# Diurnal Patterns in the Observed Cloud Liquid Water Path Response to Droplet Number Perturbations

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November 22, 2023

#### Abstract

A key uncertainty in Aerosol-cloud interactions is the cloud liquid water path (LWP) response to increased aerosols  $(\lambda)$ . LWP can either increase due to precipitation suppression or decrease due to entrainment-drying. Previous research suggests that precipitation suppression dominates in thick clouds, while entrainment-drying prevails in thin clouds. The time scales of the two competing effects are vastly different, requiring temporally resolved observations. We analyze 3-day Lagrangian trajectories of stratocumulus clouds over the southeast Pacific using geostationary data. We find that clouds with a LWP exceeding 200 g m-2 exhibit a positive response, while clouds with lower LWP show a negative response. We observe a significant diurnal cycle in  $\lambda$ , indicating a more strongly negative daytime adjustment driven by entrainment-drying. In contrast, at night, precipitation suppression can occasionally fully counteract the entrainment-drying mechanism. The time-integrated adjustment appears weaker than previously suggested in studies that do not account for the diurnal cycle.









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### 6 Key Points:

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# The adjustment of cloud liquid water path to aerosols in stratocumulus clouds is generally negative, except for the thickest clouds.

• There is a strong diurnal cycle in the adjustment of cloud liquid water path to aerosols.

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### 10 Abstract

A key uncertainty in Aerosol-cloud interactions is the cloud liquid water path (LWP) re-11 sponse to increased aerosols ( $\lambda$ ). LWP can either increase due to precipitation suppres-12 sion or decrease due to entrainment-drying. Previous research suggests that precipita-13 tion suppression dominates in thick clouds, while entrainment-drying prevails in thin clouds. 14 The time scales of the two competing effects are vastly different, requiring temporally 15 resolved observations. We analyze 3-day Lagrangian trajectories of stratocumulus clouds 16 over the southeast Pacific using geostationary data. We find that clouds with a LWP ex-17 ceeding 200 g  $m^{-2}$  exhibit a positive response, while clouds with lower LWP show a neg-18 ative response. We observe a significant diurnal cycle in  $\lambda$ , indicating a more strongly 19 negative daytime adjustment driven by entrainment-drying. In contrast, at night, pre-20 cipitation suppression can occasionally fully counteract the entrainment-drying mech-21 anism. The time-integrated adjustment appears weaker than previously suggested in stud-22 ies that do not account for the diurnal cycle. 23

### 24 Plain Language Summary

We examine how aerosols affect cloud properties, specifically cloud liquid water path (LWP). We find that the impact of aerosols on LWP varies with cloud thickness and time of day. Thicker clouds are more influenced by entrainment-drying during the day and precipitation suppression at night, while thinner clouds that are less likely to precipitate and tend to produce less intense precipitation are much more susceptible to entrinment drying no matter time of day. Overall, this nuanced understanding of how LWP responds to aerosols may help constrain the influence of aerosol-cloud-interactions on climate.

### 32 1 Introduction

The effective radiative forcing from cloud-aerosol interactions  $(ERF_{ACI})$  in marine 33 boundary layer clouds are a leading source of uncertainty in future climate projections 34 (Mülmenstädt & Feingold, 2018; Seinfeld et al., 2016). These uncertainties result from 35 ambiguity in how individual cloud properties (i.e. cloud liquid water path (LWP), cloud 36 fraction, and cloud drop size) adjust to aerosols (e.g. Christensen et al., 2020; Douglas 37 & L'Ecuyer, 2020). Whereas the sensitivity of cloud drop size to aerosol perturbation 38 is fairly well understood (Twomey, 1977), the response of LWP and cloud fraction is less 39 well constrained. In particular, LWP adjustment can follow two pathways: 1) LWP may 40

increase due to suppressed precipitation following the second-indirect aerosol effect in
more polluted environments (Albrecht, 1989) or 2) LWP may decrease in response to more
efficient entrainment-drying near cloud top (Ackerman et al., 2004; Bretherton et al., 2007).
The relative magnitude of both processes significantly affects aerosol impacts on cloud
radiative properties.

The sensitivity of LWP to changes in cloud droplet number concentration is quan-46 tified as  $\lambda = \frac{dlnLWP}{dlnN_d}$ . Previous high-resolution large-eddy simulation (LES) and ob-47 servational studies generally agree that  $\lambda$  is positive in precipitating clouds but negative 48 in non-precipitating clouds (e.g. Ackerman et al., 2004; Fons et al., 2023; Glassmeier et 49 al., 2021; Gryspeerdt et al., 2022; Hill et al., 2009; Lebsock et al., 2008; Lee et al., 2009; 50 Michibata et al., 2016; Prabhakaran et al., 2023; Toll et al., 2019). The implication of 51 this finding is that the  $\text{ERF}_{ACI}$  is largest in regimes that tend to produce precipitation, 52 whereas regimes that tend not to produce precipitation tend to have an  $\text{ERF}_{ACI}$  that 53 is less than the Twomey effect (Twomey, 1977). To visualize this, Equation 1 shows how 54 albedo (A<sub>c</sub>) changes with N<sub>d</sub> in response to changes in  $\lambda$  (Boers & Mitchell, 1994; Plat-55 nick & Twomey, 1994). This shows that  $\lambda$  needs to be less than -0.4 to fully counter-56 act the Twomey effect. Interestingly, Qiu et al. (2023) and Zhou and Feingold (2023) ob-57 served  $\lambda$  values below -0.4 in thick non-precipitating and the smallest closed-cell stra-58 tocumulus. These findings, while intriguing, represent exceptions compared to most es-59 timates, which rarely fall below -0.4. This suggests that the negative adjustment due to 60 entrainment-drying often falls short of fully countering the Twomey Effect. 61

$$\frac{dA_c}{dN_d} = \frac{A_c(1-A_c)}{3N_d} \left(1 + \frac{5}{2}\lambda\right) = \begin{cases} \text{Brightening} & \text{if } \lambda > -0.4\\ \text{Darkening} & \text{if } \lambda < -0.4 \end{cases}$$
(1)

Adding to the complexity of interpreting  $\lambda$  is the diurnal cycle of both entraine-62 ment and precipitation in stratocumulus. Of note, Diamond et al. (2020) compared morn-63 ing  $\lambda$  estimated from the Moderate Resolution Imaging Spectroradiometer (MODIS) on-64 board Terra to afternoon  $\lambda$  estimated from MODIS onboard Aqua within the southeast 65 Atlantic shipping corridors. They found that  $\lambda$  generally becomes more negative from 66 morning to afternoon, implying that the influence of entrainment-drying on LWP increases 67 throughout the day. From an LES perspective, Sandu et al. (2008) found that LWP in-68 creases at night and decreases during the day, with diurnal changes being much larger 69

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in the most polluted environments. Their diurnal cycle in LWP sensitivity coincides with 70 the diurnal cycle in stratocumulus precipitation, which peaks before sunset and gener-71 ally decreases throughout the day (Burleyson et al., 2013). Based on this limited evi-72 dence, one could speculate that a diurnal cycle of  $\lambda$  that is modulated by the diurnal cy-73 cle of precipitation may exist. The observed magnitude of  $\lambda$  and its potential diurnal cy-74 cle may shed light on the practicality of marine-cloud brightening - a proposed solar ra-75 diation management strategy (Diamond et al., 2022; Hoffmann & Feingold, 2021; Prab-76 hakaran et al., 2023; Wood, 2021). However, comprehensive observations are necessary 77 78 to validate this hypothesis.

Given that  $\lambda$  may change throughout the day, and the time-scale of the adjustment 79 is on the order 20 hours Glassmeier et al. (2021), using polar-orbiting satellites for anal-80 ysis limit our ability to provide observational constraints on the diurnal cycle of  $\lambda$ . Pre-81 vious studies have developed innovative techniques to make inferences about  $\lambda$  using MODIS 82 measurements from Terra and Aqua. For instance, Gryspeerdt et al. (2022) identified 83 a weak LWP-N<sub>d</sub> relationship that is highly dependent on the initial cloud state. How-84 ever, a limitation is that Terra and Aqua provide only two data points at a given loca-85 tion every 24 hours, and those samples are limited to two discrete times of day. To ad-86 dress this limitation, Christensen et al. (2023) combined geostationary satellite obser-87 vations with polar orbiters and ground-based stations to quantify  $\lambda$  in the U.S. Depart-88 ment of Energy's Energy Exascale Earth System Model. Consistent with other research, 89 they found that  $\lambda$  is typically negative during the day. However, their approach used geo-90 stationary observations to track changes in cloud state, rather than directly measuring 91 LWP. In our study, we employ a combination of geostationary LWP dataset that has been 92 corrected for scattering geometry related biases and microwave imagery which is insen-93 sitive to solar geometery to assess the diurnal variations of  $\lambda$ . 94

- 95 **2** Data and Methods
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### 2.1 Corrected ABI Cloud Liquid Water Path

<sup>97</sup> We use cloud-optical depth ( $\tau$ ) and cloud-top effective radius ( $r_e$ ) pixels retrieved <sup>98</sup> from the liquid-only (Pavolonis, 2020) GOES-16 Advanced-Baseline Imager (ABI) (Walther <sup>99</sup> & Straka, 2020) from 2019 to 2021 (downloaded from NOAA CLASS; https://www.avl .class.noaa.gov) to calculate low-cloud LWP using equation 17 from (Grosvenor et

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al., 2018) for solar zenith angles  $< 70^{\circ}$ . This method assumes an adiabatic increase in 101 liquid-water content with height and a constant number concentration. We apply the 102 corrections described in Smalley and Lebsock (2023a) to mitigate the scattering geom-103 etry bias, which are ubiquitous features of bi-spectral cloud microphysical retrievals. These 104 corrections adjust the LWP over a  $1^{\circ}x1^{\circ}$  degree area to that which would be observed 105 by that of microwave imagers, that do not suffer scattering biases. Smalley and Lebsock 106 (2023a) demonstrate that the corrected ABI LWP is able to reproduce the diurnal cy-107 cle of LWP observed by the fleet of microwave imagers but with the benefit of the 10-108 minute temporal resolution of ABI. 109

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### 2.2 Microwave Cloud Liquid Water Path

For solar zenith angles  $> 70^{\circ}$ , we supplement the corrected daytime ABI data with 111 LWP derived from the passive microwave imagers listed in Table S1. These data are down-112 loaded from Remote Sensing Systems (RSS; https://www.remss.com). RSS utilizes 37-113 GHz brightness temperatures for each satellite to derive LWP at a resolution of  $0.25^{\circ}$ 114  $x 0.25^{\circ}$ , employing the same algorithm and calibration procedure (e.g. Wentz, 1997; Wentz 115 & Spencer, 1998) to mitigate biases between sensors. Five of the six satellites are sun-116 synchronous but have varying equatorial-crossing times. This, in conjunction with the 117 Global Precipitation Measurement Microwave Imager (which operates in a processing 118 orbit), enables us to sample throughout the nocturnal portion of the diurnal cycle. To 119 ensure the use of LWP data in regions free from potential bias caused by ice clouds or 120 precipitation, we follow the procedure described by Smalley and Lebsock (2023a). 121

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#### 2.3 MODIS Cloud Droplet Number Concentration

We use cloud droplet number derived from the level-2 Terra and Aqua MODIS col-123 lection 6.1 cloud product optical depth and effective radius at 2.1  $\mu$ m (Platnick et al., 124 2015) co-located with GOES-16. Although number concentration from MODIS and ABI 125 are based on similar theoretical bases, Figure S1 shows that ABI has a significant low 126 bias relative to lidar observations (Hu et al., 2021), that MODIS does not have. We spec-127 ulate that the bias in the ABI data relates to the larger footprint of the ABI compared 128 to MODIS and the presence of ABI effective radii larger than 30  $\mu$ m which is the cut-129 off value for the MODIS retrieved liquid  $r_e$ . 130

### 131 2.4 Lagrangian Analysis

Following Smalley et al. (2022), we generate trajectories initiated from every 50 MODIS 132 pixels during each Terra overpass. To focus the results on Stratocumulus conditions, tra-133 jectories are filtered for initial conditions with an estimated inversion strength (Wood 134 & Bretherton, 2006) greater than 9.5 K. The median initial cloud fraction of these tra-135 jectories is 80%, which is consistent with the Stratocumulus cloud type. We calculate 136 the average initial  $N_d$  within a 1°x1° gridbox around each trajectory point from the MODIS 137 pixel level optical depth and effective radius. We then calculate the time evolution of the 138 LWP using either the bias corrected ABI during the day or the microwave data, when 139 available, at night. Note that the combined LWP is either directly derived from the mi-140 crowave imager retreivals or from ABI retreivals that have been corrected to reproduce 141 that same microwave product ensuring consistency across the diurnal cycle. 142

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### 2.5 AMSR-2 Precipitation Rates

We utilize the Advanced Microwave Scanning Radiometer 2 (AMSR-2) warm precipitation product developed by Eastman et al. (2019) to measure changes in precipitation intensity along all trajectories. The product relies on the statistical relationship between 3x5 km<sup>2</sup> AMSR-2 89-GHz brightness temperatures and collocated CloudSat precipitation rates. Because this dataset cannot precisely discriminate precipitation, we employ a threshold of 0.1 mm day<sup>-1</sup> to distinguish between raining and non-raining AMSR-2 pixels, in a manner similar to Smalley et al. (2022).

Unfortunately, the AMSR-2 data alone cannot be employed to determine how pre-151 cipitation intensity varies along any single trajectory, as it operates in a sun-synchronous 152 orbit. Therefore, we undertake the following steps: 1) colocate  $1^{\circ}x1^{\circ}$  unconditionally (in-153 cluding non-precipitating pixels) averaged 2019 AMSR-2 precipitation intensity over the 154 southeast Pacific with GOES-16, and 2) create a lookup table of mean precipitation in-155 tensity for a given ABI LWP and ABI N<sub>d</sub> (as shown in Figure S2). Subsequently, we de-156 termine the expected precipitation rate at each point by mapping observed ABI LWP 157 and  $N_d$  values back to the lookup table to find the mean precipitation at all time points 158 along all trajectories. Note that this analysis is limited to daytime-only precipitation rates 159 because the ABI LWP and  $N_d$  are unavailable at night. 160

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### $_{161}$ 2.6 Cloud Liquid Water Path Sensitivity to initial $N_d$

# We assume that any potential changes in $\lambda$ that may result from entrainment-drying or precipitation suppression are small and might be masked by the diurnal and seasonal cycles in LWP. Therefore, we remove the geographical, seasonal and diurnal cycles (using 2019 – 2021 observations) from LWP (Eq. 2) before calculating $\lambda$ .

$$CLWP' = \ln(CLWP[\text{time} = t]) - \overline{\ln(CLWP[\text{local hour, month, lat, lon]})}$$
 (2)

As demonstrated in Figure S3, we calculate  $\lambda[t]$  as the slope of the fit between  $\ln (LWP[t]) - \ln (LWP[t=0])$  and  $\ln (N_d)$  [t=0])). All estimates of  $\lambda[t]$  are then grouped by initial LWP and averaged to determine how  $\lambda$  varies over time as a function of initial LWP. To reduce noise, we smooth each calculated curve by applying a 6-hour centered running mean.

### <sup>170</sup> 3 Cloud Liquid Water Path Adjustment

Figure 1 demonstrates that, except for the thickest clouds (initial LWP  $\gtrsim 200 \text{ g m}^{-2}$ ), 171  $\lambda$  generally tends to be negative, with values decreasing during the day and then increas-172 ing at night. Notably the strength of the diurnal cycle of  $\lambda$  is modulated by the initial 173 LWP, with the trajectories with highest LWP, and therefore the greatest tendency to pre-174 cipitate, having the largest diurnal cycle. While entrainment-drying and precipitation 175 are both likely to maximize during the nightime hours (Chun et al., 2023), the obvious 176 diurnal cycle suggests a stronger diurnal amplitude of the precipitation suppression mech-177 anism relative to the entrainment-drying mechanism, which results in a near balance in 178 the two processes in the early morning hours for several of the LWP curves. Specifically, 179  $\lambda$  starts to rise at night and becomes positive in some cases, likely due to stratocumu-180 lus thickening and an increased likelihood of intense precipitation (Burleyson et al., 2013). 181 For the highest LWP bin,  $\lambda$  is nearly always positive indicating the dominance of the pre-182 cipitation suppression mechanism over the entrainment-drying mechanism for these clouds 183 which are most likely to precipitate regardless of time of day. 184

<sup>185</sup> To quantify the diurnal cycle, Figure 2 displays the autocorrelation function for all <sup>186</sup>  $\lambda$  values shown in Figure 1. Note that linear interpolation was used to fill gaps at night <sup>187</sup> before computing the autocorrelation function (see Figure S4). For the thickest clouds <sup>188</sup> (initial LWP > 50 g m<sup>-2</sup>), Figure 2 reveals a statistically significant diurnal cycle, with



Figure 1. The sensitivity in cloud liquid water path (CLWP) to initial cloud-droplet number concentration  $(N_d)$  conditioned by initial CLWP (line colors). The white-filled regions represent day, and the grey-filled regions represent night.

autocorrelation peaking approximately every 24 hours. For thinner clouds, the autocorrelation drops to zero within 12 hours and does not have a clearly statistically significant diurnal cycle. The sensitivity of the diurnal cycle to LWP strongly suggests that the precipitation suppression process is a critical factor in establishing the diurnal variation in  $\lambda$ .

We have already speculated that the susceptibility of precipitation to aerosol drives the diurnal cycle in  $\lambda$ . Figure 3 substantiates this claim by showing the diurnal (daytime) pattern in precipitation rates binned by LWP. Unsurprisingly there is a strong diurnal cycle in the precipitation rates with a minimum in the late afternoon. However, the amplitude of the diurnal variability increases with initial LWP. Although there is a diurnal cycle in precipitation rates among the thinnest clouds, it is less pronounced com-

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Figure 2. The autocorrelation function in the  $\lambda$  curves shown in Figure 1, where each curve represents clouds conditioned by initial CLWP (line colors), where the solid black line represents an autocorrelation of zero, the grey dashed lines represent the 99<sup>th</sup> percentile, and the grey solid lines represent the 95<sup>th</sup> percentile. The white-filled regions represent day, and the grey-filled regions represent night.

pared to the thickest clouds. This, combined with the fact that the thinnest clouds are less likely to produce rainfall (see Figure S2) likely explains the tendency of the diurnal cycle observed in  $\lambda$  to increase with increasing LWP.

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### 4 ABI Cloud Water Path Adjustment Comparison to Prior Studies

As demonstrated in Equation 1, stratocumulus clouds can brighten even when  $\lambda$ is negative. Only a few prior observational (Qiu et al., 2023; Zhou & Feingold, 2023) and LES (Glassmeier et al., 2021) studies have found  $\lambda$  values small enough to darken stratocumulus. Figure 4a compares the mean  $\lambda$  along each composite trajectory with previous studies (Supplemental Table S2). It shows that the mean  $\lambda$  is generally negative for all but the thickest clouds (initial LWP > 200 g m<sup>-2</sup>), indicating that entrainmentdrying is dominant over precipitation suppression most of the time. While consistent with



Figure 3. The statistical precipitation intensity as determined by using collocated AMSR-2 warm rain rates and GOES-16 ABI CLWP and  $N_d$  (Figure S4) along all trajectories composited by initial ABI CLWP (line colors).

prior studies, our  $\lambda$  values are typically much closer to zero than in most earlier estimates. Specifically we find values of  $\lambda$  that suggest a less significant entrainment-drying mechanism than many prior observational and LES studies.

The diurnal variation of  $\lambda$  suggests that the integrated daytime adjustment should 214 be stronger than the weak overall adjustment shown in Figure 4a. Figure 4b illustrates 215 how the daytime mean  $\lambda$  compares to the mean  $\lambda$  integrated along the full composite 216 trajectories. It reveals that the daytime mean  $\lambda$  is consistently smaller than the full mean 217  $\lambda$  in all cases. However, even though the negative adjustment appears stronger during 218 the day, it never reaches values indicative of a complete offset of the Twomey effect. The 219 differences between  $\lambda$  calculated from the diurnal vs the daytime only data explain some 220 of the difference between this study and prior studies which are based on daytime ob-221 servations from visible/near infrared imagery. 222



Figure 4. Dots (first column) represent the mean  $\lambda$  conditioned along each initial LWP curve shown in Figure 1. Boxplots represent the distribution of  $\lambda$  split between non-raining (turquoise), raining (yellow), and indiscriminate (orange) cases from prior observation-based (second column) and LES (third column) studies. All values are given in table S1. The filled-blue region represents situations where the cloud-field should darken, the grey-filled region represents situations where the cloud-field should brighten despite  $\lambda$  being negative, and the red-filled region represents situations where the cloud-field should brighten.

### <sup>223</sup> 5 Conclusions

Consistent with previous work we find that the sensitivity of stratocumulus LWP to changes in number concentration depends on the initial LWP. Clouds with the largest LWP tend to have a positive sensitivity to increased N<sub>d</sub>, whereas regimes with LWP < about 200 g m<sup>-2</sup> tend to have a negative sensitivity. The difference is presumably related to the dominant mechanism being different for thick clouds (precipitation suppression) and thin clouds (entrainment drying). For analogous reasons, our results emphasize that the adjustment of LWP to increasing aerosol concentrations has a distinct di-

### manuscript submitted to Geophysical Research Letters

urnal pattern. Throughout the day, we observe a general decrease in  $\lambda$ , with the most 231 negative values occurring late in the afternoon when precipitation is at its lightest. Af-232 ter sunset,  $\lambda$  begins to increase as clouds thicken and precipitation intensity increases. 233 This diurnal variation in  $\lambda$  suggests that while entrainment-drying remains active at night, 234 it is most observable during the day when stratocumulus clouds thin out, and precip-235 itation becomes less frequent and intense. At night, as the stratocumulus deck thickens, 236 the impact of precipitation suppression on  $\lambda$  becomes more pronounced relative to entrainment-237 drying. 238

Outside of the stratocumulus clouds most likely to produce the most intense precipitation, the increases in  $\lambda$  at night are insufficient to completely counterbalance the predominantly negative adjustment observed during the day. This typically results in a weak negative adjustment over a period of several days. However, we find that it never reaches magnitudes significant enough to completely offset the Twomey effect.

Our results suggest that marine-cloud brightening is most effective when aerosols are used to seed clouds that are already producing precipitation. This is because the negative impact on LWP in non-precipitating clouds helps counteract the Twomey effect. Furthermore, the significant diurnal cycle in  $\lambda$  implies that the efficacy of any intentional aerosol injection will likely be sensitive to the time of day when it occurs and this dependence will be cloud regime dependent.

### <sup>250</sup> 6 Open Research

GOES-16 Advanced-Baseline Imager cloud optical properties can be downloaded 251 from https://www.avl.class.noaa.gov, and ABI liquid water path was corrected us-252 ing lookup tables available at Zenodo (Smalley & Lebsock, 2023b). The following MERRA-253 2 products: inst3\_3d\_asm\_Np (Global Modeling and Assimilation Office (GMAO), 2015b) 254 and inst1\_2d\_asm\_Nx (Global Modeling and Assimilation Office (GMAO), 2015a) can 255 be downloaded from the Goddard Space Flight Center Distributed Active Archive Cen-256 ter. Passive Microwave liquid water path can be downloaded from Remote Sensing Sys-257 tems (https://www.remss.com). MODIS Level-2 cloud optical properties can be down-258 loaded from the Level-1 and Atmosphere Archive & Distribution System Distributed Ac-259 tive Archive Center (https://ladsweb.modaps.eosdis.nasa.gov). The AMSR-2 warm 260 rain product can be downloaded from the CloudSat Data Processing Center (https:// 261

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- 262 www.cloudsat.cira.colostate.edu/community-products/warm-rain-rate-estimates
- -from-amsr-89ghz-and-cloudsat). The code used to create and plot the output tra-
- jectory data used in this study are permanently archived at Zenodo (doi: XX.YY.ZZ).

### 265 Acknowledgments

- The research was carried out at the Jet Propulsion Laboratory, California Institute of
- <sup>267</sup> Technology, under a contract with the National Aeronautics and Space Administration

(80NM0018D0004). Ryan Eastman is supported by XX Grant.

### 269 References

- Ackerman, A. S., Kirkpatrick, M. P., Stevens, D. E., & Toon, O. B. (2004). The
   impact of humidity above stratiform clouds on indirect aerosol climate forcing.
   *Nature*, 432(7020), 1014–1017. doi: 10.1038/nature03174
- Albrecht, B. A. (1989). Aerosols, cloud microphysics, and fractional cloudiness. *Science*, 245(4923), 1227–1230. doi: 10.1126/science.245.4923.1227
- Boers, R., & Mitchell, R. M. (1994). Absorption feedback in stratocumulus clouds:
   Influence on cloud top albedo. *Tellus A*, 46(3), 229–241. doi: 10.1034/j.1600
   -0870.1994.00001.x
- Bretherton, C. S., Blossey, P. N., & Uchida, J. (2007). Cloud droplet sedimentation,
   entrainment efficiency, and subtropical stratocumulus albedo. *Geophysical Research Letters*, 34(3). doi: https://doi.org/10.1029/2006GL027648
- Burleyson, C. D., de Szoeke, S. P., Yuter, S. E., Wilbanks, M., & Brewer, W. A.
- (2013). Ship-based observations of the diurnal cycle of southeast pacific marine
   stratocumulus clouds and precipitation. Journal of the Atmospheric Sciences,
   70(12), 3876–3894. doi: https://doi.org/10.1175/JAS-D-13-01.1
- Christensen, M. W., Jones, W. K., & Stier, P. (2020). Aerosols enhance cloud
  lifetime and brightness along the stratus-to-cumulus transition. *Proceed- ings of the National Academy of Sciences*, 117(30), 17591–17598. doi:
  10.1073/pnas.1921231117
- <sup>289</sup> Christensen, M. W., Ma, P.-L., Wu, P., Varble, A. C., Mülmenstädt, J., & Fast,
- J. D. (2023). Evaluation of aerosol-cloud interactions in e3sm using a lagrangian framework. Atmospheric Chemistry and Physics, 23(4), 2789–2812. doi: 10.5194/acp-23-2789-2023

293	Chun, JY., Wood, R., Blossey, P., & Doherty, S. J. (2023). Microphysical, macro-					
294	physical, and radiative responses of subtropical marine clouds to aerosol					
295	injections. Atmospheric Chemistry and Physics, 23(2), 1345–1368. Re-					
296	trieved from https://acp.copernicus.org/articles/23/1345/2023/ doi:					
297	10.5194/acp-23-1345-2023					
298	Diamond, M. S., Director, H. M., Eastman, R., Possner, A., & Wood, R. (2020).					
299	Substantial cloud brightening from shipping in subtropical low clouds. $AGU$					
300	Advances, 1(1), e2019AV000111.doi: 10.1029/2019AV000111					
301	Diamond, M. S., Gettelman, A., Lebsock, M. D., McComiskey, A., Russell, L. M.,					
302	Wood, R., & Feingold, G. (2022). To assess marine cloud brightening's tech-					
303	nical feasibility, we need to know what to study—and when to stop. Pro-					
304	ceedings of the National Academy of Sciences, $119(4)$ , e2118379119. doi:					
305	10.1073/pnas.2118379119					
306	Douglas, A., & L'Ecuyer, T. (2020). Quantifying cloud adjustments and the radia-					
307	tive forcing due to aerosol–cloud interactions in satellite observations of warm					
308	marine clouds. Atmospheric Chemistry and Physics, $20(10)$ , $6225-6241$ . doi:					
309	10.5194/acp-20-6225-2020					
310	Eastman, R., Lebsock, M., & Wood, R. (2019). Warm rain rates from amsr-e 89-ghz					
311	brightness temperatures trained using clouds at rain-rate observations. $Jour$ -					
312	nal of Atmospheric and Oceanic Technology, 36(6), 1033-1051. doi: https://doi					
313	.org/10.1175/JTECH-D-18-0185.1					
314	Fons, E., Runge, J., Neubauer, D., & Lohmann, U. (2023). Stratocumulus adjust-					
315	ments to aerosol perturbations disentangled with a causal approach. $npj$ Cli-					
316	mate and Atmospheric Science, $6(1)$ , 130. doi: 10.1038/s41612-023-00452-w					
317	Glassmeier, F., Hoffmann, F., Johnson, J. S., Yamaguchi, T., Carslaw, K. S., & Fein-					
318	gold, G. (2021). Aerosol-cloud-climate cooling overestimated by ship-track					
319	data. Science, 371(6528), 485–489. doi: 10.1126/science.abd3980					
320	Global Modeling and Assimilation Office (GMAO). (2015a). $inst1_2d_asm_nx: 2d, 1-d_asm_nx: 2d$					
321	$hourly, instantaneous, single-level, assimilation, single-level diagnostics \ 0.625 \ x$					
322	0.5 degree v5.12.4. Goddard Space Flight Center Distributed Active Archive					
323	Center (GSFC DAAC), Greenbelt, MD, USA. (Accessed January 20, 2023)					
324	doi: 10.5067/3Z173KIE2TPD					
325	Global Modeling and Assimilation Office (GMAO). (2015b). <i>inst3_3d_asm_np:</i>					

326	$Merra\mathchar`2\mathchar`3d, 3-hourly, instantaneous, pressure-level, assimilation, assimilated$					
327	meteorological fields v5.12.4 (m2i3npasm). Goddard Space Flight Center					
328	Distributed Active Archive Center (GSFC DAAC), Greenbelt, MD, USA.					
329	(Accessed January 17, 2023) doi: 10.5067/QBZ6MG944HW0					
330	Grosvenor, D. P., Sourdeval, O., Zuidema, P., Ackerman, A., Alexandrov, M. D.,					
331	Bennartz, R., Quaas, J. (2018). Remote sensing of droplet num-					
332	ber concentration in warm clouds: A review of the current state of knowl-					
333	edge and perspectives. Reviews of Geophysics, $56(2)$ , 409–453. doi:					
334	https://doi.org/10.1029/2017 RG000593					
335	Gryspeerdt, E., Glassmeier, F., Feingold, G., Hoffmann, F., & Murray-Watson, R. J.					
336	(2022). Observing short-timescale cloud development to constrain aerosol–					
337	cloud interactions. Atmospheric Chemistry and Physics, 22(17), 11727–11738.					
338	doi: 10.5194/acp-22-11727-2022					
339	Hill, A. A., Feingold, G., & Jiang, H. (2009). The influence of entrainment and mix-					
340	ing assumption on aerosol–cloud interactions in marine stratocumulus. $Journal$					
341	of the Atmospheric Sciences, 66(5), 1450–1464. doi: https://doi.org/10.1175/					
342	2008JAS2909.1					
343	Hoffmann, F., & Feingold, G. (2021). Cloud microphysical implications for					
344	marine cloud brightening: The importance of the seeded particle size dis-					
345	tribution. Journal of the Atmospheric Sciences, 78(10), 3247–3262. doi:					
346	10.1175/JAS-D-21-0077.1					
347	Hu, Y., Lu, X., Zhai, PW., Hostetler, C. A., Hair, J. W., Cairns, B., Wood,					
348	R. (2021). Liquid phase cloud microphysical property estimates from					
349	calipso measurements. Frontiers in Remote Sensing, 2. Retrieved from					
350	https://www.frontiersin.org/articles/10.3389/frsen.2021.724615					
351	doi: $10.3389/\text{frsen}.2021.724615$					
352	Lebsock, M. D., Stephens, G. L., & Kummerow, C. (2008). Multisensor					
353	satellite observations of aerosol effects on warm clouds. Journal of Geo-					
354	physical Research: Atmospheres, 113(D15). Retrieved from https://					
355	agupubs.onlinelibrary.wiley.com/doi/abs/10.1029/2008JD009876 doi:					
356	https://doi.org/10.1029/2008JD009876					
357	Lee, S. S., Penner, J. E., & Saleeby, S. M. (2009). Aerosol effects on liquid-water					
358	path of thin stratocumulus clouds. Journal of Geophysical Research: Atmo-					

359	spheres, $114(D7)$ . doi: https://doi.org/10.1029/2008JD010513						
360	Michibata, T., Suzuki, K., Sato, Y., & Takemura, T. (2016). The source of dis-						
361	crepancies in aerosol–cloud–precipitation interactions between gcm and a-train						
362	retrievals. Atmospheric Chemistry and Physics, 16(23), 15413–15424. doi:						
363	10.5194/acp-16-15413-2016						
364	Mülmenstädt, J., & Feingold, G. (2018). The radiative forcing of aerosol–cloud inter-						
365	actions in liquid clouds: Wrestling and embracing uncertainty. Current Climate						
366	Change Reports, $4(1)$ , 23–40. doi: 10.1007/s40641-018-0089-y						
367	Pavolonis, M. (2020). Enterprise algorithm theoretical basis document for cloud						
368	type and cloud phase (NOAA Tech. Rep. No. 113 pp). NOAA. Retrieved from						
369	https://www.star.nesdis.noaa.gov/goesr/rework/documents/ATBDs/						
370	Enterprise/Enterprise_ATBD_Cloud_CldType_G17_Mitigation_Jun2020						
371	.pdf						
372	Platnick, S., Ackerman, S. A., King, M. D., Meyer, K., Menzel, W. P., Holz, R. E.,						
373	Yang, P. (2015). Modis atmosphere l2 cloud product (06_l2). NASA MODIS						
374	Adaptive Processing System, Goddard Space Flight Center. (Last accessed:						
375	September 26, 2023) doi: dx.doi.org/10.5067/MODIS/MOD06\_L2.006						
376	Platnick, S., & Twomey, S. (1994). Determining the susceptibility of cloud albedo						
377	to changes in droplet concentration with the advanced very high resolution						
378	radiometer. Journal of Applied Meteorology and Climatology, 33(3), 334–347.						
379	doi: $10.1175/1520-0450(1994)033(0334:DTSOCA)2.0.CO;2$						
380	Prabhakaran, P., Hoffmann, F., & Feingold, G. (2023). Evaluation of pulse						
381	aerosol forcing on marine stratocumulus clouds in the context of marine cloud						
382	brightening. Journal of the Atmospheric Sciences, $80(6)$ , 1585–1604. doi:						
383	https://doi.org/10.1175/JAS-D-22-0207.1						
384	Qiu, S., Zheng, X., Painemal, D., Terai, C., & Zhou, X. (2023). Diurnal variation						
385	of aerosol indirect effect for warm marine boundary layer clouds in the eastern						
386	north atlantic. EGUsphere, 2023, 1–30. doi: 10.5194/egusphere-2023-1676						
387	Sandu, I., Brenguier, J. L., Geoffroy, O., Thouron, O., & Masson, V. (2008).						
388	Aerosol impacts on the diurnal cycle of marine stratocumulus. Journal of						
389	the Atmospheric Sciences, $65(8)$ , 2705–2718. doi: https://doi.org/10.1175/						
390	2008JAS2451.1						

<sup>391</sup> Seinfeld, J. H., Bretherton, C., Carslaw, K. S., Coe, H., DeMott, P. J., Dunlea,

392	E. J., Wood, R. (2016). Improving our fundamental understand-
393	ing of the role of aerosol-cloud interactions in the climate system. Pro-
394	ceedings of the National Academy of Sciences, $113(21)$ , $5781-5790$ . doi:
395	10.1073/pnas.1514043113
396	Smalley, K. M., & Lebsock, M. (2023b). Corrections for geostationary cloud liquid
397	water path using microwave imagery $(1.0.0)$ [data set]. Zenodo. (Last accessed:
398	November 7, 2023) doi: $10.5281/zenodo.7647786$
399	Smalley, K. M., & Lebsock, M. D. (2023a). Corrections for geostationary cloud liq-
400	uid water path using microwave imagery. Journal of Atmospheric and Oceanic
401	Technology, 40(9), 1049-1061. doi: https://doi.org/10.1175/JTECH-D-23-0030
402	.1
403	Smalley, K. M., Lebsock, M. D., Eastman, R., Smalley, M., & Witte, M. K.
404	(2022). A lagrangian analysis of pockets of open cells over the southeast-
405	ern pacific. Atmospheric Chemistry and Physics, 22(12), 8197-8219. doi:
406	10.5194/acp-22-8197-2022
407	Toll, V., Christensen, M., Quaas, J., & Bellouin, N. (2019). Weak average liquid-
408	cloud-water response to anthropogenic aerosols. Nature, $572(7767)$ , $51-55$ . doi:
409	10.1038/s41586-019-1423-9
410	Twomey, S. (1977). The influence of pollution on the shortwave albedo of
411	clouds. Journal of Atmospheric Sciences, 34(7), 1149–1152. doi: 10.1175/
412	1520-0469(1977)034(1149:TIOPOT)2.0.CO;2
413	Walther, A., & Straka, W. (2020). Algorithm theoretical basis document for daytime
414	cloud optical and microphysical properties $(dcomp)$ (NOAA Tech. Rep. No. 64
415	pp). NOAA. Retrieved from https://www.star.nesdis.noaa.gov/goesr/
416	documents/ATBDs/Enterprise/ATBD_Enterprise_Daytime_Cloud_Optical
417	_and_Microphysical_Properties(DCOMP)_v1.2_2020-10-09.pdf
418	Wentz, F. J. (1997). A well-calibrated ocean algorithm for special sensor mi-
419	crowave/imager. Journal of Geophysical Research: Oceans, 102(C4), 8703-
420	8718. doi: https://doi.org/10.1029/96JC01751
421	Wentz, F. J., & Spencer, R. W. (1998). Ssm/i rain retrievals within a unified all-
422	weather ocean algorithm. Journal of the Atmospheric Sciences, $55(9)$ , 1613-
423	1627. doi: https://doi.org/10.1175/1520-0469(1998)055 $\langle 1613:SIRRWA \rangle 2.0.CO;$
424	2

425	Wood, R. (2021). Assessing the potential efficacy of marine cloud brightening for
426	cooling earth using a simple heuristic model. Atmospheric Chemistry and
427	Physics, 21(19), 14507–14533. doi: 10.5194/acp-21-14507-2021
428	Wood, R., & Bretherton, C. S. (2006). On the relationship between stratiform low
429	cloud cover and lower-tropospheric stability. Journal of Climate, $19(24)$ , $6425$
430	- 6432. Retrieved from https://journals.ametsoc.org/view/journals/
431	clim/19/24/jcli3988.1.xml doi: https://doi.org/10.1175/JCLI3988.1
432	Zhou, X., & Feingold, G. (2023). Impacts of mesoscale cloud organization on
433	aerosol-induced cloud water adjustment and cloud brightness. Geophysical
434	Research Letters, $50(13)$ , e2023GL103417. doi: 10.1029/2023GL103417



Geophysical Research Letters

Supporting Information for

# Diurnal Patterns in the Observed Cloud Liquid Water Path Response to Droplet Number Perturbations

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Tables S1 to S2 Figures S1 to S4

# Additional Supporting Information (Files uploaded separately)

Captions for Table S2 Caption: The  $\lambda$  values calculated in prior observational and LES studies shown in Figure 4. They are colored by if they represent non-precipitating, precipitating, or indiscriminate situations (*Table\_S2\_lwp\_adj\_GRL\_manuscript.xlsx*).

# Introduction

Figure S1 shows how cloud-droplet number concentration retrieved using the Advanced-Baseline Imager (ABI) onboard GOES-16 compares to MODIS and CALIPSO. Each data product is discussed in detail and this figure is explained in Section 2.3 of the main text.

Figure S2 shows the lookup table we used to statistically determine the probability of precipitation and precipitation rate along all trajectories. This was done by colocating AMSR-2 warm precipitation rates (described in section 2.5 of the main text) with ABI, and then binning precipitation occurrence and precipitation rate (including non-raining pixels i.e. 0 mm Day<sup>-1</sup>) by corrected ABI LWP and uncorrected ABI N<sub>d</sub>. We then map each LWP and N<sub>d</sub> value back to this lookup table to determine statistically the precipitation probability and rate. In general, we find that the likelihood of precipitation and precipitation rates increase mostly as a function of increasing LWP, with rain likelihood approaching 100% at LWP > 100 g m<sup>-2</sup> and maximum rain rates occurring at LWP > 200 g m<sup>-2</sup>.

Figure S3 demonstrates how we calculate  $\frac{dln(CLWP)}{dln(N_d)}$  ( $\lambda$ ). Specifically, we bin all trajectories by their starting LWP, and fit a line to all (ln(LWP[at each time]) - ln(LWP[time = 0])) and ln(N<sub>d</sub>[time = 0]) values within that bin. We then consider the slope of each fitted line, at each time as  $\lambda$ . This is described in detail in Section 2.6 in the main text.

Figure S4 shows each individual curve shown in Figure 1 of the main text. Note, the red points represent linearly interpolated  $\lambda$  values at night. This was necessary, because, although we are analyzing three years of data, there is not enough microwave LWP data to fill in all times at night. Therefore, to calculate the autocorrelation function for each sensitivity curve shown in Figure 2, we needed to fill in the gaps at night along each curve.

Table S1 shows the microwave imagers that are colocated with all trajectories analyzed to determine how cloud liquid water path (LWP) changes at night. This data and how we process it are described in Section 2.2 of the main text.

Table S2 shows how  $\lambda$  vary among prior literature. Each study is initially separated by whether they are conducted using observations or large-eddy simulations (LES), what each  $\lambda$  value (second column) represents is detailed in column 3, and if each  $\lambda$  value is representative of precipitating or non-precipitating conditions are colored in red and black respectively. Note, any  $\lambda$  colored blue represents situations where we could not confidently determine if it represents non-precipitating or precipitating conditions. In the main text, the observed values are binned in the Obs. column of Figure, and the LES values are binned in the LES column of Figure 4.



**Figure S1.** Collocated Aqua MODIS and Calipso cloud-top  $N_d$  are shown in (a), and collocated GOES-16 ABI and CALIPSO  $N_d$  are shown in (b).



**Figure S2.** Colocated GOES-16 and AMSR-2 mean precipitation rate and probability of precipitation within a  $1^{\circ} \times 1^{\circ}$  gridbox binned by corrected ABI LWP and uncorrected ABI N<sub>d</sub>. Mean precipitation rates are unconditional (i.e. include non-raining regions).



**Figure S3:** The difference between ln(LWP[time = 4 hrs]) - ln(LWP[time = 0 hrs]) for all trajectories conditioned by starting LWP are plotted against  $ln(N_d[time = 0 hrs])$ . s represents the slope of a fitted line which represents the  $\lambda$  we calculate and show at each time in Figure 1.



**Figure S4:** Each panel shows each individual curve in Figure 1, with the red points representing interpolated values. The white-filled regions represent day, and the grey-filled regions represent night.

Satellite	Orbit	Crossing Time (Local)	Duration
GMI	Non-Sun Synchronous	NA	2014-Prezent
AMSR-2	Sun Synchronous	13:33	2012-Present
SSMIS-F16	Sun Synchronous	16:27	2003-Present
SSMIS-F17	Sun Synchronous	18:35	2006-Present
SSMIS-F18	Sun Synchronous	16:30	2010-Present
WindSat	Sun Synchronous	18:10	2003-2020

Table S1.	This table	contains	the six	different	microwav	e imagers (	used.

**Table S2:** The  $\lambda$  values calculated in prior observational and LES studies shown in Figure 4. They are colored by if they represent non-precipitating (black), precipitating (red), or indiscriminate (blue) situations.