Manuscript prepared for Atmos. Chem. Phys. with version 3.2 of the LATEX class copernicus.cls. Date: 20 August 2014

On the relationship between responses in cloud water and precipitation to changes in aerosol

Z. J. Lebo^{1,2} and G. Feingold²

¹Cooperative Institute for Research in Environmental Sciences, University of Colorado Boulder, Boulder, Colorado, USA ²Chemical Sciences Division, NOAA Earth System Research Laboratory, Boulder, Colorado, USA *Correspondence to:* Z. J. Lebo (zach.lebo@noaa.gov)

Abstract. Climate models continue to exhibit strong sensitivity to the representation of aerosol effects on cloud reflectance and cloud amount. This paper evaluates a proposed method to constrain modeled cloud liquid water path (LWP) adjustments in response to changes in aerosol concentration N_a using observations of precipitation susceptibility. Recent climate modeling has suggested a linear

- 5 relationship between relative LWP responses to relative changes in N_a , i.e., $\lambda = d \ln LWP/d \ln N_a$ and the precipitation frequency susceptibility S_{pop} , which is defined as the relative change in the probability of precipitation for a relative change in N_a . Using large eddy simulations (LES) of marine stratocumulus and trade wind cumulus clouds, we show that these two cloud regimes exhibit qualitatively different relationships between λ and S_{pop} ; in stratocumulus clouds λ increases with
- 10 S_{pop} , while in trade wind cumulus, λ decreases with S_{pop} . The LES-derived relationship for marine stratocumulus is qualitatively similar, but quantitatively different than that derived from climate model simulations of oceanic clouds aggregated over much larger spatial scales. We explore possible reasons for variability in these relationships, including the selected precipitation threshold and the various definitions of precipitation susceptibility that are currently in use. Because aerosol-cloud-
- 15 precipitation interactions are inherently small-scale processes, we recommend that when deriving the relationship between λ and S_{pop} , careful attention be given to the cloud regime, the scale, and the extent of aggregation of the model output or the observed data.

1 Introduction

Like its predecessors, the IPCC Fifth Assessment Report (AR5; IPCC 2013) continues to point to 20 aerosol effects on clouds as a major source of uncertainty in our predictive climate modeling capability. Recognizing that cloud systems constantly adjust to aerosol perturbations, AR5 chose to combine both cloud albedo and LWP responses to aerosol changes into one term, i.e., the effective radiative forcing associated with aerosol-cloud interactions (ERFaci). The representation of the underlying microphysical processes associated with cloud formation and albedo and precipitation

- 25 modification must be improved to better quantify ERFaci. Attempts to constrain ERFaci with observations is an important part of this quantification. Early efforts (e.g., Quaas et al., 2006, 2009) used satellite-based measurements of drop concentration (or size) responses to changes in aerosol (Bréon et al., 2002) to constrain the albedo effect (Twomey, 1977). More detailed analysis using surface-based remote sensing and proxy data from cloud resolving models pointed to the scale dependence
- 30 of these relationships (McComiskey and Feingold, 2008, 2012) and called for a clear distinction between the cloud process scale and the satellite data aggregation scale before such observational constraints are applied.

In this paper, we shift attention to observational constraints on aerosol effects on cloud amount, or the "lifetime effect" (Albrecht, 1989), via precipitation modifications. The most direct approach

- 35 would be to quantify λ (= dlnLWP/dlnN_a, or similar); however, λ is almost impossible to measure because of the rapid adjustments resulting from both aerosol and meteorological drivers. A somewhat related quantity, precipitation susceptibility, i.e., $S_{\circ} = -d \ln R/d \ln N_d$, where R is the precipitation rate and N_d is the droplet number concentration (Feingold and Siebert, 2009; Sorooshian et al., 2009) has been introduced as a means of quantifying the influence of aerosol changes on the ambient
- 40 rain rate. Because of the high spatial variability in R, other definitions of precipitation susceptibility, such as the susceptibility of the probability of precipitation (POP) to changes in aerosol (S_{pop}), have been proposed: $S_{pop} = -d\ln \text{POP}/d\ln N_a$ (e.g., Wang et al., 2012; Terai et al., 2012). Several studies have attempted to quantify S_{pop} or S_{\circ} using satellite remote sensing (e.g., Sorooshian et al., 2009; L'Ecuyer et al., 2009), surface remote sensing (Mann et al., 2014), and in-situ aircraft (Terai
- 45 et al., 2012) observations. The values vary considerably depending on several factors, including the definition of precipitation susceptibility, averaging scale (Duong et al., 2011), phase of the cloud lifecycle (Duong et al., 2011; Feingold et al., 2013), and aerosol loading (Feingold et al., 2013). There is disagreement in the literature not only on the values of S_{pop} and S_{\circ} but also on how they depend on important controlling parameters, such as cloud depth and LWP. While quantifying the
- 50 precipitation susceptibility is not the focus of this paper, we refer to two values as guidance. The first, $S_{pop} = 0.12$ (Wang et al., 2012), was derived from satellite remote sensing data over global oceans (based on a reflectivity threshold of 0 dBZ). The second, $S_o \approx 1$ (Mann et al., 2014), was calculated from surface-based remote sensing observations in the northeastern Atlantic Ocean and continental Europe with a spatial scale of approximately 12 km (using 20-min-averaged LWP data
- and assuming a nominal wind speed of 10 m s^{-1}).

Wang et al. (2012) proposed using measurements of S_{pop} as a means of constraining LWP responses to aerosol changes in a climate model. The authors used a series of climate model simula-

tions with the NCAR Community Atmosphere Model version 5 (CAM5) and the ECHAM5-HAM2 to derive a linear relationship between λ and S_{pop} with an intercept at approximately (0,0). In-

- 60 terestingly, the model output from the Multiscale Modeling Framework (MMF) version of CAM5, which resolves clouds and precipitation more reliably than the standard CAM5 simulations, also conforms to this linear relationship. The authors proposed a method for constraining λ that proceeds as follows. The output from a series of GCM simulations is used to define $\lambda = f(S_{pop})$; then, a measurement of S_{pop} combined with the model-derived $f(S_{pop})$ yields an observational constraint
- 65 on λ . Wang et al. (2012) showed that because $f(S_{pop})$ has an intercept close to (0,0) and the measured S_{pop} is small, it follows that λ , which is the cloud LWP adjustment portion of ERFaci, is also small. However, the authors noted that more work must be performed to test these relationships in higher resolution models. The current work directly addresses this point. Specifically, this study addresses the generality of the λ - S_{pop} relationship. The relationship is examined at the cloud
- 70 scale through analysis of previously published work and more rigorously via an analysis of large eddy simulation (LES) of warm (liquid phase only) cloud systems. Observations of S_{pop} and S_{\circ} are then used to provide LES constraints on λ ; the implications for albedo susceptibility (Platnick and Twomey, 1994) are also explored.

The remainder of the paper is organized as follows. Section 2 introduces the methods used to
evaluate λ based on both extant literature and LES. The primary results are presented and discussed in Section 3. Finally, the important conclusions of this work are enumerated in Section 4.

2 Methods

2.1 Analysis of Extant Literature

If there exists a robust relationship between λ and S_{pop} (or S_o), one might expect this to emerge in the extant literature. Therefore, we surveyed published results from a wide range of studies that simulated cases based on various field campaigns. The details of these studies are listed in Table 1. In building this table (and the accompanying Fig. 1), we were faced with a lack of information regarding the rain fraction (or POP) in previously published studies. Therefore, the results are presented in terms of S_o. The potential effect of this substitution is discussed later.

85 2.2 LES Simulations

Two different cloud regimes are explored: (i) stratocumulus, based on the Second Dynamics and Chemistry of Marine Stratocumulus (DYCOMS-II) Research Flight 2 (RF02) and (ii) trade wind cumulus, based on the Rain in Cumulus over the Ocean (RICO) field experiment. The two different warm cloud regimes provide the opportunity to explore the robustness of both the $\lambda - S_{pop}$ and λ –

90 S_{\circ} relationships for different cloud regimes.

Stratocumulus clouds: DYCOMS-II, RF02 2.2.1

A suite of 25 simulations is performed using the Weather Research and Forecasting (WRF) model to explicitly examine the relationships between λ and S_{pop} (or S_{\circ}). For the purposes of this study, WRF is coupled with a two-moment, bin-emulating microphysical model that has been widely used

- 95 to examine aerosol-cloud interactions (Feingold et al., 1998; Wang and Feingold, 2009a). The simulations comprise 5 different initial aerosol number mixing ratios (i.e., $N_a = 25, 50, 75, 100,$ and 125 mg⁻¹). Note that because simulations often use different initialization procedures, N_a is used interchangeably in this paper to denote both the aerosol number concentration (units of cm^{-3}) and mixing ratio (units of mg^{-1}). Given that the air density is approximately 1 kg m^{-3} for the considered domains, $1 \text{ mg}^{-1} \approx 1 \text{ cm}^{-3}$.
- 100

While the aerosol concentration is a prognostic variable in these simulations, the shape of the distribution is invariant with time and assumed to be lognormal with a median radius of 0.2 μ m and a geometric standard deviation of 1.5. The aerosol is assumed to be composed of ammonium sulfate. The supersaturation is calculated and prognostically treated in the model; droplets are formed on

- 105 the aerosol particles with radii above the critical supersaturation required for activation following Köhler theory. The activated aerosol particles are removed from the aerosol population. Particles are regenerated upon evaporation of droplets assuming that one drop regenerates one aerosol particle (Mitra et al., 1992). Thus, collision-coalescence and surface rain provide an avenue for reduction in the aerosol concentration.
- For each N_a , a control simulation is performed based on DYCOMS-II RF02, which readily pro-110 duced drizzle (Stevens et al., 2003). The WRF-LES setup described by Yamaguchi and Feingold (2012) is used. Four additional simulations are performed to explore the sensitivity to environmental conditions and microphysical process rates, i.e., increased surface latent heat flux (140 W m⁻², Hi-LHF), decrease surface latent heat flux (46.5 W m⁻², Lo-LHF), increased collision-coalescence rate
- (110% of the predicted rate, Hi-CC), and decreased collision-coalescence rate (80% of the predicted 115 rate, Lo-CC).

All simulations are performed with a horizontal grid spacing of 50 m and a vertical grid spacing of 12 m. The domain is 6.4 km by 6.4 km in the horizontal and 1.5 km in the vertical direction. A time step of 0.2 s is used to ensure numerical stability and convergence (see Yamaguchi and Feingold,

- 2012). The total simulation time is 6 h; however, the initial 1 h of all simulations is discarded to 120 allow sufficient time for turbulence to develop. The rapid radiative transfer model (RRTM) is used to calculate the longwave radiative fluxes. The simulations are assumed to be nocturnal, i.e., shortwave radiative fluxes are not included. The necessary model information is recorded at 1-min intervals, yielding nearly 5 million x, y pairs for each simulation. Although the decorrelation time for cloud
- fields has been shown to be much longer than 1 min (e.g., \approx 15 min according to McComiskey et al., 125 2009), the 1-min resolution is necessary to capture the rare, high precipitation rate events.

2.2.2 Trade Wind Cumulus: RICO

The RICO simulations used in this study are adopted from Jiang et al. (2010). These simulations were performed using the Regional Atmospheric Modeling System (RAMS) version 6.0 with a bin

- 130 (size-resolving) microphysics scheme (Feingold et al., 1996; Stevens et al., 1996). The aerosol treatment in these simulations is very similar to that of the stratocumulus simulations (see Section 2.2.1). The domain size is 25.6 km × 25.6 km × 6 km with a horizontal grid spacing of 100 m and vertical grid spacing of 40 m. The Global Energy and Water Cycle Experiment Cloud System (GCSS) boundary layer working group initial sounding is modified to initiate heavier rainfall by
- increasing the ambient water vapor mixing ratio and decreasing the potential temperature above 1 km. The model top is also extended in Jiang et al. (2010) to 6 km to allow for deeper convection. The simulations are performed for 8 h with 5 different aerosol number concentrations, namely, 100, 200, 300, 400, and 500 cm⁻³. As in the case of the stratocumulus simulations, model output at 1-min intervals is used. For additional information on these simulations, the reader is referred to
- 140 Jiang et al. (2010).

2.3 λ Calculation

The LWP is first calculated for every column and for every output time by including only cloud water – consistent with Wang et al. (2012). Here, λ is approximated as follows:

$$\lambda = \frac{d \ln L WP}{d \ln N_a} \approx \frac{\Delta \ln L WP}{\Delta \ln N_a} = \left\langle \frac{\ln L WP^{(2)} - \ln L WP^{(1)}}{\ln N_a^{(2)} - \ln N_a^{(1)}} \right\rangle,\tag{1}$$

145 where the overbars represent spatial (horizontal) means and the brackets represent temporal means. The superscripts correspond to low (1) and high (2) aerosol loading scenarios. For reference, all variables are also defined in Table 2. The results are found to be qualitatively (and nearly quantitatively) insensitive to the order in which the calculations were performed, i.e., taking the temporal average of the relative differences (as in Eq. 1) or taking the relative difference of the temporal averages.

150 2.4 S_{pop} Calculation

To calculate S_{pop} , we first determine if it is raining at the surface in a given grid cell and assign the grid cell POP = 1 if it is raining and POP = 0 otherwise – namely, the precipitation probability POP(t) as a function of time t is conditional on a threshold rain rate:

$$\operatorname{POP}_{i,j}^{(k)}(t) = \begin{cases} 1 \text{ if } R_{i,j}^{(k)}(t) \ge T_h \\ 0 \text{ if } R_{i,j}^{(k)}(t) < T_h \end{cases}$$
(2)

155 where T_h represents a predefined threshold in mm day⁻¹ and the superscript k corresponds to the specific simulation. The surface rain rate is used for the calculations herein. Then, S_{pop} is calculated

similar to λ , i.e.,

$$S_{pop} = -\frac{d\ln \text{POP}}{d\ln N_a} \approx -\frac{\Delta \ln \text{POP}}{\Delta \ln N_a} = -\left\langle \frac{\ln \overline{POP^{(2)}} - \ln \overline{POP^{(1)}}}{\ln N_a^{(2)} - \ln N_a^{(1)}} \right\rangle.$$
(3)

For calculating POP, 10 thresholds are applied to R, ranging from 10^{-6} to 20 mm day⁻¹. Only a representative subset of these calculations is presented.

2.5 S_{\circ} Calculation

Here, S_{\circ} is computed by conditionally averaging the rain rate over the aforementioned rain rate thresholds. However, in keeping with Feingold and Siebert (2009), the denominator is $d \ln N_d$ instead of $d \ln N_a$; therefore, we have

$$165 \quad S_{\circ} = -\frac{d\ln R}{d\ln N_d} \approx -\frac{\Delta \ln R}{\Delta \ln N_d} = -\left\langle \frac{\ln \overline{R^{(2)}} - \ln \overline{R^{(1)}}}{\ln \overline{N_d^{(2)}} - \ln \overline{N_d^{(1)}}} \right\rangle.$$
(4)

2.6 $S_{\circ,mod}$ and $S_{pop,mod}$ Calculations

Two additional parameters are also computed, i.e., $S_{\circ,mod}$ and $S_{pop,mod}$; $S_{\circ,mod}$ is the same as in Eq. 4 except that N_a replaces N_d in the denominator. Similarly, $S_{pop,mod}$ replaces N_a with N_d in the denominator of Eq. 3. These modified parameters are useful for analyzing the sensitivity of

170 the results to the use of N_a or N_d , in which the latter evolves with time and the former is used to represent the response in the system to an initial change in aerosol loading (similar to the approach used in global climate simulations). The simulations also help to examine the robustness of the results to multiple representations of precipitation susceptibility.

2.7 A_f Calculations

175 While values of λ that are constrained by $f(S_{pop})$ and/or $f(S_{o,mod})$ are far from certain, the estimates discussed below for the different cloud regimes can be used to estimate the potential effects of changes in aerosol loading on albedo susceptibility A'_{\circ} . We begin with the definition of A'_{\circ} from, e.g., Feingold and Siebert (2009):

$$A'_{\circ} = A_{\circ} \left[1 + \frac{5}{2} \frac{d \ln \text{LWP}}{d \ln N_d} + \dots \right],\tag{5}$$

180 where A_{\circ} represents the albedo susceptibility under constant LWP conditions, i.e.,

$$A_{\circ} = \frac{\partial \ln A}{\partial \ln N_d} \bigg|_{LWP} = \frac{1-A}{3}.$$
(6)

The ellipsis on the right hand side of Eq. 5 represents additional terms that have been excluded in this study. These terms include such effects as changes in the breadth of the drop size distribution (Feingold et al., 1997). However, Eq. 5 is provided in terms of incremental changes in N_d , whereas,

185 the LWP susceptibility, i.e., λ , is defined relative to incremental changes in N_a . Therefore, we make use of a power law relationship between N_d and N_a :

$$N_d \propto N_a^c, \tag{7}$$

where c is theoretically ≤ 1 . Previous studies have provided a broad range of values for c. For example, Shao and Liu (2009) suggested a range of 0.25 to 0.85 based on direct measurements of

both polluted and clean clouds. Other studies have shown that c is likely on the higher end of this range in relatively clean conditions, i.e., $N_a < 500 \text{ cm}^{-3}$ (e.g. Conant et al., 2004; Twohy et al., 2005). Without being prescriptive, we choose a characteristic value of 3/4 for c in this study. As a result, the relationship presented in Eq. 7 can be rewritten as

$$\frac{d\ln N_d}{d\ln N_a} = c = \frac{3}{4}.$$
(8)

195 Then, by rewriting Eq. 5 as

$$A'_{\circ} = A_{\circ} \left[1 + \frac{5}{2} \frac{d \ln LWP}{d \ln N_a} \frac{d \ln N_a}{d \ln N_d} + \dots \right],\tag{9}$$

and incorporating Eq. 8, we get

$$A'_{\circ} = A_{\circ} \left[1 + \frac{10}{3} \lambda + \dots \right].$$
(10)

Because we are not necessarily concerned here with the specific values of either A'_{\circ} or A_{\circ} , we define 200 the albedo susceptibility enrichment factor A_f as follows:

$$A_f = \frac{A'_o}{A_o} = \left[1 + \frac{10}{3}\lambda + \dots\right].$$
 (11)

Thus $\lambda = 0.3$ corresponds to a doubling of the albedo susceptibility relative to the value under constant LWP conditions. Note that A_f can be calculated following Eq. 11 without any knowledge of the actual albedo. A further cautionary note: because A_f is an enhancement factor, in practice it

- 205 must be multiplied by the absolute albedo susceptibility A_{\circ} . As the latter approaches zero, A_f has a diminishing *absolute* effect. Values of A_f are shown in the subsequent section alongside those of λ for the two cloud types. Given that shortwave radiation is not treated in the simulations, these results should be regarded as qualitative. Previous studies have provided estimates of both S_{pop} (0.12, Wang et al., 2012) and $S_{\circ,mod}$ (0.66, Mann et al., 2014) using large satellite- and ground-based observa-
- 210 tional datasets, respectively. Mann et al. (2014) reported the precipitation susceptibility in terms of incremental changes in N_a , which corresponds to $S_{\circ,mod}$ in this study. However, precipitation susceptibility has been previously defined in numerous studies relative to incremental changes in N_d (i.e., S_{\circ}). Therefore, using Eq. 8, one finds that $S_{\circ} \approx 1$ based on the findings of Mann et al. (2014).

3 Results

215 3.1 Analysis of Extant Literature

An initial review of the literature revealed several interesting components relevant to understanding the relationship between λ and S_{pop} (or S_{\circ}). First, the lack of detailed information regarding the rain fraction or POP made it impossible to determine accurate values of S_{pop} from previously published modeling results. Therefore, we chose to use S_{\circ} in our analysis of the published literature. However,

even with this assumption, several studies still lacked the necessary details to determine a relationship between λ and S_o due to either the lack of information regarding N_d (needed to calculate S_o) or the lack of information regarding the initial aerosol number concentration (needed to calculate λ). As a result, we show the findings from the published literature (Fig. 1) for λ' as a function of S'_o, where the "prime" denotes that the axes are not necessarily the same for *all* points. Specifically, S'_o
is dln R/dln N_a in Jiang et al. (2010) and λ' is dlnLWP/dln N_d in Berner et al. (2011). For all other

references, $\lambda' = \lambda$ and $S'_{\circ} = S_{\circ}$, as defined in Eqs. 1 and 4, respectively.

Because the model output was unavailable from many of these studies, every effort was made to carefully read off the relevant values of LWP, R and N_a (or a similar aerosol measurement, such as the number concentration of cloud condensation nuclei N_{CCN} or N_d) from the published figures.

- 230 Although a consistent methodology was applied to calculate λ' and S'_{\circ} , we make no claims on the accuracy of these results. The main point is to see whether any trends in λ' vs. S'_{\circ} emerge from different models and for different environmental conditions. Figure 1 shows substantial variability in the $\lambda'-S'_{\circ}$ relationship. Depending upon which subset of points are selected, one can find a negative slope (e.g., green squares Wang and Feingold, 2009a), nearly no slope (e.g., red closed
- 235 circles, Berner et al., 2011), and a positive slope (e.g., blue crosses, Wang and Feingold, 2009a). Interestingly, Wang and Feingold (2009a) suggests either a positive or a negative slope, depending upon how the LWP and R are averaged over the domain (i.e., averaging all of the grid points or conditionally averaging grid points that exceed some predefined threshold). In the context of Fig. 1, a positive slope corresponds to increasing LWP and decreasing R for an increase in N_a . On the
- 240 other hand, a negative slope corresponds to decreasing LWP and decreasing R for an increase in N_a . None of the slopes predicted by the individual high resolution model studies exhibit an intercept near (0,0), and the slopes of these lines tend to be negative or nearly 0. A more in-depth analysis is clearly warranted.

3.2 Stratocumulus LES (DYCOMS-II)

245 3.2.1 Rain Rates

The LES results are presented below in the context of three specific thresholds on R. These thresholds mimic minimum detectable limits for R from current satellite- and ground-based retrievals.

The three values for T_h are 0.001, 0.5, and 5 mm day⁻¹. For perspective, the minimum detectable radar reflectivity for CloudSat is -30 dBZ (e.g., Haynes et al., 2009), while the minimum for the

- Tropical Rainfall Measuring Mission (TRMM) is 17 dBZ. These reflectivities correspond roughly 250 to precipitation rates of 0.5 and 5 mm day⁻¹, respectively. While inherent uncertainties in the Z-R relationships (emanating from, e.g., assumed drop size distributions and attenuation) can contribute to small variations in the lowest detectable rain rates, we use T_h of 0.5 and 5 mm day⁻¹ to represent of CloudSat and TRMM precipitation rate observations, respectively. While very low, the
- $0.001 \text{ mm day}^{-1}$ rain rate threshold is included for a broader perspective and to encompass the 255 range of rain rates presented in Mann et al. (2014).

Before delving into the relative changes in LWP, R, and POP, an analysis of the absolute range of R produced in the simulations is informative. Figure 2 depicts the mean (solid) and median (dashed) rain rates for T_h of 0.001 (gray), 0.5 (blue), and 5 (red) mm day⁻¹ for the DYCOMS-II simulations.

The shaded area encompasses the 10^{th} to the 90^{th} percentiles. Figure 2a shows that the average R 260 is approximately 1-2 mm day⁻¹ for T_h of 0.001 and 0.5 mm day⁻¹ and $N_a = 25 \text{ mg}^{-1}$; the 90th percentile approaches 10 mm day⁻¹. The values decrease as N_a increases (Figs. 2b-d).

In general, there is a small increase in the mean and median R as T_h increases from 0.001 to 0.5 mm day⁻¹. However, the increase is much more substantial for a further increase in T_h to

- 5 mm day⁻¹. At this high threshold, the mean R is close to the 90th percentile for T_h of 0.001 and 265 0.5 mm day^{-1} ; therefore, most of the lightly drizzling grid points are excluded by choosing such a high T_h . The importance of these thresholds on R will be discussed in more detail below with respect to incremental increases in N_a . Figure 2 excludes the model results for $N_a = 125 \text{ mg}^{-1}$ because R was too small for all but the smallest T_h to be confident in the averaged values of POP and R.
- 270

275

3.2.2 $\lambda - S_{pop}$ Relationship and A_f

Figure 4 presents λ vs. S_{pop} for three different rain rate thresholds (i.e., T_h). λ increases with increasing S_{pop} for all T_h ; however, the slope tends to decrease as T_h increases, especially when only examining relatively small changes in N_a (i.e., black and red points). In fact, for $T_h = 0.001$ mm day⁻¹, $S_{pop} \simeq 0$ for a change in N_a from 25 mg⁻¹ to 50 mg⁻¹. In those relatively clean

- conditions, with such a low T_h , nearly all grid points are precipitating; a small absolute change in N_a is not sufficient to decrease R to the point that R becomes less than T_h for a substantial subset of the domain; hence, little if any change is found in POP in response to increases in N_a . This finding suggests that for low T_h , POP may be largely insensitive to changes in N_a in relatively
- clean environments, especially for these stratocumulus simulations. However, for higher T_h , even in 280 relatively clean conditions, a doubling of N_a produces an increase in S_{pop} (Fig. 4c) because in these conditions, even a change in N_a from 25 mg⁻¹ to 50 mg⁻¹ is sufficient to reduce R such that R becomes less than $T_h = 5 \text{ mm day}^{-1}$ for a substantial subset of the domain.

As mentioned above, $T_h = 0.5 \text{ mm day}^{-1}$ corresponds roughly to the threshold that is commonly

- 285 used to determine precipitating locations in the CloudSat dataset. This threshold tends to suppress the LWP response to changes in N_a (i.e., λ), i.e., the intercept approaches (0,0) as $T_h \longrightarrow 5 \text{ mm}$ day⁻¹ for these stratocumulus clouds. Physically, an intercept of ≈ 0 seems unlikely. Hypothetically, if an increase in N_a results in no change in POP ($S_{pop} = 0$), the LWP should increase as the cloud droplets become smaller and more numerous, and rain formation becomes less efficient. Therefore,
- in readily precipitating clouds, one would expect that the LWP should increase in response to in-290 creasing N_a ($\lambda > 0$), as suggested in Figs. 4a and b. Both observational studies (Christensen and Stephens, 2011) and LES (e.g., Wang et al., 2003; Ackerman et al., 2004; Xue et al., 2008) have confirmed $\lambda > 0$ for readily precipitating clouds. The high-resolution LES results for stratocumulus clouds presented herein suggest that for an observed value of $S_{pop} = 0.12$ (the average, global ocean 295 value), λ varies between nearly 0 for high T_h to approximately 0.5 for lower T_h .

Figure 4a suggests that for marine stratocumulus, λ likely does not increase indefinitely as S_{pop} increases. Instead, an asymptotic behavior is suggested whereby any further increase in S_{pop} produces a smaller or nearly no change in λ . It is at this point that the change in N_a is sufficiently large to permit aerosol-induced evaporation-entrainment or sedimentation-entrainment effects to play a role.

- 300 In other words, a further suppression in POP does not lead to an additional increase in LWP because the much smaller droplets evaporate more readily (e.g., Wang et al., 2003; Ackerman et al., 2004; Xue and Feingold, 2006) or because weaker sedimentation enhances both evaporation and cloud-top cooling, both of which increase entrainment (Bretherton et al., 2007). This asymptotic behavior is challenging to discern for higher T_h due to an insufficient number of points for which R exceeds
- 305 T_h in the presence of higher aerosol loadings. However, the inability of λ to increase indefinitely as POP is further reduced should be expected given previously published findings. For example, Ackerman et al. (2004) demonstrated that the LWP first increases with increasing N_a ($\lambda > 0$); however, further increases in N_a result in $\lambda = 0$, and for a strong enough aerosol perturbation, λ becomes negative. Under these high aerosol conditions, clouds are likely not precipitating and λ is dominated
- 310 by processes other than collision-coalescence.

Figure 4 also provides a useful estimate of A_f for marine stratocumulus by applying Eq. 11 to the simulated values of λ . The right axes of the plots in Fig. 4 demonstrate the range of possible A_f . For a value of S_{pop} of 0.12, or by simply choosing the results for small changes in N_a , the DYCOMS-II RF02 simulations suggest that A_f is between 1.8 and 2.5, i.e., the albedo susceptibly may be 80% to 150% higher than expected under constant LWP conditions.

315

3.2.3 $\lambda - S_{pop,mod}$ Relationship

Figure 5 shows the relationship between λ and $S_{pop,mod}$, in which the denominators of the x and y axes are no longer the same. For low T_h , changing the denominator has little to no effect on the relationship between relative changes in LWP and POP (Fig. 5a). However, for higher T_h , i.e., val-

- 320 ues that better reflect the higher detection limits of satellite retrievals, the inconsistent denominator causes the relationship to become less linear and more scattered, especially for $T_h = 5 \text{ mm day}^{-1}$. The reason for this discrepancy is related to the fact that the relative changes in LWP and POP due to changes in N_a reflect a response due to the prescribed aerosol perturbation, i.e., the changes are relative to only the initial aerosol loading, whereas relative changes in LWP and POP due to changes in
- 325 N_d reflect the effects of numerous microphysical processes (e.g., activation, collision-coalescence, and scavenging). Because N_d is not constant in time, the relative change in N_d tends to vary as a function of time. This transient nature produces the scatter in Figs. 5b and c.

3.2.4 $\lambda - S_{\circ,mod}$ Relationship and A_f

- As discussed above, S_o is typically represented in terms of relative changes in N_d. However, the
 previous subsection demonstrated how inconsistencies in the denominator can cause the relationship between λ and S_{pop} to lose its coherency. Therefore, we show the relationship between λ and S_{o,mod}, i.e., where the denominators in the x and y axes are both a function of the relative change in N_a (Fig. 6). As mentioned in Section 3.2.2, small changes in N_a exhibit little to no effect on POP when a low threshold on R is applied to determine raining and non-raining locations. However,
- 335 the same does not hold true for R, i.e., even at low thresholds, R still changes due to increases in aerosol loading, even for small absolute changes. Therefore, the stratocumulus clouds continue to precipitate throughout most of domain for imposed increases in N_a , yet the average R is slightly reduced. This effect is demonstrated in Fig. 6a, where we see that $S_{o,mod}$ is greater than 0 (unlike the case for S_{pop} , Fig. 4a).
- 340 A comparison between Figs. 4 and 6 suggests that the relationships are qualitatively the same (i.e., λ tends to increase as either S_{pop} or $S_{\circ,mod}$ increases), although the slopes can be quite different. The difference in slopes is related to the aforementioned point that changes in N_a act differently on R and POP. In the case of $S_{\circ,mod}$, small changes in N_a do little to affect the average R in the heavily drizzling regions, i.e., the high threshold is inclusive enough to maintain a relatively constant
- 345 average R for all aerosol perturbations. On the other hand, for low T_h , nearly the entire domain is considered to be drizzling and a small change in N_a reduces R. However, because this reduction is not sufficient to convert many drizzling locations into non-drizzling points, S_{\circ} increases (Fig. 6a) while S_{pop} (Fig. 4a) remains nearly constant for small changes in N_a .

Using the $S_{\circ,mod} = 0.66$ observational constraint from Mann et al. (2014) (recall that $S_{\circ} \approx 1$ for realistic values of c) for this scenario, one arrives at values of λ ranging from 0.4 to 1.0 for $T_h =$ 0.001 mm day⁻¹ and $T_h = 0.5$ mm day⁻¹, respectively. For $T_h = 5$ mm day⁻¹, Fig. 6c suggests that λ would be substantially larger; however, the simulations do not extend to large enough N_a to quantify this effect. The right hand axes of Fig. 6 provide equivalent estimates of A_f derived from Eq. 11 and point to enhancements in the albedo susceptibility of 2.5 (4) for $T_h = 0.001$ mm day⁻¹

355 $(0.5 \text{ mm day}^{-1}).$

3.3 Trade Wind Cumulus: RICO LES

3.3.1 Rain Rates

Figure 3a shows that the average R for N_a = 100 cm⁻³ is approximately 10-20 mm day⁻¹ for T_h of 0.001 and 0.5 mm day⁻¹ in the simulated trade wind clouds. The domain average is naturally
much less than this; however, the threshold removes very small values of R such that the average R is higher. The average R for all thresholds tends to decrease as N_a increases (Figs. 3b-e); the largest change occurs when N_a increases from 300 to 400 cm⁻³ (Figs. 3c and d). The changes in R for increasing N_a are similar to those shown for the stratocumulus case (Fig. 2) except that R tends to change more rapidly in the trade wind cumulus, especially for higher aerosol loadings.
Moreover, Fig. 3 demonstrates that the clouds precipitate for all aerosol loading scenarios and under all threshold values in the RICO case; therefore, the analysis that follows incorporates all 5 RICO simulations.

3.3.2 $\lambda - S_{pop}, S_{pop,mod}$, and $S_{\circ,mod}$ Relationships and A_f

- The RICO simulations elicit an important finding that was alluded to earlier, namely, λ is not nec-370 essarily positive. Figure 7 demonstrates that λ is negative for changes in N_a that are a factor of 3 or larger. Moreover, Fig. 7a shows that in the case of these shallow trade wind cumulus clouds, λ decreases as S_{pop} increases. This downward trend is related to the balance between aerosol perturbations acting to decrease R and to increase entrainment and evaporation of cloud water. The former acts to increase S_{pop} , while the latter decreases λ . However, the simulations also suggest that λ
- saturates, as suggested earlier in the case of stratocumulus clouds (Fig. 4). For progressively larger changes in N_a , S_{pop} continues to increase while λ remains relatively constant. This asymptotic behavior results from the fact that the changes in droplet size for increases in aerosol loading beyond 400 cm⁻³ are small relative to an increase in N_a from 100 to 200 mg⁻¹ and thus limit additional evaporation-entrainment feedbacks on the cloud system. This is analogous to the findings of Xue and
- 380 Feingold (2006) (Figs. 3 and 5 therein), who showed that several cloud characteristics (e.g., LWP and cloud fraction) asymptote for high aerosol number concentrations. This effect is largely related to the system converging on the saturation adjustment limit, which precludes further decreases in λ . The results of the RICO simulations for small changes in N_a (i.e., from 100 to 200 cm⁻³) suggest that A_f may be similar for both marine stratocumulus and trade wind cumulus (i.e., approximately
- 385 1.7 for the RICO simulations). However, whereas A_f was shown to increase for larger changes in N_a in marine stratocumulus (Fig. 4), A_f decreases in the case of trade wind cumulus for large enough aerosol perturbations. In this case, the LWP response to an aerosol perturbation acts to *decrease* the albedo susceptibility (A_f is less than 1).

The DYCOMS-II stratocumulus simulations demonstrated that the consistency in the denomina-390 tor of the x and y axes is important for increasing the coherency in the $\lambda - S_{pop}$ or $\lambda - S_{\circ,mod}$ relationships. However, in the trade wind cumulus case, this effect is not noticeable (Figures 7a and b are very similar). To explore this further, we consider the relative droplet number concentration $N_d/N_{d,0}$, where $N_{d,0}$ is the drop concentration associated with the lowest aerosol perturbation simulation. For the trade wind cumulus case, an increase in N_a results in an increase in N_d that does not

- 395 produce a noticeable trend in $N_d/N_{d,0}$ over the course of the 8-h simulations (Fig. 8a). This is not so in the case of drizzling stratocumulus clouds, where $N_d/N_{d,0}$ increases as a result of the efficient removal of aerosol from the domain, especially relative to the more polluted cases (i.e., $N_a = 100$ and 125 mg⁻¹; Fig. 8b). The difference is related to the difference in the cloud systems. In the case of trade wind cumulus, only a small fraction of the domain contains condensed cloud water at any 400 given time; therefore, the time required to scavenge a large portion of the ambient aerosol is much
- longer than in the case of stratocumulus clouds where the cloud fraction is often close to 1.

Figures 7a and b suggest that λ decreases more rapidly with increased aerosol loading for lower T_h. For T_h = 0.001 mm day⁻¹, λ decreases from approximately 0.2 to -0.8 for an increase in S_{pop} of only 0.8. However, for T_h = 5 mm day⁻¹, λ decreases from approximately 0.2 to -0.8 for an increase
405 in S_{pop} of 2.5. This has important implications for constraining λ using observations of S_{pop}. For example, if the former trend is true, then small values of S_{pop} result in small values of λ, as suggested by Wang et al. (2012). However, if the latter trend is true, i.e., λ decreases gradually with increasing N_a (and increasing S_{pop}), then a small value of S_{pop} implies that λ is larger. For reference, if S_{pop} is 0.12 (Wang et al., 2012), then λ is approximately 0 for T_h = 0.001 mm day⁻¹ (Fig. 7a, closed

- 410 circles), while λ increases to approximately 0.3 for $T_h = 5 \text{ mm day}^{-1}$ (determined by extrapolating the points to smaller values of S_{pop} in Fig. 7a, diamonds). Alternatively, if $S_{\circ,mod} = 0.66$ (Mann et al., 2014), then Fig. 7c indicates that λ ranges from 0.3 ($T_h = 0.001 \text{ mm day}^{-1}$) down to 0.05 (T_h = 5 mm day⁻¹). The equivalent range of A_f is an enhancement of 20% to 100% relative to constant LWP conditions. However, for even slightly higher $S_{\circ,mod}$ or S_{pop} , λ quickly becomes negative and
- 415 A_f becomes less than 1.

4 Conclusions

relationship at the large eddy scale.

Given the difficulty in observationally constraining the LWP response to an increase in aerosol loading λ , Wang et al. (2012) explored the relationship between λ and the precipitation frequency susceptibility S_{pop} for a set of climate model simulations. A robust relationship between λ and S_{pop} would provide a useful way to constrain λ via S_{pop} observations. The current work examines this



First, a review of the literature shows no clear relationship between λ and S_{\circ} ; however, these results exhibit little quantitive power given the paucity of the model output from the published studies. To explore this relationship in more detail, a set of large eddy simulations of a drizzling stratocumu-

425 lus case is performed, and a previously published set of trade wind cumulus simulations is analyzed.

In each of these sets, the simulations differ only in their initial aerosol concentration, which also allows us to explore albedo susceptibility in an idealized framework for two important shallow cloud regimes.

The following important findings are drawn from this analysis. For brevity, the findings are for-430 mulated with respect to S_{pop} ; however, the conclusions also apply more generally to $S_{o,mod}$.

- The *y*-intercept of the λ − S_{pop} relationship is likely > 0 for both stratocumulus and trade wind cloud systems. This result differs from the climate model-derived *y*-intercept of ≈ 0 in Wang et al. (2012)
- λ does not necessarily increase linearly as a function of S_{pop}. Instead, λ exhibits an asymptotic
 behavior at sufficiently large S_{pop}. In the case of stratocumulus, aerosol-induced evaporationentrainment and/or sedimentation-entrainment effects limit further increases in the LWP. This effect is schematically represented in Fig. 9 (red, stippled). For trade wind cumulus clouds, λ is shown to *decrease* with increasing S_{pop} due to the effects of entrainment and evaporation (Fig. 9; blue, dotted). These different trends in λ are important if one wishes to diagnose λ from observations of S_{pop} or S_{o,mod}, especially for small aerosol perturbations, which are reflected by larger changes in λ and small changes in S_{pop} (Fig. 9; crossed).

3. At the $S_{pop} = 0$ intercept, λ is approximately 0.2-0.3 in both the stratocumulus and trade wind cumulus cases. However, the simulations suggest that λ may increase or decrease with increased aerosol loading (and increasing S_{pop}) depending on the cloud type and dominant microphysical processes.

445

450

455

460

- 4. To gauge the influence of these results on albedo susceptibility, A_f, which is defined as the fractional enhancement in the albedo susceptibility relative to constant LWP conditions, is calculated. For the stratocumulus cloud case, A_f ranges from enhancements of 80% to 150% if S_{pop} = 0.12 (Wang et al., 2012) is used as a constraint, or approximately 150% to 300% if S_{0,mod} = 0.66 (Mann et al., 2014) is the constraint. In the case of the trade wind cumulus clouds, the values of A_f are 20-80% for S_{pop} = 0.12 and 70% for S_{0,mod} = 0.66. However, for slightly higher S_{pop} or S_{0,mod}, the albedo susceptibility may actually decrease relative to constant LWP conditions due to the strong leverage of λ in Eq. 11. These values are approximate given that solar radiation is not explicitly included in the simulations and because the simulations are relatively short and somewhat idealized.
- 5. The importance of using a consistent denominator in the λ and S_{pop} calculations is also demonstrated by calculating S_{pop} (but not λ) in terms of N_d rather than N_a (i.e., $S_{pop,mod}$). The introduced inconsistency is important in the case of stratocumulus clouds in which N_d decreases (quite rapidly in relatively clean conditions) as a function of time. This effect produces an ill-defined relationship between λ and $S_{pop,mod}$.

14

6. The slope and intercept of the $\lambda - S_{pop}$ relationship is largely dependent upon the selected rain rate threshold. This dependency is because determining POP is a binary option, i.e., it is either raining or it is not, which is dependent on some threshold for what is considered "raining".

The current study indicates that the $\lambda - S_{pop}$ relationship is likely related to the resolution of cloud processes, the scales at which the aerosol interacts with clouds, and the type of system being analyzed (i.e., stratocumulus versus trade wind cumulus). Based on our earlier work (McComiskey and Feingold, 2012), we surmise that even if convection *and* aerosol-cloud processes are adequately resolved, the $\lambda - S_{pop}$ relationship will also be dependent on the scale at which the data are aggregated. (The influence of aggregation was also discussed by Wang et al., 2012, .) More specifically,

- 470 the true global λS_{pop} relationship is an aggregation of local relationships in different cloud *and* aerosol regimes. Because measurements of λ are not practical, a productive avenue would be to pursue regime-based measurements of S_{pop} or $S_{\circ,mod}$ combined with large eddy simulations of the type performed here to assess λ at a range of scales. The aggregation of these local relationships would provide a more direct comparison with the global ocean relationship derived by Wang et al.
- 475 (2012). A breakdown of GCM results for different cloud regimes would provide interesting comparison. In conclusion, we caution that these scale, threshold, and aerosol proxy sensitivities be carefully considered before $\lambda - S_{pop}$ relationships are universally applied.

Acknowledgements. The authors thank the Department of Energy's Atmospheric System Research Program and NOAA's Climate Goal for funding. Hongli Jiang is thanked for providing the RICO simulations.

480 References

- Ackerman, A. S., Toon, O. B., and Hobbs, P. V.: A model for particle microphysics, turbulent mixing, and radiative transfer in the stratocumulus-topped marine boundary layer and comparisons with measurements, J. Atmos. Sci., 52, 1204–1236, 1995.
- 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, 2004.
 - Albrecht, B.: Aerosols, cloud microphysics, and fractional cloudiness, Science, 245, 1227–1230, doi:10.1126/ science.245.4923.1227, 1989.
 - Berner, A. H., Bretherton, C. S., and Wood, R.: Large-eddy simulation of mesoscale dynamics and entrainment around a pocket of open cells observed in VOCALS-REx RF06, Atmos. Chem. Phys., 11, 10525–10540,
- doi:10.5194/acp-11-10525-2011, 2011.
 - Bréon, F., Tanré, D., and Generoso, S.: Aerosol Effect on cloud droplet size monitored from satellite, Science, 295, 834–838, doi:10.1126/science.1066434, 2002.
 - Bretherton, C. S., Blossey, P. N., and Uchida, J.: Cloud droplet sedimentation, entrainment efficiency, and subtropical stratocumulus albedo, Geophys. Res. Let., 34, doi:10.1029/2006GL027648, 2007.
- 495 Christensen, M. W. and Stephens, G. L.: Microphysical and macro physical responses of marine stratocumulus polluted by underlying ships: Eevidence of cloud deepening, J. Geophys. Res., 116, doi: 10.1029/2010JD014638, 2011.
 - Conant, W. C., VanReken, T. M., Rissman, T. A., Varutbangkul, V., Jonsson, H. H., Nenes, A., Jimenez, J. L., Delia, A. E., Bahreini, R., Robets, G. C., Flagan, R. C., and Seinfeld, J. H.: Aerosol-cloud drop concentration
- closure in warm cumulus, J. Geophys. Res., 109, doi:10.1029/2003JD004324, 2004.
 Cotton, W. R., Pielke Sr., R. A., Walko, R. L., Liston, G. E., Tremback, C. J., Jiang, H., McAnelly, R. L., Harrington, J. Y., Nicholls, M. E., Carrio, G. G., and McFadden, J. P.: RAMS 2001: Current status and future directions, Meteor. Atmos. Phys., 82, 5–29, doi:10.1007/s00703-001-0584-9, 2003.
- Duong, H. T., Sorooshian, A., and Feingold, G.: Investigating potential biases in observed and modeled
 metrics of aerosol-cloud-precipitation interactions, Atmos. Chem. Phys., 11, 4027–4037, doi:10.5194/ aco-11-4027-2011, 2011.
 - Feingold, G. and Siebert, H.: Cloud-Aerosol Interactions from the micro to cloud scale, in: Clouds in the perturbed climate system: their relationship to energy balance, atmospheric dynamics, and precipitation, edited by Heintzenberg, J. and Charlson, R. J., MIT Press, 2009.
- 510 Feingold, G., Stevens, B., Cotton, W. R., and Frisch, A. S.: The Relationship between Drop Incloud Residence Time and Drizzle Production in Numerically Simulated Stratocumulus Clouds, J. Atmos. Sci., 53, 1108– 1122, 1996.
 - Feingold, G., Boers, R., Stevens, B., and Cotton, W. R.: A modeling study of the effect of drizzle on cloud optical depth and susceptibility, J. Geophys. Res., 102, 13 52713 534, doi:10.1029/97JD00963, 1997.
- 515 Feingold, G., Walko, R. L., Stevens, B., and Cotton, W. R.: Simulations of marine stratocumulus using a new microphysical parameterization scheme, Atmos. Res., 47, 505–528, 1998.
 - Feingold, G., McComiskey, A., Rosenfeld, D., and Sorooshian, A.: On the relationship between cloud contact time and precipitation susceptibility to aerosol, J. Geophys. Res., 118, doi:10.1002/jgrd.50819, 2013.
 - Haynes, J. M., L'Ecuyer, T. S., Stephens, G. L., Miller, S. D., Mitrescu, C., Wood, N. B., and Tanelli, S.: Rainfall

- 520 retrieval over the ocean with spaceborne W-band radar, J. Geophys. Res., 114, doi:10.1029/2008JD009973, 2009.
 - Jiang, H., Feingold, G., and Sorooshian, A.: Effect of aerosol on the susceptibility and efficiency of precipitation in warm trade cumulus clouds, J. Atmos. Sci., 67, 3525–3540, 2010.
- Khairoutdinov, M. F. and Randall, D. A.: Cloud resolving modeling of the ARM summer 1997 IOP: Model
 formulation, results, uncertainties, and sensitivities, J. Atmos. Sci., 60, 607–625, 2003.
 - L'Ecuyer, T. S., Berg, W., Haynes, J., Lebsock, M., and Takemura, T.: Global observations of aerosol impacts on precipitation occurrence in warm maritime clouds, J. Geophys. Res., 114, doi:10.1002/2008JD011273, 2009.
- Lee, S., Feingold, G., and Chuang, P. Y.: Effect of Aerosol on CLoud-Environmental Interactions in Trade cumulus, J. Atmos. Sci., 69, 3607–3632, doi:10.1175/JAS-D-12-026.1, 2012.
- Mann, J. A., Chiu, J. C., Hogan, R. J., O'Connor, E. J., L'Ecuyer, T. S., Stein, T. H. M., and Jefferson, A.: Aerosol impacts on drizzle properties in warm clouds from ARM Mobile Facility maritime and continental deployments, J. Geophys. Res., 119, doi:10.1002/2013JD021339, 2014.
- McComiskey, A. and Feingold, G.: Quantifying error in the radiative forcing of the first aerosol indirect effect, 535 Geophys. Res. Let., 35, doi:10.1029/2007GL032667, 2008.
 - McComiskey, A. and Feingold, G.: The scale problem in quantifying aerosol indirect effects, Atmos. Chem. Phys., 12, 1031–1049, doi:10.5194/acp-12-1031-2012, 2012.
 - McComiskey, A., Feingold, G., Frisch, A. S., Turner, D. D., Miller, M. A., Chiu, J. C., Min, Q., and Ogren, J. A.: An assessment of aerosol-cloud interactions in marine stratus clouds based on surface remote sensing,
- J. Geophys. Res., 114, doi:10.1029/2008JD011006, 2009.
 Mitra, S. K., Brinkmann, J., and Pruppacher, H. T.: A wind tunnel study on the drop-to-particle conversion, J. Aerosol Sci., 23, 245–256, 1992.
 - Morrison, H. and Gettelman, A.: A new two-moment bulk stratiform cloud microphysics scheme in the community atmosphere model, version 3 (CAM3). Part I: Description and numerical tests, J. Climate, 21, 3642– 3659, 2008.
 - Ogura, Y. and Phillips, N.: Scale analysis of deep and shallow convection in the atmosphere, J. Atmos. Sci., 19, 173–179, 1962.

545

- Platnick, S. and Twomey, S.: Determining the susceptibility of cloud albedo to changes in droplet concentration with the advanced very high resolution radiometer, J. Appl. Meteor. Clim., 33, 334–347, 1994.
- 550 Quaas, J., Boucher, O., and Lohmann, U.: Constraining the total aerosol indirect effect in the LMDZ and ECHAM4 GCMs using MODIS satellite data, Atmos. Chem. Phys., 6, 947–955, 2006.
 - Quaas, J., Ming, Y., Menon, S., Takemura, T., M.Wang, Penner, J. E., Gettelman, A., Lohmann, U., Bellouin, N., Boucher, O., Sayer, A. M., Thomas, G. E., McComiskey, A., Feingold, G., Hoose, C., Kristjansson, J. E., Liu, X., Balkanski, Y., Donner, L. J., Ginoux, P. A., Stier, P., Grandey, B., Feichter, J., Sednev, I., Bauer,
- 555 S. E., Koch, D., Grainger, R. G., Kirkevag, A., Iversen, T., Seland, ., Easter, R., Ghan, S. J., Rasch, P. J., Morrison, H., Lamarque, J.-F., Iacono, M. J., Kinne, S., and Schulz, M.: Aerosol indirect effects - general circulation model intercomparison and evaluation with satellite data, Atmos. Chem. Phys., 9, 8697–8717, 2009.
 - Shao, H. and Liu, G.: A critical examination of the observed first aerosol indirect effect, J. Atmos. Sci., 66,

560 1018–1032, 2009.

585

- Skamarock, W. C., Klemp, J. B., Dudhia, J., Gill, D. O., Barker, D. M., Duda, M. G., Huang, X.-Y., Wang, W., and Powers, J. G.: A description of the advanced research WRF Version 3, National Center for Atmospheric Research, Boulder, Colorado, USA, 2008.
- Sorooshian, A., Feingold, G., Lebsock, M. D., Jiang, H., and Stephens, G. L.: On the precipitation susceptibility
 of clouds to aerosol perturbations, Geophys. Res. Let., 36, doi:10.1029/2009GL038993, 2009.
 - Stevens, B., Feingold, G., Cotton, W. R., and Walko, R. L.: Elements of the microphysical structure of numerically simulated nonprecipitating stratocumulus, J. Atmos. Sci., 53, 980–1006, 1996.
 - Stevens, B., Lenschow, D. H., Vali, G., Gerber, H., Bandy, A., Blomquist, B., Brenguier, J., Bretherton, C. S., Burnet, F., Campos, T., Chai, S., Faloona, I., Friesen, D., Haimov, S., Laursen, K., Lilly, D. K., Loehrer,
- 570 S. M., Malinowski, S. P., Morley, B., Petters, M. D., Rogers, D. C., Russel, L., Savic-Jovcic, V., Snider, J. R., Sraub, D., Szumowski, M. J., Takagi, H., Thornton, D. C., Tschudi, M., Twohy, C., Wetzel, M., and van Zanten, M. C.: Dynamics and Chemistry of Marine Stratocumulus–DYCOMS-II, Bull. Amer. Meteor. Soc., 84, 579–593, doi:10.1175/BAMS-84-5-579, 2003.
 - Stevens, B., Moeng, C.-H., Ackerman, A. S., Bretherton, C. S., Chlond, A., De Roode, S., Edwards, J., Go-
- - Stevens, D. E., Ackerman, A. S., and Bretherton, C. S.: Effects of domain size and numerical resolution on the simulation of shallow cumulus convection, J. Atmos. Sci., 59, 3285–3301, 2002.
- 580 Terai, C. R., Wood, R., Leon, D. C., and Zuidema, P.: Does precipitation susceptibility vary with increasing cloud thickness in marine stratocumulus?, Atmos. Chem. Phys., 12, 4567–4583, doi:10.5194/ acp-12-4567-2012, 2012.
 - Twohy, C. H., Petters, M. D., Snider, J. R., Stevens, B., Tahnk, W., Wetzel, M., Russel, L., and Burnet, F.: Evaluation of the aerosol indirect effect in marine stratocumulus clouds: Droplet number, size, liquid water path, and radiative impact, J. Geophys. Res., 110, doi:10.1029/2004JD005116, 2005.
 - Twomey, S.: The Influence of Pollution on the Shortwave Albedo of Clouds, J. Atmos. Sci., 34, 1149–1152, 1977.
 - 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, J. Atmos. Sci., 66, 3237–3256, 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, J. Atmos. Sci., 66, 3257–3275, 2009b.
 - Wang, M., Ghan, S., Liu, X., L'Ecuyer, T. S., Zhang, K., Morrison, H., Ovchinnikov, M., Easter, R., Marchand, R., Chand, D., Qian, Y., and Penner, J. E.: Constraining cloud lifetime effects of aerosols using A-Train
- satellite measurements, Geophys. Res. Let., 39, doi:10.1029/2012GL052204, 2012.
 - Wang, S., Wang, Q., and Feinfold, G.: Turbulence, condensation, and liquid water transport in numerically simulated nonprecipitating stratocumulus clouds, J. Atmos. Sci., 60, 262–278, 2003.
 - Xue, H. and Feingold, G.: Large-eddy simulations of trade wind cumulus: Investigation of aerosol indirect effects, J. Atmos. Sci., 63, 1605–1622, 2006.

- 600 Xue, H., Feingold, G., and Stevens, B.: Aerosol effects on clouds, precipitation, and the organization of shallow cumulus convection, J. Atmos. Sci., 65, 392–406, 2008.
 - Yamaguchi, T. and Feingold, G.: Technical note: Large-eddy simulation of cloudy boundary layer with the Advanced Research WRF model, J. Adv. Model. Earth Syst., 3, doi:10.1029/2012MS000164, 2012.

Campaign	Location	Reference	Cloud Type(s)	Model	Microphysics
ASTEX ^a	Northeastern Atlantic	Ackerman et al. (2004)	Nocturnal drizzling stratocumulus	Stevens et al. (2002), Ogura and	Ackerman et al. (1995)
				Phillips (1962)	
$DYCOMS-II^b$	Coastal Southern California	Ackerman et al. (2004)	Marine stratocumulus	Stevens et al. (2002), Ogura and	Ackerman et al. (1995)
				Phillips (1962)	
FIRE-I ^c	Coastal Southern California	Ackerman et al. (2004)	Cloud field	Stevens et al. (2002), Ogura and	Ackerman et al. (1995)
				Phillips (1962)	
\mathbf{RICO}^d	Antigua and Barbuda (Caribbean)	Lee et al. (2012)	Precipitating shallow cumulus	WRF ¹ (Skamarock et al., 2008)	2-moment bulk (Feingold et al.,
					1998; Wang and Feingold, 2009a)
VOCALS-REx ^e	Southeast Pacific	Berner et al. (2011)	POCs	SAM 2 (Khairoutdinov and Ran-	2-moment bulk (Morrison and Get-
				dall, 2003)	telman, 2008)
$DYCOMS-II^b$	Coastal Southern California	Wang and Feingold (2009a)	Marine stratocumulus	WRF ¹ (Skamarock et al., 2008)	2-moment bulk (Feingold et al.,
					1998)
$DYCOMS-II^b$	Coastal Southern California	Wang and Feingold (2009b)	Marine stratocumulus, ship tracks	WRF ¹ (Skamarock et al., 2008)	2-moment bulk (Feingold et al.,
					1998)
$RICO^d$	Antigua and Barbuda (Caribbean)	Jiang et al. (2010)	Precipitating shallow cumulus	RAMS ³ (Cotton et al., 2003)	bin (Feingold et al., 1996; Stevens
					et al., 1996)
^a Atlantic Stratocumulus	Transition Experiment				

Table 1: Data source description.

 $^{b}\,$ second Dynamics and Chemistry of Marine Stratocumulus (Stevens et al., 2003, 2005)

^c First ISCCP (International Satellite Cloud Climatology Project) Regional Experiment

 d Rain in Cumulus over the Ocean

e VAMOS (Variability of the American Monsoon System) Ocean-Cloud-Atmosphere-Land Study - Regional Experiment

¹ Weather Research and Forecasting

² System for Atmospheric Modeling

³ Regional Atmospheric Modeling System

Table 2: Variable names and definitions.

Variable	Name	Description
R	Rain Rate	
N_a	Aerosol Number Concentration or Mixing Ratio	
N_d	Droplet Number Concentration	
$N_{d,0}$	Droplet Number Concentration for Cleanest Simulati	ion
ρ	Air Density	
z	Height	
q_c	Cloud Water Mixing Ratio	
POP	Probability of Precipitation/Precipitation Frequency	
LWP	Liquid Water Path	$\int_0^\infty q_c ho dz$
S_{pop}	Precipitation Frequency Susceptibility	$\frac{dln POP}{dln N_a}$
S_{\circ}	Precipitation Susceptibility	$\frac{dlnR}{dlnN_d}$
$S_{pop,mod}$	Modified Precipitation Frequency Susceptibility	$\frac{dln POP}{dln N_d}$
$S_{\circ,mod}$	Modified Precipitation Susceptibility	$\frac{dlnR}{dlnN_a}$
λ	LWP Susceptibility	$\frac{dln L \overline{W} P}{dln N_a}$
A_f	Albedo Susceptibility Enrichment Factor	-
$N_d/N_{d,0}$	Relative Droplet Number Concentration	



Fig. 1: Scatterplot of λ' versus S'_{\circ} from previously published studies. The legend provides the reference that corresponds to each symbol. Note here that "prime" notation is used because not all these studies provide enough detail to determine λ and S_{\circ} . Specifically, S'_{\circ} is $d\ln R/d\ln N_a$ in Jiang et al. (2010) and λ' is $d\ln LWP/d\ln N_d$ in Berner et al. (2011). For all other references, $\lambda' = \lambda$ and $S'_{\circ} = S_{\circ}$.



Fig. 2: Mean (solid) and median (dashed) rain rates for the 3 rain rate thresholds, i.e., T_h of 0.001 (gray), 0.5 (blue), and 5 (red) mm day⁻¹ for four different aerosol loadings. The shaded region encompasses the 10th percentile to the 90th percentile. R is depicted as equal to to T_h for the first hour as a reference point for the minimum R that is possible under each T_h condition. The model output is for the DYCOMS-II case.



Fig. 3: As in Fig. 2 except for the RICO case (the model output is from Jiang et al., 2010).



Fig. 4: Scatterplot of λ (and A_f , right axis) versus S_{pop} for thresholds T_h of (a) 0.001, (b) 0.5, and (c) 5 mm day⁻¹. These thresholds are representative of the set of 10 thresholds analyzed. Here, the following colors denote changes in N_a from 25 mg⁻¹ to 50 mg⁻¹ (black), 75 mg⁻¹ (red), and 100 mg⁻¹ (blue) for the DYCOMS-II case. The symbols signify the control (solid circles), Hi-LHF (open circles), Lo-LHF (crosses), Lo-CC (open squares), and Hi-CC (open triangles) simulations. Note that not all symbols appear, especially for larger changes in N_a and high threshold values because for those conditions no points met the criterion for calculating λ and/or S_{pop} . The thin dashed line shows the linear relationship determined by Wang et al. (2012) for the $\lambda - S_{pop}$ relationship and the vertical dashed line corresponds to the satellite-measured value of S_{pop} , i.e., 0.12 (Wang et al., 2012). Note that the previously reported value of S_{pop} is used for reference and may not directly correspond to the rain rate thresholds used in the current $\frac{25}{25}$.



Fig. 5: As in Fig. 4 except for λ versus $S_{pop,mod}$, i.e., where the denominator in Eq. 3 is N_d .



Fig. 6: As in Fig. 4 except for λ versus $S_{o,mod}$, i.e., where the denominators of the x and y axes are the same. The vertical dashed line corresponds to the surface remotely measured value of $S_{o,mod}$, i.e., 0.66 (Mann et al., 2014). Note that the previously reported value of $S_{o,mod}$ is used for reference and may not directly correspond to the rain rate thresholds used in the current study.



Fig. 7: (a) λ (and A_f) vs. S_{pop} , (b) λ vs. $S_{pop,mod}$, and (c) λ vs. $S_{o,mod}$ for the RICO simulations from Jiang et al. (2010). The colors correspond to increasing N_a from 100 mg⁻¹ to 200 (black), 300 (red), 400 (blue), and 500 (green) cm⁻³. The symbols denote the different thresholds used to conditionally average R and POP, i.e., $T_h = 0.001$ (closed circle), 0.5 (downward pointing triangle, and 5 (diamond) mm day⁻¹. In (a), the thin dashed line shows the linear relationship determined by Wang et al. (2012) for the $\lambda - S_{pop}$ relationship and the vertical dashed line corresponds to (a) the satellite-measured value of S_{pop} , i.e., 0.12 (Wang et al., 2012) and (c) the surface-based estimate of $S_{o,mod}$, i.e., 0.66 (Mann et al., 2014). Note that the pre28usly reported values of S_{pop} and $S_{o,mod}$ are used for reference and may not directly correspond to the rain rate thresholds used in the current study.



Fig. 8: N_d relative to N_d for the lowest aerosol number concentration scenario (i.e., $N_{d,0}$) for both the (a) RICO and (b) DYCOMS-II RF02 simulations. Doubling (red), tripling (blue), quadrupling (green), and quintupling (orange) N_a are depicted for both sets of simulations. corresponding to $N_a = 200, 300, 400, \text{ and } 500 \text{ cm}^{-3}$ relative to 100 cm^{-3} , respectively, for RICO and $N_a = 50, 75, 100, \text{ and } 125 \text{ cm}^{-3}$ relative to 25 cm^{-3} , respectively, for DYCOMS-II RF02.



Fig. 9: Schematic representation of the results presented herein. The red (blue) curve corresponds to the trajectory in the $\lambda - S_{pop}$ parameter space for increasing changes in N_a (i.e., ΔN_a) in marine stratocumulus (trade wind cumulus). The dotted region corresponds to the area of the parameter space where further increases in S_{pop} result in smaller changes in λ due to entrainment effects. The dashed region corresponds to the area of the area in which the cloud microphysical characteristics asymptote to nearly constant values for larger ΔN_a . The crossed area represents the region in which λ changes rapidly relative to small changes in S_{pop} .