainment rate.

   This is true, and is generally considered in the entrainment rate parameterization (8) and its components. Here, however, we are considering the general behavior of Δb with L as represented by (11), and not the entrainment rate directly.

Stronger buoyancy jumps can be created by more effective long-wave cooling (McMichael et al., 2019). Also, low cloud fraction environments may have diffuse inversion layers but reduced entrainment in the absence of radiatively- and evaporatively-driven turbulence.

These interactions are considered in (11) via Δs and Δq_t. However, these are assumed constant for Figs. 1 and 2. The influence of Δq_t is discussed in Sec. 3.3.

7. Line 270: Negative buoyancy near cloud base may not guarantee a decrease in entrainment, but may aid in decoupling. Turbulence production near cloud-top is the more critical quantity.

   As indicated by (5) to (8) and many detailed studies on this subject (e.g., Yamaguchi and Randall 2008), entrainment is not determined by (the small-scale) turbulence production at the cloud-top but the boundary layer's large-scale dynamics. Thus, changes in the integrated buoyancy flux ⟨B⟩ primarily determine the entrainment rate.

8. Lines 336-341: This paragraph makes it seem like the diurnal cycle was taken into consideration. I would recommend emphasizing the diurnal uncertainties that still exist in m.

   The interaction of clouds with solar radiation is probably the strongest motivation to study aerosol-cloud interactions. Not mentioning solar radiation in the summary/conclusion feels sacrilegious. To clarify that solar radiation has been neglected for our study, we amended the following statement: *"[…] Still, the additional nuance provided by these models should not be disregarded, as they help quantifying the effects that have been neglected here (e.g., interactive surface fluxes, changes in boundary layer depth, or the diurnal cycle of shortwave radiation). Thus, we advocate that the assessment of aerosol-cloud interactions should balance the use of complex and simple approaches by substantiating quantitative understanding with qualitative insight."*

References:

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McMichael LA, Mechem DB, Wang S, Wang Q, Kogan L, Teixeira J. Assessing the mechanisms governing the daytime evolution of marine stratocumulus using large-eddy simulation. Q J R Meteorol Soc. 2019; 145: 845–866. https://doi.org/10.1002/qj.3469

Yamaguchi, T., & Randall, D. A. (2008). Large-eddy simulation of evaporatively driven entrainment in cloud-topped mixed layers. *Journal of the Atmospheric Sciences*, *65*(5), 1481-1504.