

# Diurnal evolution of non-precipitating marine stratocumuli in an LES ensemble

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## Abstract.

We explore the cloud system evolution of non-precipitating marine stratocumuli with a focus on the impacts of the diurnal cycle and free-tropospheric (FT) humidity based on an ensemble of 244 large-eddy simulations generated by perturbing initial thermodynamic profiles and aerosol conditions. Cases are categorized based on their degree of decoupling and cloud liquid water path ( $LWP_c$ , based on model columns with cloud optical depths greater than 1). A budget analysis method is proposed to analyze the evolution of cloud water in both coupled and decoupled boundary layers. More coupled clouds start with relatively low  $LWP_c$  and cloud fraction ( $f_c$ ) but experience the least decrease in  $LWP_c$  and  $f_c$  during the daytime. More decoupled clouds undergo greater daytime reduction in  $LWP_c$  and  $f_c$ , especially those with higher  $LWP_c$  at sunrise because they suffer from faster weakening of net radiative cooling. During the nighttime, a positive correlation between FT humidity and  $LWP_c$  emerges, consistent with higher FT humidity reducing both radiative cooling and the humidity jump, both of which reduce entrainment and increase  $LWP_c$ . The  $LWP_c$  is more likely to decrease during the nighttime for larger  $LWP_c$  and greater inversion base height ( $z_i$ ), conditions under which entrainment dominates as turbulence develops. In the morning, the rate of the  $LWP_c$  reduction depends on the  $LWP_c$  at sunrise,  $z_i$ , and the degree of decoupling, with distinct contributions from subsidence and radiation.

## 1 Introduction

Subtropical marine stratocumuli cover vast areas of Earth's surface and play an important role in Earth's energy balance by reflecting solar radiation back to space. A cloud reflects more solar radiation when its liquid water is distributed among a larger number of aerosol particles to form more numerous and smaller cloud droplets (Twomey, 1974, 1977). This initial effect propagates to other cloud properties through a series of complex processes, e.g., suppression of precipitation formation (Albrecht, 1989; Pincus and Baker, 1994), enhancement of cloud-top entrainment (Bretherton et al., 2007; Wang et al., 2003), and an increase in solar absorption (Boers and Mitchell, 1994). These processes, all considered part of aerosol–cloud interactions (ACIs), may offset one another and their importance depends on the cloud's properties, its environment, and the time scale of interest (Stevens and Feingold, 2009).

From observations alone, it is difficult to identify and quantify the details of the aforementioned processes (e.g., Gryspeerdt et al., 2019; Wall et al., 2023), given the incomplete information of observed clouds and their environments, including co-  
25 varying meteorology and aerosols, and often in the form of snapshots rather than temporal evolution of the same cloud field (Stevens and Feingold, 2009; Mülmenstädt and Feingold, 2018). Despite recent efforts in inferring processes after constraining such co-variations (e.g., Zhang et al., 2022; Zhang and Feingold, 2023) and in quantifying the temporal evolution in the cloud responses to aerosol perturbations (e.g., Qiu et al., 2024; Smalley et al., 2024; Gryspeerdt et al., 2022), causality or process attribution remains a challenge. While opportunistic experiments, such as ship tracks, provide a way to observe the adjustment  
30 of cloud properties to additional aerosol, they are often limited in their ability to represent the wide range of conditions the marine stratocumuli reside in (e.g., Manshausen et al., 2022; Yuan et al., 2023; Toll et al., 2019).

Meanwhile, fine-scale numerical modeling has been used to provide process-level understanding of ACIs. Many previous works focused on case studies with aerosol perturbation experiments (Sandu et al., 2008; Caldwell and Bretherton, 2009; Wang and Feingold, 2009b; Wang et al., 2010; Chen et al., 2011; Yamaguchi et al., 2015; Possner et al., 2018; Kazil et al., 2021;  
35 Prabhakaran et al., 2023; Chun et al., 2023). Although much has been learned from these studies, they do not cover the wide range of real-world conditions.

Recent work by Feingold et al. (2016) and Glassmeier et al. (2019) took a different approach: exploring ACIs in large-eddy simulation (LES) ensembles of marine stratocumuli. Instead of performing aerosol perturbation experiments for each combination of meteorological factors, they used experiment design techniques to optimize the sampling of the initial condition space  
40 and later distilled the information regarding ACIs from both the individual and collective behaviors of ensemble members. The methodology is as follows: LESs of large numbers of idealized cases are each set up with different initial profiles of thermodynamic variables and aerosol that are generated by perturbing six parameters. These parameters, which we will introduce in detail in Section 2, were drawn independently from ranges of reasonable values, although the co-variability between parameters was not constrained to match the co-variability in nature. Other configurations are more idealized. For example, all cases  
45 share the same fixed SST, subsidence profile, and prescribed surface fluxes, but all are based on an observed case (DYCOMS-II RF02; Ackerman et al., 2009). The resulting clouds are realistic in terms of the range of LWP.

This approach has proved to be fruitful. Based on an LES ensemble of more than 150 nocturnal marine stratocumulus simulations, Glassmeier et al. (2019) found that several cloud properties (cloud fraction, cloud albedo, and relative cloud radiative effect) of ensemble members can be well described in the state-space of liquid water path (LWP) and cloud droplet number  
50 concentration ( $N_d$ ). Using the same LES ensemble, Hoffmann et al. (2020) showed that all non-precipitating cases in this ensemble approach a steady state LWP band from different parts of the state space: clouds starting with high LWP thin over time and clouds starting with low LWP, and possibly partial cloudiness, thicken over time. The authors further performed a budget analysis based on mixed-layer theory (MLT; Lilly, 1968) and demonstrated how the balance between radiative cooling, cloud-top entrainment warming and drying, and other processes shaped the  $N_d$ -dependence of steady state LWP. Glassmeier et al.  
55 (2021) estimated the magnitude and time scale of the LWP adjustment to an  $N_d$  perturbation from the collective behavior of the ensemble members and used them to infer biases in using ship-track to estimate the climatological forcing of anthropogenic aerosol. Hoffmann et al. (2023) explored the evolution of precipitating and non-precipitating stratocumuli in the space of albedo

and cloud fraction with another LES ensemble of 127 cases that used ERA5 climatology to constrain the initial thermodynamic profiles and employed interactive surface fluxes to improve the realism of the simulations.

60 The environmental conditions covered in the LES ensembles used by these works can be expanded. For instance, the free-troposphere (FT) in these simulations was fairly dry, while in reality a moister FT reduces cloud-top radiative cooling and modulates cloud-top entrainment warming and drying (Ackerman et al., 2004; Eastman and Wood, 2018). The ERA5 climatology used in Hoffmann et al. (2023) is based on all months, while the conditions during the months when the stratocumuli prevail are more relevant (Wood, 2012). In addition, the surface fluxes in those simulations were either constants prescribed  
65 following DYCOMS-II RF02 (Ackerman et al., 2009) or interactive but only responding to local wind fluctuations with calm mean winds, leading to relatively weak (but not unrealistic) surface fluxes (Hoffmann et al., 2023). Lastly, despite the insights gained from nocturnal simulations, the daytime behavior of marine stratocumulus population needs to be explored to understand the shortwave radiative effects of these clouds, which are more relevant to aerosol–cloud climate forcing and issues like marine cloud brightening (Latham, 1990; Feingold et al., 2024).

70 In this study, we explore the cloud system evolution within an LES ensemble that includes more realistic interactive surface fluxes, with a focus on the impacts of the diurnal cycle and FT humidity. The rest of this manuscript is organized as follows. We first introduce the model and simulation configurations in Section 2 and then provide an overview of the LES ensemble in Section 3. Next, we introduce a budget analysis method and present results in Section 4. With this method, we examine the nighttime and daytime evolution of individual cases in Section 5. A few specific issues will be discussed in Section 6, after  
75 which we end the paper with a summary in Section 7.

## 2 Model and simulations

All LESs for this study are performed using the System for Atmospheric Modeling (SAM; Khairoutdinov and Randall, 2003), version 6.10.10. SAM solves the anelastic Navier-Stokes equations in finite difference representation for the atmosphere on the Arakawa C grid. Similar to recent work by Yamaguchi et al. (2017) and Glassmeier et al. (2019), SAM is configured  
80 with a fifth-order advection scheme by Yamaguchi et al. (2011) and Euler time integration scheme for scalars, a second-order center advection scheme and with the third-order Adams-Bashforth time integration scheme for momentum, a 1.5-order TKE-based subgrid-scale (SGS) model (Khairoutdinov and Randall, 2003; Deardorff, 1980), a bin-emulating bulk two-moment microphysics parameterization (Feingold et al., 1998) assuming a log-normal aerosol size distribution with fixed size and width parameters, and the Rapid Radiative Transfer Model (RRTMG; Mlawer et al., 1997; Iacono et al., 2008) that is modified  
85 to take into account background profiles of temperature and moisture above the model domain top (Yamaguchi et al., 2015), which is critical for radiative transfer in shallow domain simulations.

Different from Yamaguchi et al. (2017) and Glassmeier et al. (2019), the SAM used for this work uses the total water mixing ratio (sum of vapor and hydrometeors) and the total number concentration (sum of aerosol and drop number concentrations) as prognostic variables to ensure better closure of the budgets associated with these two quantities for advection and several  
90 other physical processes (Morrison et al., 2016; Ovtchinnikov and Easter, 2009). As a result, the water vapor mixing ratio is

diagnosed from the total water and hydrometeor mixing ratios and the aerosol number concentration is diagnosed from the total, cloud droplet, and rain drop number concentrations. See the last paragraph of Section 2 in Yamaguchi et al. (2019) for a comprehensive summary of the advantages and disadvantages of this method.

As in Feingold et al. (2016) and Glassmeier et al. (2019), the LES ensemble members are generated from perturbed initial profiles. The initial profiles of liquid water potential temperature ( $\theta_l$ ) and total water mixing ratio ( $q_t$ ) are each constructed from two parts: a well-mixed boundary layer (BL) profile including a sharp jump at the top of the BL and a FT profile based on ERA5 climatology (Hersbach et al., 2020) and the Marine ARM GPCI Investigation of Clouds (MAGIC) campaign (Lewis et al., 2012; Zhou et al., 2015) observations. The initial BL  $\theta_l$  and  $q_t$  profiles are controlled by five parameters:  $\theta_l$  and  $q_t$  in the BL and their jumps,  $\Delta\theta_l$  and  $\Delta q_t$ , across the inversion base at the height of  $h_{\text{mix}}$ . See Appendix A for details on the FT profiles and the construction of the complete profiles. The initial aerosol number mixing ratio, specified by a sixth parameter,  $N_a$ , is uniform throughout the domain. The initial horizontal wind speed is  $0 \text{ m s}^{-1}$  everywhere. With this simplified configuration, there is no shear in the mean wind profile in our simulations to produce TKE, making the turbulence closer to a free convection.

Hundreds of initial profiles are set up from sets of these six parameters randomly and independently drawn from their ranges: BL  $\theta_l$  is drawn from 284 to 294 K, BL  $q_t$  from 6.5 to 10.5  $\text{g kg}^{-1}$ ,  $\Delta\theta_l$  from 6 to 10 K,  $\Delta q_t$  from  $-10$  to  $0 \text{ g kg}^{-1}$ ,  $h_{\text{mix}}$  from 500 to 1300 m, and  $N_a$  from 30 to 500  $\text{mg}^{-1}$ . Compared with the parameter ranges used in Glassmeier et al. (2019), the range for  $\Delta q_t$  now covers  $-6$  to  $0 \text{ g kg}^{-1}$  to include conditions with more humid FT. All initial profiles with (1) height of lifted condensation level ( $z_{\text{LCL}}$ ) between around 225 m and 1075 m, (2) a saturated layer (i.e.,  $h_{\text{mix}} > z_{\text{LCL}}$ ), and (3) FT  $\theta_l$  and  $q_t$  profiles falling between the minimum and maximum of the ERA5 climatological profiles are simulated with the lower boundary conditions and large-scale forcings described below, which are the same for all simulations.

First, the surface fluxes of sensible heat, latent heat, and momentum are computed based on Monin-Obukhov similarity. The sea surface temperature (SST) is fixed for all simulations at 292.4 K. Since the mean horizontal wind speed is close to  $0 \text{ m s}^{-1}$  in the lowest model level as a result of the simulation setup, a constant horizontal wind speed of  $7 \text{ m s}^{-1}$  is added to the surface local wind fluctuation when calculating sensible and latent heat fluxes to obtain realistic flux values. Both this wind speed and the aforementioned SST are based on the ERA5 climatology from the same region and time period as described in Appendix A. This wind speed is also comparable to that in Kazil et al. (2016), which is produced by specifying the geostrophic wind velocity following DYCOMS-II RF01 (Stevens et al., 2005). Second, a constant surface aerosol flux of  $70 \text{ cm}^{-2} \text{ s}^{-1}$ , based on estimates by Kazil et al. (2011), is prescribed to offset the loss of aerosol through coalescence scavenging (Wang et al., 2010). Lastly, a time-invariant subsidence profile is imposed as

$$w_s = \begin{cases} -Dz, & z < 2000 \text{ m} \\ -0.0075 \text{ m s}^{-1}, & z \geq 2000 \text{ m}, \end{cases} \quad (1)$$

where the divergence  $D = 3.75 \times 10^{-6} \text{ s}^{-1}$ . No other large-scale forcing is applied in the simulations.

The simulation domain is  $48 \times 48 \times 2.5 \text{ km}^3$  in the x-, y-, and z-dimensions with 200-m horizontal and 10-m vertical grid spacings. The horizontal grid spacing is relatively coarse while the simulations for typical marine stratocumuli do not converge until much finer resolution (Stevens et al., 2000; Mellado et al., 2018; Matheou and Teixeira, 2019); it leaves a

significant fraction of small-scale turbulence to be parameterized by the SGS model. However, there is empirical evidence that  
125 simulations with such a coarse resolution could be useful. Wang and Feingold (2009a) showed that the differences between  
closed- and open-cell stratocumuli captured by simulations using a 300-m horizontal grid spacing are similar to those using  
a 100-m horizontal grid spacing. Also, Pedersen et al. (2016) found that anisotropic grids may perform better in simulating  
the anisotropic turbulence in the inversion layer. Their findings are consistent with Mellado et al. (2018) who reported that  
LES of the DYCOMS-II RF01 case (Stevens et al., 2005) with a coarser vertical resolution requires a larger aspect ratio to  
130 match observations. The capability of the coarse resolution and large aspect ratio to reproduce a single case certainly does not  
guarantee good performance for a range of environmental conditions, which essentially requires a correct representation of the  
sensitivity of clouds to environmental conditions. However, considering all these factors, we choose to use a 200-m horizontal  
grid spacing to pair with the 10-m vertical grid spacing. With this choice we are able to afford a larger number of ensemble  
members.

135 The simulation domain uses periodic lateral boundary conditions and has a damping layer from 2 km to domain top. The  
domain resides at 25°N, 235°W. All simulations are initialized at 18:40 local time (LT; 03:00Z) and then advanced for 24 h  
with a 1-s time step. Sunrise occurs between 05:23 and 05:24 LT and sunset occurs between 18:36 and 18:37 LT, following the  
diurnal cycle on May 16 at the location of the domain. The location of the domain and the day-of-year of the simulation are  
selected based on the centers of the region and the time period during which the ERA5 climatology is used to configure the  
140 simulations. (See Appendix A.)

For this study, we focus on non-precipitating cases, defined by a cloud-base precipitation rate of less than 0.5 mm day<sup>-1</sup>  
(Wood, 2012). We further exclude simulations with multi-layer clouds, including surface fog. Finally, we discard simulations  
where the cloud top ever reaches 1.9 km, 100 m below the lower bound of the damping layer, where the damping could  
potentially interfere with the dynamics in the hydrometeor-free Entrainment Interfacial Layer (Moeng et al., 2005; Haman  
145 et al., 2007; J. Kurowski et al., 2009). This leaves 244 cases for further investigation. The first 2-h of each simulation is  
excluded as the spin-up.

### 3 Overview of LES ensemble behavior

In this section, we present an overview of the evolution of the 244 non-precipitating cases in our LES ensemble. Following  
Glassmeier et al. (2019), we start with the trajectories in the plane of cloud droplet number concentration ( $N_d$ ) and cloud liquid  
150 water path ( $LWP_c$ ) (Figure 1). Both variables are based on “cloudy columns”, which are defined as columns with cloud optical  
depths greater than 1. During the nighttime, the cases that start with low  $LWP_c$  experience an increase in  $LWP_c$ , while the  
behavior of the high  $LWP_c$  cases is not immediately clear. The nighttime cloud fractions ( $f_c$ ) are usually high. At sunrise, 67%  
of cases have  $f_c > 0.99$  and 86% cases have  $f_c > 0.95$ . During the daytime, all cases start to lose  $LWP_c$  and  $f_c$  right after  
sunrise or in the early morning. Between noon and 15:00, about 89% cases reach their lowest daytime  $LWP_c$ . In the last hour  
155 of the simulation, 95% cases are gaining  $LWP_c$ . Very low  $f_c$  occurs for many cases in the afternoon. The variation in  $N_d$  is  
rather weak for most cases.

### 3.1 Categorization of cases

To provide a more consolidated view of the evolution, we categorize the cases by their degree of decoupling in the morning because the diurnal decoupling (Nicholls, 1984; Turton and Nicholls, 1987) is a common feature of the cloud-topped marine BL diurnal cycle and we expect different diurnal cycles between more coupled and more decoupled cases. We compute the relative decoupling index (denoted with  $\mathcal{D}$ ) defined by Kazil et al. (2017),

$$\mathcal{D} = \frac{\overline{z_{cb}} - \overline{z_{LCL}}}{\overline{z_{LCL}}}, \quad (2)$$

where  $\overline{z_{cb}}$  and  $\overline{z_{LCL}}$  are the mean cloud base height and mean lifting condensation level (LCL, determined from conditions in the lowest model level), both averaged for cloudy columns. This index is a variant of the subcloud decoupling index,  $\overline{z_{cb}} - \overline{z_{LCL}}$ , originally proposed by Jones et al. (2011). A small value of  $\mathcal{D}$  is more likely to be coupled while a large value of  $\mathcal{D}$  is more decoupled.

Figure 2a shows  $\mathcal{D}$  at 09:40 LT in the plane of  $LWP_c$  and domain-mean inversion base height ( $z_i$ , based on levels with the greatest vertical gradient of liquid water static energy in individual columns) at sunrise. Clouds with greater  $\mathcal{D}$  tend to occur in deeper BLs; many of these clouds experience very low daytime  $f_c$  minima (Figure 2b) unless they start with very high  $LWP_c$  at sunrise, although most cases have daytime  $f_c$  maxima that are close to overcast (not shown). Based on this finding, we divide the cases into three categories based on  $\mathcal{D}$  at 09:40 and  $LWP_c$  at sunrise (05:22): (1) loDloL ( $\mathcal{D} \leq 1$ ), (2) hiDloL ( $\mathcal{D} > 1$  and  $LWP_c \leq 180 \text{ g m}^{-2}$ , the highest  $LWP_c$  for the loDloL category), and (3) hiDhiL ( $\mathcal{D} > 1$  and  $LWP_c > 180 \text{ g m}^{-2}$ ) for further analysis (Figure 2c). Figure 2d shows the time series of  $\mathcal{D}$  by category. During the nighttime, the medians of  $\mathcal{D}$  for all three categories are relatively small, suggesting more coupled conditions. Some cases in the hiDloL and hiDhiL categories always exhibit a higher degree of decoupling during the night. During the daytime,  $\mathcal{D}$  for all three categories increases into the afternoon. Overall, cases in the loDloL category experience weaker decoupling with their  $\mathcal{D}$  start to increase at a slower rate from a later time, compared with other two categories. Figure 2e shows the time series of median  $\overline{z_{cb}}$  and median  $\overline{z_{LCL}}$  by category. During the daytime, the median  $\overline{z_{LCL}}$  decreases for both hiDloL and hiDhiL, consistent with a strengthening decoupling limiting the surface based mixed layer. This does not happen to loDloL. Also, both hiDloL and hiDhiL categories experience dramatic diurnal changes in median  $\overline{z_{cb}}$  and the cloud depth, approximated with  $z_i - \overline{z_{cb}}$ . Even though the categorization is based on  $\mathcal{D}$  at 09:40 LT, it nicely separates the loDloL category from the other two categories through the daytime (Figure 2e).

We include the profiles at sunrise and 13:30 LT from two example cases from loDloL and hiDloL in the supplementary material (Figure S1). They show many features consistent with the observed profiles for coupled and decoupled marine stratocumulus-topped BLs (e.g., Nowak et al., 2021), especially the decoupled conditions at 13:30 LT for the case from the hiDloL category: the stratified layer between the relatively well-mixed cloud layer and surface-based mixed layer, the pronounced stratification in the  $q_t$  profile (Figure S1f), and the weak fluxes above the surface-based mixed layer through cloud-top (Figure S1g).

### 3.2 Cloud evolution by category

Figures 3a and 3b display the average time series of  $LWP_c$  and  $f_c$  for three categories. Among the three categories, the loDloL category shows the lowest nighttime  $LWP_c$  and  $f_c$ . However, this category also has the smallest decrease in  $LWP_c$  and  $f_c$  during the day. By contrast, the hiDloL category has greater  $LWP_c$  and nearly overcast conditions ( $f_c > 0.99$ ) at sunrise but experiences a much more dramatic decrease in both  $LWP_c$  and  $f_c$ . The hiDhiL category has the highest  $LWP_c$  and  $f_c$  at sunrise among all three categories. This category also shows diurnal fluctuations of large amplitude in both  $LWP_c$  and  $f_c$  with the daytime minimum between the loDloL and hiDloL categories for both variables. It reaches its lowest  $LWP_c$  and lowest  $f_c$  latest in the day among all three categories. At the end of the simulation, all three categories experience a recovery of both  $LWP_c$  and  $f_c$ . At this stage, they all have similar  $LWP_c$ , indicating that the diurnal cycle imposes a strong constraint to narrow the range of  $LWP_c$ , consistent with previous findings (e.g., van der Dussen et al., 2013). In contrast,  $f_c$  differs significantly: the loDloL category has the highest  $f_c$  and the hiDloL category the lowest  $f_c$ .

When plotted in the plane of  $f_c$  and the cloud depth ( $z_i - z_{cb}$ ) (Figure 3c), the mean trajectories of the three categories produce loops of different sizes. The trajectory of the loDloL category makes the smallest loop, which can be interpreted as the least diurnal variation in cloud aspect ratio (the ratio between the cloud depth and  $f_c$ ). Clouds in the hiDloL and hiDhiL categories experience greater variation in the aspect ratio, more so for the hiDloL categories. We examine the 3-D cloud fields for selected cases from these two categories and find that clouds in both categories evolve into a cumulus-rising-into-stratocumulus structure by noon (not shown). The cloud bases of the cumuli lower slightly while the stratocumuli continue to thin and lose  $f_c$ . This transition lowers  $\overline{z_{cb}}$  and leads to the segments in the trajectories where  $f_c$  decreases but cloud depth starts to recover. As the clouds develop towards sunset, they regain  $f_c$  to become stratiform again.

These behaviors agree with observed diurnal cycles of marine stratocumuli. As summarized in Section 2.b.5 in Wood (2012), the marine stratocumuli near the coast show weaker diurnal variability in LWP and cloud fraction and are more coupled to the sea surface in shallower BLs, while the clouds observed downwind of the subtropical maxima show stronger diurnal variability in deeper and more decoupled BLs. The similar range of evolution between our LES ensemble and the observations are not necessarily driven by the same mechanisms because our ensemble is limited by the experiment design, especially the simplified treatment of the wind profile and the lack of realistic co-variability between the environmental conditions in simulation configurations, e.g., between SST and BL depth, between subsidence and inversion strength (Wood and Bretherton, 2006), and between FT  $\theta_1$  and FT  $q_t$  (Eastman and Wood, 2018). Still, the capability of our LES ensemble to capture a variety of features in observed diurnal cycles of marine stratocumuli suggests that analysis of the statistical behavior of this data set is valuable.

### 3.3 Surface fluxes

To end this overview, we summarize the surface fluxes in the simulations (Figures S2a and S1b). At the end of the first 2-h of the simulations, both the ranges of surface sensible heat flux (SHF) and latent heat flux (LHF) from all simulations encompass the values prescribed in the DYCOMS-II RF02 case (i.e., 16 and 93 W m<sup>-2</sup>, respectively). Afterwards, the SHF decreases over

time until late afternoon as the SHF effectively brings the BL air temperature towards the SST. The SHF is the strongest in the loDloL category, followed by the hiDhiL and then the hiDloL categories. This is because the shallower BLs in our ensemble also tend to be colder due to the criteria applied in the initial profiles. (For example, for a shallow BL to be initially saturated, its  $z_{\text{LCL}}$  needs to be lower, which is more likely when the initial BL  $\theta_1$  is low. See more in Section 2.) LHF shows a smaller  
 225 relative change throughout the day. During the nighttime, the LHF for the loDloL category remains quite steady and that for the hiDloL category even increases as the turbulence spins up. The LHF is also the strongest in the loDloL category, while the LHF from the other two categories are comparable at all times.

Following Eq. 1 in Lilly (1968), the domain-mean surface sensible and latent heat fluxes (SHF and LHF) can be written as

$$\text{SHF} = C_T U (\theta_{\text{SST}} - \theta_{\text{air}}), \text{LHF} = C_q U (q_{\text{sat}}(\text{SST}) - q_{\text{v,air}}), \quad (3)$$

230 where the wind speed used for surface flux calculations ( $U$ ), lowest model level air temperature and water vapor mixing ratio ( $\theta_{\text{air}}$  and  $q_{\text{v,air}}$ ) are also the domain-means. Recall that in our simulations, the SST is 292.4 K and equivalent to a potential temperature,  $\theta_{\text{SST}}$ , of 290.9 K given the surface pressure used in the simulations. (See Appendix A.) The saturation mixing ratio at SST ( $q_{\text{sat}}(\text{SST})$ ) is approximately constant due to the negligible drift of surface pressure. Comparing Figures S2c–S2f with Figures S2a–S2b, it is clear that the evolutions of the SHF and LHF in our simulations are driven primarily by  $(\theta_{\text{SST}} - \theta_{\text{air}})$   
 235 and  $(q_{\text{sat}}(\text{SST}) - q_{\text{v,air}})$ , respectively. On average, the transfer coefficients for SHF ( $C_T$ ) and for LHF ( $C_q$ ) that are diagnosed from Eq. (3) decrease slightly over time, although cases with  $\theta_{\text{air}}$  very close to  $\theta_{\text{SST}}$  see larger fluctuations in  $C_T$ .  $U$  mostly ranges between 7 than  $7.3 \text{ m s}^{-1}$  throughout the day (Figure S3) because they result from the summation of relatively weak local wind velocities and a large constant wind speed ( $7 \text{ m s}^{-1}$ , see Section 2). Our results are consistent with the findings reported by Kazil et al. (2014) for a closed-cell stratocumulus case.

#### 240 4 Budget analysis for evolution of LWP<sub>c</sub>

We perform a detailed budget analysis to understand the simulated LWP<sub>c</sub> evolution. Previous studies used mixed-layer theory (MLT) to calculate the LWP<sub>c</sub> tendency from the tendencies of BL mean liquid water potential temperature ( $\theta_l$ ) and total water mixing ratio ( $q_t$ ) as well as the motion of  $z_i$  (Wood, 2007; Caldwell and Bretherton, 2009; Ghonima et al., 2015; Hoffmann et al., 2020). In particular, van der Dussen et al. (2014) derived the LWP budget equations focusing only on the adiabatic  
 245 cloud layer by replacing the surface flux term with a cloud-base term. Many clouds in our simulations occur in decoupled BLs with partial cloudiness, especially during the daytime. Therefore, we apply the MLT-based approach to the “cloud volume” (CV), which we define for a given time  $t$  as the volume consisting of all cloudy columns between  $z_i(t)$  and the first grid box interface below  $\overline{z_{\text{cb}}}(t)$  (Figure S4). The choice of this volume is inspired by previous work showing success in assuming the cloud layer being well-mixed in decoupled BLs (Turton and Nicholls, 1987; Bretherton and Wyant, 1997). It is also based on  
 250 our observation that in our simulations the entrainment velocity, diagnosed as

$$w_e = \frac{dz_i}{dt} - w_s(z_i), \quad (4)$$



is rarely negative, even at its weakest point in the late afternoon, meaning there is always some turbulent motion near the cloud top that mixes the air between the cloud layer and the FT. Different from these two previously mentioned papers and van der Dussen et al. (2014), we only assume the cloudy region of the cloud layer is well-mixed to deal with partial cloudiness. This is an alternative method to Chun et al. (2023), where the authors diagnosed the LWP budget by first assuming an overcast cloud in a well-mixed BL and then attributed the difference between actual LWP tendency and the sum of diagnosed terms to partial cloudiness and deviation from adiabaticity clouds. The specific definition of the CV base takes full advantage of quantities reported by SAM at the grid box interface to reduce the impacts of vertical interpolation. The CV depth defined this way is within a few percent of the actual cloud depth. We first show the derivation of CV budgets and then show results from both the BL and CV budgets.

#### 4.1 Derivation

Consider a scalar quantity  $\phi$  (in our case  $\theta_1$  or  $q_t$ ) at time  $t$  in a volume consisting of a set of model columns covering a fraction of the domain area ( $f(t)$ , which is 1 for the BL budget and  $f_c$  for the CV budget) between the volume base height  $z_0(t)$  (which is 0 for the BL budget and CV base height for the CV budget) and  $z_i(t)$ . Inspired by the BL total water budget in Appendix B in Kazil et al. (2016), we build a budget for the mean scalar quantity in this volume,  $\langle\phi\rangle$ , from the budgets of the total amount of this scalar quantity,  $\Phi$ , and total air mass,  $M$ , in this volume. Since SAM solves the anelastic equations of motion, where the air density  $\rho_0$  only changes with height,

$$\Phi(t) = f(t) \int_{z_0(t)}^{z_i(t)} \rho_0(z) \phi(z, t) dz, \quad (5)$$

and

$$M(t) = f(t) \int_{z_0(t)}^{z_i(t)} \rho_0(z) dz = \langle\rho_0\rangle(t) f(t) h(t), \quad (6)$$

where  $\langle\rho_0\rangle(t)$  is the mean air density of the volume,  $\phi(z, t)$  is the time-dependent mean  $\phi$  profile of the volume, and  $h(t) = z_i(t) - z_0(t)$  is the volume thickness. Then,

$$\langle\phi\rangle = \Phi/M \quad (7)$$

$$\Rightarrow \frac{d\langle\phi\rangle}{dt} = \frac{1}{M} \frac{d\Phi}{dt} - \frac{\Phi}{M^2} \frac{dM}{dt} = \frac{1}{M} \frac{d\Phi}{dt} - \frac{\langle\phi\rangle}{M} \frac{dM}{dt}. \quad (8)$$

(Starting from Eq. (7), we omit “(t)” for most time-dependent variables to simplify the notation.) The  $\langle\phi\rangle$  tendency can also be decomposed into the contributions from various processes

$$\frac{d\langle\phi\rangle}{dt} = \sum_P \left. \frac{d\langle\phi\rangle}{dt} \right|_P = \sum_P \left( \left. \frac{1}{M} \frac{d\Phi}{dt} \right|_P - \left. \frac{\langle\phi\rangle}{M} \frac{dM}{dt} \right|_P \right), \quad (9)$$

where the processes  $P$  include volume-top entrainment (ENTR), processes at volume sides (LAT for lateral), radiation (RAD),  
 280 subsidence (SUBS), and processes at the volume base: transport flux at volume base (BASE), precipitation flux at volume base  
 (PRCP), and a term tracking the impacts of the rising or lowering of the volume base (BM, standing for “base motion”). The  
 $d\langle\phi\rangle/dt$  due to each of these seven processes can be calculated from  $d\Phi/dt$  and  $dM/dt$  due to the same process via Eq. (9).

When we apply this approach to the budget of  $\langle\phi\rangle$  in a CV,  $f$  is equivalent to cloud fraction  $f_c$  and several terms are quite  
 straightforward to estimate accurately. The RAD and BASE terms for  $\Phi$  are directly computed from the 3-D modeled fields of  
 285 radiative heating rate, vertical velocity, and  $\phi$ , and neither process modifies  $M$ . Although we are dealing with non-precipitating  
 cases, we retain the PRCP terms to minimize the residual. The BM term is calculated following

$$\left.\frac{d\langle\phi\rangle}{dt}\right|_{\text{BM}} = \frac{1}{M} \left.\frac{d\Phi}{dt}\right|_{\text{BM}} - \frac{\langle\phi\rangle}{M} \left.\frac{dM}{dt}\right|_{\text{BM}} = -\frac{\rho_0(z_0)\phi(z_0,t)f_c}{M} \frac{dz_0}{dt} + \frac{\rho_0(z_0)\langle\phi\rangle f_c}{M} \frac{dz_0}{dt}. \quad (10)$$

The SUBS term for  $\Phi$  is diagnosed by applying the Reynolds Transport Theorem (RTT),

$$\begin{aligned} \left.\frac{d\Phi}{dt}\right|_{\text{SUBS}} &= f_c \int_{z_0}^{z_i} \rho_0(z) \left.\frac{d\phi(z,t)}{dt}\right|_{\text{SUBS}} dz + \rho_0(z_i)\phi(z_i,t)f_c \left.\frac{dz_i}{dt}\right|_{\text{SUBS}} \\ 290 \quad &= f_c \int_{z_0}^{z_i} \rho_0(z) \left.\frac{d\phi(z,t)}{dt}\right|_{\text{SUBS}} dz + \rho_0(z_i)\phi(z_i,t)f_c w_s(z_i), \end{aligned} \quad (11)$$

where  $d\phi(z,t)/dt|_{\text{SUBS}}$  is calculated by applying SAM’s subsidence subroutine to the  $\phi(z,t)$  profile. Note that although the  
 CV base is defined to be close to  $\overline{z_{\text{cb}}}$ , which evolves due to many processes, this choice of CV base is to avoid applying MLT  
 later to deeper stratified layers. In other words, as long as the CV base sits in a well-mixed layer, there is no need to update  
 its height based on the cloud base height, and our choice to move it following the cloud base height is arbitrary. So, physical  
 295 processes do not directly move the CV base and there is no  $dz_0/dt$  in the terms for any processes but the BM term. The SUBS  
 term for  $M$  is

$$\left.\frac{dM}{dt}\right|_{\text{SUBS}} = \rho_0(z_i)f_c w_s(z_i). \quad (12)$$

The ENTR flux of  $\Phi$  can be parameterized as

$$\left.\frac{d\Phi}{dt}\right|_{\text{ENTR}} = \rho_{0,e}\phi_e f_c w_e, \quad (13)$$

300 where  $w_e$  is the entrainment velocity estimated from Eq. (4) and  $\rho_{0,e}$  and  $\phi_e$  are an air density and a  $\phi$  value that are relevant  
 to the entrainment flux of  $\phi$ . (Subscript “e” stands for “entrainment”, as in  $w_e$ .) Combined with the ENTR term for  $M$ , the  
 contribution of entrainment to the  $\langle\phi\rangle$  tendency is

$$\left.\frac{d\langle\phi\rangle}{dt}\right|_{\text{ENTR}} = \frac{1}{M} \left.\frac{d\Phi}{dt}\right|_{\text{ENTR}} - \frac{\langle\phi\rangle}{M} \left.\frac{dM}{dt}\right|_{\text{ENTR}} = \frac{\rho_{0,e}\phi_e f_c w_e}{M} - \frac{\rho_0(z_i)\langle\phi\rangle f_c w_e}{M}. \quad (14)$$

Assuming constant  $\rho_0$  and overcast conditions ( $f_c = 1$ ), Eq. (14) reduces to

$$305 \quad \left.\frac{d\langle\phi\rangle}{dt}\right|_{\text{ENTR}} = \frac{1}{h} w_e \Delta\phi, \quad (15)$$

where  $\Delta\phi$  is the  $\phi$  jump at the volume top. Previous work used  $\phi$  values at certain levels above and below  $z_i$  (usually denoted as  $z_+$  and  $z_-$ ) to calculate the jump (Yamaguchi et al., 2011; Bretherton et al., 2013). Comparing Eqs. (14) and (15), it seems that we can follow a similar method to find a level above  $z_i$  and use the  $\phi$  and  $\rho_0$  at this level in place of  $\phi_e$  and  $\rho_{0,e}$ . However, it is unclear what formula can be used to reliably find this level for all coupled and decoupled conditions in our simulations.

310 With Eq. (14), the challenging part is the entrainment flux term,  $d\Phi/dt|_{\text{ENTR}}$ . For now, we approximate it with the entrainment flux term for the BL. We first apply Eq. (9) to the whole BL. In this case, the BM and LAT terms vanish and the BASE term is calculated from the surface fluxes reported by SAM (denoted with SURF term). Because all terms other than the ENTR term are relatively easy to estimate directly and accurately, we don't keep a residual term, essentially lumping any residual into the ENTR term. So,

$$315 \quad \left. \frac{d\langle\phi\rangle_{\text{BL}}}{dt} \right|_{\text{ENTR}} = \left. \frac{d\langle\phi\rangle_{\text{BL}}}{dt} \right|_{\text{RAD}} + \left. \frac{d\langle\phi\rangle_{\text{BL}}}{dt} \right|_{\text{SUBS}} + \left. \frac{d\langle\phi\rangle_{\text{BL}}}{dt} \right|_{\text{SURF}} + \left. \frac{d\langle\phi\rangle_{\text{BL}}}{dt} \right|_{\text{PRCP}}. \quad (16)$$

Then,

$$\left. \frac{d\Phi_{\text{BL}}}{dt} \right|_{\text{ENTR}} = \langle\phi\rangle_{\text{BL}} \left. \frac{dM_{\text{BL}}}{dt} \right|_{\text{ENTR}} + M_{\text{BL}} \left. \frac{d\langle\phi\rangle_{\text{BL}}}{dt} \right|_{\text{ENTR}} = \rho_0(z_i) \langle\phi\rangle_{\text{BL}} w_e + M_{\text{BL}} \left. \frac{d\langle\phi\rangle_{\text{BL}}}{dt} \right|_{\text{ENTR}}. \quad (17)$$

We use this term in place of  $d\Phi/dt|_{\text{ENTR}}$  in the CV budget.

Regarding the LAT term, we can write

$$320 \quad \left. \frac{\langle\phi\rangle}{M} \frac{dM}{dt} \right|_{\text{LAT}} = \frac{\langle\phi\rangle h \langle\rho_0\rangle}{M} \frac{df_c}{dt} = \frac{\langle\phi\rangle}{f} \frac{df_c}{dt}. \quad (18)$$

Finally, we attribute all the remaining  $\langle\phi\rangle$  tendency to  $d\Phi/dt|_{\text{LAT}}$  to close the budget without the need for a residual term.

Thus far, we have been tracking the budget of  $\langle\theta_1\rangle$  and  $\langle q_t\rangle$  and have not invoked MLT. Next, we apply the following equation for the  $\text{LWP}_c$  tendency, derived based on MLT, to the CV,

$$\frac{d\text{LWP}_c}{dt} = \Gamma_1 \langle\rho_0\rangle (z_i - z_{\text{cb}}) \left[ \frac{dz_i}{dt} - \left( \frac{dz_{\text{cb}}}{d\langle q_t\rangle} \frac{d\langle q_t\rangle}{dt} + \frac{dz_{\text{cb}}}{d\langle\theta_1\rangle} \frac{d\langle\theta_1\rangle}{dt} \right) \right], \quad (19)$$

325 where  $z_{\text{cb}}$  is the mean cloud base height,  $\Gamma_1$  is the liquid water adiabatic lapse rate, and  $dz_{\text{cb}}/d\langle\theta_1\rangle$  and  $dz_{\text{cb}}/d\langle q_t\rangle$  are based on the derivation in Ghonima et al. (2015) and follow similar notations in Hoffmann et al. (2020). In the calculation of  $\Gamma_1$ ,  $dz_{\text{cb}}/d\langle\theta_1\rangle$ , and  $dz_{\text{cb}}/d\langle q_t\rangle$ , the actual cloud-base air temperature and pressure are used. We decompose  $dz_i/dt$  into the sum of  $w_e$  and  $w_s$ , substitute  $d\langle q_t\rangle/dt$  and  $d\langle\theta_1\rangle/dt$  with the sum of individual budget terms diagnosed earlier, and finally group the  $dz_i/dt$ ,  $d\langle q_t\rangle/dt$ , and  $d\langle\theta_1\rangle/dt$  terms on the right-hand side of Eq. (19) by processes. Budget terms are diagnosed at the end of  
330 each simulation hour (local time 40 min past each hour). A residual (RES) term is required to close the  $\text{LWP}_c$  budget.

## 4.2 BL budgets

Before presenting results for the CV budgets, we briefly introduce the diurnal cycles of the BL budgets, including the BL  $\langle\theta_1\rangle$  and  $\langle q_t\rangle$  budgets and the  $\text{LWP}_c$  budget when they are used in Eq. (19). These results serve as a reference for the CV budgets in the next subsection.

335 The BL  $\langle \theta_1 \rangle$  and  $\langle q_t \rangle$  budgets share similarity between the three categories, i.e., loDloL, hiDloL, and hiDhiL (Figure 4). For the BL  $\langle \theta_1 \rangle$  budget (left column in Figure 4), RAD and ENTR are the leading terms during the nighttime. After sunrise, RAD quickly changes from cooling to warming, while ENTR warming weakens at a slower rate, leading to a peak in positive net BL  $\langle \theta_1 \rangle$  tendency in the morning. For the BL  $\langle q_t \rangle$  budget (right column Figure 4), ENTR and SURF are the leading terms throughout the day. After sunrise, ENTR drying weakens faster than SURF moistening, leading to a peak in positive net BL  
 340  $\langle q_t \rangle$  tendency between noon and 15:00 LT. Recall that in MLT, the subsidence has zero contributions to the tendencies of both the mixed-layer  $\langle \theta_1 \rangle$  and mean  $\langle q_t \rangle$ . In our case, the contributions are not zero but still small compared with leading terms.

Figure 5 shows the  $LWP_c$  budgets when the BL  $\langle \theta_1 \rangle$  and  $\langle q_t \rangle$  budgets are used in Eq. (19). Comparing the actual  $LWP_c$  tendency diagnosed from the time series of  $LWP_c$ , the sum of the ENTR, RAD, SUBS, and SURF terms, and the RES term in the right column of Figure 5, applying MLT to the BL achieves fairly good closure during the nighttime for the loDloL category  
 345 and between 02:00 and sunrise for the hiDloL category, but not during the daytime when the BLs are more decoupled.

The left column in Figure 5 shows the actual  $LWP_c$  tendency as well as the contributions from the RAD, ENTR, SUBS, and SURF terms. During the nighttime, the most distinct feature is that the SUBS term is much more important relative to other terms in the  $LWP_c$  budget than in the BL  $\langle \theta_1 \rangle$  and  $\langle q_t \rangle$  budgets. This is due to the strong negative contribution by the subsidence to the  $dz_i/dt$  term in Eq. (19). It is more negative for the hiDloL and hiDhiL categories because cases in these two categories  
 350 have a higher  $z_i$  and thus a stronger subsidence due to the subsidence profile we impose. The ENTR term is comparable to other terms because its strong warming and drying effect (Figure 4) is offset by its positive contribution to the  $dz_i/dt$  term. See Figure S5. We do not discuss the results for the daytime due to the large residual.

### 4.3 CV $\langle \theta_1 \rangle$ and $\langle q_t \rangle$ budgets

We first present the diurnal cycles of CV  $\langle \theta_1 \rangle$  and  $\langle q_t \rangle$  budgets averaged by category (Figure 6). Similar to the BL budgets,  
 355 the ENTR and RAD terms are the leading terms for the CV  $\langle \theta_1 \rangle$  budget during the nighttime. Both weaken after sunrise, with RAD cooling weakening faster. The ENTR warming decreases steadily towards late afternoon and becomes stronger before sunset. The main difference from the BL budgets in the left column of Figure 4 is that RAD is mostly cooling during the daytime because much of the warming effect by RAD occurs in the subcloud layer and is excluded in the CV  $\langle \theta_1 \rangle$  budget. This warming strengthens the stratification of the subcloud layer, weakens the turbulent motion, and limits its impacts on the CV.  
 360 The remaining effects of this subcloud warming on the CV are accounted as transport in BASE and LAT terms. (See Figure S6 for an example.) The RAD cooling becomes stronger after around 09:00 or 10:00. It continues to strengthen through the rest of the day for the loDloL and hiDhiL categories, even though the  $LWP_c$  does not recover until afternoon (Figure 3a). This trend is dominated by the trend in CV-integrated radiative heating rates (not shown). For the hiDloL category, there is a second weakening-strengthening cycle. This is a signature of the rapid lowering of  $\overline{z_{cb}}$  in this category as the stratiform parts of the  
 365 clouds shrink and cumulus parts dominate (see Section 3 and Figure 2e) and, as a result, the total radiative divergence for the CV is distributed over a deeper layer. Note that due to subsidence and the growing of  $z_i$ , the FT in all our simulations becomes drier over time. (FT  $q_t$  values at the end of the simulations are between 64% and 85% of those at sunrise.) This effect likely also modulates the balance between longwave cooling and shortwave absorption.

As the ENTR term for the CV  $\langle \theta_1 \rangle$  continues to decrease after the radiation passes its morning weakest point, the BASE-n-LAT term starts to play a more significant role (left column in Figure 6). This term is defined as the sum of the BASE and LAT terms. It represents the processes associated with the interface between the CV and the rest of the BL (i.e., CV base and lateral sides). It shows an opposite trend from the RAD term and becomes the main term balancing the radiation in the afternoon. This can be interpreted as follows: while there is not enough kinetic energy for mixing across the inversion base, the radiative cooling in the CV still couples with the dynamics inside the BL.

For  $\langle q_t \rangle$ , the ENTR and BASE-n-LAT terms are the leading terms (right column of Figure 6). Unlike the BASE-n-LAT term for the  $\langle \theta_1 \rangle$  budget, which can warm or cool the CV at different times, the BASE-n-LAT term mostly moistens the CV.

As mentioned before, the base motion (BM) term comes from the arbitrary choice of CV base height, although it is related to the actual cloud base height evolution. When the BL is stratified, a rising CV base means the air mass near cloud base, which has lower  $\theta_1$  than the CV mean, is excluded from the CV. This results in an increase in  $\langle \theta_1 \rangle$  in the CV. Similarly we can infer the sign of this term for  $\langle \theta_1 \rangle$  and  $\langle q_t \rangle$  budgets under other conditions. This BM term is near zero during the nighttime when the BL is close to being well-mixed. Its relative importance peaks between 13:00 and 15:00 for both  $\langle \theta_1 \rangle$  and  $\langle q_t \rangle$  when the cloud base averaged for all cases starts to lower, accompanying the recovery of  $LWP_c$ . The magnitudes of cooling and moistening during this time are greater than the magnitudes of warming and drying between 09:00 and noon, primarily because the layer near the cloud base is more stratified in the afternoon.

The SUBS term always warms and dries the CV. Its effect peaks in the early afternoon around the time when the clouds are the thinnest.

#### 4.4 $LWP_c$ budget based on CV $\langle \theta_1 \rangle$ and $\langle q_t \rangle$ budgets

Figure 7 shows the  $LWP_c$  budget by category, with the actual  $LWP_c$  tendency and ENTR, RAD, SUBS, and BASE-n-LAT terms in the left column and the BM and RES terms in the right column. The PRCP terms are negligible and omitted.

We start with the terms in the right column. For all three categories, it is encouraging that the RES term in the  $LWP_c$  budget is fairly small. The improvement over the results based on the BL budgets (Figure 5) is dramatic for all three categories between sunrise and early afternoon; it is also evident for the hiDhiL category during the nighttime. Although the BM term is overall not important until early afternoon, quantifying it for CV  $\langle \theta_1 \rangle$  and  $\langle q_t \rangle$  budgets makes the LAT term (and thus the BASE-n-LAT term) slightly more accurate. Interestingly, the sum of the BM and RES term is even closer to zero. Qualitatively, the correlation between the BM term and the RES is expected considering that more stratified conditions simultaneously lead to a larger BM term and less applicability of MLT.

Moving to the terms in the left column of Figure 7, we know based on the small sum of the BM and RES term that the ENTR, RAD, SUBS, and BASE-n-LAT terms collectively explain the actual evolution of the  $LWP_c$  very well until early afternoon. In particular, we can infer from the small sum of the BM and RES term that the sum of these four terms captures the reduction of  $LWP_c$ , most rapid for the hiDhiL category and least for the loDloL category, in the morning, as is evident in the time series of the actual  $LWP_c$  tendency.

The ENTR, RAD, and BASE-n-LAT terms are expected to be the leading terms simply based on their roles in the CV  $\langle\theta_1\rangle$  and  $\langle q_t\rangle$  budgets. By contrast with the results in Figure 5, the SUBS terms are less important relative to the ENTR term. This is because the  $dz_i/dt$  term in Eq. (19) is constant in the two versions of  $LWP_c$  budget but the  $d\langle\theta_1\rangle/dt$  and  $d\langle q_t\rangle/dt$  terms are  
405 strongly affected by the depth over which the volume-integrated forcing is distributed.

The SUBS term has the smallest diurnal fluctuation among the four terms. As a result, one can infer that the net effect of the ENTR, RAD, and BASE-n-LAT terms would approximately follow the trend of the actual  $LWP_c$  tendency for each category. Among these three terms, the ENTR and RAD terms always begin to weaken right after sunrise. The BASE-n-LAT term remains near its maximum strength until 09:00 for the loDloL category, but it starts to weaken right after sunrise for the  
410 other two categories. This delay is likely the signature of better coupling with the surface. Due to this delay, although the rate of ENTR weakening for the loDloL category is slower than for the hiDloL category, the combined negative effect from ENTR and BASE-n-LAT terms (pink dash-dotted lines) diminishes faster between sunrise and 09:40 for loDloL. Since the change in the RAD term from sunrise to between 09:00 and 10:00 is about the same between these two categories, the delayed decrease in the BASE-n-LAT term explains the slower  $LWP_c$  reduction for the loDloL category. The weakening of the BASE-n-LAT term  
415 balances that of the ENTR term closely for the hiDhiL category and the net effect (the pink dash-dotted lines) only weakens very slowly. As a result, the line for the RAD term is nearly parallel to the line for the actual  $LWP_c$  tendency. Interestingly, when the actual  $LWP_c$  tendency becomes the most negative in the morning for the loDloL and hiDloL categories, its value is very close to the SUBS term, meaning the ENTR, RAD, and BASE-n-LAT terms sum to about zero. It is unclear whether this is by accident but this is different for the hiDhiL category, where the actual  $LWP_c$  tendency can be much more negative than  
420 the SUBS term, driven by the dramatic change in the RAD term.

To summarize, applying the MLT to the CV achieves satisfactory closure for the  $LWP_c$  budget from nighttime to early afternoon. In the morning, the coupling to the surface, evident in the BASE-n-LAT term, explains the relatively smaller loss of  $LWP_c$  for the loDloL category. The strong reduction of the RAD cooling causes the rapid reduction of  $LWP_c$  for the hiDhiL category. In the next section, we will use the budget analysis to understand the evolution of individual LES ensemble members,  
425 not just the mean evolution by category.

## 5 Nighttime and daytime evolution of LES ensemble members

With the categorization of cases and the budget analysis presented, we can now examine the nighttime and daytime evolution of simulations in detail.

### 5.1 Nighttime evolution of individual cases

430 Figure 8 highlights several aspects of the nighttime evolution. Overall, the nighttime evolution is characterized by the establishment of a positive correlation between  $LWP_c$  and a characteristic FT  $q_t$ . (Since subsidence is the only process that modifies the FT  $q_t$  profile in our simulations, the characteristic FT  $q_t$  is determined as follows. For a given time, we track the air mass at 20 m above  $z_i$  back in time using the subsidence profile, Eq. (1), to calculate its height at the beginning of the simulation,

and represent the current FT  $q_t$  with the initial  $q_t$  at that height.) This can be seen by comparing the trajectories, colored by FT  $q_t$ , during the first three hours after the start of the simulations (Figure 8a) and during the three hours before sunrise (Figure 8b). It is also evident in the time series of the correlation coefficient between  $LWP_c$  and FT  $q_t$  (Figure 8c). At the beginning of each simulation,  $LWP_c$  is determined by three of the six prescribed parameters: BL  $\theta_1$ , BL  $q_t$ , and  $h_{\text{mix}}$ . As a result of the random sampling of the initial conditions, it is largely uncorrelated with the FT  $q_t$  even after we exclude cases based on criteria described in Section 2. FT  $q_t$  acts as a boundary condition for the simulated clouds. It affects  $LWP_c$  by modulating entrainment drying and the downward longwave radiation reaching the cloud top, two effects that compete with each other (Eastman and Wood, 2018). Based on the way we specify FT  $q_t$  profiles, the FT humidity controlling the longwave radiation positively correlates with the FT humidity that is relevant to the entrainment. For example, a case with a dry FT in our ensemble would experience greater entrainment drying; at the same time, it experiences strong radiative cooling because the FT is more transparent to longwave radiation. Although this strong radiative cooling favors high  $LWP_c$ , it also drives the clouds to entrain more, potentially reducing  $LWP_c$ . The positive correlation between  $LWP_c$  and FT  $q_t$  in our simulations suggests that the entrainment effect dominates.

Figures 8d–f show the  $LWP_c$  velocity, defined as the ratio between  $LWP_c$  change and mean  $LWP_c$  over a period of time, for the three hours before sunrise in  $LWP_c$ – $z_i$ ,  $N_d$ – $z_i$ , and  $N_d$ – $LWP_c$  planes, where the locations of dots are based on states at sunrise. Most cases with  $LWP_c$  less than  $60 \text{ g m}^{-2}$  at sunrise gain  $LWP_c$  during the three hours before sunrise (Figures 8d and 8f). This qualitatively agrees with Hoffmann et al. (2020) and Glassmeier et al. (2021). However, the sign of the  $LWP_c$  velocity is mixed for cases with greater  $LWP_c$ , where only 56% cases are gaining  $LWP_c$ . Among these cases, there is a weak negative correlation between  $z_i$  and  $LWP_c$  velocity, i.e., shallower/deeper BLs tend to see increasing/decreasing  $LWP_c$ , possibly because deeper BLs are more likely to be decoupled from the surface. When projected onto the  $N_d$ – $LWP_c$  plane (Figure 8f), cases with low  $LWP_c$  and low  $N_d$  mostly gain  $LWP_c$ , while cases losing  $LWP_c$  only occur under high  $LWP_c$  and high  $N_d$  conditions. To some extent, this is consistent with the findings in Hoffmann et al. (2020) and Glassmeier et al. (2021).

However, due to some potentially realistic yet complicated correlations among  $LWP_c$ ,  $N_d$ ,  $z_i$ , and FT  $q_t$ , we cannot simply attribute the correlation between  $LWP_c$  velocity and  $N_d$  to  $N_d$ . First, there is a positive correlation between  $LWP_c$  and  $N_d$  because we focus on the non-precipitating conditions and high  $LWP_c$  cases are only possible if  $N_d$  is sufficiently high to suppress the cloud-base precipitation (Figure 8f). Second, due to the positive correlation between  $LWP_c$  and  $z_i$  (deeper  $z_i$  supporting higher  $LWP_c$ , Figure 8d), there is also a positive correlation between  $z_i$  and  $N_d$  (notice very few cases in the upper left corner of Figures 8e). Similarly, because of the positive correlation between  $LWP_c$  and FT  $q_t$  (Figures 8b and 8c), there is a positive correlation between FT  $q_t$  and  $N_d$  (not shown).

We examine the correlation between radiative cooling and  $LWP_c$  to assess the impacts of the positive correlation between FT  $q_t$  and  $LWP_c$  on the  $LWP_c$  tendency (Figure 9). Recall that to calculate the RAD term for the  $LWP_c$  budgets, we first calculate the CV-integrated radiative heating rate, then assume it evenly distributes in the CV to calculate the RAD term for the CV  $\langle \theta_1 \rangle$  budget, and then use Eq. (19) to calculate the RAD term for the  $LWP_c$ . The CV-integrated radiative heating rate strongly depends on FT  $q_t$  while the cloud-top temperature (approximated using the lowest temperature in the mean temperature profile for the CV) explains a small portion of its variance (i.e., lower cloud-top temperature associates with less integrated radiative

cooling, Figure 9a). The sensitivity of the CV-integrated radiative heating rate to FT  $q_t$  increases for FT  $q_t$  below  $3 \text{ g kg}^{-1}$ .  
 470 More than 90% of cases have  $\text{LWP}_c$  greater than  $40 \text{ g m}^{-2}$  at this time and the emissivity of these clouds should have saturated  
 (Garrett et al., 2002; Petters et al., 2012). (Our integrated radiative heating rate with FT  $q_t$  of  $4.5 \text{ g kg}^{-1}$ , the FT  $q_t$  estimated  
 from Figure 2 in Petters et al. (2012) is very close to the saturated cloud-integrated radiative heating for longwave radiation  
 in their Figure 1.) However, the RAD contribution to the CV  $\langle \theta_1 \rangle$  budget strongly and positively correlates with  $\text{LWP}_c$  (filled  
 circles in Figure 9b) due to correlation between  $\text{LWP}_c$  and  $\langle q_t \rangle$  as well as the scaling by CV depth. Earlier, we showed that the  
 475 MLT-based budget works well for the loDloL and hiDloL categories during the nighttime (Figure 5). One might argue that it is  
 more appropriate to assume the CV-integrated radiative heating rate is distributed from the surface to  $z_i$ . This scaling reduces  
 the slope but not the sign of the correlation between the scaled RAD term and  $\text{LWP}_c$  (hollow circles in Figure 9b). It is only  
 when we use the CV-integrated radiative cooling rate scaled with  $z_i$  in Eq. (19) that we find a positive correlation between the  
 scaled RAD term for  $\text{LWP}_c$  tendency and  $\text{LWP}_c$  (hollow circles in Figure 9c; compare with hollow circles in Figure 9b).

480 The ratio between the scaled RAD term for the  $\text{LWP}_c$  tendency and for the CV  $\langle \theta_1 \rangle$  tendency depends on  $\Gamma_1$ ,  $\langle \rho_0 \rangle$ , cloud  
 depth, and  $dz_{cb}/d\langle \theta_1 \rangle$ . Both the positive correlations between the cloud depth and  $\text{LWP}_c$ , as discussed in Hoffmann et al.  
 (2020), and between other prefactors and  $\text{LWP}_c$  (not shown) contribute to this change in the sign of the correlation. For the  
 $\text{LWP}_c$  velocity, the division by  $\text{LWP}_c$  itself further modifies the correlation and the slope between a budget term and  $\text{LWP}_c$   
 (Figure 9d). In summary, not only the FT  $q_t$  but also the  $z_i$ , the coupling state, and other factors (e.g., the prefactors in Eq. 19)  
 485 shape the correlation between the radiative contribution to  $\text{LWP}_c$  tendency or velocity and the  $\text{LWP}_c$ .

We show the behavior of other terms for the  $\text{LWP}_c$  tendency in Figure 10a. The BASE-n-LAT term positively contributes to  
 the  $\text{LWP}_c$  tendency. It negatively correlates with  $\text{LWP}_c$  for greater  $\text{LWP}_c$ , but positively correlates with it for lower  $\text{LWP}_c$ , prob-  
 ably because cases with lower  $\text{LWP}_c$  at sunrise, mostly in the loDloL category, have weaker boundary layer circulation. The  
 ENTR term negatively contributes to the  $\text{LWP}_c$  tendency. It positively correlates with  $\text{LWP}_c$  for greater  $\text{LWP}_c$ , but negatively  
 490 correlates with it for lower  $\text{LWP}_c$ . Compared with the RAD and BASE-n-LAT terms, this correlation suggests that, to the first  
 order, the entrainment is determined by the driving force for the turbulence, e.g., the radiative cooling and the boundary layer  
 circulation. The SUBS term negatively contributes to the  $\text{LWP}_c$  velocity and positively correlates with  $\text{LWP}_c$ . After scaling  
 by  $z_i$ , the BASE-n-LAT, ENTR, and SUBS terms show a much tighter positive, negative, and negative correlation with  $\text{LWP}_c$   
 (Figure 10b).

## 495 5.2 Daytime evolution of individual cases

Figures 11a and 11b show the most distinct feature of the daytime evolution of the individual cases. More decoupled cases tend  
 to lose  $\text{LWP}_c$  more rapidly between sunrise and 12:00. For cases with  $z_i$  greater than about 0.9 km, the positive correlation  
 between  $\text{LWP}_c$  and  $z_i$  at sunrise (dots in Figure 8b) becomes negative by 12:00 (dots in Figure 11a). In the afternoon, the  $\text{LWP}_c$   
 recovers for most cases and a positive correlation between  $\text{LWP}_c$  and  $z_i$  is restored by the end of the simulation.

500 To understand the factors controlling the evolution of  $\text{LWP}_c$  in the  $\text{LWP}_c$ - $z_i$  plane, we investigate the behavior of four groups  
 of cases with different properties: (1) loDloL cases with  $\text{LWP}_c$  at sunrise between  $75$  and  $90 \text{ g m}^{-2}$  (2) hiDloL cases with  
 $\text{LWP}_c$  at sunrise in the same range (hiDloL Group 1), (3) hiDloL cases with  $\text{LWP}_c$  at sunrise between  $150$  and  $180 \text{ g m}^{-2}$



(hiDloL Group 2), and (4) hiDhiL cases with  $LWP_c$  at sunrise between 240 and 300  $g\ m^{-2}$ . Comparing Figures 11c and 11d, all four groups develop negative slopes between  $LWP_c$  and  $z_1$  between sunrise and 09:40, the least negative for the loDloL group and the most negative for the hiDhiL group. Figure 12a shows the  $LWP_c$  tendencies and budget terms for each case in these four groups. The mean  $LWP_c$  tendency between sunrise and 09:40 differs between groups, by  $z_1$ , and by degree of coupling. For example, the loss of the  $LWP_c$  is faster/slower for groups with higher/lower  $LWP_c$  at sunrise; within each group, cases with greater  $z_1$  tend to lose  $LWP_c$  faster; the hiDloL Group 1 loses  $LWP_c$  faster than the loDloL group. Across different  $z_1$ , the RAD term positively correlates with the actual  $LWP_c$  tendency and shows similar spread (Figure 12b). The variation of the RAD term between groups is consistent with both the nighttime behavior of the RAD term (i.e., more positive RAD term for low  $LWP_c$  and low FT  $q_t$ , e.g., cases with higher  $z_1$  in the loDloL group and hiDloL Group 1; also see Figures 8b and 9c) and the anticipated greater absorption of shortwave radiation for cases with higher  $LWP_c$  (e.g., the hiDhiL group). Unfortunately, we do not have separate longwave and shortwave radiative output to quantify the relative importance of longwave cooling and shortwave warming at this point. The ENTR and BASE-n-LAT terms are larger in magnitude than the RAD term (Figures 12c and 12d). The SUBS term shows negative  $z_1$ -dependence with small differences between groups (Figure 12e). The sum of the BM and RES terms is very small, compared with other terms and the actual  $LWP_c$  tendency (Figure 12f). Based on these results, it is reasonable to take the sum of the SUBS, the BM, the PRCP, and the RES terms as a baseline and investigate how much the RAD, the ENTR, and the BASE-n-LAT terms drive the actual  $LWP_c$  tendency to deviate from this baseline. Figures 12g and 12h shows the sum of the RAD, the ENTR, and the BASE-n-LAT terms as well as the sum of the ENTR and the BASE-n-LAT terms. Combined with the RAD term in Figure 12b, we conclude that the differences in  $LWP_c$  tendency between groups with different  $LWP_c$  at sunrise are more associated with the RAD term, and the other details derive from a subtle balance between the RAD, ENTR, and BASE-n-LAT terms.

## 6 Discussion

In this section, we discuss an uncertainty in our budget analysis method, and then address the role of the interactive surface fluxes in the simulations.

### 6.1 Uncertainty in ENTR term for $\langle\theta_1\rangle$ and $\langle q_t\rangle$ budgets

As described earlier, we use the entrainment fluxes (i.e.,  $d\Phi/dt|_{\text{ENTR}}$ ) from the BL  $\langle\theta_1\rangle$  and  $\langle q_t\rangle$  budgets to calculate the ENTR term for the CV. However, because the cloudy region of a domain is more turbulent than the clear-sky region, one would expect a higher entrainment flux in the cloudy region than the domain-mean for partially cloudy scenes. Underestimating the magnitude of entrainment fluxes for the CV budget will cause a compensating error in the BASE-n-LAT term because the latter holds the residual between the actual CV  $\langle\theta_1\rangle$  and  $\langle q_t\rangle$  tendencies and the sum of the other terms.

In this subsection, we resort to the jump-based method (Eq. (15)) to assess the potential bias in our ENTR term. We first repeat the budget analysis for all clear-sky columns between the same base and top as the CV (denoted with “nCV”, meaning “not CV”), and then partition the total entrainment warming and drying in the CV and the nCV with the cloudy region jump

535  $\Delta\phi_{\text{CV}}$  and clear-sky jump  $\Delta\phi_{\text{nCV}}$ . This alternative estimate of the entrainment tendency for the CV is

$$\left. \frac{d\langle\phi\rangle}{dt} \right|_{\text{ENTR,alt}} = \frac{f_c (d\langle\phi\rangle/dt|_{\text{ENTR}}) + (1 - f_c) (d\langle\phi\rangle_{\text{nCV}}/dt|_{\text{ENTR}})}{f_c + (1 - f_c)\Delta\phi_{\text{nCV}}/\Delta\phi_{\text{CV}}}, \quad (20)$$

where “alt” stands for “alternative” and, again,  $\phi$  represents either  $\theta_l$  or  $q_l$ . The question becomes how to define  $z_+$  and  $z_-$  separately for  $\phi$  profiles averaged in the cloudy and clear-sky regions to calculate the jumps. We follow Yamaguchi et al. (2011), where the authors check the domain-wide liquid water static energy ( $s_l$ ) variance profile and define  $z_+$  and  $z_-$  as the levels with  $s_l$  variance falling to 5% of the peak value. This method works reasonably well for DYCOMS-II RF02, the case simulated in Yamaguchi et al. (2011). (See Appendix C in that work.) We apply a constant absolute  $s_l$  variance threshold of 0.235 K<sup>2</sup> (5% of 4.7 K<sup>2</sup>, the peak  $s_l$  variance in Yamaguchi et al., 2011) to search for  $z_+$  and  $z_-$  to qualitatively capture the idea that the jump is smaller when turbulence mixing is weaker (lower peak  $s_l$  variance).

We take a few extra steps to handle potential outliers. We exclude all time steps with  $f_c < 0.01$  (1.9% of all time steps) and keep the entrainment tendencies with  $f_c > 0.99$  unchanged. Sometimes, the peak  $s_l$  variance of a profile (usually the clear-sky ones) is below 0.235 K<sup>2</sup> and no  $z_+$  or  $z_-$  are identified. For this situation, we keep a data point if only  $\Delta\phi_{\text{CV}}$  can be calculated (about 6.4% of all time steps) and set its  $\Delta\phi_{\text{nCV}}$  to 0, which actually exaggerates the difference between the cloudy and clear-sky region. We exclude a data point if neither  $\Delta\phi_{\text{CV}}$  nor  $\Delta\phi_{\text{nCV}}$  can be calculated, which rarely occurs.

For all three categories, we find no significant difference between the current and the alternative ENTR terms until the afternoon (Figure 13). These results certainly depend on details of our method, e.g., the value of the  $s_l$  variance threshold. However, without a more solid foundation for an alternative choice of the threshold, sensitivity tests would not provide more reliable quantification of the bias.

One other method is to partition the entrainment flux using Eq. (13), such that

$$\left. \frac{d\Phi}{dt} \right|_{\text{ENTR,alt}} = \frac{1}{f_c + (1 - f_c)(\rho_{0,e}\phi_e)_{\text{nCV}}/(\rho_{0,e}\phi_e)_{\text{CV}}} \left. \frac{d\Phi}{dt} \right|_{\text{ENTR}}. \quad (21)$$

If we use  $\rho_0\phi$  at  $z_+$  identified earlier as an estimate of  $\rho_{0,e}\phi_e$ , the resulting ENTR terms are even closer to our current estimates.

These results do not necessarily mean that our current ENTR term is accurate. They simply suggest that, the two alternative methods we test to introduce contrast between cloudy region and clear-sky entrainment produce limited “correction” to current ENTR estimates. While these results provide some confidence in the robustness of current ENTR estimates, it seems to be inconsistent with the argument that the cloudy region is more turbulent and thus should entrain more. We argue that this inconsistency is partially rooted in the assumption that the movement of  $z_i$  is the result of the entrainment and the subsidence (Eq. (4)). We find that the air is on average descending/ascending at speeds around a few mm s<sup>-1</sup> near the mean  $z_i$  in the cloudy/clear-sky region, which are indeed at very similar heights, despite the mean updraft/downdraft for the bulk of BL in the cloudy/clear-sky region (Figure 13c). This is probably the signature of a mesoscale (instead of large-scale, e.g., the prescribed subsidence, which is horizontally uniform in the domain) mean circulation in the FT, similar to the one shown in Zhou and Bretherton (2019). (See their Figure 9.) In other words, the cloudy/clear-sky region is more/less turbulent, but there may be a mesoscale downdraft/updraft limiting/promoting the growth of  $z_i$ . With Eq. (4), the effect of this mesoscale mean air motion is

lumped into the entrainment. This finding suggests that our current ENTR term should be interpreted as a *collective effect* of processes except the prescribed subsidence that move the  $z_i$ .

## 6.2 Sensitivity of cloud evolution to SST

570 As mentioned earlier, our LES ensemble covers a wide range of conditions by perturbing initial profiles. However, all simulations are configured with the same fixed SST and subsidence profile. As a result, the initial BL  $\theta_1$  could be more than 6.8 K colder than  $\theta_{\text{SST}}$ , which is not very realistic for marine stratocumuli. Also, the correlation that the initially shallower BLs in our ensemble tend to be colder and drier (see Section 3.3) means shallower BLs tend to be colder than the SST and experience greater surface fluxes. To capture more realistic co-variability between environment conditions, one may consider simulating  
575 marine stratocumuli as they are advected towards warmer SST (e.g., Sandu and Stevens, 2011; Teixeira et al., 2011; Bretherton and Blossey, 2014; Yamaguchi et al., 2015; de Roode et al., 2016; Kazil et al., 2021) or as they reach equilibrium with different environmental conditions along this transition (Chung et al., 2012).

For now, we assess the impacts of this correlation on our results by re-running all simulations with the SST set to 0.5 K warmer than the initial lowest model level air temperature *for each case*. (Hereafter, we refer to this LES ensemble as the  
580 “SST0.5K+” set and the original LES ensemble as the “fSST” set, where “f” stands for “fixed”.) Compared with the fSST set, the surface fluxes in the SST0.5K+ set are much weaker (Figures S9a and S9b), which are consistent with the overall more decoupled conditions, especially in the afternoon (cf. Figures S8d and 2d). The  $\text{LWP}_c$  at sunrise increase in the SST0.5K+ set; they are less correlated with  $z_i$  but more positively correlated with FT  $q_t$  (Figures S14b and S14c). This behavior broadly agrees with De Roode et al. (2014) where warmer SST causes thinning of stratocumuli when the response in entrainment is strong.  
585 Other results are similar between SST0.5K+ and fSST. In particular, in the morning, clouds in deeper BLs still experience dramatic loss in  $\text{LWP}_c$  such that a negative correlation develops between  $\text{LWP}_c$  and  $z_i$ . (Compare Figures S17a and 11a.) We present other figures based on the SST0.5K+ set from in the supplementary material.

## 7 Summary

In this work, we explore the cloud system evolution of non-precipitating marine stratocumuli with a focus on the impacts of the  
590 diurnal cycle and free-tropospheric (FT) humidity by analyzing 244 cases in an LES ensemble generated by perturbing initial profiles and aerosol conditions.

We separate the cases into three categories with distinct behavior based on their relative decoupling index ( $\mathcal{D}$ ) at 09:40 and cloud liquid water paths ( $\text{LWP}_c$ ) at sunrise: a loDloL category ( $\mathcal{D} \leq 1$ ), a hiDloL category ( $\mathcal{D} > 1$  and  $\text{LWP}_c \leq 180 \text{ g m}^{-2}$ , the highest  $\text{LWP}_c$  for the loDloL category), and a hiDhiL category ( $\mathcal{D} > 1$  and  $\text{LWP}_c > 180 \text{ g m}^{-2}$ ). Cases in the loDloL  
595 category are commonly associated with lower  $z_i$ . They start with the lowest  $\text{LWP}_c$  and cloud fraction ( $f_c$ ) among the three categories and may not ever become overcast. However, on average, they also experience the least reduction in  $\text{LWP}_c$  and  $f_c$  during the daytime. Clouds in the hiDloL category occur in deeper BLs, start with more  $\text{LWP}_c$ , and tend to be overcast during the nighttime. On average, they experience dramatic  $\text{LWP}_c$  and  $f_c$  reductions during the day. These clouds tend to evolve into

a cumulus-rising-into-stratocumulus structure in the afternoon. Clouds in the hiDhiL category share many features with those  
600 in the hiDloL category but show different timing and amplitude of daytime  $LWP_c$  and  $f_c$  fluctuations. The diurnal cycles of  
 $LWP_c$  and  $f_c$  for three categories are closely related to the diurnal cycles of their coupling states.

We perform a budget analysis to understand the diurnal cycle of  $LWP_c$  by tracking the mean  $\theta_1$  and  $q_t$  budgets for the “cloud  
volume” (CV), a volume consisting of all cloudy columns between the first grid box base below the mean cloud base and  
 $z_i$ , and then applying the LWP budget equation (Eq. (19)) to the CV, assuming it is well-mixed. By focusing on the cloudy  
605 region of the cloud layer, this method closes the budget with a very small residual (RES) until early afternoon. In particular, it  
adequately captures the rapid  $LWP_c$  reduction in the morning for all categories. A delayed decrease in the positive contribution  
to  $LWP_c$  from the BASE-n-LAT term, a term that tracks the impacts of the processes associated with the interface between the  
CV and the rest of the BL (i.e., CV base and lateral sides), after sunrise explains the slower  $LWP_c$  reduction in the loDloL  
category than in the hiDloL category. For the hiDhiL category, the strong decrease in the radiative (RAD) cooling results in  
610 the most rapid  $LWP_c$  reduction in this category.

The impact of a humid FT on the evolution of simulations during the nighttime is distinct. A positive correlation between  
FT  $q_t$  and  $LWP_c$  emerges and strengthens towards sunrise. Because the longwave emissivity of clouds is saturated in most  
cases, the FT  $q_t$  strongly affects the CV-integrated radiative heating rate. As a result, there is stronger radiative cooling for  
cases with lower  $LWP_c$  through the correlation between the FT  $q_t$  and  $LWP_c$ . This illustrates how the co-variability among  
615 state variables and cloud controlling factors modifies the distribution of  $LWP_c$  tendency in state variable spaces. During the  
daytime, clouds in deeper BLs lose  $LWP_c$  faster in the morning, again suggesting that state variables beyond  $LWP_c$  and  $N_d$  are  
necessary to understand the  $LWP_c$  tendency. A closer analysis reveals that the  $LWP_c$  tendency in the morning varies with the  
 $LWP_c$  at sunrise,  $z_i$ , and the degree of decoupling. A budget analysis for  $LWP_c$  shows that the subsidence term (SUBS) causes  
a more negative  $LWP_c$  tendency at deeper  $z_i$  and this effect is similar for cases with different  $LWP_c$  at sunrise and degree of  
620 decoupling. The entrainment (ENTR) and BASE-n-LAT terms closely balance each other, and there is a weak dependence of  
the net effect on  $z_i$ . It is the RAD term that differentiates cases with similar  $z_i$  in terms of the  $LWP_c$  tendency.

In the design of the current LES ensemble, SST and subsidence profiles are not perturbed. Also, the natural co-variability  
between different environmental conditions is not captured. To partially address these limitations, we perform additional runs  
for all cases with the SST set to 0.5 K warmer than the initial lowest model level air temperature for each case. The statistical  
625 behavior of the clouds with this configuration is similar to the LES ensemble with fixed SST, although the correlation be-  
tween  $LWP_c$  and  $z_i$  at sunrise becomes weaker. Future simulations should use more realistic forcings and naturally co-varying  
thermodynamic and aerosol conditions to improve the realism of the LES ensemble. A related issue is that the 24-hr length  
of current simulations, although covering one diurnal cycle, is insufficient for the mesoscale organization of clouds to fully  
develop (Kazil et al., 2017). This limitation should be addressed in future work.

630 We demonstrate the emergence of the correlations among environmental conditions and state variables as the clouds evolve.  
All these correlations project onto the correlations with  $N_d$  and need to be carefully considered when we distill the causality  
between  $N_d$  and variables like the  $LWP_c$  tendency or the  $LWP_c$  velocity. We pursue this task in Zhang et al. (2024).

## Appendix A: Constructing initial thermodynamic profiles

In this appendix, we describe the method for (1) creating the upper air  $\theta_1$  and  $q_1$  profiles and (2) connecting them with the initial  
635 BL  $\theta_1$  and  $q_1$  profiles (described in Section 2) to construct the initial  $\theta_1$  and  $q_1$  profiles.

To prepare for the upper air profiles, we generate ERA5-based climatological profiles in a few steps. First, we produce mean  
profiles from all ERA5 profiles in the Californian stratocumulus region (i.e., the  $10^\circ$  by  $10^\circ$  box between  $20^\circ\text{N}$ ,  $30^\circ\text{N}$ ,  $120^\circ\text{W}$ ,  
and  $130^\circ\text{W}$  as defined in Klein and Hartmann, 1993) during April, May, and June (the months with highest stratocumulus cover  
in the region; Wood, 2012) from 2000 to 2011. Then, we search for the height with the maximum  $\theta_1$  gradient below 2 km and  
640 keep the mean profile segments between this height and 35.8 km, the top of the mean profiles.

When we connect the  $\theta_1$  climatological profile produced this way to the initial BL profiles, some simulations experience  
very rapid growth in the inversion base height ( $z_i$ ) in the first few hours, suggesting that the  $\theta_1$  gradient across the inversion  
is too weak. To solve this issue, we prepare a transitional profile for  $\theta_1$ . We average the observed  $\theta_1$  profiles during the warm  
season legs of the MAGIC campaign after translating them vertically to line up at inversion bases and having their BL values  
645 subtracted at all heights. We keep the first 1.5 km of this mean profile above the inversion base.

To construct an initial  $\theta_1$  profile, we first translate the transitional profile so that its lowest point attaches to point right above  
the inversion base. Next, we scale the ERA5-based  $\theta_1$  climatological profile so that its lowest point attaches to the highest point  
of the transitional profile (now sitting at 1.5 km above  $h_{\text{mix}}$ ) while its highest point stays fixed at 35.8 km. For an initial  $q_1$   
profile, we scale the ERA5-based  $q_1$  climatological profile so that its lowest point directly attaches to the point right above  
650 the inversion base while its highest point stays fixed at 35.8 km. A constant surface pressure of 1018.52 mb, based on ERA5  
climatology, is used for all initial profiles. See Figure A1 for an illustration.

*Code and data availability.* The System for Atmospheric Modeling (SAM) code is publicly available at <http://rossby.msrc.sunysb.edu/SAM.html>. The ERA5 data is archived at Copernicus Climate Change Service (C3S) Climate Data Store (CDS) (Hersbach et al., 2017). The  
MAGIC data is available via ARM Data Discovery (Atmospheric Radiation Measurement (ARM) user facility, 2012). Data for reproducing  
655 the results will be provided following acceptance.

*Author contributions.* GF, TY, and YC initiated this study. TY, FG, and YC designed the LES ensemble. TY and YC performed the sim-  
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manuscript.

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## References

- Ackerman, A. S., Kirkpatrick, M. P., Stevens, D. E., and Toon, O. B.: The impact of humidity above stratiform clouds on indirect aerosol climate forcing, *Nature*, 432, 1014–1017, <https://doi.org/10.1038/nature03174>, 2004.
- Ackerman, A. S., vanZanten, M. C., Stevens, B., Savic-Jovicic, V., Bretherton, C. S., Chlond, A., Golaz, J.-C., Jiang, H., Khairoutdinov, M., Krueger, S. K., Lewellen, D. C., Lock, A., Moeng, C.-H., Nakamura, K., Petters, M. D., Snider, J. R., Weinbrecht, S., and Zulauf, M.: Large-eddy simulations of a drizzling, stratocumulus-topped marine boundary layer, *Monthly Weather Review*, 137, 1083–1110, <https://doi.org/10.1175/2008MWR2582.1>, 2009.
- Albrecht, B. A.: Aerosols, cloud microphysics, and fractional cloudiness, *Science*, 245, 1227–1230, <https://doi.org/10.1126/science.245.4923.1227>, 1989.
- 680 Atmospheric Radiation Measurement (ARM) user facility: Balloon-Borne Sounding System (SONDEWNP). 2012-10-01 to 2013-10-03, ARM Mobile Facility (MAG) Los Angeles, CA to Honolulu, HI - container ship Horizon Spirit; AMF2 (M1)., Compiled by Even Keeler, Ken Burk, and Jenni Kyrouac. ARM Data Center: Oak Ridge, Tennessee, USA., <https://doi.org/10.5439/1595321>, last accessed: September 19, 2022, 2012.
- Boers, R. and Mitchell, R. M.: Absorption feedback in stratocumulus clouds influence on cloud top albedo, *Tellus A: Dynamic Meteorology and Oceanography*, 46, 229, <https://doi.org/10.3402/tellusa.v46i3.15476>, 1994.
- Bretherton, C. S. and Blossey, P. N.: Low cloud reduction in a greenhouse-warmed climate: Results from Lagrangian LES of a subtropical marine cloudiness transition, *Journal of Advances in Modeling Earth Systems*, 6, 91–114, <https://doi.org/10.1002/2013MS000250>, 2014.
- Bretherton, C. S. and Wyant, M. C.: Moisture transport, lower-tropospheric stability, and decoupling of cloud-topped boundary layers, *Journal of the Atmospheric Sciences*, 54, 148–167, [https://doi.org/10.1175/1520-0469\(1997\)054<0148:MTLTSA>2.0.CO;2](https://doi.org/10.1175/1520-0469(1997)054<0148:MTLTSA>2.0.CO;2), 1997.
- 690 Bretherton, C. S., Blossey, P. N., and Uchida, J.: Cloud droplet sedimentation, entrainment efficiency, and subtropical stratocumulus albedo, *Geophysical Research Letters*, 34, <https://doi.org/10.1029/2006GL027648>, 2007.
- Bretherton, C. S., Blossey, P. N., and Jones, C. R.: Mechanisms of marine low cloud sensitivity to idealized climate perturbations: A single-LES exploration extending the CGILS cases, *Journal of Advances in Modeling Earth Systems*, 5, 316–337, <https://doi.org/10.1002/jame.20019>, 2013.
- 695 Caldwell, P. and Bretherton, C. S.: Large eddy simulation of the diurnal cycle in Southeast Pacific stratocumulus, *Journal of the Atmospheric Sciences*, 66, 432–449, <https://doi.org/10.1175/2008JAS2785.1>, 2009.
- Chen, Y.-C., Xue, L., Lebo, Z. J., Wang, H., Rasmussen, R. M., and Seinfeld, J. H.: A comprehensive numerical study of aerosol-cloud-precipitation interactions in marine stratocumulus, *Atmospheric Chemistry and Physics*, 11, 9749–9769, <https://doi.org/10.5194/acp-11-9749-2011>, 2011.
- 700 Chun, J.-Y., Wood, R., Blossey, P., and Doherty, S. J.: Microphysical, macrophysical, and radiative responses of subtropical marine clouds to aerosol injections, *Atmospheric Chemistry and Physics*, 23, 1345–1368, <https://doi.org/10.5194/acp-23-1345-2023>, 2023.
- Chung, D., Matheou, G., and Teixeira, J.: Steady-state large-eddy simulations to study the stratocumulus to shallow cumulus cloud transition, *Journal of the Atmospheric Sciences*, 69, 3264–3276, <https://doi.org/10.1175/JAS-D-11-0256.1>, 2012.
- De Roode, S. R., Siebesma, A. P., Dal Gesso, S., Jonker, H. J. J., Schalkwijk, J., and Sival, J.: A mixed-layer model study of the stratocumulus response to changes in large-scale conditions, *Journal of Advances in Modeling Earth Systems*, 6, 1256–1270, <https://doi.org/10.1002/2014MS000347>, 2014.

- de Roode, S. R., Sandu, I., van der Dussen, J. J., Ackerman, A. S., Blossey, P., Jarecka, D., Lock, A., Siebesma, A. P., and Stevens, B.: Large-eddy simulations of EUCLIPSE–GASS Lagrangian stratocumulus-to-cumulus transitions: Mean state, turbulence, and decoupling, *Journal of the Atmospheric Sciences*, 73, 2485–2508, <https://doi.org/10.1175/JAS-D-15-0215.1>, 2016.
- 710 Deardorff, J. W.: Stratocumulus-capped mixed layers derived from a three-dimensional model, *Boundary-Layer Meteorology*, 18, 495–527, <https://doi.org/10.1007/bf00119502>, 1980.
- Eastman, R. and Wood, R.: The competing effects of stability and humidity on subtropical stratocumulus entrainment and cloud evolution from a Lagrangian perspective, *Journal of the Atmospheric Sciences*, 75, 2563–2578, <https://doi.org/10.1175/JAS-D-18-0030.1>, 2018.
- Feingold, G., Walko, R. L., Stevens, B., and Cotton, W. R.: Simulations of marine stratocumulus using a new microphysical parameterization scheme, *Atmospheric Research*, 47–48, 505–528, [https://doi.org/10.1016/S0169-8095\(98\)00058-1](https://doi.org/10.1016/S0169-8095(98)00058-1), 1998.
- 715 Feingold, G., McComiskey, A., Yamaguchi, T., Johnson, J. S., Carslaw, K. S., and Schmidt, K. S.: New approaches to quantifying aerosol influence on the cloud radiative effect, *Proceedings of the National Academy of Sciences*, 113, 5812–5819, <https://doi.org/10.1073/pnas.1514035112>, 2016.
- Feingold, G., Ghatge, V. P., Russell, L. M., Blossey, P., Cantrell, W., Christensen, M. W., Diamond, M. S., Gettelman, A., Glassmeier, F., Gryspeerdt, E., Haywood, J., Hoffmann, F., Kaul, C. M., Lebsock, M., McComiskey, A. C., McCoy, D. T., Ming, Y., Mülmenstädt, J., Possner, A., Prabhakaran, P., Quinn, P. K., Schmidt, K. S., Shaw, R. A., Singer, C. E., Sorooshian, A., Toll, V., Wan, J. S., Wood, R., Yang, F., Zhang, J., and Zheng, X.: Physical science research needed to evaluate the viability and risks of marine cloud brightening, *Science Advances*, 10, eadi8594, <https://doi.org/10.1126/sciadv.adi8594>, 2024.
- 720 Garrett, T. J., Radke, L. F., and Hobbs, P. V.: Aerosol effects on cloud emissivity and surface longwave heating in the Arctic, *Journal of the Atmospheric Sciences*, 59, 769–778, [https://doi.org/10.1175/1520-0469\(2002\)059<0769:AEOCEA>2.0.CO;2](https://doi.org/10.1175/1520-0469(2002)059<0769:AEOCEA>2.0.CO;2), 2002.
- 725 Ghonima, M. S., Norris, J. R., Heus, T., and Kleissl, J.: Reconciling and validating the cloud thickness and liquid water path tendencies proposed by R. Wood and J. J. van der Dussen et al., *Journal of the Atmospheric Sciences*, 72, 2033–2040, <https://doi.org/10.1175/jas-d-14-0287.1>, 2015.
- Glassmeier, F., Hoffmann, F., Johnson, J. S., Yamaguchi, T., Carslaw, K. S., and Feingold, G.: An emulator approach to stratocumulus susceptibility, *Atmospheric Chemistry and Physics*, 19, 10 191–10 203, <https://doi.org/10.5194/acp-19-10191-2019>, 2019.
- 730 Glassmeier, F., Hoffmann, F., Johnson, J. S., Yamaguchi, T., Carslaw, K. S., and Feingold, G.: Aerosol-cloud-climate cooling overestimated by ship-track data, *Science*, 371, 485–489, <https://doi.org/10.1126/science.abd3980>, 2021.
- Gryspeerdt, E., Goren, T., Sourdeval, O., Quaas, J., Mülmenstädt, J., Dipu, S., Unglaub, C., Gettelman, A., and Christensen, M.: Constraining the aerosol influence on cloud liquid water path, *Atmospheric Chemistry and Physics*, 19, 5331–5347, <https://doi.org/10.5194/acp-19-5331-2019>, 2019.
- 735 Gryspeerdt, E., Glassmeier, F., Feingold, G., Hoffmann, F., and Murray-Watson, R. J.: Observing short-timescale cloud development to constrain aerosol–cloud interactions, *Atmospheric Chemistry and Physics*, 22, 11 727–11 738, <https://doi.org/10.5194/acp-22-11727-2022>, 2022.
- Haman, K. E., Malinowski, S. P., Kurowski, M. J., Gerber, H., and Brenguier, J.: Small scale mixing processes at the top of a marine stratocumulus—a case study, *Quarterly Journal of the Royal Meteorological Society*, 133, 213–226, <https://doi.org/10.1002/qj.5>, 2007.
- 740 Hersbach, H., Bell, B., Berrisford, P., Hirahara, S., Horányi, A., Muñoz Sabater, J., Nicolas, J., Peubey, C., Radu, R., Schepers, D., Simmons, A., Soci, C., Abdalla, S., Abellan, X., Balsamo, G., Bechtold, P., Biavati, G., Bidlot, J., Bonavita, M., De Chiara, G., Dahlgren, P., Dee, D., Diamantakis, M., Dragani, R., Flemming, J., Forbes, R., Fuentes, M., Geer, A., Haimberger, L., Healy, S., Hogan, R., Hólm, E., Janisková, M., Keeley, S., Laloyaux, P., Lopez, P., Lupu, C., Radnoti, G., de Rosnay, P., Rozum, I., Vamborg, F., Vil-



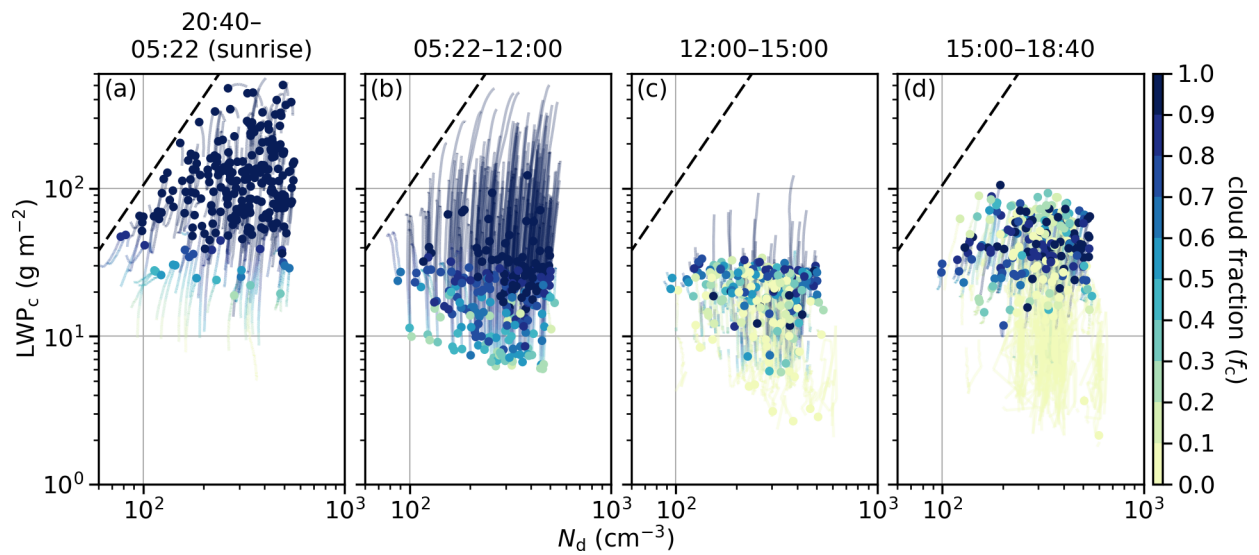
- 745 laume, S., and Thépaut, J.-N.: Complete ERA5 from 1940: Fifth generation of ECMWF atmospheric reanalyses of the global climate, <https://doi.org/10.24381/cds.143582cf>, 2017.
- Hersbach, H., Bell, B., Berrisford, P., Hirahara, S., Horányi, A., Muñoz-Sabater, J., Nicolas, J., Peubey, C., Radu, R., Schepers, D., Simmons, A., Soci, C., Abdalla, S., Abellan, X., Balsamo, G., Bechtold, P., Biavati, G., Bidlot, J., Bonavita, M., Chiara, G. D., Dahlgren, P., Dee, D., Diamantakis, M., Dragani, R., Flemming, J., Forbes, R. M., Fuentes, M., Geer, A., Haimberger, L., Healy, S., Hogan, 750 R. J., Hólm, E., Janisková, M., Keeley, S., Laloyaux, P., Lopez, P., Lupu, C., Radnoti, G., de Rosnay, P., Rozum, I., Vamborg, F., Villaume, S., and Thépaut, J.-N.: The ERA5 global reanalysis, *Quarterly Journal of the Royal Meteorological Society*, 146, 1999–2049, <https://doi.org/10.1002/qj.3803>, 2020.
- Hoffmann, F., Glassmeier, F., Yamaguchi, T., and Feingold, G.: Liquid water path steady states in stratocumulus: Insights from process-level emulation and mixed-layer theory, *Journal of the Atmospheric Sciences*, 77, 2203–2215, <https://doi.org/10.1175/JAS-D-19-0241.1>, 2020.
- 755 Hoffmann, F., Glassmeier, F., Yamaguchi, T., and Feingold, G.: On the roles of precipitation and entrainment in stratocumulus transitions between mesoscale states, *Journal of the Atmospheric Sciences*, 80, 2791–2803, <https://doi.org/10.1175/JAS-D-22-0268.1>, 2023.
- Iacono, M. J., Delamere, J. S., Mlawer, E. J., Shephard, M. W., Clough, S. A., and Collins, W. D.: Radiative forcing by long-lived greenhouse gases: Calculations with the AER radiative transfer models, *Journal of Geophysical Research*, 113, <https://doi.org/10.1029/2008jd009944>, 2008.
- 760 J. Kurowski, M., P. Malinowski, S., and W. Grabowski, W.: A numerical investigation of entrainment and transport within a stratocumulus-topped boundary layer, *Quarterly Journal of the Royal Meteorological Society*, 135, 77–92, <https://doi.org/10.1002/qj.354>, 2009.
- Jones, C. R., Bretherton, C. S., and Leon, D.: Coupled vs. decoupled boundary layers in VOCALS-REx, *Atmospheric Chemistry and Physics*, 11, 7143–7153, <https://doi.org/10.5194/acp-11-7143-2011>, 2011.
- Kazil, J., Wang, H., Feingold, G., Clarke, A. D., Snider, J. R., and Bandy, A. R.: Modeling chemical and aerosol processes in the transition 765 from closed to open cells during VOCALS-REx, *Atmospheric Chemistry and Physics*, 11, 7491–7514, <https://doi.org/10.5194/acp-11-7491-2011>, 2011.
- Kazil, J., Feingold, G., Wang, H., and Yamaguchi, T.: On the interaction between marine boundary layer cellular cloudiness and surface heat fluxes, *Atmospheric Chemistry and Physics*, 14, 61–79, <https://doi.org/10.5194/acp-14-61-2014>, 2014.
- Kazil, J., Feingold, G., and Yamaguchi, T.: Wind speed response of marine non-precipitating stratocumulus clouds over a diurnal cycle 770 in cloud-system resolving simulations, *Atmospheric Chemistry and Physics*, 16, 5811–5839, <https://doi.org/10.5194/acp-16-5811-2016>, 2016.
- Kazil, J., Yamaguchi, T., and Feingold, G.: Mesoscale organization, entrainment, and the properties of a closed-cell stratocumulus cloud, *Journal of Advances in Modeling Earth Systems*, 9, 2214–2229, <https://doi.org/10.1002/2017MS001072>, 2017.
- Kazil, J., Christensen, M. W., Abel, S. J., Yamaguchi, T., and Feingold, G.: Realism of Lagrangian large eddy simulations driven by reanalysis 775 meteorology: Tracking a pocket of open cells under a biomass burning aerosol layer, *Journal of Advances in Modeling Earth Systems*, 13, e2021MS002664, <https://doi.org/10.1029/2021MS002664>, 2021.
- Khairoutdinov, M. F. and Randall, D. A.: Cloud Resolving Modeling of the ARM Summer 1997 IOP: Model formulation, results, uncertainties, and sensitivities, *Journal of the Atmospheric Sciences*, 60, 607–625, [https://doi.org/10.1175/1520-0469\(2003\)060<0607:CRMOTA>2.0.CO;2](https://doi.org/10.1175/1520-0469(2003)060<0607:CRMOTA>2.0.CO;2), 2003.
- 780 Klein, S. A. and Hartmann, D. L.: The seasonal cycle of low stratiform clouds, *Journal of Climate*, 6, 1587–1606, [https://doi.org/10.1175/1520-0442\(1993\)006<1587:TSCOLS>2.0.CO;2](https://doi.org/10.1175/1520-0442(1993)006<1587:TSCOLS>2.0.CO;2), 1993.
- Latham, J.: Control of global warming?, *Nature*, 347, 339–340, <https://doi.org/10.1038/347339b0>, 1990.

- Lewis, E. R., Wiscombe, W. J., Albrecht, B. A., Bland, G. L., Flagg, C. N., Klein, S. A., Kollias, P., Mace, G., Reynolds, R. M., Schwartz, S. E., Siebesma, A. P., Teixeira, J., Wood, R., and Zhang, M.: MAGIC: Marine ARM GPCI investigation of clouds, Tech. Rep. DOE/SC-ARM-12-020, U.S. Department of Energy, last accessed: July 15, 2023, 2012.
- 785 Lilly, D. K.: Models of cloud-topped mixed layers under a strong inversion, *Quarterly Journal of the Royal Meteorological Society*, 94, 292–309, <https://doi.org/10.1002/qj.49709440106>, 1968.
- Manshausen, P., Watson-Parris, D., Christensen, M. W., Jalkanen, J.-P., and Stier, P.: Invisible ship tracks show large cloud sensitivity to aerosol, *Nature*, 610, 101–106, <https://doi.org/10.1038/s41586-022-05122-0>, 2022.
- 790 Matheou, G. and Teixeira, J. a.: Sensitivity to physical and numerical aspects of large-eddy simulation of stratocumulus, *Monthly Weather Review*, 147, 2621–2639, <https://doi.org/10.1175/MWR-D-18-0294.1>, 2019.
- Mellado, J. P., Bretherton, C. S., Stevens, B., and Wyant, M. C.: DNS and LES for simulating stratocumulus: Better together, *Journal of Advances in Modeling Earth Systems*, 10, 1421–1438, <https://doi.org/10.1029/2018MS001312>, 2018.
- Mlawer, E. J., Taubman, S. J., Brown, P. D., Iacono, M. J., and Clough, S. A.: Radiative transfer for inhomogeneous atmospheres: RRTM, a validated correlated-k model for the longwave, *Journal of Geophysical Research: Atmospheres*, 102, 16 663–16 682, <https://doi.org/10.1029/97JD00237>, 1997.
- 795 Moeng, C.-H., Stevens, B., and Sullivan, P. P.: Where is the interface of the stratocumulus-topped PBL?, *Journal of the Atmospheric Sciences*, 62, 2626–2631, <https://doi.org/10.1175/JAS3470.1>, 2005.
- Morrison, H., Jensen, A. A., Harrington, J. Y., and Milbrandt, J. A.: Advection of coupled hydrometeor quantities in bulk cloud microphysics schemes, *Monthly Weather Review*, 144, 2809–2829, <https://doi.org/10.1175/MWR-D-15-0368.1>, 2016.
- 800 Mülmenstädt, J. and Feingold, G.: The radiative forcing of aerosol–cloud interactions in liquid clouds: Wrestling and embracing uncertainty, *Current Climate Change Reports*, 4, 23–40, <https://doi.org/10.1007/s40641-018-0089-y>, 2018.
- Nicholls, S.: The dynamics of stratocumulus: aircraft observations and comparisons with a mixed layer model, *Quarterly Journal of the Royal Meteorological Society*, 110, 783–820, <https://doi.org/10.1002/qj.49711046603>, 1984.
- 805 Nowak, J. L., Siebert, H., Szodry, K.-E., and Malinowski, S. P.: Coupled and decoupled stratocumulus-topped boundary layers: Turbulence properties, *Atmospheric Chemistry and Physics*, 21, 10 965–10 991, <https://doi.org/10.5194/acp-21-10965-2021>, 2021.
- Ovtchinnikov, M. and Easter, R. C.: Nonlinear advection algorithms applied to interrelated tracers: Errors and implications for modeling aerosol–cloud interactions, *Monthly Weather Review*, 137, 632–644, <https://doi.org/10.1175/2008MWR2626.1>, 2009.
- Pedersen, J. G., Malinowski, S. P., and Grabowski, W. W.: Resolution and domain-size sensitivity in implicit large-eddy simulation of the stratocumulus-topped boundary layer, *Journal of Advances in Modeling Earth Systems*, 8, 885–903, <https://doi.org/10.1002/2015MS000572>, 2016.
- 810 Petters, J. L., Harrington, J. Y., and Clothiaux, E. E.: Radiative–dynamical feedbacks in low liquid water path stratiform clouds, *Journal of the Atmospheric Sciences*, 69, 1498–1512, <https://doi.org/10.1175/JAS-D-11-0169.1>, 2012.
- Pincus, R. and Baker, M. B.: Effect of precipitation on the albedo susceptibility of clouds in the marine boundary layer, *Nature*, 372, 250–252, <https://doi.org/10.1038/372250a0>, 1994.
- 815 Possner, A., Wang, H., Wood, R., Caldeira, K., and Ackerman, T. P.: The efficacy of aerosol extendashcloud radiative perturbations from near-surface emissions in deep open-cell stratocumuli, *Atmospheric Chemistry and Physics*, 18, 17 475–17 488, <https://doi.org/10.5194/acp-18-17475-2018>, 2018.
- Prabhakaran, P., Hoffmann, F., and Feingold, G.: Evaluation of pulse aerosol forcing on marine stratocumulus clouds in the context of marine cloud brightening, *Journal of the Atmospheric Sciences*, 80, 1585–1604, <https://doi.org/10.1175/JAS-D-22-0207.1>, 2023.
- 820

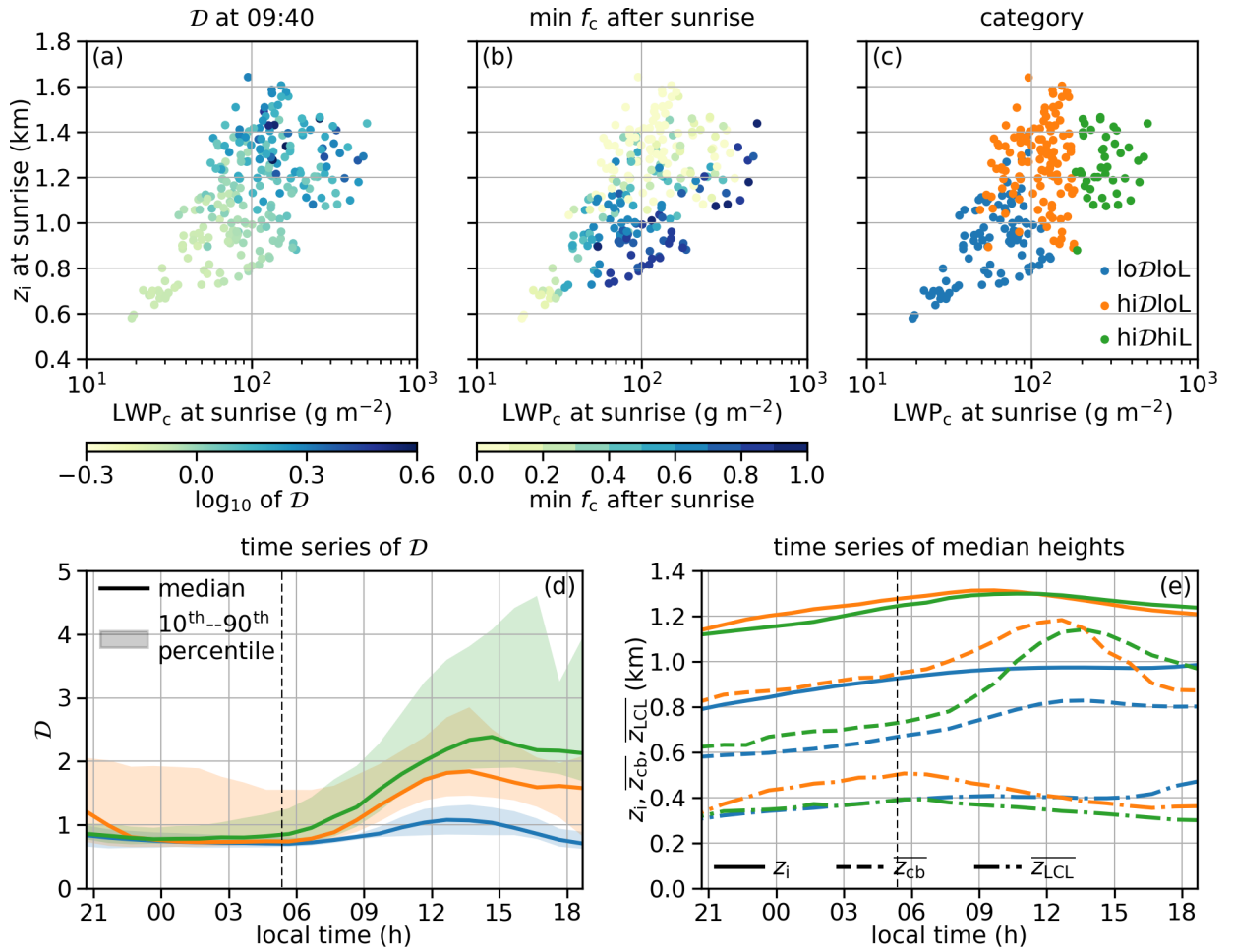
- Qiu, S., Zheng, X., Painemal, D., Terai, C. R., and Zhou, X.: Daytime variation in the aerosol indirect effect for warm marine boundary layer clouds in the eastern North Atlantic, *Atmospheric Chemistry and Physics*, 24, 2913–2935, <https://doi.org/10.5194/acp-24-2913-2024>, 2024.
- Sandu, I. and Stevens, B.: On the factors modulating the stratocumulus to cumulus transitions, *Journal of the Atmospheric Sciences*, 68, 1865–1881, <https://doi.org/10.1175/2011JAS3614.1>, 2011.
- Sandu, I., Brenguier, J.-L., Geoffroy, O., Thouron, O., and Masson, V.: Aerosol impacts on the diurnal cycle of marine stratocumulus, *Journal of the Atmospheric Sciences*, 65, 2705–2718, <https://doi.org/10.1175/2008JAS2451.1>, 2008.
- Smalley, K. M., Lebsock, M. D., and Eastman, R.: Diurnal patterns in the observed cloud liquid water path response to droplet number perturbations, *Geophysical Research Letters*, 51, <https://doi.org/10.1029/2023GL107323>, 2024.
- 830 Stevens, B. and Feingold, G.: Untangling aerosol effects on clouds and precipitation in a buffered system, *Nature*, 461, 607–613, <https://doi.org/10.1038/nature08281>, 2009.
- Stevens, B., Moeng, C.-H., Ackerman, A. S., Bretherton, C. S., Chlond, A., de Roode, S., Edwards, J., Golaz, J.-C., Jiang, H., Khairoutdinov, M., Kirkpatrick, M. P., Lewellen, D. C., Lock, A., Müller, F., Stevens, D. E., Whelan, E., and Zhu, P.: Evaluation of large-eddy simulations via observations of nocturnal marine stratocumulus, *Monthly Weather Review*, 133, 1443–1462, <https://doi.org/10.1175/MWR2930.1>, 2005.
- 835 Stevens, D. E., Bell, J. B., Almgren, A. S., Beckner, V. E., and Rendleman, C. A.: Small-scale processes and entrainment in a stratocumulus marine boundary layer, *Journal of the Atmospheric Sciences*, 57, 567–581, [https://doi.org/10.1175/1520-0469\(2000\)057<0567:SSPAEI>2.0.CO;2](https://doi.org/10.1175/1520-0469(2000)057<0567:SSPAEI>2.0.CO;2), 2000.
- Teixeira, J., Cardoso, S., Bonazzola, M., Cole, J., DelGenio, A., DeMott, C., Franklin, C., Hannay, C., Jakob, C., Jiao, Y., Karlsson, J., Kitagawa, H., Köhler, M., Kuwano-Yoshida, A., LeDrian, C., Li, J., Lock, A., Miller, M. J., Marquet, P., Martins, J., Mechoso, C. R., v. Meijgaard, E., Meinke, I., Miranda, P. M. A., Mironov, D., Neggers, R., Pan, H. L., Randall, D. A., Rasch, P. J., Rockel, B., Rossow, W. B., Ritter, B., Siebesma, A. P., Soares, P. M. M., Turk, F. J., Vaillancourt, P. A., Engeln, A. V., and Zhao, M.: Tropical and subtropical cloud transitions in weather and climate prediction models: The GCSS/WGNE Pacific Cross-section Intercomparison (GPCI), *Journal of Climate*, 24, 5223–5256, <https://doi.org/10.1175/2011JCLI3672.1>, 2011.
- 840 Toll, V., Christensen, M., Quaas, J., and Bellouin, N.: Weak average liquid-cloud-water response to anthropogenic aerosols, *Nature*, 572, 51–55, <https://doi.org/10.1038/s41586-019-1423-9>, 2019.
- Turton, J. D. and Nicholls, S.: A study of the diurnal variation of stratocumulus using a multiple mixed layer model, *Quarterly Journal of the Royal Meteorological Society*, 113, 969–1009, <https://doi.org/10.1002/qj.49711347712>, 1987.
- Twomey, S.: Pollution and the planetary albedo, *Atmospheric Environment*, 8, 1251–1256, [https://doi.org/10.1016/0004-6981\(74\)90004-3](https://doi.org/10.1016/0004-6981(74)90004-3), 1974.
- 850 Twomey, S.: The influence of pollution on the shortwave albedo of clouds, *Journal of the Atmospheric Sciences*, 34, 1149–1152, [https://doi.org/10.1175/1520-0469\(1977\)034<1149:TIOPOP>2.0.CO;2](https://doi.org/10.1175/1520-0469(1977)034<1149:TIOPOP>2.0.CO;2), 1977.
- van der Dussen, J. J., de Roode, S. R., Ackerman, A. S., Blossey, P. N., Bretherton, C. S., Kurowski, M. J., Lock, A. P., Neggers, R. A. J., Sandu, I., and Siebesma, A. P.: The GASS/EUCLIPSE model intercomparison of the stratocumulus transition as observed during ASTEX: LES results, *Journal of Advances in Modeling Earth Systems*, 5, 483–499, <https://doi.org/10.1002/jame.20033>, 2013.
- 855 van der Dussen, J. J., de Roode, S. R., and Siebesma, A. P.: Factors controlling rapid stratocumulus cloud thinning, *Journal of the Atmospheric Sciences*, 71, 655–664, <https://doi.org/10.1175/JAS-D-13-0114.1>, 2014.

- Wall, C. J., Storelvmo, T., and Possner, A.: Global observations of aerosol indirect effects from marine liquid clouds, *Atmospheric Chemistry and Physics*, 23, 13 125–13 141, <https://doi.org/10.5194/acp-23-13125-2023>, 2023.
- 860 Wang, H. and Feingold, G.: Modeling mesoscale cellular structures and drizzle in marine stratocumulus. Part I: Impact of drizzle on the formation and evolution of open cells, *Journal of the Atmospheric Sciences*, 66, 3237–3256, <https://doi.org/10.1175/2009JAS3022.1>, 2009a.
- Wang, H. and Feingold, G.: Modeling mesoscale cellular structures and drizzle in marine stratocumulus. Part II: The microphysics and dynamics of the boundary region between open and closed cells, *Journal of the Atmospheric Sciences*, 66, 3257–3275, <https://doi.org/10.1175/2009JAS3120.1>, 2009b.
- 865 Wang, H., Feingold, G., Wood, R., and Kazil, J.: Modelling microphysical and meteorological controls on precipitation and cloud cellular structures in Southeast Pacific stratocumulus, *Atmospheric Chemistry and Physics*, 10, 6347–6362, <https://doi.org/10.5194/acp-10-6347-2010>, 2010.
- Wang, S., Wang, Q., and Feingold, G.: Turbulence, condensation, and liquid water transport in numerically simulated nonprecipitating stratocumulus clouds, *Journal of the Atmospheric Sciences*, 60, 262–278, [https://doi.org/10.1175/1520-0469\(2003\)060<0262:TCALWT>2.0.CO;2](https://doi.org/10.1175/1520-0469(2003)060<0262:TCALWT>2.0.CO;2), 2003.
- 870 Wood, R.: Cancellation of aerosol indirect effects in marine stratocumulus through cloud thinning, *Journal of the Atmospheric Sciences*, 64, 2657–2669, <https://doi.org/10.1175/JAS3942.1>, 2007.
- Wood, R.: Stratocumulus clouds, *Monthly Weather Review*, 140, 2373–2423, <https://doi.org/10.1175/MWR-D-11-00121.1>, 2012.
- 875 Wood, R. and Bretherton, C. S.: On the relationship between stratiform low cloud cover and lower-tropospheric stability, *Journal of Climate*, 19, 6425–6432, <https://doi.org/10.1175/JCLI3988.1>, 2006.
- Yamaguchi, T., Randall, D. A., and Khairoutdinov, M. F.: Cloud modeling tests of the ULTIMATE–MACHO scalar advection scheme, *Monthly Weather Review*, 139, 3248–3264, <https://doi.org/10.1175/MWR-D-10-05044.1>, 2011.
- Yamaguchi, T., Feingold, G., Kazil, J., and McComiskey, A.: Stratocumulus to cumulus transition in the presence of elevated smoke layers, *Geophysical Research Letters*, 42, 10–478, <https://doi.org/10.1002/2015GL066544>, 2015.
- 880 Yamaguchi, T., Feingold, G., and Kazil, J.: Stratocumulus to cumulus transition by drizzle, *Journal of Advances in Modeling Earth Systems*, 9, 2333–2349, <https://doi.org/10.1002/2017MS001104>, 2017.
- Yamaguchi, T., Feingold, G., and Kazil, J.: Aerosol-cloud interactions in trade wind cumulus clouds and the role of vertical wind shear, *Journal of Geophysical Research: Atmospheres*, 124, 12 244–12 261, <https://doi.org/10.1029/2019JD031073>, 2019.
- 885 Yuan, T., Song, H., Wood, R., Oreopoulos, L., Platnick, S., Wang, C., Yu, H., Meyer, K., and Wilcox, E.: Observational evidence of strong forcing from aerosol effect on low cloud coverage, *Science Advances*, 9, <https://doi.org/10.1126/sciadv.adh7716>, 2023.
- Zhang, J. and Feingold, G.: Distinct regional meteorological influences on low-cloud albedo susceptibility over global marine stratocumulus regions, *Atmospheric Chemistry and Physics*, 23, 1073–1090, <https://doi.org/10.5194/acp-23-1073-2023>, 2023.
- Zhang, J., Zhou, X., Goren, T., and Feingold, G.: Albedo susceptibility of northeastern Pacific stratocumulus: The role of covarying meteorological conditions, *Atmospheric Chemistry and Physics*, 22, 861–880, <https://doi.org/10.5194/acp-22-861-2022>, 2022.
- 890 Zhang, J., Chen, Y.-S., Yamaguchi, T., and Feingold, G.: Cloud water adjustments to aerosol perturbations are buffered by solar heating in non-precipitating marine stratocumuli, *Atmospheric Chemistry and Physics*, 24, 10 425–10 440, <https://doi.org/10.5194/acp-24-10425-2024>, 2024.
- Zhou, X. and Bretherton, C. S.: Simulation of mesoscale cellular convection in marine stratocumulus: 2. Nondrizzling conditions, *Journal of Advances in Modeling Earth Systems*, 11, 3–18, <https://doi.org/10.1029/2018MS001448>, 2019.
- 895

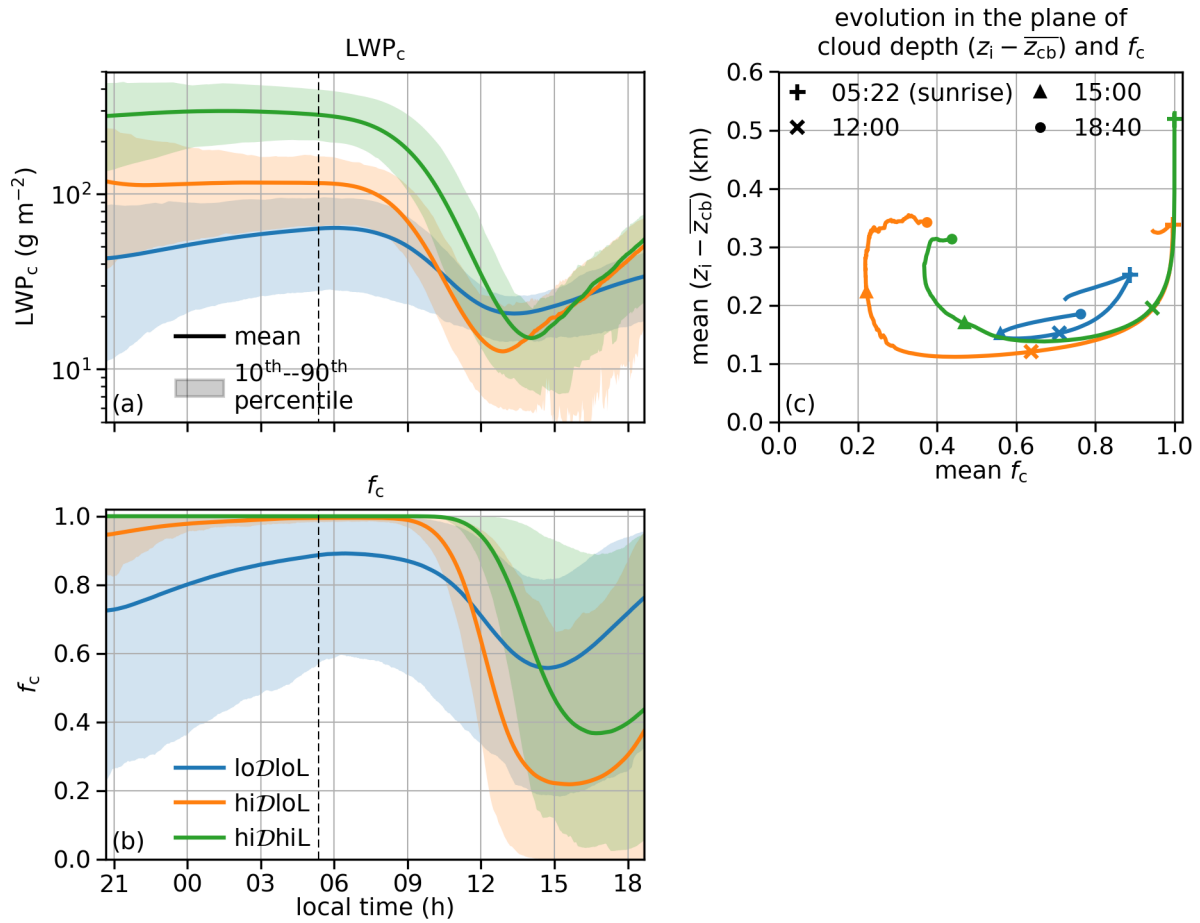
Zhou, X., Kollias, P., and Lewis, E. R.: Clouds, precipitation, and marine boundary layer structure during the MAGIC field campaign, *Journal of Climate*, 28, 2420–2442, <https://doi.org/10.1175/JCLI-D-14-00320.1>, 2015.



**Figure 1.** Evolution of the simulations in the plane of cloud droplet number concentration ( $N_d$ ) and cloud LWP ( $LWP_c$ ), split in to four time periods as shown in the panel titles. Curves indicate the trajectories over the time period and dots indicate the states at the end of the time period. The thick black dashed lines correspond to a characteristic mean drop radius of  $12 \mu\text{m}$ , below which precipitation is inhibited.

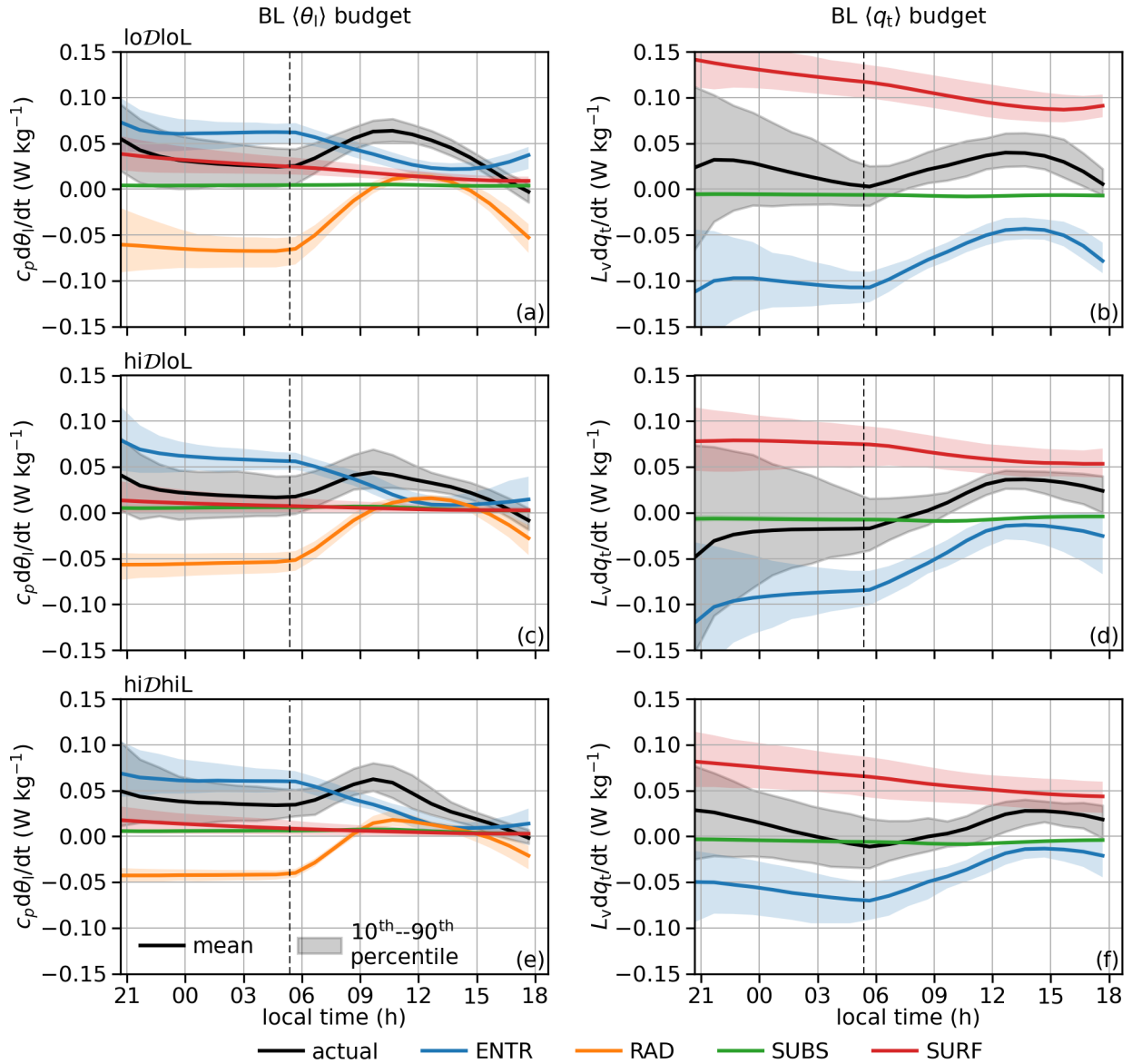


**Figure 2.** (a) Relative decoupling index ( $\mathcal{D}$ ) at 09:40 and (b) minimum cloud fraction ( $\min f_c$ ) after sunrise in the plane of inversion base height ( $z_i$ ) and cloud LWP ( $LWP_c$ ) at sunrise; (c) categories based on  $\mathcal{D}$  at 09:40 and  $LWP_c$  at sunrise: (1) loDloL ( $\mathcal{D} \leq 1$ ), (2) hiDloL ( $\mathcal{D} > 1$  and  $LWP_c \leq 180 \text{ g m}^{-2}$ ), and (3) hiDhiL ( $\mathcal{D} > 1$  and  $LWP_c > 180 \text{ g m}^{-2}$ ); time series of (d) median and quantiles of  $\mathcal{D}$  and (e) medians of  $z_i$ ,  $\bar{z}_{cb}$ , and  $\bar{z}_{LCL}$  by category. The vertical dashed black lines in Panels (d) and (e) indicate sunrise.

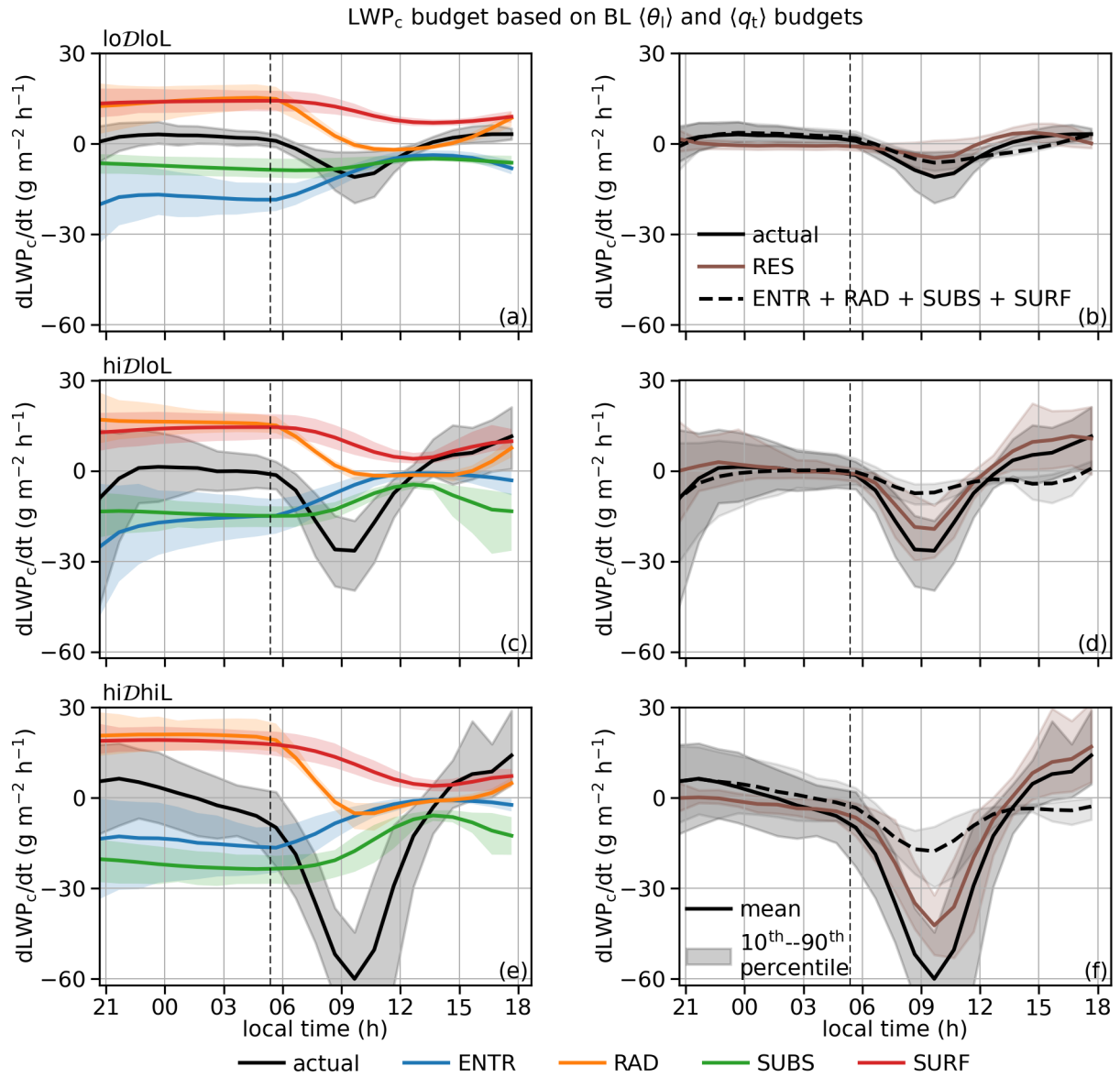


**Figure 3.** Time series of (a) cloud LWP ( $LWP_c$ ) and (b) cloud fraction ( $f_c$ ); (c) evolution by category in the plane of cloud depth ( $z_i - \bar{z}_{LCL}$ ). The vertical dashed black lines in Panels (a) and (b) indicate sunrise.

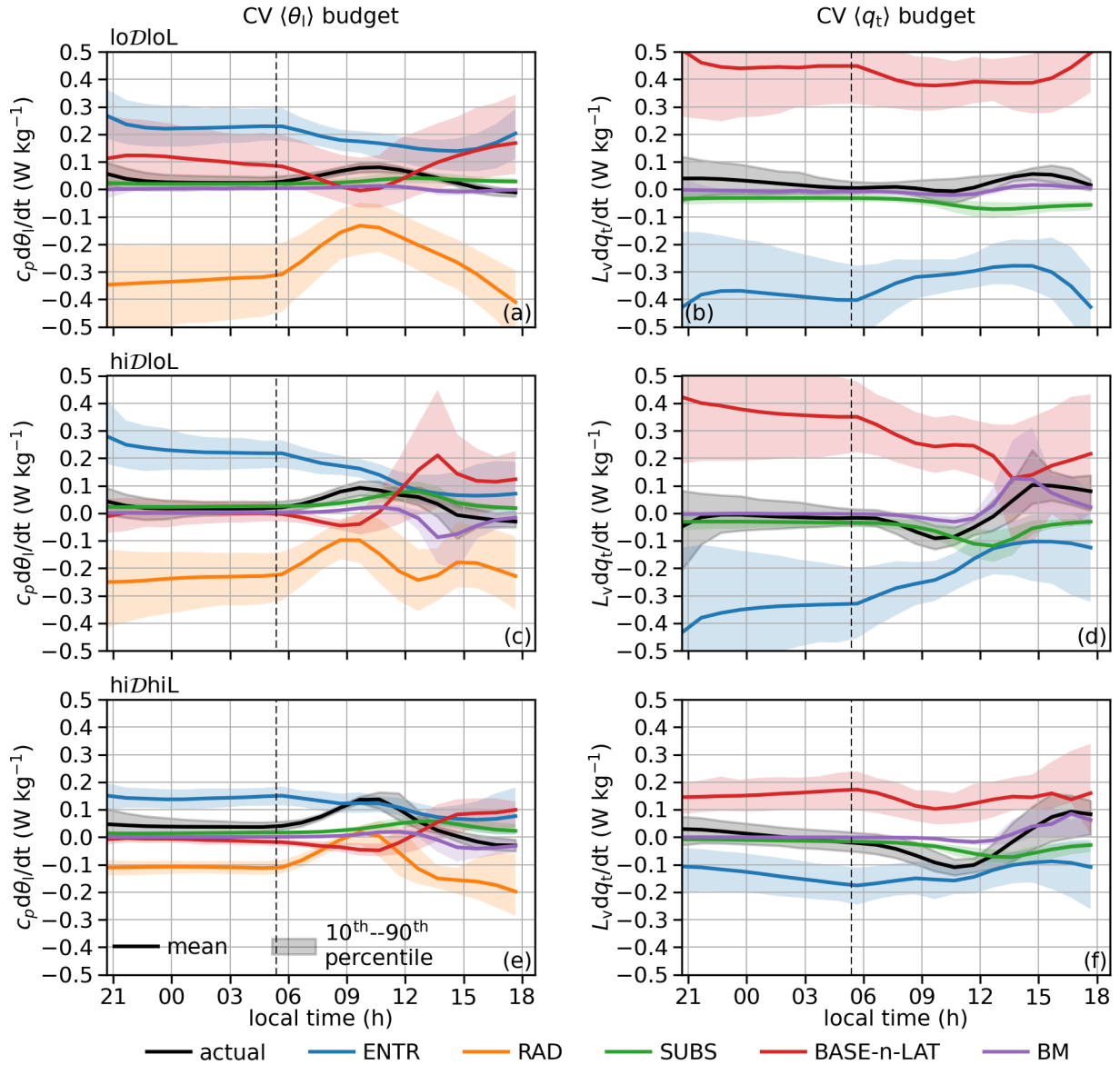




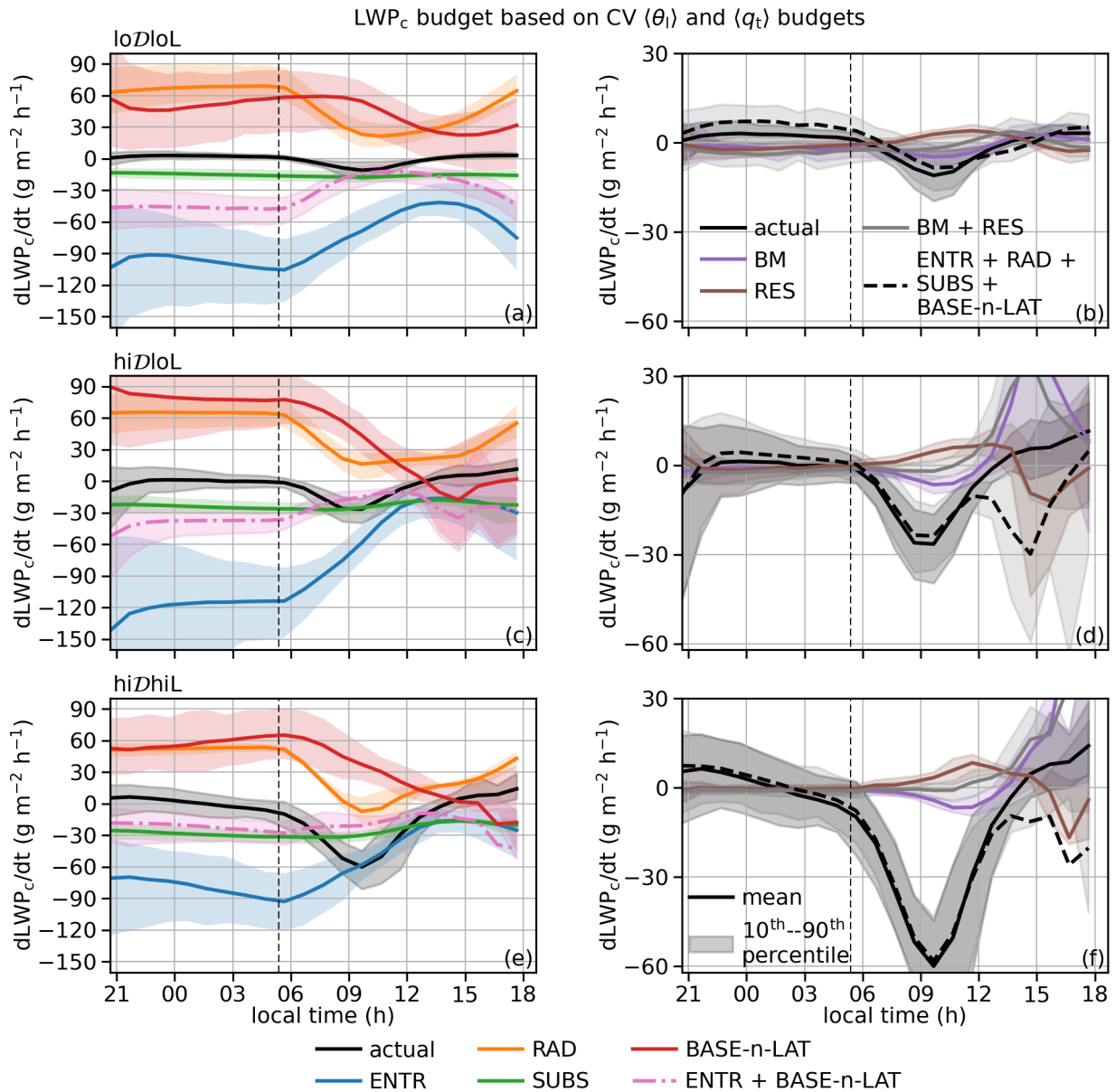
**Figure 4.** Time series of actual BL  $\langle \theta_t \rangle$  and  $\langle q_t \rangle$  tendencies and budget terms due to individual processes by category. The vertical dashed black lines indicate sunrise.



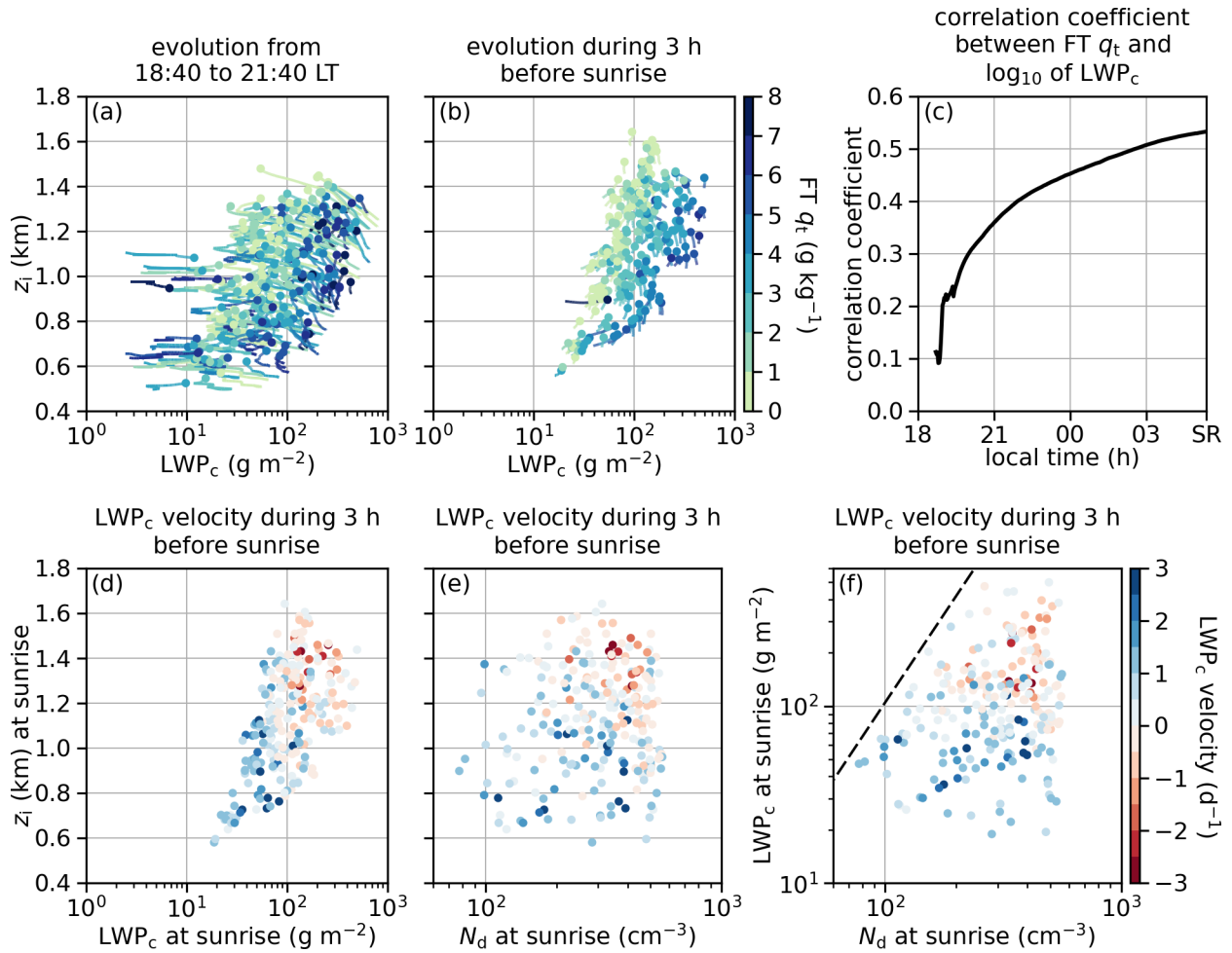
**Figure 5.** Time series of LWP<sub>c</sub> tendencies and budget terms due to individual processes by category, based on BL  $\langle \theta_l \rangle$  and  $\langle q_t \rangle$  budgets. The actual LWP<sub>c</sub> tendencies are shown in both the left and right columns for easier comparison with individual budget terms. The vertical dashed black lines indicate sunrise.



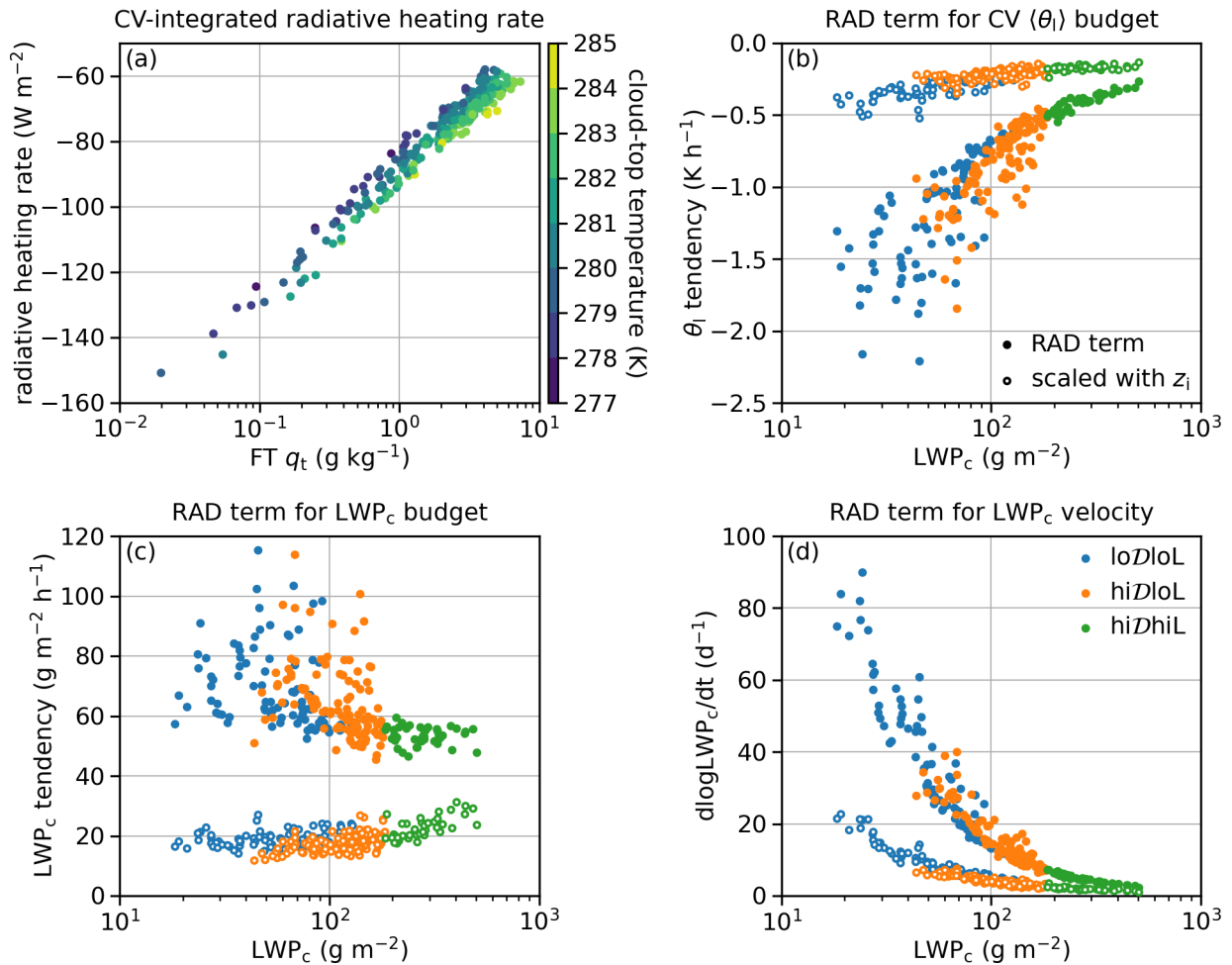
**Figure 6.** Time series of actual CV  $\langle \theta_t \rangle$  and  $\langle q_t \rangle$  tendencies and budget terms due to individual processes by category. The vertical dashed black lines indicate sunrise.



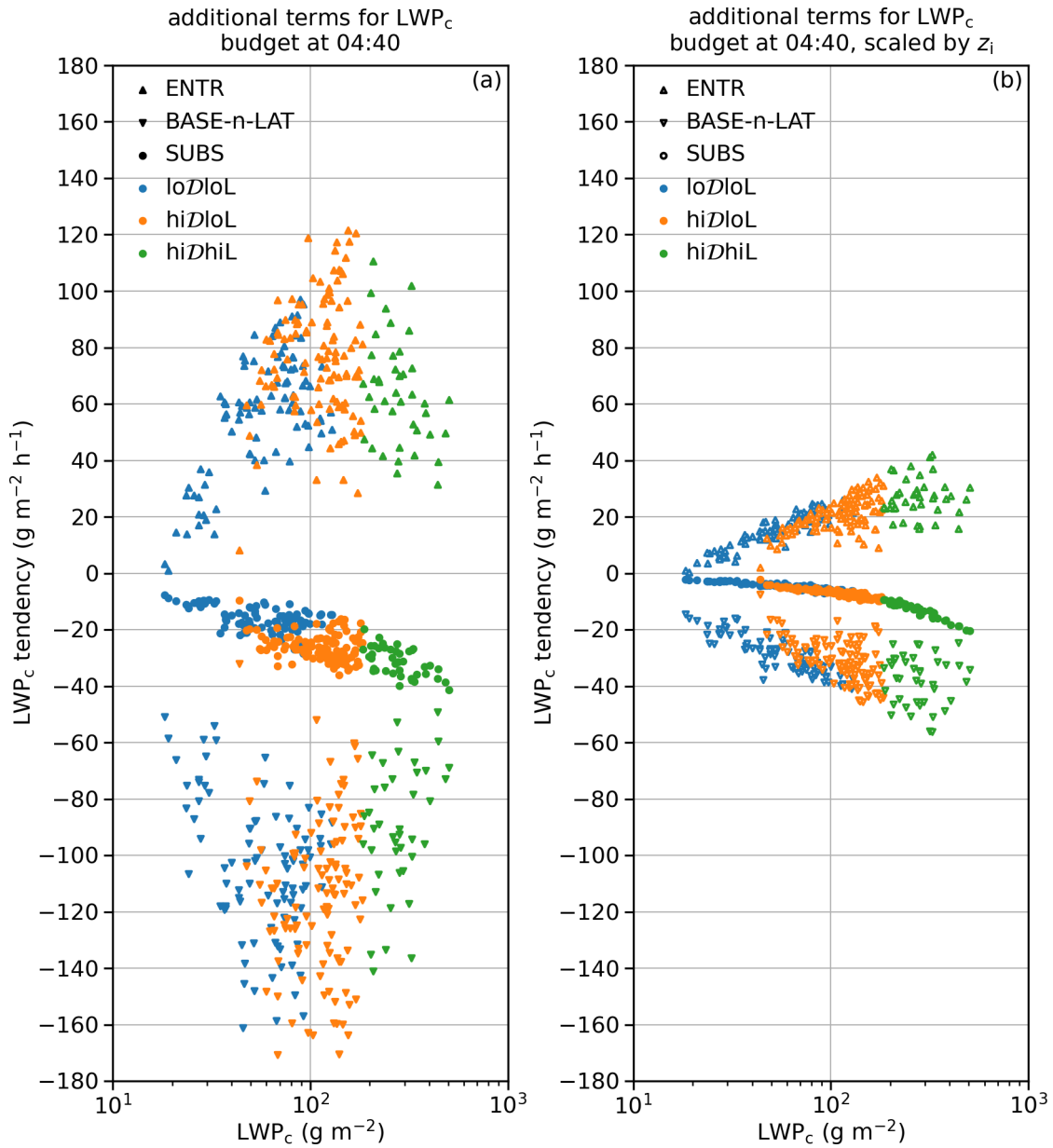
**Figure 7.** Time series of LWP<sub>c</sub> tendencies and budget terms due to individual processes by category, based on CV  $\langle\theta_t\rangle$  and  $\langle q_t\rangle$  budgets. The actual LWP<sub>c</sub> tendencies are shown in both the left and right columns for easier comparison with individual budget terms. The vertical dashed black lines indicate sunrise.



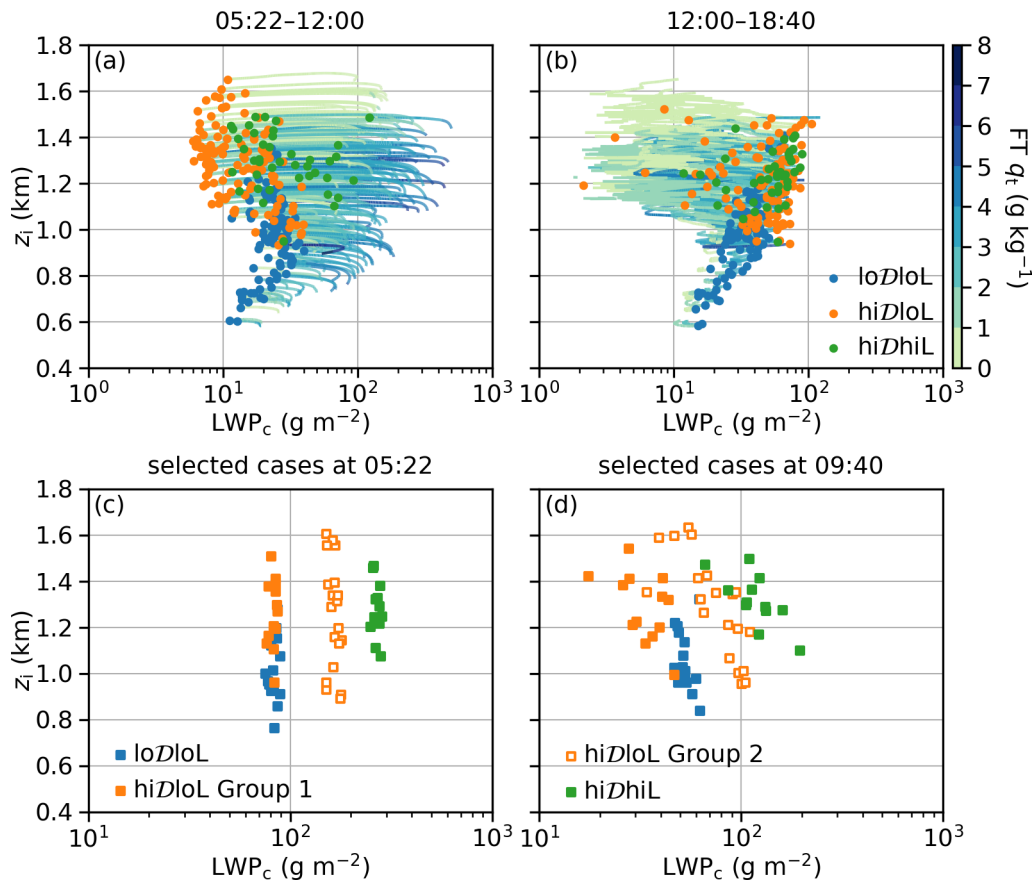
**Figure 8.** Evolution of LES ensemble members during nighttime. In Panels (a) and (b), curves indicate the trajectories over the time period, and dots indicate the states at the end of the time period shown in the panel titles. “SR” indicates sunrise.



**Figure 9.** Radiative cooling at 04:40 LT. (a) CV-integrated radiative heating rate, (b) RAD term for CV  $\langle \theta_i \rangle$  budget, (c) RAD term for LWP<sub>c</sub> budget, (d) radiative contribution to LWP<sub>c</sub> velocity. Hollowed circles in Panels (b) and (c) represent the tendencies when the CV-integrated radiative heating rate is hypothetically uniformly distributed over the entire BL depth.

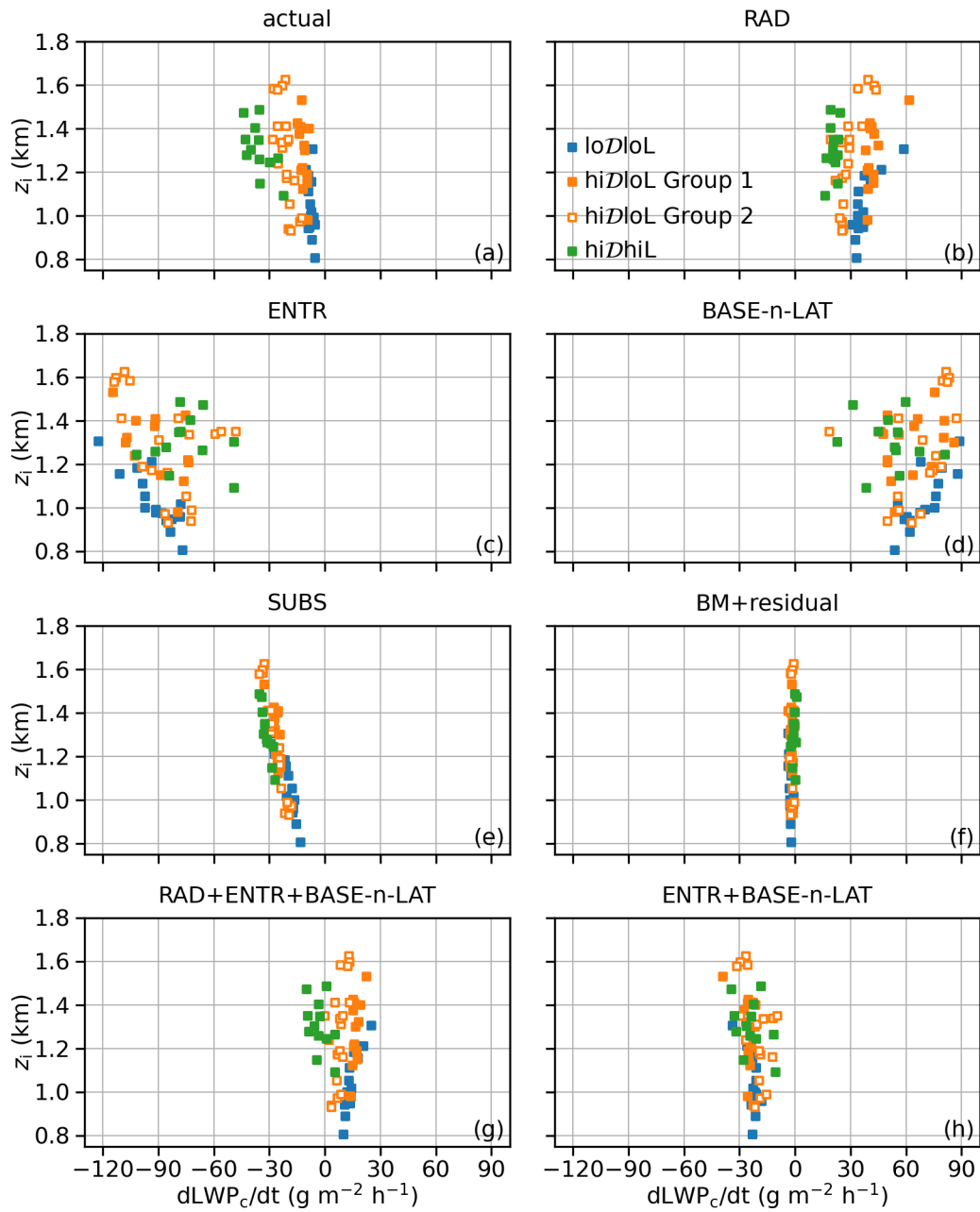


**Figure 10.** Co-variability between ENTR, BASE-n-LAT, and SUBS terms for LWP<sub>c</sub> budget and LWP<sub>c</sub> at 04:40 LT.

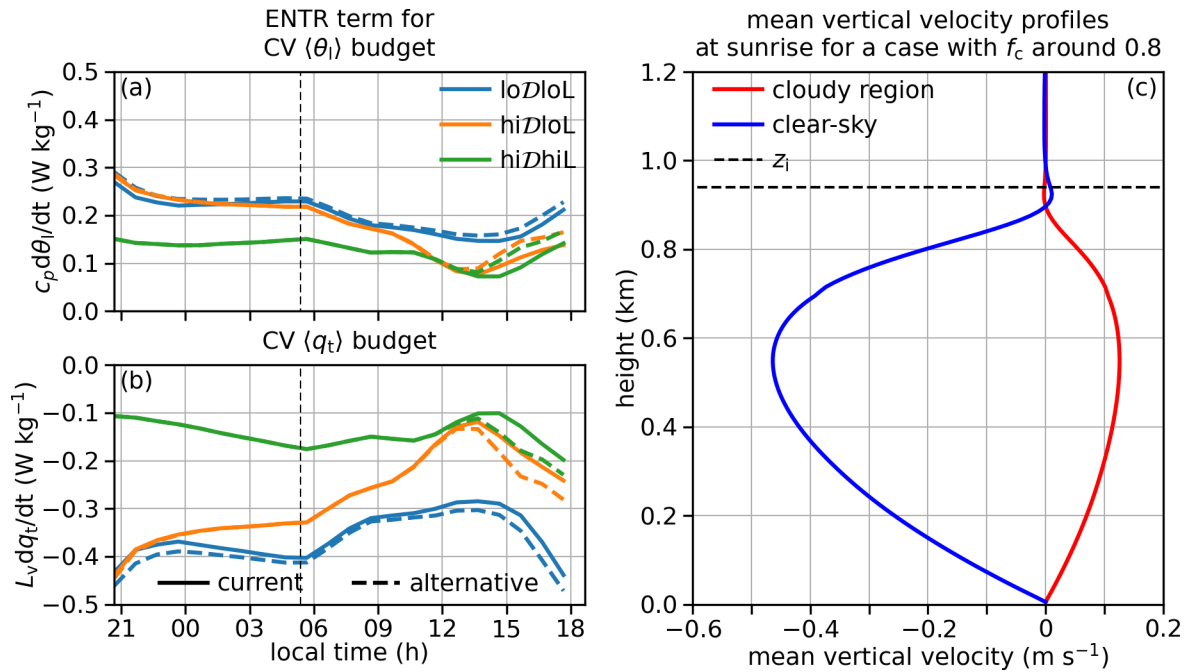


**Figure 11.** Evolution of LES ensemble members during daytime. In Panels (a) and (b), curves indicate the trajectories over the time period and dots indicate the states at the end of the time period, shown in the panel titles. Symbols in Panels (c) and (d) indicate groups of cases that are selected for further examination: (1) loDloL cases with  $LWP_c$  at sunrise between  $75$  and  $90 \text{ g m}^{-2}$  (2) hiDloL cases with  $LWP_c$  at sunrise in the same range (hiDloL Group 1), (3) hiDloL cases with  $LWP_c$  at sunrise between  $150$  and  $180 \text{ g m}^{-2}$  (hiDloL Group 2), and (4) hiDhiL cases with  $LWP_c$  at sunrise between  $240$  and  $300 \text{ g m}^{-2}$ .

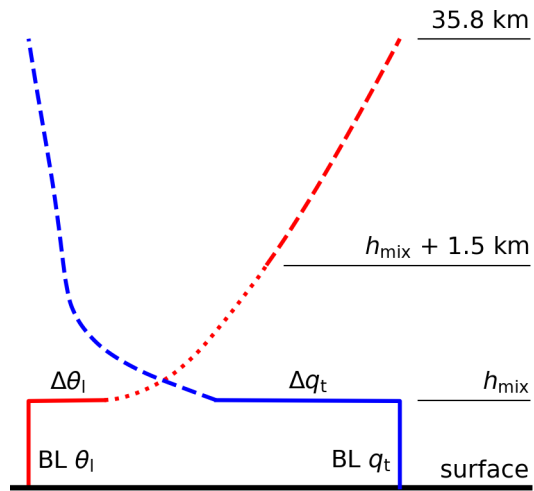




**Figure 12.** Mean  $LWP_c$  tendencies and budget terms due to individual processes for selected cases between sunrise and 09:40.



**Figure 13.** Time series of current and alternative estimates of the entrainment contribution to CV (a)  $\langle \theta_t \rangle$  and (b)  $\langle q_t \rangle$  budgets. The vertical dashed black lines indicate sunrise. Panel (c) shows an example to facilitate the discussions near the end of Section 6.1.



**Figure A1.** A sketch showing the construction of initial  $\theta_1$  and  $q_t$  profiles (in red and blue, respectively) from initial BL profiles (solid segments), ERA5-based climatological profiles (dashed segments) and the MAGIC-based transitional  $\theta_1$  profile (dotted segment).