Distinct regional meteorological influences on low cloud albedo susceptibility over global marine stratocumulus regions

Jianhao Zhang¹ and Graham Feingold²

¹CIRES at the University of Colorado Boulder; Chemical Sciences Laboratory at NOAA ²CSD, ESRL, NOAA, Boulder

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Abstract

Marine stratocumuli cool the Earth effectively due to their high reflectance of incoming solar radiation, and persistent occurrence. The susceptibility of cloud albedo to droplet number concentration perturbations depends strongly on large-scale meteorological conditions. Studies focused on the meteorological dependence of cloud adjustments often overlook the covariability among meteorological factors and their geographical and temporal variability. We use 8 years of satellite and reanalysis data sorted by day and geographical location to show that large-scale meteorological factors, including lower-tropospheric stability, free-tropospheric relative humidity, sea surface temperature, and boundary layer depth, have distinct covariabilities over each of the eastern subtropical ocean basins where marine stratocumulus prevail. This leads to markedly different monthly evolution in albedo susceptibility over each basin. Our results stress the importance of considering the geographical distinctiveness of temporal meteorological covariability when scaling up the local-to-global response of cloud albedo to aerosol perturbations.

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Jianhao Zhang^{1,2}and Graham Feingold²

5	¹ Cooperative Institute for Research in Environmental Sciences (CIRES), University of Colorado, Boulder,
6	CO, USA
7	² Chemical Sciences Laboratory, National Oceanic and Atmospheric Administration (NOAA), Boulder,
8	CO, USA

Key Points:

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10	• Annual mean cloud brightening potential is the highest over subtropical coastal
11	regions and the equatorial eastern Pacific.
12	• Features in regional relationships between key meteorological factors and albedo
13	susceptibilities are absent in a global analysis.
14	• Monthly evolution of cloud radiative susceptibility and the co-varying large-scale
15	meteorological conditions are regionally distinct.

 $Corresponding \ author: \ Jianhao \ Zhang, \ \texttt{jianhao.zhang} \texttt{Qnoaa.gov}$

16 Abstract

Marine stratocumuli cool the Earth effectively due to their high reflectance of incoming 17 solar radiation, and persistent occurrence. The susceptibility of cloud albedo to droplet 18 number concentration perturbations depends strongly on large-scale meteorological con-19 ditions. Studies focused on the meteorological dependence of cloud adjustments often 20 overlook the covariability among meteorological factors and their geographical and tem-21 poral variability. We use 8 years of satellite and reanalysis data sorted by day and ge-22 ographical location to show that large-scale meteorological factors, including lower-tropospheric 23 stability, free-tropospheric relative humidity, sea surface temperature, and boundary layer 24 depth, have distinct covariabilities over each of the eastern subtropical ocean basins where 25 marine stratocumulus prevail. This leads to markedly different monthly evolution in albedo 26 susceptibility over each basin. Our results stress the importance of considering the ge-27 ographical distinctiveness of temporal meteorological covariability when scaling up the 28 local-to-global response of cloud albedo to aerosol perturbations. 29

³⁰ Plain Language Summary

Bright, warm marine clouds help cool the Earth by reflecting a good fraction of sun-31 light. Their brightness is modulated by the amount of tiny particles in the air (aerosol) 32 that can be induced by natural and/or human activities, e.g., volcanic eruptions and/or 33 ship exhaust. Using 8 years of satellite observations, we show maps of potential cloud 34 brightness changes associated with increases in the number of cloud droplets. The re-35 sults suggest strong cloud brightening potential over subtropical coastal regions and weak 36 darkening potential (reduction in cloud brightness) over some parts of the remote oceans. 37 We find that the environmental conditions in which these cloud reside co-vary in time 38 differently from one part of the world to another, leaving distinct regional fingerprints 39 of cloud brightness changes. Such distinct fingerprints are not evident when data is ag-40 gregated globally. These findings imply that environmental conditions, especially the way 41 they co-vary with each other, and their frequency of occurrence in space and time, is key 42 to assessment of the overall brightness changes of marine low clouds. 43

44 1 Introduction

Marine warm (liquid) clouds cover about one third of the global ocean surface in 45 annual mean (Chen et al., 2014). They prevail over low-latitude to mid-latitude oceans, 46 more pronouncedly over the eastern subtropical oceans where the Earth's major semi-47 permanent marine stratocumulus decks form (Klein & Hartmann, 1993; Wood, 2012). 48 These bright and blanket-like stratiform clouds reflect a good fraction of the incident so-49 lar radiation (ranging from 0.35 to 0.42 in annual mean; Bender et al., 2011) that would 50 otherwise (in the absence of these clouds) be largely absorbed by the dark ocean ($\sim 94\%$), 51 effectively cooling the Earth (e.g., Stephens et al., 2012). For warm clouds exhibiting con-52 stant macrophysical properties (e.g., liquid water path (LWP) and cloud cover), their 53 brightness, or cloud albedo, quantified as the ratio of the reflected shortwave flux to the 54 incoming solar radiation at the top of atmosphere, is particularly sensitive to the droplet 55 concentration (N_d) , such that higher N_d accompanied by smaller drops makes the cloud 56 more reflective (cloud brightening; Twomey, 1974, 1977). However, cloud macrophys-57 ical properties do change with time as the system evolves, through precipitation, evap-58 oration, and/or entrainment mixing processes (Wood, 2012). Microphysical changes in 59 N_d and droplet sizes induced by aerosol perturbations can substantially modulate the 60 rate and efficiency of these processes and thereby cause further adjustments in macro-61 physical properties and cloud albedo (e.g., Ackerman et al., 2004; Bretherton et al., 2007; 62 Jiang et al., 2006). 63

In nature, the responses of cloud macrophysical properties to N_d perturbations are always complicated by the variability driven by local meteorology, and for decades, the

stated challenge and focus has been to untangle aerosol effects from covarying meteo-66 rological conditions (Stevens & Feingold, 2009). Simulations of marine boundary layer 67 (MBL) clouds, in which meteorology can be easily controlled, indicate a bidirectional LWP 68 adjustment to increasing N_d , such that for precipitating clouds, an increase in N_d induces 69 smaller droplets that suppress condensate removal, eventually leading to an increase in 70 LWP (brighter clouds; Albrecht, 1989), whereas for non-precipitating clouds, the reduced 71 droplet sizes lead to weaker sedimentation fluxes at cloud tops, (Bretherton et al., 2007) 72 and faster evaporation (Wang et al., 2003; Xue & Feingold, 2006), which both cause stronger 73 entrainment mixing that reduce cloud LWP, resulting in less reflective clouds. 74

Observations of cloud adjustments following anthropogenic aerosol perturbations 75 confirm the bidirectional LWP responses (e.g., Chen et al., 2012; Trofimov et al., 2020). 76 while the aggregated response remains uncertain (Malavelle et al., 2017; Toll et al., 2019; 77 Christensen et al., 2022). This means that cloud LWP responses to increased N_d can ei-78 ther enhance or offset the microphysical brightening depending on the meteorological con-79 ditions. Progress has been made over the years towards establishing fundamental knowl-80 edge of the environmental state/regime dependence of cloud adjustments to aerosol per-81 turbations. For inversion-capped MBL clouds, the budget of cloud condensate is regu-82 lated mainly by entrainment drying at cloud tops and the fraction of precipitation that 83 reaches the surface, which are strongly dependent on the humidity in the free-troposphere 84 and the lower-tropospheric stability (Ackerman et al., 2004; Chen et al., 2014; Gryspeerdt 85 et al., 2019). In part related to the atmospheric stability, clouds exhibit a much more 86 negative LWP response to increased N_d in deep MBLs than those that reside in shallower 87 MBLs (e.g., Possner et al., 2020; Toll et al., 2019). Furthermore, Dagan et al. (2015) show 88 that the direction in which cloud condensate responds to an increase in aerosol depends on an optimal aerosol concentration which is determined by thermodynamic conditions 90 (e.g., temperature and humidity). Wood (2007) shows that cloud-base height is the sin-91 gle most important determinant of whether cloud thickness changes will enhance or off-92 set the Twomey brightening. 93

Clearly, the spatiotemporally integrated response of cloud albedo (A_c) to aerosol 94 perturbations depends crucially on the frequency of occurrence of the environmental states 95 that characterize cloud adjustments. However, we lack quantitative and even qualita-96 tive characterization of the way meteorological conditions influence aerosol effects in the 97 real world. Mülmenstädt and Feingold (2018) state the need for a shift in attention from 98 untangling aerosol effects from covarying meteorology towards embracing and understand-99 ing the covariabilities between them. The focus of this study is exactly on this point. In 100 addition to characterizing the geographical distribution of warm cloud albedo suscep-101 tibility using satellite observations over global oceans from 60° S to 60° N, we illustrate 102 the dependence of large-scale meteorological influences on albedo susceptibility in dif-103 ferent stratocumulus basins (Section 4) and the role of temporal covariabilities in gov-104 erning the observed susceptibility (Section 5); the latter are understudied and often ig-105 nored in "untangling" studies. We find distinct fingerprints (in terms of monthly evo-106 lution) of albedo susceptibility in different stratocumulus basins, consistent with the cor-107 responding temporally covarying meteorological conditions. We show that a frequency-108 weighted aggregation of these regional fingerprints obscures these regional differences and 109 therefore gives a biased view of albedo susceptibility. 110

Data and Methods

¹¹² We obtain coincident marine low-cloud properties, including cloud optical depth ¹¹³ (τ), cloud top effective radius (r_e), low-cloud fraction (f_c), cloud LWP, cloud top height ¹¹⁴ (CTH), and top-of-atmosphere (TOA) shortwave (SW) fluxes from the MODerate res-¹¹⁵ olution Imaging Spectroradiometer (MODIS) (Platnick et al., 2003) and the Clouds and ¹¹⁶ the Earth's Radiant Energy Systems (CERES; Wielicki et al., 1996) sensors onboard the ¹¹⁷ Terra and Aqua satellite (overpass ~10:30 and ~13:30 local time, respectively), which

are integrated into the CERES Single Scanner Footprint (SSF) product Edition 4 (level 118 2) with a footprint resolution of 20 km (Su et al., 2015). N_d is calculated following Zhang 119 et al. (2022) for all CERES footprints with $f_c > 0.8$, in order to minimize retrieval bi-120 ases (Grosvenor et al., 2018), while including some partially cloudy footprints. Footprint 121 cloud properties are aggregated to 1° spatial resolution, to match susceptibilities calcu-122 lated for individual $1^{\circ} \times 1^{\circ}$ satellite snapshots. At this scale, the confounding effect of 123 meteorology is negligible (Goren & Rosenfeld, 2012, 2014). Linear least-squares log-log 124 regressions of footprint properties are used to calculate albedo susceptibility $S_0 = dln(A_c)/dln(N_d)$ 125 and radiative susceptibility $F_0 = d(A_c)/dln(N_d) \times f_c \times SW_{dn}$; for both metrics, posi-126 tive values indicate more reflected sunlight, thereby cooling, following Zhang et al. (2022). 127 Note that in calculating S_0 we do not stratify by LWP. The logarithmic transformation 128 alleviates the dependence of S_0 on the absolute value of N_d . 129

Meteorological conditions, including sea surface temperature (SST), lower tropo-130 spheric temperature, humidity, and wind profiles, are obtained from the European Cen-131 tre for Medium-Range Weather Forecasts (ECMWF) fifth-generation atmospheric re-132 analysis (ERA5; Hersbach et al., 2020), and interpolated and aggregated to the Terra 133 and Aqua overpass times at 1° spatial resolution. Lower-tropospheric-stability (LTS) is 134 calculated as the difference in potential temperature between 700 hPa and 1000 hPa. Free-135 tropospheric relative humidity (RH_{ft}) is defined as the the mean relative humidity be-136 tween inversion top and 700 hPa, following Eastman and Wood (2018). 137

The datasets span 60° S to 60° N, covering global oceans, from 2005 to 2012 (8 years). 138 We screen for cloudy satellite scenes over open water when only single layer liquid cloud 139 (SLLC) is present. Analyses using the Aqua observations are shown in the main text, 140 and those using the Terra observations are shown in the supplementary material, in or-141 der to assess robustness of our findings (qualitatively), and to explore the role of the di-142 urnal cycle (quantitatively). Regional annual maxima in SLLC fractional coverage and 143 frequency of occurrence are used to identify 5 major marine stratus/stratocumulus re-144 gions $(20^{\circ} \times 20^{\circ})$, Fig. S1, magenta boxes). 145

¹⁴⁶ 3 Global map of albedo susceptibility

The climatology of geographical distribution of marine low-cloud S_0 (Fig. 1a) is 147 represented by an aggregation of susceptibilities derived from individual satellite snap-148 shots over the 8-year period, taking into account the frequency of occurrence of differ-149 ent cloudy scenes and meteorological regimes. It is clear that over most parts of the global 150 ocean (60° S to 60° N), low clouds have a brightening potential (positive S_0) in annual 151 mean, more pronouncedly off the coast of continental land masses where N_d is climato-152 logically higher (Fig. S2) and the MBL is shallower (Fig. S3), compared to those over 153 remote oceans. Only over the remote subtropical southeast Pacific/Atlantic regions, do 154 the data show weak darkening potential (negative S_0) in annual mean. The darkening 155 potential means that the brightening of the clouds via the Twomey effect -i.e., more 156 particles lead to more droplets and brighter clouds – is more than compensated by liq-157 uid water losses. 158

One can then translate the S_0 map into an annual flux perturbation potential map 159 (Fig. 1b), which highlights the high annual cooling potential over subtropical stratocu-160 mulus regions even more, by taking into account the cloud fraction and frequency and 161 amount of incoming solar radiation at a given geographical location. In the remote parts 162 of the subtropical stratocumulus decks, warmer SSTs deepen the MBL and encourage 163 entrainment of free-tropospheric air at cloud tops (Fig. S3 and Bretherton, 1992; Wyant 164 et al., 1997), favoring entrainment-feedback-driven LWP decreases with increasing N_d 165 (Bretherton et al., 2007; Wang et al., 2003). The prevalence of this MBL condition leads 166 to a frequently occurring cloud darkening regime that offsets the Twomey brightening, 167



Figure 1. Geographical distribution of marine low-cloud (a) albedo susceptibility (S₀) and (b) the product of radiative susceptibility (F₀) and annual frequency of occurrence of single layer liquid cloud (SLLC). Spatial-temporal averages of $5^{\circ} \times 5^{\circ}$ areas are shown. Only areas with SLLC frequency of occurrence greater than 0.1 are shown in (a).

consistent with the net warming potential observed over the southeast Pacific/Atlantic (Fig. 1b).

Contributions from the three susceptibility regimes, namely non-precipitating bright-170 ening, darkening, and precipitating brightening, to the total F_0 are observed to have dis-171 tinct geographical preferences (Fig. 2). The regime separation is performed for each 5° 172 \times 5° area to ensure robustness, based on the sign of S₀ and a r_e of 12 μ m (above which 173 clouds are more likely to drizzle) that manifest in the LWP- N_d variable space, similarly 174 to Zhang et al. (2022). Each regime represents a cluster of cloud states that is dominated 175 by brightening or darkening, and precipitating or non-precipitating potentials. Contri-176 butions from the non-precipitating brightening cloud states tend to dominate the shal-177 low, often polluted, stratus/stratocumulus off the coast of continents (Fig. 2a). The pre-178 cipitating brightening cloud states, attributed to rain suppression (Albrecht, 1989), con-179 tribute substantially over most parts of the remote, clean oceans and the equatorial east-180 ern Pacific (Fig. 2c). Inbetween the geographical preferences of the above two regimes 181 lies the region where non-precipitating darkening cloud states become the leading con-182 tributor to the overall F_0 , especially over the southeast Pacific and Atlantic (Fig. 2b), 183 where net warming potentials are observed (Fig. 1). 184

4 Distinct fingerprints of S_0 in meteorological space at regional scale

Local adjustments of low clouds to aerosol perturbations are strongly dependent on the depth of the stratocumulus-topped MBL (approximated by CTH; e.g., Possner et al., 2020; Toll et al., 2019) and RH_{ft} (e.g., Chen et al., 2014; Gryspeerdt et al., 2019) Figure 3 shows S₀ under different MBL and free-troposphere (FT) states, as a function of CTH and RH_{ft} . Globally (60° S - 60° N), positive S₀ is found everywhere across the



Figure 2. Geographical distribution of marine low-cloud radiative susceptibility (F₀) separated into 3 regimes: (a) non-precipitating brightening, (b) darkening, and (c) precipitating brightening. The 3 regimes are separated based on the sign of S₀ and a r_e of 12 μ m in the LWP-N_d variable space, for each 5° × 5° area, similarly to Zhang et al. (2022). Only areas with SLLC frequency of occurrence greater than 0.1 are shown.



Figure 3. Mean S₀ under different meteorological conditions, namely free-tropospheric relative humidity (RH_{ft} ; x-axis) and cloud top height (CTH; y-axis; a proxy for the marine boundary layer depth), for (a) global oceans (60° S - 60° N), (b) NE Pacific, (c) SE Pacific, (d) SE Atlantic, (e) NE Atlantic, and (f) Australian stratocumulus regions. Bin sizes for CTH and RH_{ft} are 0.2 km and 10%, respectively. The size of the square indicates the frequency of occurrence of a meteorological state. Bins with less than 0.1% frequency of occurrence (or less than 100 samples) are not shown.

two meteorological state-spaces, with less susceptible conditions occurring under drier 191 FT and intermediate MBL depth (~ 1.5 km; Fig. 3a). This is consistent with the entrain-192 ment feedback argument that reduced droplet sizes and the subsequent reduced cloud-193 top sedimentation flux enhance evaporation and thereby cloud-top entrainment mixing 194 (Bretherton et al., 2007; Xue & Feingold, 2006), which is further facilitated by the deeper 195 MBL and the drier air above cloud tops. Cloud brightening potentials associated with 196 rain-suppression overwhelm these entrainment-feedback-consistent signals, as the clouds 197 become even deeper (>2 km) and are more likely to precipitate (Fig. 3a). 198

199 When the stratocumulus regime is singled out (Fig. 3b-f), the S_0 distribution in the two meteorological states is in qualitative agreement with the global analysis, how-200 ever, cloud darkening (negative S_0) appears under the deep-MBL, dry-FT atmospheric 201 states, more pronouncedly over the southeast Pacific stratocumulus deck (Fig. 3c), while 202 weak brightening potential is observed under those conditions over the NE Atlantic (Fig. 203 3e). This is likely because clouds over the NE Atlantic precipitate more often than those 204 over the SE Pacific, under these MBL and FT conditions (discussed further in Section 205 5).206

Distinct "fingerprints" of S_0 in the CTH-RH_{ft} variable space are evident, when in-207 dividual basins are being compared. This manifests in two ways; first, the frequency of 208 occurrence (indicated by the square sizes) of the FT and MBL conditions varies from 209 basin to basin. For example, deep MBL (>2 km) or humid FT conditions rarely occur 210 under the large-scale subsidence-dominated regions (Fig. 3b-d), compared to the NE At-211 lantic or the Australian basins (Fig. 3e-f). Second, different S_0 , at least in magnitude, 212 and in some cases even in sign, are observed across basins. This suggests cloud states 213 214 (defined in LWP, N_d space) are not necessarily the same for the same MBL and FT states, implying that other meteorological factors co-evolve with MBL and FT states differently 215 from region to region, leaving distinct imprints on S_0 . These distinct regional fingerprints 216 of S_0 -meteorology relationships are lost in the global analysis (Fig. 3a) due to the merg-217 ing of different cloud/meteorology regimes besides MBL depth and RH_{ft} . 218

5 Seasonal covariability between albedo susceptibility and large-scale meteorology

Four key large-scale meteorological factors evolve and co-vary distinctly across basins 221 (Fig. 4, right column), leading to markedly different monthly evolution in F_0 (Fig. 4, left column). Even among regions strongly influenced by large-scale subsidence (Fig. 4a-223 c), large-scale meteorological conditions vary in magnitude and do not covary the same 224 way temporally (e.g., RH_{ft} tracks SST except over the SE Atlantic, LTS anti-correlates 225 with SST except over the NE Pacific). As a result of the complex and distinct regional 226 covariability in meteorological conditions, the temporal rise and fall of a single meteo-227 rological factor leads to markedly different responses in F_0 across basins. For instance, 228 when SST peaks over the NE Pacific, F_0 is at its annual minimum, owing to the coin-229 cident relatively strong LTS, keeping the high-LWP (color of the circle) clouds from deepening-230 precipitating while being susceptible to entrainment drying (Fig. 4a, Jul-Aug). In con-231 trast, over the Australian stratus region, the annual maximum in F_0 coincides with the 232 warmest SST, owing to enhanced Twomey brightening potential associated with more frequent non-precipitating conditions, compared to the other months over this region (Fig. 234 4e, Feb). Taking CTH as another example, high CTHs (deep MBLs) lead to strong precipitation-235 suppression brightening over the NE Atlantic, whereas deep MBLs over the SE Pacific 236 show very weak brightening potentials, due to high stability and dry FT conditions, in 237 striking contrast to the NE Atlantic (Fig. 4b and d). 238

The covariability among large-scale meteorological factors over the SE Atlantic follows that over the SE Pacific, although the ocean surface is warmer, LTS is weaker, and the FT is moister in general over the SE Atlantic (Fig. 4c). This leads to qualitatively



Figure 4. Left column: Monthly mean radiative susceptibility (F₀; black; positive values indicate cooling) and frequency of occurrence of SLLC (magenta). Color of the circle indicates monthly mean LWP. Size of the circle indicates monthly mean N_d. Open (closed) circles indicate likely precipitating (non-precipitating) condition, based on $r_e = 12 \ \mu\text{m}$. Middle column: Monthly mean F₀ broken into 3 regimes: non-precipitating brightening (dotted green), darkening (brown), and precipitating brightening (solid green). Right column: Monthly mean meteorological conditions: LTS (blue), RH_{ft} (dark green), SST (red), and CTH (cyan). Rows (a–e) represent results for the NE Pacific, SE Pacific, SE Atlantic, NE Atlantic, and the Australian stratocumulus regions, respectively.

similar F_0 evolutions between the two basins; that is high F_0 during austral winter and 242 low F_0 during austral summer. An exception occurs during late fall to winter (June-July). 243 when precipitating clouds over the SE Pacific exhibit relatively weak positive F_0 whereas 244 non-precipitating high N_d clouds occur and exhibit strong F_0 over the SE Atlantic. This 245 difference can be attributed to an aerosol source that is unique to the SE Atlantic basin, 246 in the form of a large amount of biomass burning aerosol that is advected by the co-occurring 247 African Easterly Jet in the FT during the southern African burning season (June-October; 248 Adebiyi & Zuidema, 2016). The elevated aerosol is likely to be entrained into the MBL 249 during June-July when the FT jet is not yet at its full strength (Zhang & Zuidema, 2021). 250

Among five subtropical stratocumulus/stratus regions, the SE Pacific hosts the least 251 susceptible conditions overall and is the only basin with monthly mean cloud darken-252 ing potential (Fig. 4b). This is attributed to the extremely dry free-tropospheric con-253 ditions, under which entrainment feedbacks acting to reduce cloud LWP, and associated 254 with reduced droplet sizes (due to increasing aerosol) are enhanced. Low clouds over the 255 NE Atlantic indicate the highest cloud brightening potential among the five regions, es-256 pecially during March-September when the MBL is shallow and FT is relatively moist, 257 giving rise to thin, non-precipitating clouds with low LWP, relatively high N_d , and the 258 lowest entrainment-driven cloud darkening potential (Fig. 4d). During October-February 259 over the NE Atlantic, when CTH is high (deep MBL), clouds precipitate often, leading 260 to a high potential for precipitation-suppression related cloud brightening. Given the deep MBLs, precipitating conditions occur fairly frequently over the Australian stratus region 262 almost throughout the year, except for the January-March period when increasing LTS 263 leads to lower LWP and shallower MBL (Fig. 4e). This leads to an almost all-year-round 264 precipitation-suppression driven cloud brightening potential, enhanced during the January-March period with a contribution from the non-precipitating brightening. 266

Albedo susceptibility, cloud frequency and areal coverage, and aerosol conditions 267 (indicated by N_d) collectively determine the SW flux budget at TOA in response to an 268 aerosol perturbation. The temporal covariability among these variables (Fig. 4, left col-269 umn) can lead to a muted SW flux perturbation even when highly susceptible clouds oc-270 cur, due to a coinciding low frequency of cloud occurrence and/or high aerosol condi-271 tions (e.g., the NE Pacific and the SE Atlantic). This stresses the necessity of taking such 272 temporal covariability into account when assessing the climatological radiative effect of 273 aerosol-cloud interactions. Furthermore, not only do various spatial-temporal averages 274 applied in satellite-based approaches lead to biased susceptibilities (Feingold et al., 2022), 275 temporal covariabilities among multiplicands (F_0 , cloud frequency, and aerosol condi-276 tion) also bias their product (TOA SW flux perturbation) if temporal averages are ap-277 plied to multiplicands before multiplication (Fig. S4). Biases associated with individ-278 ual stratocumulus regions vary in sign and magnitude, indicating that these quantities 279 do not necessarily co-vary the same way temporally across basins (e.g., the NE versus 280 SE Pacific). 281

²⁸² 6 Discussion and implications

To illustrate our findings in a conceptual framework one may ideally construct a 283 manifold that depicts S_0 in a 4-dimensional (SST, CTH, RH_{ft} , and LTS) space by pop-284 ulating the 4-D space with a large body of realizations, i.e., climatological observations. 285 However, often ignored is that each realization used to construct the 4-D manifold is as-286 sociated with a specific combination of longitude, latitude, and time. In other words, given 287 a certain geographical region and time, realizations will only populate parts of the 4-D 288 space, leading to local constructions (distinct fingerprints) that looks different from a 289 global construction. 290

Our work is highly relevant to assessment of the radiative effect of aerosol-cloud interactions for climate applications. In addition, our findings have direct implications

for Marine Cloud Brightening (MCB), which has been proposed as a way to mitigate the 293 worst effects of the ongoing global warming crisis by creating more reflective (in the SW) 294 MBL clouds, ideally with expanded areal coverage and prolonged lifetime, through de-295 liberate aerosol injections (Latham et al., 2012). The bright, linear cloud features seen 296 in satellite images, referred to as ship tracks (Coakley et al., 1987), are examples of ideal 297 outcomes of an MCB experiment, and the conditionality of such an outcome on mete-298 orological conditions is one of the key issues underpinning the viability of MCB. This 299 study underscores two key points for the MCB community: 1) understanding or eval-300 uating the impact of meteorology on cloud albedo susceptibility needs to be done at lo-301 cal/regional scales, where meteorological covariability is accounted for; 2) When scaling 302 up the flux perturbation, it is crucial to consider the natural covariability between me-303 teorology and aerosol, to which cloud responses to aerosol perturbation are sensitive. 304

Last but not least, S_0 and the regime-specific F_0 derived from the morning obser-305 vations (Terra; Fig. S5-S6) have the same geographical distributions as those observed 306 in the afternoon (Aqua; Fig. 1-2), except that Terra observations indicate a slightly higher 307 global mean S_0 of 0.14, compared to 0.13 for Aqua observations. While the qualitative 308 distributions of S_0 in the CTH-RH_{ft} state space remain the same, regardless of the ob-309 serving time (morning versus afternoon), the morning observations (Terra; Fig. S7) do 310 indicate a general shift towards $S_0 > 0$, which replaces the cloud darkening regime re-311 lated to the deep-MBL dry-FT conditions (Fig. 3) with a weak brightening regime. This 312 stresses the importance of cloud diurnal evolution for S_0 in that the same meteorolog-313 ical conditions may lead to opposing susceptibility regimes (i.e., brightening versus dark-314 ening) depending on the time of the day. Except for generally higher cloud LWP and 315 higher F_0 , the characterized covariability among cloud LWP, N_d , SLLC frequency of oc-316 currence, F_0 , and large-scale meteorological conditions using the Terra observations agrees 317 well with those using the Aqua observations (Fig. S8 and Fig. 4). 318

7 Concluding Remarks

Marine warm cloud albedo susceptibility is derived from satellite-retrieved cloud 320 microphysical properties and radiative fluxes, and sorted by day and geographical loca-321 tion. Geographical distributions of albedo susceptibility and the contributions from three 322 susceptibility regimes (non-precipitating brightening, darkening, precipitating brighten-323 ing) are shown over global oceans (60° S to 60° N). Monthly evolutions in cloud radia-324 tive susceptibility, meteorological conditions (from ERA5 reanalysis), warm cloud fre-325 quency of occurrence, LWP and N_d are shown for five primary eastern subtropical stra-326 tus/stratocumulus regions $(20^{\circ} \times 20^{\circ})$, to illustrate the covariabilities among them. The 327 key findings are as follows: 328

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1. An overall annual mean cloud brightening potential is observed for global marine warm clouds – most pronounced over subtropical coastal regions where shallow marine stratocumulus prevail along with high annual-mean N_d , and over the equatorial eastern Pacific where clouds rain more often (Fig. 1).

- 2. Cloud darkening associated with entrainment-driven negative LWP adjustments offsets the cloud brightening potential over remote parts of the stratocumulus regions where deeper MBLs favor cloud top entrainment, especially over the SE Pacific/Atlantic where darkening overcomes brightening in annual mean (Fig. 1 and Fig. 2).
- 338 3. The distinct regional fingerprints of S_0 in CTH and RH_{ft} variable space are absent in the global analysis because the latter merges different cloud/meteorology regimes (Fig. 3).
- 4. Meteorological conditions have distinct regional covariabilities, leading to markedly different monthly evolutions in F_0 (Fig. 4).

- 5. The SE Pacific, a region with the driest free-tropospheric conditions, hosts the least susceptible clouds exhibiting cloud darkening potential over several months during austral winter. Frequently occurring non-precipitating low-LWP, high-N_d clouds, found in shallow MBLs (March-September) over the NE Atlantic, represent the highest potential radiative responses to N_d perturbations among the five stratocumulus regions (Fig. 4).
- 6. While the qualitative agreement between Terra and Aqua underscores the robustness of our findings, their quantitative disagreement points to the important role of cloud diurnal evolution in determining albedo susceptibility (Figs S5-S8).

When the influence of meteorological conditions on low cloud S_0 are studied, it may 352 seem tempting to try to disentangle effects of individual meteorological factors on S_0 by 353 controlling for the others. Our results, however, indicate that this may not be the best 354 approach since it is the natural covariability among meteorological conditions that dic-355 tates the regionally distinct temporal evolution in S_0 . These results convey the impor-356 tance of spatiotemporal variability in S_0 as a basis for both understanding the limita-357 tion in scale-up of the meteorological influences on the radiative effect of aerosol-cloud 358 interactions from regional to global, as well as for making decisions regarding when, where, 359 and if marine cloud brightening efforts should be attempted. 360

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368 Data Availability Statement

The CERES SSF data were obtained from the Atmospheric Science Data Center 369 at the NASA Langley Research Center (https://asdc.larc.nasa.gov/project/CERES/ 370 CER_SSF_Aqua-FM4-MODIS_Edition4A; NASA/LARC/SD/ASDC (2014a); https://asdc 371 .larc.nasa.gov/project/CERES/CER_SSF_Terra-FM2-MODIS_Edition4A; NASA/LARC/SD/ASDC 372 (2014b)). The fifth-generation ECMWF (ERA5) atmospheric reanalyses of the global 373 climate data are available through the Copernicus Climate Change Service (C3S, https:// 374 doi.org/10.24381/cds.bd0915c6; Hersbach et al. (2020)). The 1° albedo susceptibil-375 ity data derived and shown in this study can be found at https://csl.noaa.gov/groups/ 376 csl9/datasets/data/cloud_phys/2022-Zhang-Feingold/. 377

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