



Regime-dependent impacts of aerosol vertical placement on liquid water path adjustment in southeastern Atlantic marine boundary layer clouds

Xiaojian Zheng^{1,2}, Yan Feng², David Painemal³, Shaocheng Xie⁴, Zhujun Li^{3,5}, and Yuan Wang¹

¹Department of Earth System Science, Stanford University, Stanford, CA, USA

²Environmental Science Division, Argonne National Laboratory, Lemont, IL, USA

³Science Directorate, NASA Langley Research Center, Hampton, VA, USA

⁴Lawrence Livermore National Laboratory, Livermore, CA, USA

⁵Analytical Mechanics Associates, Hampton, VA, USA

Correspondence: Xiaojian Zheng (xjzheng@stanford.edu)

Abstract

5 The adjustment of cloud liquid water path (LWP) to droplet number concentration (N_d) remains a leading uncertainty in the aerosol-induced cloud radiative effect. We combine nine years (2006–2014) of collocated, vertically resolved CALIOP aerosol extinction and MODIS cloud retrievals over the southeastern Atlantic around Ascension Island, and partition the meteorology into three weather regimes (weakened-high, background, and strengthened-high) with a deep-learning
 10 clustering of ERA5 circulation, to quantify the LWP adjustment to droplet concentration (L_0) by regime and by aerosol vertical placement relative to the cloud top. In the strengthened-high regime, which encompasses the smoky dry season, increasing separated free-tropospheric aerosol steepens L_0 from -0.07 to -0.31 , as the shortwave absorption of smoke aerosol enhances the inversion that caps the cloud. This steepening persists despite the moisture co-transported with
 15 the plume, which partly offsets the entrainment-driven loss of cloud water. Contacting absorbing aerosol thins the cloud in both the strengthened-high and background regimes, but the LWP– N_d coupling weakens toward zero in the humid background regime. Combining all days without regime distinction weakens both signals. In contrast, the nudged Energy Exascale Earth System Model version 2 (E3SMv2) produces a uniformly positive LWP adjustment to N_d across the
 20 regimes and aerosol placements, consistent with a precipitation-suppression response. We suggest that cloud-water adjustments in smoke-topped stratocumulus be quantified by weather regime and with vertically resolved aerosol predictors, and that the rain-mass state and sub-cloud updraft in E3SMv2 be calibrated before the simulated adjustments are used to constrain aerosol forcing.

25



1. Introduction

Marine boundary layer (MBL) clouds over the southeastern Atlantic (SEA) constitute one of the largest and most radiatively influential subtropical low-cloud regimes (Adebiyi et al., 2020). Their persistence is maintained by broad subsidence, sharp capping inversions, and cool upwelled waters of the Benguela current, with additional modulation from the southern African easterly jet (AEJ-S) in the mid-troposphere and the coastal low-level jet (Ryoo et al., 2021, 2022). The resulting cloud deck exerts a strong shortwave cooling on the regional energy budget and is climatologically most persistent during the dry-season months when smoke export from the African continent peaks (July–October) (Redemann et al., 2021; Ryoo et al., 2021).

A prominent feature of the SEA system is the seasonal westward transport of biomass-burning aerosols from southern Africa. Fires over the Namibian and Angolan highlands inject smoke rich in black and organic carbon that is lofted to 3–6 km and advected thousands of kilometers over the stratocumulus deck by the mid-level easterlies, frequently producing elevated smoke-over-cloud conditions (Adebiyi and Zuidema, 2016; Redemann et al., 2021). This configuration produces complex aerosol–cloud interactions (ACIs). Smoke aloft can modify lower-tropospheric thermodynamics by absorbing solar radiation and heating the atmosphere (the semi-direct effect; Ackerman et al., 2000; Johnson et al., 2004; Wilcox, 2012), and, when aerosol mixes into or below the cloud layer, it can act as cloud condensation nuclei (CCN) and alter the cloud droplet number concentration (N_d), precipitation, and radiative properties (e.g., Costantino and Bréon, 2013; Diamond et al., 2018; Kacarab et al., 2020; Zhang and Zuidema, 2021). Importantly, the smoke does not travel alone. The plume air is systematically moister than the surrounding free troposphere, so that smoke events over the SEA are simultaneously moisture events, with consequences for cloud maintenance that can oppose the aerosol effects themselves (Zhang and Zuidema, 2021; Diamond et al., 2022). The smoke plume is moist relative to the subsiding free-tropospheric air it displaces, while the smoky season itself remains the driest state of the free troposphere in the regime mean, a distinction quantified in Sect. 3.4.

Over the SEA, ground-based measurements at Ascension Island (8°S, 14.5°W) during the U.S. Department of Energy (DOE) Layered Atlantic Smoke Interactions with Clouds (LASIC) campaign documented strong diurnal modulation of boundary-layer thermodynamics, cloud, and precipitation properties under smoky conditions relative to clean periods (Zuidema et al., 2018; de Graaf et al., 2023), a cycle that also modulates the regional aerosol warming (Chang et al., 2025), and de Graaf et al. (2023) further isolated a first indirect effect (Twomey, 1977) of African smoke at the base of the marine cumulus clouds downwind of the island. Dedrick et al. (2024, 2025) showed that the activation link between aerosol and droplet number weakens in smoky air because available supersaturation is consumed by particle competition, so the Twomey response saturates precisely where aerosol is most abundant. Aircraft profiling during



the ObseRvations of Aerosols above CLouds and their intEractionS (ORACLES) campaign
 65 demonstrated that the vertical separation between the smoke layer and the cloud top is decisive.
 The same control appears in long-range-transported plumes over the eastern North Atlantic,
 where a layer whose base lies more than about 100 m above the cloud top leaves the cloud
 microphysics unchanged, and aerosol within the boundary layer alters the droplets more than an
 equal amount entrained from above (Y. Wang et al., 2020). Clouds in contact with overlying
 70 smoke carry higher N_d , smaller effective radii, and substantially reduced drizzle relative to
 vertically separated cases, and the contact air itself is more humid, which offsets the evaporative
 effects of cloud-top entrainment (Redemann et al., 2021; Gupta et al., 2021, 2022). Satellite
 analyses corroborate and extend these findings, though aerosol and cloud fields over this
 stratocumulus deck vary strongly in space and time (Chang et al., 2021), and these records have
 75 been used to assess the radiative impact of the smoke layers directly (Logan et al., 2024). Over
 the southeastern Atlantic, the low-cloud fraction reaches its seasonal maximum in the same dry-
 season months in which the smoke loading peaks, and Zhang and Zuidema (2021) showed that
 the light-absorbing aerosols above the clouds amplify this seasonal cycle and modify cloud
 optical properties and precipitation.

80 Modeling studies likewise find that the cloud response depends on where the absorbing aerosol
 layer is located relative to the inversion. High-resolution large-eddy simulations demonstrate
 enhanced entrainment-driven thinning when the heated layer is separated from the cloud top and
 a muted response in cloud thickness, tied to reduced cloud-top radiative cooling, when the
 absorbing aerosol resides at or within the inversion (Zhou et al., 2017; Herbert et al., 2020). Baró
 85 Pérez et al. (2024) found that heating by smoke residing within the boundary layer tends to
 reduce cloud cover and accelerate the stratocumulus-to-cumulus transition. On larger scales,
 Diamond et al. (2022) showed that cloud adjustments arising from smoke–circulation
 interactions strongly modulate that same transition. Meanwhile, regional and global models often
 misplace the smoke layer and underestimate above-cloud aerosol loading and its organic
 90 fraction, biasing above-cloud aerosol optical depth and, consequently, simulated cloud
 adjustments (Das et al., 2020; Shinozuka et al., 2020).

Despite the progress, significant knowledge gaps remain. Though observations and models
 largely agree that above-cloud smoke can warm and stabilize the lower troposphere and
 influence cloud microphysics, albedo, and drizzle, the magnitude and even the sign of the
 95 inferred cloud adjustments vary across studies. Two persistent difficulties limit what satellite
 ACI statistics over the SEA can claim. The first is the lack of vertical context. Most satellite ACI
 studies rely on column-integrated metrics such as aerosol optical depth (AOD) or aerosol index,
 without information on where the aerosol actually resides, even though the cloud response
 depends critically on whether the smoke lies above the cloud or is embedded in it (Zhang and
 100 Zuidema, 2021). Stier (2016) found in an aerosol–climate model that AOD explains only about
 25 % of the variability in boundary-layer CCN globally, and Painemal et al. (2020) demonstrated



the limitations of using AOD to infer ACI. Matching CALIOP-derived extinction near the cloud top with MODIS droplet number yields far stronger correlations than using column AOD, which explains only a small fraction of the variance. To our knowledge, sustained region-scale observational constraints that explicitly resolve aerosol height relative to the cloud top remain limited over the SEA, which in turn hampers targeted model evaluation and process examination. The second difficulty is meteorological confounding. Smoke arrives over the SEA embedded in particular circulation states, and the loading, plume altitude, co-transported moisture, and cloud-controlling environment (stability, subsidence, and free-tropospheric humidity) all covary with the synoptic regime (Adebiyi and Zuidema, 2018). The aerosol–cloud relationship itself also shifts with the boundary-layer humidity and turbulence that accompany each state, as ground-based measurements over the eastern North Atlantic stratocumulus have shown (Zheng et al., 2022b). A bulk relationship between above-cloud smoke and any cloud property therefore mixes aerosol effects and meteorological influences, and the mixture proportions are seasonal. Over the SEA, this confounding is structural, because the biomass-burning season coincides with a particular circulation pattern different from the background, so that a smoky-minus-clean composite over this region compares different weather as much as it compares different aerosols.

In this study, over a $10^\circ \times 10^\circ$ SEA domain centered on Ascension Island, we use the vertically resolved extinction product of the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP; Painemal et al., 2020) to classify, day by day, the placement of the aerosol relative to the Moderate Resolution Imaging Spectroradiometer (MODIS) cloud top into two aerosol–cloud configurations, aerosol separated from the cloud in the free troposphere (FT) and aerosol in contact with the cloud top (CT), with the aerosol residing within the boundary layer (MBL) treated as an additional stratifier of the droplet supply. This allows us to systematically investigate how the elevated aerosol influences the underlying cloud decks when it lies above the cloud and when it touches the cloud. In addition, we partition the meteorology of 2006–2014 into three prevailing weather regimes using a deep-learning clustering of ERA5 circulation fields, upgraded from the deep-learning-based clustering framework of our earlier work (Zheng et al., 2025) to an improved joint-optimization variant, the Improved Deep Embedded Clustering (IDEC; Sect. 2.4). Because the regimes are identified from the circulation alone, the stratification is independent of the aerosol and cloud fields whose relationships it is meant to examine. Within this framework of weather regimes and aerosol–cloud configurations we quantify the adjustment of cloud liquid water path (LWP) to droplet number, $L_0 = d\ln(\text{LWP})/d\ln(N_d)$, and evaluate its dependence on smoke against the aerosol climatology of each regime (Sect. 2.5).

The observations are then used to evaluate GCM simulations. We examine simulations with the Energy Exascale Earth System Model version 2 (E3SMv2) in a nudged 1-degree configuration, constrained by ERA5 winds and temperature and sampled at 13:00 UTC over the SEA, coincident with the satellite overpasses. After the introduction of the data and method in Sect. 2, we first analyze the satellite-observed aerosol–cloud relationships conditioned on weather regime



140 and aerosol–cloud configuration in Sect. 3, including the analyses that separate the smoke signal from its co-transported moisture and the influence of below-cloud aerosols. Section 4 examines the E3SMv2 simulations and discusses the model–observation comparison, and Sect. 5 summarizes the main conclusions with further directions.

145 2. Data and Method

2.1 Satellite retrievals of aerosol and cloud

The aerosol extinction coefficient (σ_{EXT}) used in this study is a research product derived from CALIOP aboard the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) platform (Winker et al., 2009). The retrievals are produced at a 1-km along-track resolution using the Fernald–Klett iterative approach (Klett, 1981; Fernald, 1984), constrained by an independent CALIOP-based AOD, and are limited to cloud-free pixels. A two-layer lidar-ratio version of the retrieval extends the product into the marine boundary layer and is used for the below-cloud aerosol (Painemal et al., 2020). The retrieved profiles have been evaluated against airborne high-spectral-resolution lidar (HSRL) measurements during the Caribbean 2010 field campaign and show good agreement. The detailed aerosol retrieval methodology and product evaluation are described in Painemal et al. (2019) and Li et al. (2022).

Cloud properties are from MODIS Aqua at 1 km resolution, processed with the CERES Edition 4.0 algorithms (Minnis et al., 2021). MODIS LWP agrees with ARM surface observations of marine boundary layer clouds to within about 10–15 % (Xi et al., 2014), and satellite LWP retrievals have been further validated against ship-based observations over the ocean (Painemal et al., 2016). N_d is derived with the adiabatic formulation, in which N_d scales as $\Gamma^{1/2} \tau^{1/2} r_e^{-5/2}$. The droplet effective radius (r_e) and cloud optical depth (τ) are retrieved from the 3.79 μm and 0.64 μm bands, respectively (Painemal et al., 2013, 2020); k , the ratio of volume-mean to effective radius, is set to 0.8 (Martin et al., 1994); and Γ is the adiabatic condensation rate computed from the MODIS cloud-top temperature and pressure (Albrecht et al., 1990; Painemal et al., 2020). This N_d retrieval compares well with in situ measurements from eleven aircraft campaigns over marine stratocumulus (Gryspeerdt et al., 2022).

Aerosol and cloud products are collocated as follows. Each CALIOP profile is spatially averaged to 5 km along track and matched to MODIS with 5-pixel $\times$ 5-pixel boxes, two east and two west of the CALIPSO ground track. Within each approximately 25 km along-track CALIOP segment, the MODIS boxes with a low-cloud fraction of at least 80 % are retained and averaged, and all retrievals in the segment are then averaged so that the aerosol and cloud fields are matched at comparable scales (Painemal et al., 2020; Li et al., 2026). The pairing places clear-sky extinction profiles immediately adjacent to nearby cloudy columns and enables a near-simultaneous assessment of aerosol–cloud interaction. Precipitation occurrence is from the CloudSat Cloud



Profiling Radar. Within each collocated segment, drizzle, light rain, and rain are flagged where the maximum reflectivity falls between -15 and -7 dBZ, between -7 and 0 dBZ, and above 0 dBZ, respectively (Haynes et al., 2009). The analysis domain spans roughly $10^\circ \times 10^\circ$ over the southeastern Atlantic, from 3 – 13° S and 10 – 20° W, with a local solar overpass time near 13:30. Hereafter, for brevity, we refer to all satellite-retrieved aerosol and cloud properties collectively as ‘satellite’ data. Valid cloud samples require $5 < N_d < 1000 \text{ cm}^{-3}$, $10 < \text{LWP} < 1000 \text{ g m}^{-2}$, and a segment low-cloud fraction above 0.1, applied uniformly to every analysis.

Column absorption is characterized with the CAMS EAC4 reanalysis (Inness et al., 2019). The speciated aerosol optical depths at 550 nm (black carbon, organic matter, dust, sea salt, and sulfate) are averaged over the analysis domain at 12:00 UTC daily, and the column absorption aerosol optical depth (AAOD) is reconstructed as the sum of the speciated optical depths weighted by one minus the single-scattering albedo of each species, with values consistent with the CAMS aerosol optics (0.25 for black carbon, 0.98 for organic matter, 0.96 for dust, and 1.0 for sea salt and sulfate; Bozzo et al., 2020). The column single-scattering albedo follows as $\text{SSA} = 1 - \text{AAOD}/\text{AOD}$. Note that the black-carbon single-scattering albedo is the least certain of these values, and varying it between 0.20 and 0.30 leaves the regime ordering of Sect. 3.1 unchanged (Sect. S1). Consistent with a surface comparison at Ascension Island, in which CAMS reproduces the burning-season evolution of carbon monoxide, black carbon, and sulfate but overestimates their boundary-layer mass concentrations (Tatro and Zuidema, 2026), the speciated optical depths are used here only for the relative, regime-wise contrast in column absorption.

2.2 E3SMv2 model simulations

E3SMv2 is a fully coupled Earth system model (Golaz et al., 2022); its atmospheric component EAMv2 (Rasch et al., 2019; Xie et al., 2018) uses a spectral-element dynamical core at approximately 110 km horizontal resolution with 72 vertical layers. Turbulence, shallow convection, and cloud macrophysics are handled by the Cloud Layers Unified by Binormals scheme (CLUBB; Larson, 2017), deep convection by the Zhang–McFarlane bulk mass-flux parameterization (ZM; Zhang and McFarlane, 1995) with a dilute convective available potential energy (CAPE) formulation (Neale et al., 2008) and a dynamic CAPE trigger (Xie et al., 2019), stratiform microphysics by the two-moment Morrison–Gettelman scheme (MG2; Gettelman and Morrison, 2015), and aerosols by the four-mode Modal Aerosol Module (MAM4; H. Wang et al., 2020). We run EAMv2 with the horizontal winds and temperature nudged toward ERA5 (Hersbach et al., 2020) at a 6-h relaxation time (Sun et al., 2019), the same nudged configuration as in Zheng et al. (2025), in which only the circulation and temperature are constrained and the humidity, cloud, and aerosol fields evolve freely. We archive hourly output for 2006–2014 over the same $10^\circ \times 10^\circ$ SEA domain (42 model columns) and sample it at 13:00 UTC.



Model cloud diagnostics were derived following the satellite definitions as closely as the model permits. MBL cloud samples are defined below 680 hPa, with a cloud fraction above 5 %
 215 flagging a valid cloud layer and the same threshold identifying the cloud-base height. To compare with the MODIS cloud-top height (CTH), the model cloud top is diagnosed from the inversion base (Z_{inv}), identified where the product of the vertical gradients of liquid-water potential temperature and relative humidity reaches its minimum (Zheng et al., 2025), a criterion that captures strong inversions despite the coarse vertical spacing near cloud top. The nudged
 220 EAMv2 boundary-layer inversion has been compared with LASIC radiosondes at Ascension Island, which it reproduces reasonably well while slightly overestimating inversion strength (Howes et al., 2026). The model LWP is the vertical integral of liquid water content between cloud base and cloud top, and the in-cloud N_d is the grid-mean cloud droplet number weighted by cloud fraction at each level. For the boundary-layer aerosol proxy we use the below-cloud
 225 aerosol extinction.

2.3 Aerosol–cloud configuration

We define two aerosol layers relative to the MODIS CTH of each 25 km segment, annotated in Fig. 2a: Contacting aerosol (CT) has the mean σ_{EXT} within ± 0.3 km of the cloud top, a 600 m
 230 layer centered on the cloud top, so that the sampled aerosol is in direct contact with the cloud-top region. Separated aerosol (FT) has the mean σ_{EXT} between 0.4 and 1.0 km above the cloud top, a 600 m layer offset from the cloud top by a 400 m gap. The gap places the sampled plume well above the cloud and, in most cases, above or near the top of the inversion layer, given the typical inversion thickness over the SEA (Zhang and Zuidema, 2021). Aerosol in this layer is therefore
 235 separated from the cloud and its influence must arrive through the thermodynamic environment.

For the E3SMv2 records, the aerosol-layer analogues are constructed from the diagnosed inversion base height Z_{inv} : contacting aerosol is the mean extinction within $Z_{\text{inv}} \pm 300$ m, and separated aerosol is the mean between $Z_{\text{inv}} + 400$ m and $Z_{\text{inv}} + 1000$ m, mirroring the satellite definitions. The below-cloud (MBL) aerosol, used as a stratifier in Sect. 3.6, is the mean
 240 extinction between 60 m above the surface and 60 m below the cloud top in both datasets. All aerosol amounts in this paper are layer means over one of these three bands, and no column-integrated extinction enters any analysis.

2.4 Meteorological regime clustering

245 We classify the daily 13:00 UTC circulation into recurrent weather regimes with a deep-learning clustering pipeline that couples a convolutional–recurrent autoencoder to the IDEC objective (Guo et al., 2017), hereafter referred to as the CNN–LSTM–IDEC classifier (CLIC). The classifier operates on four ERA5 fields, the 700 hPa geopotential height (Z_{700}), sea-level



pressure, and the 10 m wind components, as daily 13:00 UTC maps over an approximately $10^\circ \times$
250 10° clustering domain for 2006–2014, normalized per variable.

The pipeline proceeds in three stages. First, an autoencoder learns a compact representation of
the circulation. Each input sample consists of two consecutive days of the four fields, so that the
encoder receives both the instantaneous pattern and its day-to-day evolution; four stacked
convolutional stages extract spatial features from each day's maps, an LSTM layer fuses the two-
255 day sequence, and a dense bottleneck projects the result onto a 32-dimensional latent space, from
which a mirrored convolutional decoder reconstructs the input fields. Second, K-means
clustering of the standardized latent features provides the initial cluster centroids, with the
number of clusters chosen by comparing their silhouette scores (Rousseeuw, 1987). Third, a
clustering layer that computes soft regime memberships from a Student-t kernel over the latent
260 distances (Xie et al., 2016) is attached to the encoder, and the full network is retrained on the
joint IDEC loss, the reconstruction loss plus a Kullback–Leibler clustering loss toward a self-
sharpening target distribution, weighted by a clustering coefficient $\gamma = 0.1$. Each day is finally
assigned to the regime of its largest membership.

This framework upgrades the DEC-based classifier of our earlier work (Zheng et al., 2025) by
265 retaining the reconstruction term, which anchors the representation to the circulation fields
themselves while the cluster assignments sharpen. To assess how much the pipeline gains over
conventional clustering, we repeat the three-regime classification with three established methods
on the same input fields. The pipeline separates the regimes most cleanly, reaching a silhouette
score of 0.34, against 0.27 for K-means on the same autoencoder latent space, 0.15 for K-means
270 on the principal components of the raw circulation fields, and 0.13 for a self-organizing map
clustered from the same components, in which each method is scored in the feature space it
clusters in. The cluster count is set by the same score: $K = 3$ reaches 0.34, whereas $K = 4$ falls to
0.25, and its fourth cluster is a transition mixture drawn from the other three. We then label the
clustered regimes R1, R2, and R3 in order of increasing aerosol loading, and name them by the
275 circulation state established in Sect. 3.1: the weakened-high (R1), background (R2), and
strengthened-high (R3) phases of the subtropical high.

2.5 Cloud adjustment metric and statistical framework

The cloud-water adjustment $L_0 = d\ln(\text{LWP})/d\ln(N_d)$ is quantified as the slope of an ordinary
280 least-squares regression of $\ln(\text{LWP})$ on $\ln(N_d)$ over valid cloud samples (Gryspeerd et al., 2019).
Within each regime, the aerosol dependence of L_0 is evaluated by stratifying samples into
percentile groups of that regime's own FT or CT extinction distribution (below the 5th, below
the 25th, interquartile, above the 75th, and above the 95th percentile). The thresholds are applied
to the absolute extinction values of the 2006–2014 record, with no normalization or anomaly



285 transformation, so that the within-regime aerosol dependence is not confounded with the
 between-regime contrast.

The confidence intervals are computed with a cluster bootstrap that resamples days with
 replacement (5000 replicates), and all slopes are quoted with these day-bootstrap 95 % intervals.
 The aerosol dependence itself is tested with the trend statistic ΔL_0 , defined as the slope in the
 290 polluted group (above the 75th percentile) minus the slope in the clean group (below the 25th
 percentile), evaluated under joint day-resampling of the two groups. The resulting family of
 twelve trend tests (two datasets $\times$ two aerosol–cloud configurations $\times$ three regimes) is controlled
 with the Benjamini–Hochberg false discovery rate (Benjamini and Hochberg, 1995), and only
 trends whose p-values fall below 0.05 after this control are given mechanistic weight; all p-
 295 values quoted in this paper are the controlled values. The N_d susceptibility to an aerosol layer is
 quantified with the same method as $\beta = d\ln(N_d)/d\ln(\sigma_{EXT})$, evaluated for the FT, CT, or MBL
 layer (Sects. 3.5 and 3.6). Note that the subsets sampled on fewer than 30 distinct days are shown
 for completeness but carry no confidence interval and should be interpreted with caution.

300

3. Meteorological-regime-dependent cloud–aerosol characteristics from satellite

3.1 Weather regimes and their aerosol and cloud characteristics

Figure 1 presents the composite meteorology of the three identified regimes. Regime R1, the
 weakened-high regime (14 % of days), is the weakening phase of the subtropical high, in which
 305 the Z_{700} anomaly is uniformly negative, the absolute height contours are the lowest of the three
 columns, and the 700 hPa easterlies lose strength over the southern half of the domain. Beneath
 this weakened high, the boundary layer is anomalously warm and moist, with 925 hPa equivalent
 potential temperature (θ_{e925}) anomalies of several K above the record mean throughout the
 domain and southeasterly trades of ordinary strength. The lower-tropospheric stability (LTS;
 310 bottom row) is the weakest of the three regimes over most of the domain, and the 700 hPa
 relative humidity (RH_{700}) contours reach their highest values on the equatorward side, a
 comparatively moist free troposphere above a weakly capped boundary layer. R1 is thus a warm
 and humid configuration.

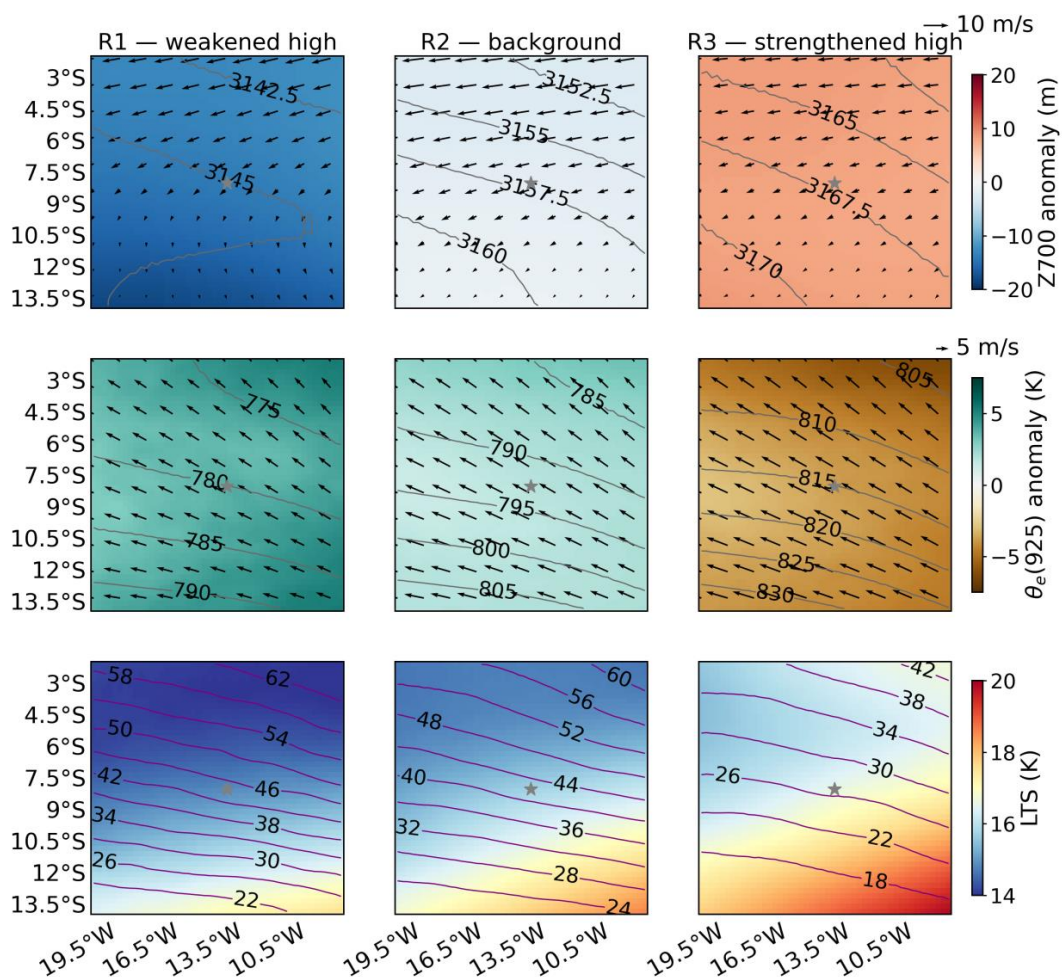


Figure 1. Composite meteorological fields of the three weather regimes (columns: R1, 14 % of days; R2, 49 %; R3, 37 %). Top row: 700 hPa geopotential height anomaly (shading, m) with the regime-mean 700 hPa winds (vectors) and absolute geopotential height (contours, m). Middle row: 925 hPa equivalent potential temperature anomaly (shading, K) with the regime-mean 925 hPa winds (vectors) and 925 hPa geopotential height (contours, m). Bottom row: lower-tropospheric stability (LTS; shading, K) with 700 hPa relative humidity (contours, %). Anomalies are computed relative to the 2006–2014 all-days mean at 13:00 UTC. The grey star marks Ascension Island.



325 Regime R3 (Fig. 1, right column), the strengthened-high regime (37 % of days), is the
strengthening phase of the same high. Its Z_{700} anomaly is positive everywhere, with the highest
mid-level heights in the record, and the 925 hPa panel shows a strengthened St. Helena High
driving intensified southeasterly trades across the domain. The θ_{e925} anomaly is correspondingly
negative by several K, marking a cool, dry boundary layer under enhanced anticyclonic
330 influence. In the bottom row, the two cloud-controlling extremes coincide. LTS reaches its
maximum, most prominently over the southeastern half of the domain, while the RH_{700} contours
drop to the lowest values across the regimes, representing the driest free troposphere in the
record. This is the canonical dry-season configuration of the SEA, a reinforced subtropical high
with strong subsidence and a sharp capping inversion (Ryoo et al., 2021), and it overlaps with
335 the biomass-burning export season (Redemann et al., 2021; Zhang and Zuidema, 2021).

Regime R2 (49 % of days; Fig. 1, middle column), the background regime, is the near-
climatological circulation state and, by a wide margin, the most common one. Its Z_{700} and θ_{e925}
anomalies are close to zero, with its absolute height field, trade-wind strength, stability, and free-
tropospheric humidity all falling between the two extremes, and it is the most frequent regime in
340 most months of the year. The three regimes are thus three states of one circulation system, as the
height pattern of the subtropical high is common to every column of Fig. 1, and only its level and
the strength of the trade winds change, with R1 the weakened phase, R3 the strengthened phase,
and R2 the mean state between them.

Figure 2 characterizes aerosol properties of each regime, and Fig. 2a illustrates the aerosol–cloud
345 configurations used for the rest of the paper. Anchored to the mean cloud top (dotted line), Fig.
2a draws the CT, FT, and MBL bands of Sect. 2.3, and every aerosol-stratified statistic in this
paper refers to one of these three bands. The median CALIOP extinction profiles of each regime
share a common shape. Extinction is largest within the boundary layer, decreases toward the
mean cloud top, and then recovers into an elevated aerosol layer whose maximum lies above the
350 mean cloud top in every regime. Within the CT band, the three regimes keep the order $R3 > R2 >$
 $R1$ but lie within 0.01 km^{-1} of one another (median-profile means of 0.039, 0.035, and 0.031
 km^{-1}). The aerosol in contact with the cloud top therefore has similar extinction characteristics
across the regimes, while its absorption and composition do not, as the column absorption in Fig.
2c and the aerosol attribution later in this section show. Above the CT band the regimes separate
355 in the amount and depth of the elevated aerosol. R3 carries the highest extinction in the FT band
and an elevated layer that remains substantial deep into the free troposphere, with a second
maximum near 4 km (0.035 km^{-1} at 4.1 km, against $0.025\text{--}0.028 \text{ km}^{-1}$ in the FT band) and the
widest interquartile spread of the record. This upper part of the R3 plume reaches Ascension still
near the altitude at which it left the continent (Adebiyi and Zuidema, 2016). It lies above the FT
360 band and shows no stratification in this paper, whereas the FT band samples the lower part of the
plume that has subsided toward the inversion. The R1 and R2 profiles, in contrast, decay
monotonically to weaker values above the CT band.

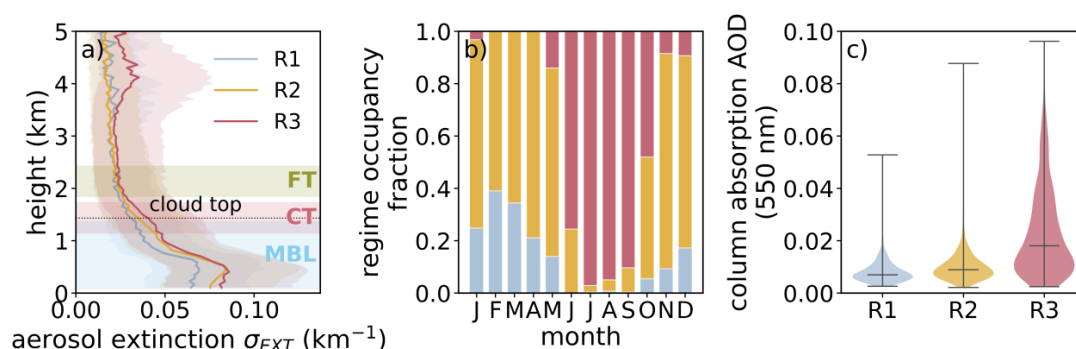


Figure 2. Aerosol characteristics of the regimes and the aerosol–cloud configurations. (a) Median CALIOP aerosol extinction coefficient (σ_{EXT}) profiles per regime (shading: interquartile range), with the separated-aerosol (FT), contacting-aerosol (CT), and boundary-layer (MBL) layers drawn relative to the mean cloud-top height (dotted line), in which CT spans ± 0.3 km of cloud top, FT spans 0.4–1.0 km above it, and MBL spans 60 m above the surface to 60 m below cloud top. (b) Monthly regime occupancy (fraction of days per calendar month assigned to each regime). (c) Distributions of the column absorption aerosol optical depth (AAOD, 550 nm) per regime, derived from the CAMS EAC4 speciated aerosol optical depths; horizontal bars denote medians.

Of the 828 June–August days in the record, 740 are classified R3, and the red bars show that R3 remains dominant through September before receding in the transition months (Fig. 2b); R1 appears almost exclusively from January through May, and R2 occupies the bulk of the remaining months.

Figure 2c shows the column AAOD derived from the CAMS EAC4 speciated optical depths (Sect. 2.1). The R3 median of 0.018 compares with 0.009 in R2 and 0.007 in R1, and the corresponding column single-scattering albedo falls to 0.906 in R3 against 0.94 in the other two regimes (Fig. S1c), so the R3 aerosol column is by far the most absorbing of the record, the optical signature of black-carbon-rich biomass-burning smoke (Redemann et al., 2021). The column aerosol lidar ratio of the CALIPSO Synergized Optical Depth of Aerosols product (CALIPSO–SODA) is type-diagnostic at 532 nm (Li et al., 2022), and in our record it rises, within every regime, from about 29 sr on the cleanest separated-aerosol days, the value of a marine-dominated column, to 58 sr in R3’s smokiest quartile, approaching the characteristic biomass-burning range. Because the column value averages the marine boundary layer into the retrieval, the elevated layer itself must sit higher still (Fig. S2c, d). An independent view of the aerosol above the clouds comes from the CALIOP depolarization-ratio retrieval over opaque



water clouds (Hu et al., 2007; Jethva et al., 2026), which involves no aerosol lidar-ratio assumption and samples the over-cloud columns directly (Sect. S2). In that record, the retrieved above-cloud optical depth is highest in R3 (median 0.39, against 0.30–0.32 in R1 and R2), and the extinction Ångström exponent increases from 0.97 in R1 to 1.47 in R3 (Fig. S2a, b), a fine-particle signature. The CAMS EAC4 composition agrees, in which carbonaceous aerosol (black carbon plus organic matter) accounts for a median 69 % of the column aerosol optical depth in R3, against 57–60 % in R1 and R2, with dust contributing only 2 % in R3, and within R3 the carbonaceous fraction climbs from 59 % on the cleanest separated-aerosol days to 83 % on the smokiest (Sect. S1).

Of these measures, only the over-cloud retrieval samples the aerosol above the clouds directly, and its above-cloud optical depth and Ångström exponent identify a fine-particle layer that is thickest in R3. Despite being column-integrated, the column absorption, the column lidar ratio, and the CAMS carbonaceous fraction all rise with the separated-aerosol percentile within every regime, and the AAOD rises from 0.010 in the clean quartile of R3 to 0.040 in its polluted quartile, with the other two as shown in Figs. S1b and S2d. Because those percentile groups are defined by the extinction of the separated layer alone, the rise is consistent with an absorbing, carbonaceous elevated layer. Taken together, the evidence indicates that the above-cloud aerosol is a biomass-burning smoke-enriched plume. This attribution is strongest for R3, while the upper tails of R1 and R2 also reach comparable carbonaceous fractions on their most polluted days (Fig. S1b). We therefore refer to the stratifying layers by the neutral term aerosol throughout the configuration analysis and reserve smoke for R3, where the attribution is unambiguous. Taken together, the regime index orders three environmental properties at once, from clean to polluted, from moist to dry aloft, and from weakly to strongly capped. The adjustment signals below are evaluated within those environments.

415

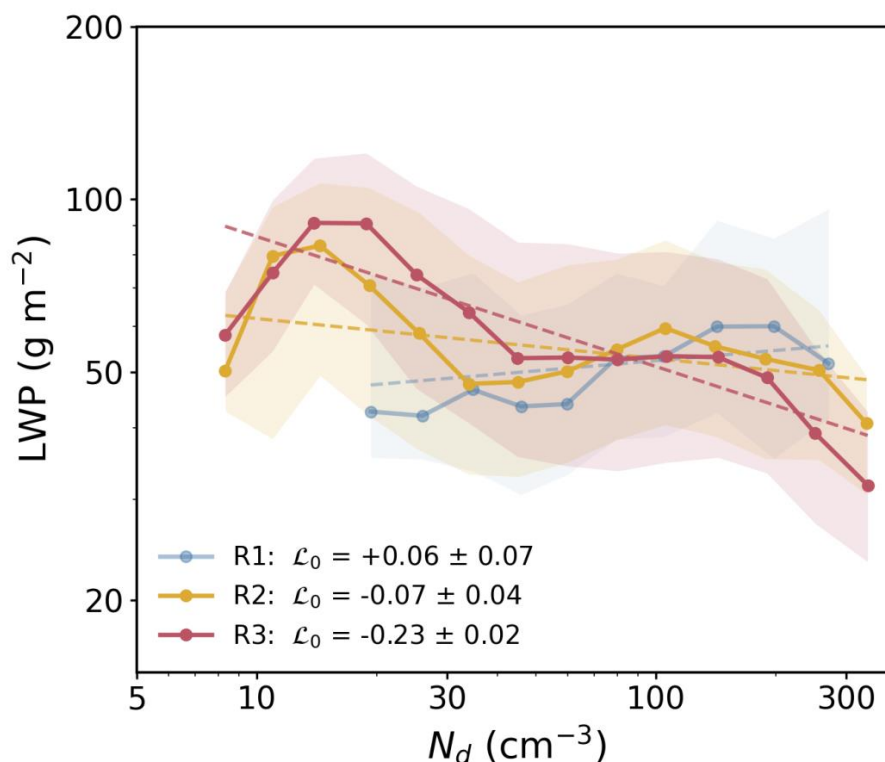
3.2 Cloud LWP responses to N_d under different regimes

Figure 3 shows the baseline LWP– N_d relation of each regime, before any aerosol stratification. The R2 and R3 curves share a similar shape in which LWP first rises with N_d in the low-droplet-number range below roughly 15 cm^{-3} , peaks, and declines beyond it. The R3 curve descends most deeply from its maximum and reaches the lowest median LWP of the figure at the largest N_d , the R2 curve follows a shallower descent, and the shorter R1 curve, which occupies mostly the moderate droplet numbers, drifts weakly upward instead. Over the full range, the whole-regime slopes separate cleanly, with $L_0 = +0.06 \pm 0.07$ in R1, -0.07 ± 0.04 in R2, and -0.23 ± 0.02 in R3, and the R1 interval spanning zero. All three pairwise differences are robust under the day bootstrap (Fig. 3). In other words, for the same fractional increase in droplet number, cloud water declines fastest in the strengthened-high regime, more gently in the background regime, and shows no detectable decline in the weakened-high regime. Note that the R2 slope of -0.07 ,

425



though separable from both neighbors, is itself near zero, an average of aerosol-stratified slopes of both signs (Fig. 4b).



430

Figure 3. LWP– N_d relationship in regimes R1 (blue), R2 (yellow) and R3 (red). Curves are binned medians of LWP in N_d bins per regime (shading: interquartile range). The legend gives the whole-regime adjustment $L_0 = d\ln(\text{LWP})/d\ln(N_d)$ with 95 % day-bootstrap half-widths, and the dashed lines are the fitted regressions of L_0 whose slopes are the legend values. All pairwise regime differences in L_0 are statistically significant under joint day-resampling.

435

This inverted-V shape is the well-documented non-monotonic LWP adjustment of the satellite record (Grypsperdt et al., 2019). In the scavenging-limited range, rising N_d suppresses the precipitation sink and LWP accumulates (Albrecht, 1989). Beyond the maximum, smaller droplets evaporate more readily in entrained air and sediment less efficiently out of the cloud-top region, so the entrainment–evaporation pathway takes over and LWP falls as N_d rises (Ackerman et al., 2004; Bretherton et al., 2007). Li et al. (2026) hypothesized that the inverted-V pattern in

440



records combined across regions partly reflects mixing samples from different cloud states. The regime decomposition supports that reading over the SEA. The regime-dependent slopes are single-signed (+0.06, −0.07, −0.23; Fig. 3) and each integrates across the two branches, with the positive R1 slope occurring at the low end of the LWP range, consistent with that study.

The regime ordering of Fig. 3 also follows the meteorological environments of Fig. 1. The strengthened-high regime combines the strongest inversion with the driest free troposphere, so the air entrained at cloud top is farthest from saturation and each entrainment event evaporates the most cloud water. The declining branch is steepest and longest there, and the whole-regime slope, which integrates across both branches, is −0.23. In the weakened-high regime, in contrast, the moist free troposphere delivers entrained air close to saturation and the evaporative effect stays small (Ackerman et al., 2004), so the declining branch is muted and the weakly positive, statistically zero slope is consistent with the precipitation-suppression branch of the inverted-V shape remaining visible (Albrecht, 1989). R2, with moderate stability and humidity, falls between the two. This ordering emerges from the cloud-controlling environment of each regime, before any aerosol stratification, in line with the environmental control of the aerosol–cloud coupling reported for the eastern North Atlantic stratocumulus (Zheng et al., 2022b), and sets the baseline against which the aerosol-stratified signals in Sect. 3.3 are read.

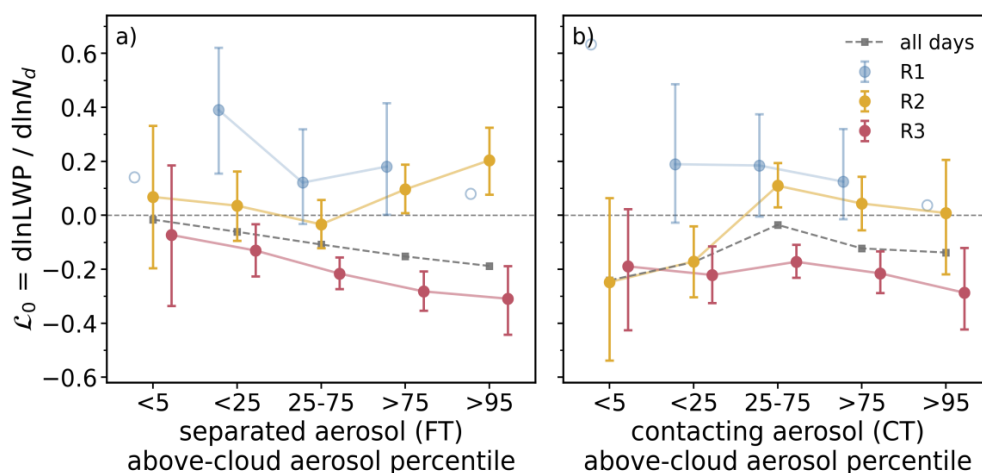


Figure 4. LWP– N_d adjustment L_0 versus above-cloud aerosol amount within each regime, for (a) separated aerosol (FT) and (b) contacting aerosol (CT). Percentile groups (below the 5th, below the 25th, interquartile, above the 75th, above the 95th) are defined within each regime’s own extinction distribution. Whiskers are 95 % day-bootstrap confidence intervals; open regime-colored markers denote groups sampled on fewer than 30 distinct days, which carry no interval and are interpreted with caution; the grey dashed line is the all-days (bulk) statistic computed without regime stratification.



470 3.3 Cloud responses to elevated aerosols in the separated and contacting configurations

We recompute the regime-specific adjustment L_0 for the samples in the five aerosol-percentile groups of Sect. 2.5, independently for the separated aerosol (Fig. 4a) and the contacting aerosol (Fig. 4b). In the separated-aerosol panel, R3 stands apart from the other regimes. Its series descends across the entire percentile range, from -0.07 in the cleanest twentieth of days through
 475 -0.22 in the interquartile range to -0.31 above the 95th percentile. The trend is statistically significant ($p = 0.037$). The LWP adjustment is nearly absent on the cleanest R3 days. Increasing droplet number leaves cloud water largely unchanged ($L_0 = -0.07$). As the separated aerosol layer above the cloud thickens optically, the fractional loss of cloud water increases progressively. Neither R1 nor R2 shows a similar dependence. R1's series is positive, with wide,
 480 overlapping intervals and no ordered progression, and both tail groups are open markers indicating limited samples. R2 hovers near zero across the percentile groups and drifts weakly above zero toward its most polluted end. The steepening is therefore specific to R3, which represents the most negative baseline LWP adjustment.

R3's baseline environment already predisposes both aerosol–cloud pairing configurations to
 485 water loss through the dry-entrainment base state of Sect. 3.2, which underlies the negative L_0 across most aerosol percentiles in the separated and the contacting series alike. The separated aerosol layer adds a radiative strengthening of the inversion. The layer typically sits a few hundred meters above the inversion (0.4 – 1.0 km above the cloud top; Fig. 2a), so its shortwave absorption warms the free troposphere above the inversion and sharpens the potential-
 490 temperature jump across it (Johnson et al., 2004; Wilcox, 2012), a heating effect whose net cloud response depends on the aerosol vertical placement relative to the inversion (Sect. 1). Under a sharper cap the entrainment–evaporation exchange concentrates at the interface (de Lozar and Mellado, 2015; Mellado, 2017), where higher droplet numbers correspond to smaller droplets that evaporate faster in the mixed filaments and sediment less out of the entrainment zone. The
 495 coupling between droplet number and water loss therefore tightens as the layer thickens, which is what the steepening negative L_0 measures. This reading is consistent with large-eddy studies of absorbing layers above stratocumulus, in which above-inversion heating alone strengthens the entrainment-driven thinning without any drying (Zhou et al., 2017; Herbert et al., 2020).

In R3, the cloud LWP adjustment under contacting aerosol (Fig. 4b) is negative in every
 500 percentile group, near -0.2 through the interquartile range, and edges from -0.19 in the cleanest twentieth to -0.29 in the smokiest, a mild steepening far weaker than the separated-layer trend in Fig. 4a. Here the direct heating by absorbing aerosol in contact with the cloud top, a semi-direct effect examined in Sect. 3.5, adds to the same dry base state, so the response stays negative regardless of the aerosol loading. In R2, by contrast, increasing contacting aerosol is
 505 accompanied by an orderly relaxation of the adjustment, from -0.17 in the clean quartile to



+0.04, statistically indistinguishable from zero, in the polluted quartile ($p = 0.026$). Whatever coupling existed between cloud water and droplet number on R2's clean days is absent on its polluted ones. We attribute this contrast to the humidity of the air in contact with the cloud top and to the droplet supply from the contacting layer, both of which differ sharply between the two regimes, and we examine both in Sect. 3.5. R1 drifts in the same direction as R2, from +0.19 in its clean quartile to +0.12 in its polluted quartile, but with confidence intervals that include zero and undersampled tail groups (open markers), so the comparison is drawn between R2 and R3.

The grey bulk curves in both panels show the LWP adjustment for the all-days sample. For separated aerosol (FT), the all-days dependence is real but diluted. The all-days series falls from −0.02 to −0.19 across the percentile range, reaching barely 60 % of R3's amplitude at the smoky end, because the all-days sample mixes R3 days with R1 and R2 days that do not share the steepening (Fig. S3). For contacting aerosol (CT), the bulk curve is non-monotonic and averages R2's relaxation against R3's flat negative response, opposite-signed behaviors that cancel each other. Separating the regimes therefore isolates the strengthened-high regime, which carries the record's strongest adjustment and its only separated-aerosol dependence, from an all-days sample in which the background regime alone supplies 49 % of the days.

3.4 Separating the aerosol signal from the co-transported moisture

The L_0 steepening in Fig. 4a mixes the influence of the aerosol with that of its accompanying moisture, because separated aerosol arrives embedded in plume air with its own thermodynamic properties (Adebisi et al., 2015; Adebisi and Zuidema, 2018). Figures 5a and 5b show the 700 hPa specific humidity of the aerosol percentile groups, per regime, for the separated and the contacting layers respectively, with day-bootstrap intervals. Both layers are shown because the co-transported moisture bears on both configurations. Figure 5a shows the separated-aerosol analysis of this section, and Fig. 5b, the humidity of the contact air, is read in Sect. 3.5. A companion check at 850 hPa, closely above the median cloud top, is shown in Fig. S4. In every regime and both aerosol–cloud configurations, q_{700} rises with the aerosol percentile. In R3 the rise is steepest, from 1.7 g kg^{−1} on the cleanest separated-aerosol days to 3.1 g kg^{−1} on the smokiest (Fig. 5a), consistent with the climatological covariability reported by Zhang and Zuidema (2021). The biomass-burning plume is moister than the free troposphere it displaces, so within each regime the smokier days are also the moister days, even though R3 itself remains the driest regime at every aerosol amount compared to R1 and R2. The contact layer behaves the same way (Fig. 5b). The regimes differ in the humidity level itself, with far moister contact air in R2 than in R3 at every aerosol amount, a contrast that Sect. 3.5 attributes to the drier subsiding air that renews the contact layer in R3.

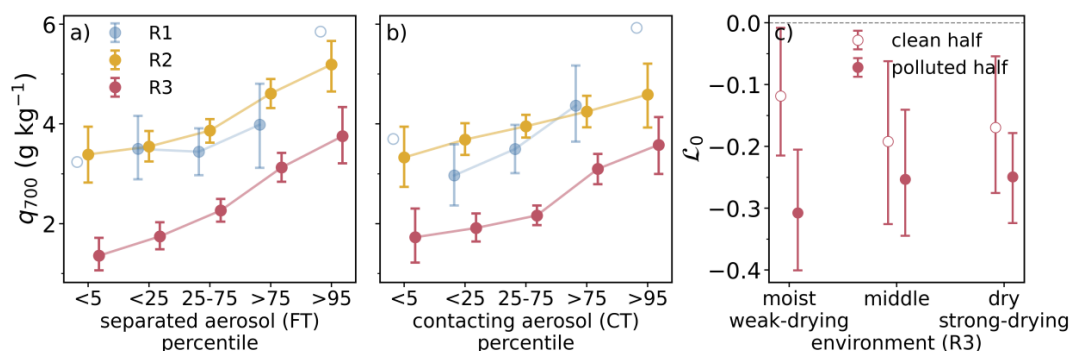


Figure 5. The moisture co-transported with the aerosol. (a, b) 700 hPa specific humidity (q_{700}) versus (a) separated-aerosol and (b) contacting-aerosol percentile group per regime (means with 95 % day-bootstrap intervals). (c) L_0 in the clean and polluted halves of the separated-aerosol distribution within three equal groups of the entrainment-drying index ($\text{EDI} = (1 - \text{RH}_{700}/100) \times \text{LTS}$), for R3 (other regimes in Fig. S5).

A moister free troposphere reduces the evaporative effect of entrainment (Ackerman et al., 2004). Entrained parcels arrive closer to saturation and evaporate less cloud water per mixing event, so the co-transported moisture moves the adjustment toward zero. This offsetting effect contributes to the muted responses of R1 and R2, whose aerosol-stratified slopes hover near or above zero, especially toward their most polluted tails (Fig. 4). In R3, the offset is weakest and the negative response stands. In general, Fig. 5 shows that the moisture traveling with the aerosol opposes the steepening, which still emerges despite the compensating effect. This offset applies to both aerosol layers, since q_{700} rises with the aerosol percentile under the separated and the contacting configurations alike (Fig. 5a, b). The remainder of this section isolates the separated-aerosol steepening of Fig. 4a from that of moisture.

To approximate the effects of entrainment drying, we further define an entrainment-drying index, $\text{EDI} = (1 - \text{RH}_{700}/100) \times \text{LTS}$, a stratifier designed for this analysis. The first factor is the subsaturation of the air available for entrainment, which sets the evaporative water loss per mixing event, and the LTS factor gauges the strength of the capping inversion under which that exchange operates. High EDI thus marks days on which the entrained air is far from saturation beneath a strong cap. Splitting R3 into three equal EDI groups and comparing the clean and polluted halves of the separated-aerosol distribution within each (Fig. 5c), the clean-to-polluted steepening is present in every group and is strongest and statistically significant in the moistest, weakest-drying one, where L_0 falls from -0.12 to -0.31 ($p = 0.005$). The middle and dry groups show the same clean-to-polluted ordering with overlapping intervals. The strongest response



appearing in the weakest-drying group is consistent with both the weak baseline coupling there, which leaves the most room for the inversion sharpening to act, and the stronger aerosol contrast that group samples, since plume moisture and smoke arrive together. Process simulations that carry the plume moisture alongside its absorption reach the same configuration, with the co-transported moisture sustaining the cloud and offsetting part of the absorption-driven loss (Yamaguchi et al., 2015; Baró Pérez et al., 2024). That said, co-transported moisture alone cannot explain the steepening. The aerosol itself also contributes, since shortwave absorption in the elevated layer strengthens the inversion (Ackerman et al., 2000; Johnson et al., 2004). The same holds for the contacting layer in R3, where the negative response of Fig. 4b persists, with its mild steepening, although the contact air moistens with the aerosol (Fig. 5b), whereas in R2 the moistening prevails and the coupling relaxes.

3.5 Cloud thinning and decoupling under cloud-contacting aerosols

In this section we further investigate the contacting-aerosol responses, because Sect. 3.3 left the R2–R3 contrast unexplained, and because R2 and R3 carry comparable contacting-aerosol extinction near the cloud top (Fig. 2a), so the contrast must arise from what that aerosol does to the droplets and from the air in which the contact occurs. Figure 6 shows two diagnostics. Figure 6a gives the N_d susceptibility $\beta = d\ln(N_d)/d\ln(\sigma_{EXT})$, computed within each regime against both elevated layers, the contacting layer (filled diamonds) as the subject of this section and the separated layer (open diamonds) as a control for the steepening of Sect. 3.3. For separated aerosol (FT), the susceptibility is statistically indistinguishable from zero in R1 and R3 and weakly negative in R2; in R3, the regime in which the L_0 steepening under the separated-aerosol configuration is strongest (Fig. 4a), the estimate is 0.03 (95 % CI -0.05 to 0.10). This null shows, first, that the separated-aerosol L_0 steepening is not an artifact of aerosol– N_d collinearity, since the amount of overlying separated aerosol does not covary with droplet number within a regime, and the slopes of Fig. 4a genuinely describe how cloud water responds per unit change in N_d . The N_d variance itself grows toward the polluted groups, so dynamic-range compression is excluded as well. Second, the separated aerosol does not appear to affect the cloud microphysics directly, consistent with aircraft cases over the eastern North Atlantic in which a plume separated from the cloud top left the droplets unchanged (Y. Wang et al., 2020). Whatever the elevated layer does to the cloud, it does by changing the strength of the LWP– N_d coupling, the slope L_0 itself.

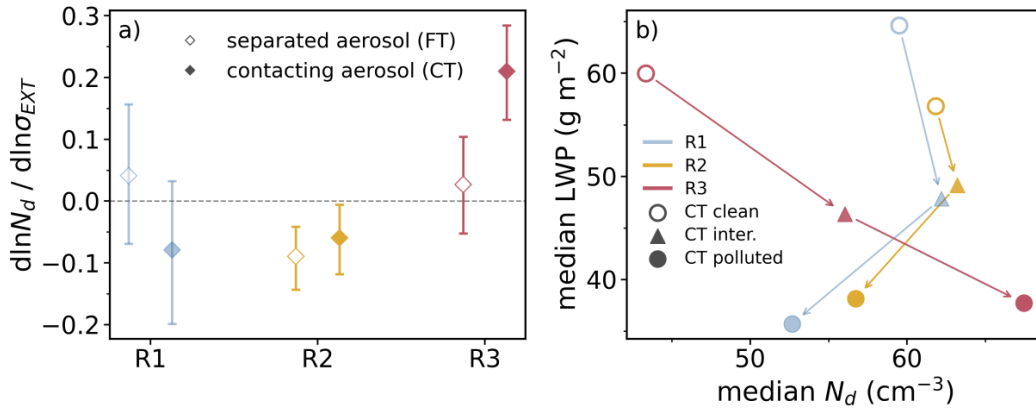


Figure 6. Cloud responses under contacting aerosol. (a) The N_d susceptibility to the elevated aerosol, $\beta = d\ln(N_d)/d\ln(\sigma_{EXT})$, per regime, computed against the layer-mean extinction of the separated (FT; open diamonds) and contacting (CT; filled diamonds) layers, with 95 % day-bootstrap intervals. (b) Median cloud state in N_d -LWP space across contacting-aerosol groups (arrows point clean $\rightarrow$ polluted). 'CT clean', 'CT inter.', and 'CT polluted' denote the clean quartile, interquartile range, and polluted quartile of each regime's contacting-aerosol distribution, drawn as open circles, triangles, and filled circles.

In R3, the contacting layer is associated with a clear droplet-number enhancement (Fig. 6a), $\beta = +0.21$ (95 % CI 0.13–0.28), consistent with the droplet-number enhancement by entrainment of the contacting aerosol sampled by ORACLES aircraft when the smoke layer touched the stratocumulus top (Diamond et al., 2018; Gupta et al., 2021). In R2, the corresponding susceptibility is slightly negative (-0.06), where the contacting aerosol modestly changes the droplet number. The R2 relaxation therefore cannot come from aerosol-driven N_d variability and may originate on the water side of the coupling, which we examine in Fig. 6b.

Figure 6b then traces the cloud state itself. It follows the median position in N_d -LWP space as the contacting-aerosol amount increases from the clean quartile to the polluted quartile. Both major regimes share the first-order feature. The cloud thins, with median LWP falling from 57 to 38 g m^{-2} in R2 and from 60 to 38 g m^{-2} in R3. Thinning under contacting aerosol is thus universal, consistent with shortwave heating by absorbing aerosol evaporating cloud water at the cloud top (the semi-direct effect; Ackerman et al., 2000; Zhang and Zuidema, 2021), the effect most visible at midday, with the longwave cloud-top warming by the overlying layer a smaller day-and-night contributor (Pandey and Adebisi, 2026). The regimes differ in how the cloud state evolves. In R2, LWP falls sharply while N_d decreases only slightly, and the day-to-day spread of



LWP narrows at the same time. With water loss no longer scaling with droplet number, the regression of $\ln(\text{LWP})$ on $\ln(N_d)$ weakens toward zero, as L_0 relaxes from -0.17 to statistical zero. The physical reason water loss decouples from N_d , the humidity of the contact air, is discussed below. In R3, median N_d rises from 43 to 67 cm^{-3} (the enhancement of Fig. 6a) while LWP falls, so droplets and water continue to move in opposite senses at every contacting-aerosol loading, and the slope stays negative throughout.

The L_0 relaxation in R2 is not seasonal. It survives when the biomass-burning months are excluded entirely ($\Delta L_0 = +0.19$, $p = 0.026$ without JJA) and re-emerges within December–May alone ($\Delta L_0 = +0.26$, $p = 0.012$), with near-identical month composition in the clean and polluted contact groups. Two conditions distinguish R2: the humidity of the air in contact with the cloud top, and the weakness of the droplet supply from the contacting layer ($\beta = -0.06$, against $+0.21$ in R3; Fig. 6a). The first is the humidity of the contact air. In R2, the air in contact with the cloud top is humid and becomes more humid as it becomes more polluted, with q_{700} rising from 3.7 to 4.3 g kg^{-1} across the contact percentiles (Fig. 5b). R3's contact air, in contrast, spans 1.9–3.1 g kg^{-1} , and even at its smokiest it remains drier than R2 at its cleanest. The same ordering holds at 850 hPa. The two humidity levels vary nearly independently from day to day, and in R3 they are weakly anticorrelated (Fig. S4d), since the plume moisture lies above the inversion while the contact layer below it is continually renewed by subsiding free-tropospheric air, consistent with the stronger subsidence on smokier R3 days limiting the moistening of the near-cloud layer even as the plume above moistens. Humid contact air weakens the evaporative pathway at its first step, so the water loss becomes insensitive to the droplet number (Ackerman et al., 2004; de Lozar and Mellado, 2015). Gupta et al. (2021) documented the same configuration from aircraft, with moister air immediately above the cloud in contact cases and a reduced evaporative effect of entrainment.

In contrast, the negative L_0 in R3 is sustained by the evaporative pathway, in which the contact air stays dry. The second condition is the droplet supply. In R2, the contacting aerosol covaries only weakly with N_d (Fig. 6a), so as the evaporative pathway weakens, there is little N_d replenishment from the elevated aerosol to re-impose an aerosol-driven anticorrelation. In contrast, in R3, the elevated aerosol keeps adding droplets while the cloud thins, so LWP and N_d keep moving in opposite senses and the negative slope persists. R1 drifts the same way as R2 (from $+0.19$ toward $+0.04$), consistent with the same decoupling, but its contact groups are sampled on too few days for any statistically significant statement.

3.6 Influences of below-cloud aerosols

Beyond the two aerosol–cloud configurations, the aerosol residing below the cloud enters our analysis as a stratifier that organizes the adjustment. Figure 7c shows the N_d susceptibility to the below-cloud aerosol, $\beta(\text{MBL}) = d\ln N_d / d\ln \sigma_{\text{EXT}}(\text{MBL})$, climbing across the regimes, from 0.12 in



R1 through 0.26 in R2 to 0.63 in R3. In the weakened-high regime, the droplet number is only weakly tied to the boundary-layer aerosol, whereas in the strengthened-high regime the below-cloud aerosol is by far the dominant control on droplet supply. This ordering is consistent with previous understanding of the smoke-influenced marine boundary layer at Ascension, where the burning-season aerosol reaches below cloud and governs activation (Zuidema et al., 2018; de Graaf et al., 2023; Dedrick et al., 2024), and with aircraft measurements over the eastern North Atlantic and the Southern Ocean in which aerosol reaching the cloud base activates readily into droplets (Zheng et al., 2024). Note that a steeper $\beta(\text{MBL})$ is compatible with the competition-limited activation reported at the same site, since competition lowers the fraction of particles that activate, whereas the regime ordering here reflects which aerosol layer supplies the droplets. From ORACLES lidar retrievals over the same stratocumulus deck, Lenhardt et al. (2026) likewise found that the cloud response to above-cloud CCN weakens as the environment stabilizes and that below-cloud aerosol dominates the droplet supply under strong inversions, in agreement with the strengthened-high regime here, in which $\beta(\text{MBL})$ reaches 0.63 while the separated-aerosol susceptibility is indistinguishable from zero.

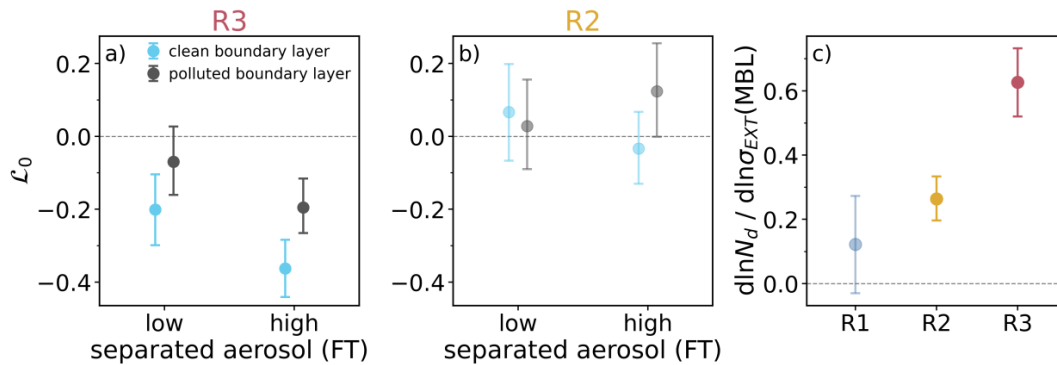


Figure 7. Influence of below-cloud aerosol. (a, b) L_0 in the 2×2 space of below-cloud aerosol (clean/polluted halves of the MBL extinction) and separated-aerosol amount (low/high halves), for R3 and R2. (c) The N_d susceptibility to the below-cloud aerosol, $\beta(\text{MBL}) = d\ln N_d / d\ln \sigma_{\text{EXT}}(\text{MBL})$, per regime.

Figures 7a and 7b set this below-cloud axis against the separated-aerosol axis, showing the $LWP-N_d$ L_0 grouped by below-cloud aerosol (clean and polluted halves of the MBL extinction) and overlying separated aerosol (low and high halves), for R3 and R2 respectively. The R3 matrix (Fig. 7a) shows a clear organization along both axes, and the two axes organize the adjustment in opposite senses. At fixed below-cloud aerosol, more overlying aerosol steepens the



adjustment, and the most negative cell in the record, $L_0 = -0.36$, occurs under heavy overlying aerosol above a clean boundary layer. At fixed overlying aerosol, a more polluted boundary layer weakens the N_d -based slope by 0.13–0.17, and the least negative cell (-0.07) occurs under low overlying aerosol above a polluted boundary layer. The R2 matrix (Fig. 7b) shows no comparable organization, its four cells straddle zero with overlapping confidence intervals, so the aerosol axes organize the adjustment only where the baseline coupling is strong.

Where below-cloud aerosol supplies the droplets, as the $\beta(\text{MBL})$ of 0.63 indicates in R3, fluctuations in N_d increasingly track fluctuations in the aerosol, and the droplet number functions partly as a below-cloud aerosol proxy. Aerosol-driven N_d variability carries no LWP signal beyond the adjustment it transmits, so it dilutes the cloud-process variability that generates the negative coupling and the fitted slope is buffered toward zero. This buffering does not affect the separated-aerosol axis. The steepening of Fig. 7a appears at fixed below-cloud aerosol, and since the buffering can only push slopes toward zero, the R3 whole-regime adjustment and its aerosol dependence are understated. The LWP– N_d responses suffer less dilution under the clean-boundary-layer condition (Fig. 7a), hence the adjustment under heavy overlying aerosol reaches -0.36 , steeper than the -0.31 at the smoky end of Fig. 4a.

The satellite snapshot itself also leans toward the buffered side of the coupling. Activation adjusts the droplet number within minutes of an aerosol perturbation, whereas large-eddy ensembles of non-precipitating stratocumulus place the LWP adjustment on a timescale of roughly 20 h, approaching a steady state near -0.64 (Glassmeier et al., 2021). A 13:00 UTC snapshot therefore records the fast N_d response in full while the slower water response is still developing, and yields a weaker slope by construction. Satellite records combined across regions and seasons yield still weaker slopes, with natural-experiment constraints placing the declining branch of the combined LWP– N_d relation near -0.15 against about -0.4 in raw regressions (Gryspeerdt et al., 2019). The regime-resolved values here, -0.31 under heavy separated aerosol and -0.36 with the below-cloud supply held clean, thus reach the steep end of the observational span while remaining understated of the R3 coupling, given the buffering above and the timescale mismatch.

4. Cloud–aerosol relationships over SEA in E3SMv2

4.1 Regime-based evaluation of the simulated cloud-water adjustment

The nudged E3SMv2 cloud and aerosol outputs are diagnosed with the same regime labels and aerosol–cloud configurations, to examine the simulated cloud-water adjustment under similar weather and smoke seasons and to compare its dependence on the aerosol vertical distribution with the observations. The nudging (Sect. 2.2; Zheng et al., 2025) largely limits the large-scale circulation as a model degree of freedom, a configuration in which E3SMv2 has been shown to



track the observed thermodynamic evolution of the marine boundary layer (Choi et al., 2025), so that the regime labels of Sect. 2.4 can be applied to the model columns. Model aerosol extinction enters these diagnostics as an average over the same vertical layers used to derive the satellite predictor (Sect. 2.3), and the below-cloud CCN concentration from the model provides an additional activation-explicit predictor (Sect. S7). Similar to the satellite observations, the model places its extinction maximum at the top of the boundary layer and captures the clean-to-polluted ordering of the extinction profiles across the regimes, with R3 characterized by the largest free-tropospheric aerosol extinction in both model and satellite datasets (Fig. S6a). The model plume decays more smoothly with height than the observed discrete elevated layer, and in every regime it keeps aerosol above the inversion, as observed. Regressing the modeled droplet number concentration against the modeled below-cloud aerosol extinction, the same predictor as Fig. 7c, the model captures the ascending regime trend of the N_d susceptibility to the MBL aerosol, with $\beta(\text{MBL}) = 0.13, 0.16, \text{ and } 0.38$ for R1–R3 against the observed 0.12, 0.26, and 0.63 (Fig. S6c). The model and the satellite agree in R1, whereas the model is biased low by 0.10 in R2 and by 0.25 in R3, and the deficit widens as the below-cloud aerosol loading increases. A similar bias has been found over the SEA in the same nudged EAMv2–MAM4 configuration as used here and is related to an underestimated sub-cloud turbulent updraft (Howes et al., 2023, 2026). In those studies, the model converts only 37 % of the below-cloud aerosol number into cloud droplets, compared with 76 % measured by the ORACLES aircraft. A capped supersaturation activates a progressively smaller fraction of the particles as the aerosol number rises, so the modeled susceptibility flattens most in the regime carrying the most below-cloud aerosol.

Figure 8a places the satellite-retrieved and model-calculated L_0 values as functions of the separated-aerosol percentile within R3. The two series lie on opposite sides of the zero line and diverge as the aerosol loading increases. The modeled response remains positive in every group and strengthens in the opposite direction to the satellite-derived response, increasing from +0.38 on the cleanest days to +0.68 on the smokiest, with the rise concentrated across the interquartile and smoky groups. The model reverses both the sign and the aerosol dependence of the observed adjustment. The regime-wide slopes behave the same way. The model simulates a positive response of LWP to N_d across all three weather regimes, with values of +0.74 in R1, +0.74 in R2, and +0.42 in R3 (Fig. 8b, circles), compared with the corresponding satellite-derived values of +0.06, −0.07, and −0.23. While the model and the observations disagree on the sign of the response, both shift toward more negative values from R1 to R3. The modeled positive response weakens in the same order in which the observed negative response strengthens, and the model's weakest thickening occurs in the regime of the strongest observed thinning. It is noteworthy that although rainwater is present in all modeled cloudy columns, the median in-cloud rainwater path is only of order 0.05 g m^{-2} (regime means of $1.0\text{--}1.5 \text{ g m}^{-2}$; Fig. S6b), three orders of magnitude below the cloud water it accompanies. Rain therefore occurs in every modeled column but contributes little water mass. This nearly rain-free state matters here because the suppression of this weak rain generates the model's positive adjustment, as developed next.

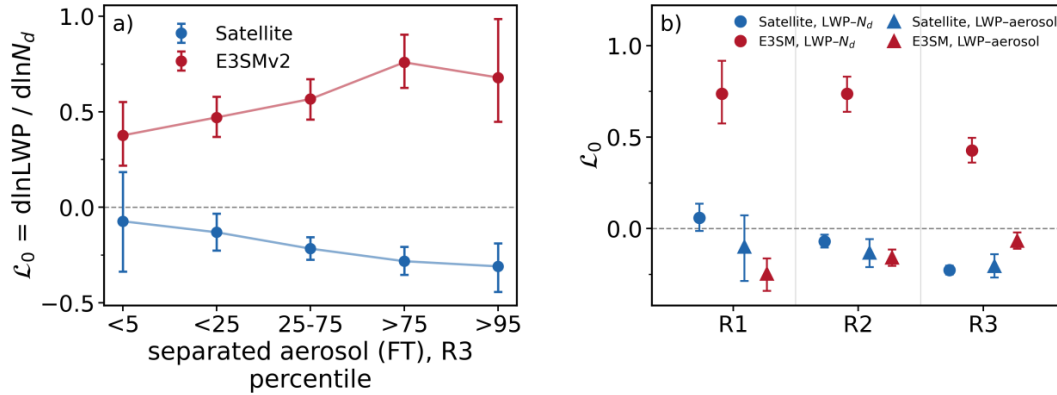


Figure 8. The E3SMv2–satellite comparison of liquid water path adjustments. (a) L_0 versus separated-aerosol percentile in R3, satellite (blue) versus E3SMv2 (red). (b) Whole-regime adjustment slopes by predictor and dataset (color: satellite blue, E3SMv2 red; symbol: circles for the N_d -based slope, triangles for the below-cloud-aerosol-extinction-based slope).

4.2 Diagnosing the model–satellite divergence

A positive LWP adjustment is a known tendency in global model simulations (Mülmenstädt et al., 2020, 2024), and our results suggest the modeled rain state is one likely origin here. In the MG2 microphysics (Gettelman and Morrison, 2015), the autoconversion of cloud water to rain follows the Khairoutdinov and Kogan (2000) scheme and decreases as N_d rises, so higher droplet number concentration suppresses the droplet-to-rain conversion sink. Because the sink term operates in every modeled cloudy column persistently even when the rainwater amount is negligible, fluctuations in N_d act on rain formation throughout the cloud field. The condensate withheld from rain is retained as cloud water, so each suppression appears directly as an LWP gain. Moreover, if the model initiates drizzle in too many cloudy columns and too lightly, an unrealistically large fraction of the cloud field becomes subject to this N_d -dependent suppression, which exaggerates the positive LWP response (Mülmenstädt et al., 2020). In single-column simulations of the eastern North Atlantic clouds with the closely related CAM6 physics, light precipitation of this kind occurs on more than 80 % of the days (Wang et al., 2023). Note that the small standing rain mass is not equivalent to a small rain-production rate. Rain forms and falls out within the sub-hourly physics integration of the model, so although the hourly snapshot retains only a residual rain mass, the autoconversion flux operates continuously between snapshots, and variations in N_d modulate this cumulative flux. The model’s regime stratification confirms that the positive LWP response weakens as precipitation becomes more active. In the



low-aerosol groups of every regime, the adjustment is more positive in the low-rain columns than in the high-rain columns, by 0.22–0.25 (Sect. S6). In the high-aerosol groups, the same ordering appears in R2 and R3, by 0.15–0.19, without reaching significance ($p = 0.10$ – 0.21), and R1 shows no difference, consistent with the buffering of the N_d axis where aerosol is abundant. The same nudged EAMv2 gives the opposite sign over the Eastern North Atlantic, where the N_d -based LWP adjustment is negative in every regime, from -0.17 to -0.36 (Zheng et al., 2025). Over that region, the modeled LWP– N_d relation spans both branches of the inverted-V, with drizzle suppression retaining cloud water at low N_d and an entrainment–evaporation feedback, triggered too rapidly, depleting it as N_d rises, so the declining branch dominates the whole-regime slope. Over the SEA, the modeled clouds never leave the accumulating branch, since the columns hold negligible rainwater, so that accretion by falling rain is negligible and the removal of cloud water to rain is carried by the N_d -dependent autoconversion. The sign of the modeled adjustment therefore follows how much cloud water the clouds lose beyond the suppression, negligible over the nearly rain-free SEA stratocumulus and large over the Eastern North Atlantic clouds, while a drizzle onset that occurs too frequently exaggerates the positive response where the accumulating branch dominates.

The EAMv2 vertical grid further limits the separated-aerosol comparison. The observed steepening arises at the inversion, where the absorbing layer sharpens a temperature jump that in nature spans a few tens of meters and the entrainment–evaporation exchange is concentrated (de Lozar and Mellado, 2015; Mellado, 2017). In the 72-layer grid, the layer spacing is 130–160 m near the median cloud top and 200–330 m in the band the separated layer occupies, 0.4–1.0 km higher (Fig. 2a), so the inversion spreads over one to two model layers, the separation between the layer and the cloud top spans only two to four layers, and the entrainment beneath the inversion is entirely parameterized (CLUBB). Low-cloud biases of this origin persist in E3SM until the vertical spacing approaches large-eddy scales (Bogenschütz et al., 2021; Lee et al., 2022). At this spacing, the model can represent neither the inversion sharpening that drives the steepening nor a clean distinction between contacting and separated aerosol, in line with its smoothly decaying plume. Part of the missing dependence on aerosol vertical placement in Fig. 8a may therefore stem from the vertical grid itself, in addition to the autoconversion covariance discussed above, and nudging the large-scale circulation cannot compensate for either of the model limitations.

Computed against the aerosol extinction, the same predictor used for the satellite, the model’s LWP–aerosol slope is negative in every regime (-0.25 , -0.16 , -0.07 for R1–R3), as is the satellite’s (-0.10 , -0.13 , -0.20 ; Fig. 8b, triangles). The same model that thickens its clouds by $+0.42$ to $+0.74$ per unit fractional N_d increase shows no positive LWP response to the aerosol that supplies those droplets. The below-cloud CCN behaves consistently, with LWP–CCN slopes of -0.20 and -0.12 in R1 and R2 and an R3 slope of -0.01 that is statistically indistinguishable from zero (Sect. S7, Fig. S7). Since an aerosol-driven adjustment must be transmitted along the



830 activation chain, if the model's positive LWP– N_d relation were an aerosol-driven adjustment, it
would survive, attenuated by the activation slope, when the adjustment is computed against the
aerosol itself. Instead, the measured slope is negative in every regime. Note that on the satellite
side the two predictors agree in sign wherever the N_d -based L_0 slope is significant, so the
comparison is diagnostic precisely because the two axes disagree in the model. The positive
835 LWP– N_d relationship in E3SMv2 is therefore hypothesized to be internal covariance generated
by the autoconversion parameterization, which moves droplet number and retained cloud water
jointly, and it disappears once the adjustment is expressed against the aerosol itself. The negative
aerosol-based slope, in turn, is consistent with the same conversion flux acting on the aerosol.
The below-cloud aerosol is the reservoir from which activation draws the droplets, and the
840 conversion of cloud water to rain removes the activated aerosol together with the condensate
(Liu et al., 2016; H. Wang et al., 2020), a coalescence scavenging that aircraft measurements
show depletes the below-cloud CCN budget as drizzle forms (Zheng et al., 2022a, 2024). This
covariance leaves the aerosol lowest where the cloud has processed the most water, and wet
scavenging of this kind can be mistaken for an aerosol effect in aerosol–cloud–precipitation
845 statistics (Gryspeerdt et al., 2015). The removal is proportional to the conversion flux, which
operates continuously while the standing rain mass stays small, and it is not transmitted along the
activation chain, so it can produce a negative aerosol-based slope while the N_d -based slope stays
positive.

Furthermore, the regime ordering supports this reading. The model slope is strongest in the
850 weakened-high regime, which carries the largest mean rain mass (1.5 g m^{-2} against 1.0 in the
other two regimes), and weakest in the strengthened-high regime, in which a similar rain mass
acts on the largest below-cloud reservoir and depletes the smallest fraction of it, whereas the
satellite slope strengthens with the regime's aerosol loading instead, and the below-cloud CCN
slopes weaken in the same order as the extinction slopes. The model's aerosol-based slope is
855 therefore hypothesized to reflect cloud removal of aerosol, and its agreement with the satellite in
sign does not indicate a reproduced adjustment. For aerosol forcing estimates, the implication is
direct. Over this aerosol-topped stratocumulus deck, a model in this state projects an LWP
adjustment of the wrong sign, and meteorological stratification cannot repair it. Bringing the
model's rain mass toward observed magnitudes is a necessary, though possibly not sufficient,
860 condition for improving the adjustment signal, which may implicate the model's process
coupling and time integration as much as its microphysical constants (Santos et al., 2021). The
sub-cloud updraft that affects droplet activation is too weak in the default EAMv2 configuration
(Howes et al., 2026). Although a weaker activation slope attenuates a transmitted adjustment
instead of flipping its sign, the representation of the updraft speed should also be calibrated with
865 caution.



5. Summary and discussion

This study examines aerosol–cloud interactions over the smoke-topped southeastern Atlantic with an analysis that differentiates the weather regimes and resolves the aerosol by its vertical placement relative to the cloud top. Nine years (2006–2014) of vertically resolved CALIOP extinction and MODIS cloud retrievals were collocated over a $10^\circ \times 10^\circ$ domain centered on Ascension Island, and the daily 13:00 UTC circulation was classified by the deep-learning clustering of ERA5 fields into a weakened-high regime (R1, 14 % of days), the background regime (R2, 49 %), and a strengthened-high regime (R3, 37 %) that contains most of the biomass-burning dry season. Within each regime, the cloud-water adjustment L_0 was evaluated as a function of separated (FT) and contacting (CT) aerosol and of the below-cloud (MBL) aerosol, and a meteorology-nudged E3SMv2 simulation sampled identically at 13:00 UTC provides the model comparison under the same weather. Three conclusions emerge.

1. The LWP– N_d adjustment is set jointly by weather regime and by the aerosol placement relative to the cloud. The baseline adjustment already spans +0.06 to –0.23 across the regimes before any aerosol stratification, ordered by each regime’s inversion strength and free-tropospheric humidity. On top of that baseline, free-tropospheric absorbing aerosol separated from the cloud steepens the R3 adjustment from –0.07 to –0.31, a dependence that survives the entrainment-drying control and that the co-transported plume moisture opposes. Aerosols in contact with the clouds are associated with cloud thinning in both the background and strengthened-high regimes (median LWP from 57 to 38 and 60 to 38 g m^{–2}), but the LWP– N_d coupling weakens toward zero in the humid background regime, while the dry strengthened-high regime maintains a negative coupling under a continuous droplet supply from the contacting aerosol. Statistics averaged across all regimes dilute the first signal, while the two opposing contacting-aerosol responses cancel in the all-sample statistic, so resolving the regimes recovers the dry-season state, where the adjustment is strongest and most aerosol-sensitive.
2. The three vertical placements of aerosols act on different terms of the same relationship. Below-cloud aerosol acts on the predictor. It supplies the droplets through activation (β (MBL) up to 0.63 in the strengthened-high regime) and, by making N_d track the aerosol, buffers the N_d -based slope toward zero. Aerosols in contact with the cloud top act on the cloud state itself. Absorbing aerosol thins the cloud through radiatively driven evaporation, and whether the LWP– N_d coupling survives that thinning is set by the contact humidity and the droplet supply, with humid contact air and little droplet replenishment relaxing the coupling in the background regime and dry contact air with a continuous droplet supply ($\beta = +0.21$) preserving it in the strengthened-high regime. Aerosol layers aloft that are separated from the cloud touch neither the droplets nor the water and instead set the strength of the LWP– N_d coupling, that is, how strongly the cloud water responds to a change in droplet number. A column-integrated burden such as the aerosol optical depth therefore integrates



over all three vertical placements, so AOD-type aerosol proxies cannot support a well-posed aerosol–cloud analysis over the SEA. The supersaturation competition documented at Ascension points in the same direction, suggesting the aerosol– N_d activation link saturates when aerosol is abundant (Dedrick et al., 2024, 2025), another way the N_d axis becomes unreliable in polluted air.

3. The regime-based E3SMv2 simulation produces the opposite LWP adjustment to the satellite observations. The L_0 is positive in every regime (+0.74, +0.74, +0.42) and strengthens with separated aerosol (+0.38 to +0.68) where the satellite steepens downward. The 72-layer vertical grid, with 130–330 m layer spacing around the inversion, leaves the inversion sharpening that drives the observed steepening unresolved, so part of the missing dependence on aerosol placement stems from the vertical grid as well as from the microphysics. Rainwater occurs in every model column but with negligible water mass (median in-cloud rainwater path of 0.05 g m^{-2}), so the suppression of drizzle and precipitation is a continuously available source of LWP gain. The model’s positive LWP– N_d relationship may partly arise from the modeled suppression of drizzle formation by increasing N_d , which constraining the large-scale meteorology alone cannot eliminate. Other mechanisms may also contribute, including weaker precipitation removal, enhanced condensation, reduced evaporation or entrainment, increased cloud occurrence, and covariance between N_d , LWP, and the meteorological conditions. However, once the adjustment is expressed against the below-cloud aerosol the sign reverses to negative values in every regime (–0.25, –0.16, –0.07). This sign change indicates that the positive LWP– N_d relation is not transmitted from the aerosol, since no LWP gain remains when the adjustment is measured against the aerosol that supplies the droplets.

For process-level understanding, the regime-resolved statistics assembled here, especially the separated-aerosol steepening in Fig. 4a, can constrain large-eddy and single-column simulations of absorbing layers over stratocumulus. In practice, quantify cloud adjustments against aerosol predictors alongside N_d , since both satellite buffering and the model’s sign reversal show that the N_d axis loses meaning exactly where aerosol is abundant. For model development, calibrating the rain-mass burden in E3SMv2 is necessary, though possibly not sufficient, to recover the observed sign of aerosol dependence over SEA, and a machine-learning emulator of the warm-rain process (Gettelman et al., 2021) offers one practical route to that calibration. Overall, the aerosol–cloud statistics developed in this study over the southeastern Atlantic are set jointly by the weather regime and by the aerosol vertical placement, and the framework of weather regimes and aerosol–cloud configurations that separates them is transferable to other stratocumulus regions under elevated absorbing aerosol, where it can be used to locate the origin of modeled LWP adjustment biases.



Code availability

945 The CNN–LSTM–IDEC classifier (CLIC) used for the regime classification is archived as CLIC v1.0.0 at Zenodo (<https://doi.org/10.5281/zenodo.23008635>; Zheng, 2026), together with the configuration of this study and the resulting daily regime labels for 2006–2014.

Data availability

950 The collocated CALIPSO–CloudSat–MODIS aerosol–cloud product is available from the NASA Langley Atmospheric Science Data Center at https://doi.org/10.5067/SATCORPS/CALIOP_CLOUDSAT_MODIS_25KM_L3.1.0 (Painemal, 2026). ERA5 data are available from the Copernicus Climate Data Store (<https://doi.org/10.24381/cds.bd0915c6> and <https://doi.org/10.24381/cds.adbb2d47>; Copernicus Climate Change Service, 2018a, b). The CAMS EAC4 reanalysis is available from the 955 Copernicus Atmosphere Data Store (<https://doi.org/10.24381/d58bbf47>; Copernicus Atmosphere Monitoring Service, 2020). The nudged E3SMv2 output over the southeastern Atlantic domain is archived at Zenodo (<https://doi.org/10.5281/zenodo.22972647>; Zheng et al., 2026).

Author contributions

960 XZ designed the study, performed the analysis, and wrote the manuscript. YF and YW supervised the study. XZ, YF, and SX performed the E3SMv2 simulations and supported the model configuration. DP and ZL developed and provided the collocated satellite product. All authors contributed to the scientific discussion, the interpretation of the results, and the revision of the manuscript.

965

Competing interests

Some authors are members of the editorial board of Atmospheric Chemistry and Physics.

Acknowledgements

970 The work at Argonne National Laboratory was supported by the U.S. DOE Office of Science under contract DE-AC02-06CH11357. Work at Lawrence Livermore National Laboratory was performed under the auspices of the U.S. DOE by Lawrence Livermore National Laboratory under contract No. DE-AC52-07NA27344. We acknowledge the computing resources provided on the Perlmutter system at the National Energy Research Scientific Computing Center 975 (NERSC), a DOE Office of Science User Facility, and the computing cluster Improv operated by



Argonne's Laboratory Computing Resource Center (LCRC). The analysis was carried out on the Sherlock cluster, maintained by the Stanford Research Computing Center. Anthropic's Claude was used to improve the grammar and streamline the writing style of the manuscript; the authors reviewed and take full responsibility for the content.

980

Financial support

This research has been supported by the CloudSat and CALIPSO Science Team Reconnect Program under the Science Mission Directorate of NASA (grant no. 80NSSC26K0136). X.Z. and Y.W. have been supported by the National Science Foundation (grant no. 2440581). Y.F. and S.X. have been supported by the Energy Exascale Earth System Model (E3SM) project, and Y.F. by the Atmospheric System Research (ASR) program, both funded by the U.S. Department of Energy (DOE), Office of Science, Office of Biological and Environmental Research.

985

References

- 990 Ackerman, A. S., Kirkpatrick, M. P., Stevens, D. E., and Toon, O. B.: The impact of humidity above stratiform clouds on indirect aerosol climate forcing, *Nature*, 432, 1014–1017, <https://doi.org/10.1038/nature03174>, 2004.
- Ackerman, A. S., Toon, O. B., Stevens, D. E., Heymsfield, A. J., Ramanathan, V., and Welton, E. J.: Reduction of tropical cloudiness by soot, *Science*, 288, 1042–1047, <https://doi.org/10.1126/science.288.5468.1042>, 2000.
- 995
- Adebisi, A. A., and Zuidema, P.: Low cloud cover sensitivity to biomass-burning aerosols and meteorology over the southeast Atlantic, *J. Climate*, 31, 4329–4346, <https://doi.org/10.1175/JCLI-D-17-0406.1>, 2018.
- Adebisi, A. A., and Zuidema, P.: The role of the southern African easterly jet in modifying the southeast Atlantic aerosol and cloud environments, *Q. J. Roy. Meteor. Soc.*, 142, 1574–1589, <https://doi.org/10.1002/qj.2765>, 2016.
- 1000
- Adebisi, A. A., Zuidema, P., and Abel, S. J.: The convolution of dynamics and moisture with the presence of shortwave absorbing aerosols over the southeast Atlantic, *J. Climate*, 28, 1997–2024, <https://doi.org/10.1175/JCLI-D-14-00352.1>, 2015.
- 1005 Adebisi, A. A., Zuidema, P., Chang, I., Burton, S. P., and Cairns, B.: Mid-level clouds are frequent above the southeast Atlantic stratocumulus clouds, *Atmos. Chem. Phys.*, 20, 11025–11043, <https://doi.org/10.5194/acp-20-11025-2020>, 2020.
- Albrecht, B. A., Fairall, C. W., Thomson, D. W., White, A. B., Snider, J. B., and Schubert, W. H.: Surface-based remote sensing of the observed and the adiabatic liquid water content



- 1010 of stratocumulus clouds, *Geophys. Res. Lett.*, 17, 89–92,
<https://doi.org/10.1029/GL017i001p00089>, 1990.
- Albrecht, B. A.: Aerosols, cloud microphysics, and fractional cloudiness, *Science*, 245, 1227–1230, <https://doi.org/10.1126/science.245.4923.1227>, 1989.
- 1015 Baró Pérez, A., Diamond, M. S., Bender, F. A.-M., Devasthale, A., Schwarz, M., Savre, J.,
 Tonttila, J., Kokkola, H., Lee, H., Painemal, D., and Ekman, A. M. L.: Comparing the
 simulated influence of biomass burning plumes on low-level clouds over the southeastern
 Atlantic under varying smoke conditions, *Atmos. Chem. Phys.*, 24, 4591–4610,
<https://doi.org/10.5194/acp-24-4591-2024>, 2024.
- 1020 Benjamini, Y., and Hochberg, Y.: Controlling the false discovery rate: a practical and powerful
 approach to multiple testing, *J. Roy. Stat. Soc. B*, 57, 289–300,
<https://doi.org/10.1111/j.2517-6161.1995.tb02031.x>, 1995.
- Bogenschutz, P. A., Yamaguchi, T., and Lee, H.: The Energy Exascale Earth System Model
 simulations with high vertical resolution in the lower troposphere, *J. Adv. Model. Earth
 Sy.*, 13, e2020MS002239, <https://doi.org/10.1029/2020MS002239>, 2021.
- 1025 Bozzo, A., Benedetti, A., Flemming, J., Kipling, Z., and Rémy, S.: An aerosol climatology for
 global models based on the tropospheric aerosol scheme in the Integrated Forecasting
 System of ECMWF, *Geosci. Model Dev.*, 13, 1007–1034, <https://doi.org/10.5194/gmd-13-1007-2020>, 2020.
- 1030 Bretherton, C. S., Blossey, P. N., and Uchida, J.: Cloud droplet sedimentation, entrainment
 efficiency, and subtropical stratocumulus albedo, *Geophys. Res. Lett.*, 34, L03813,
<https://doi.org/10.1029/2006GL027648>, 2007.
- 1035 Chang, I., Gao, L., Adebiyi, A. A., Doherty, S. J., Painemal, D., Smith, W. L., Lenhardt, E. D.,
 Fakoya, A. A., Flynn, C. J., Zheng, J., Yang, Z., Castellanos, P., da Silva, A. M., Zhang,
 Z., Wood, R., Zuidema, P., Christopher, S. A., and Redemann, J.: Regional aerosol
 warming enhanced by the diurnal cycle of low cloud, *Nat. Geosci.*, 18, 702–708,
<https://doi.org/10.1038/s41561-025-01740-1>, 2025.
- 1040 Chang, I., Gao, L., Burton, S. P., Chen, H., Diamond, M. S., Ferrare, R. A., Flynn, C. J.,
 Kacenelenbogen, M., LeBlanc, S. E., Meyer, K. G., Pistone, K., Schmidt, S., Segal-
 Rozenhaimer, M., Shinozuka, Y., Wood, R., Zuidema, P., Redemann, J., and Christopher,
 S. A.: Spatiotemporal heterogeneity of aerosol and cloud properties over the southeast
 Atlantic: an observational analysis, *Geophys. Res. Lett.*, 48, e2020GL091469,
<https://doi.org/10.1029/2020GL091469>, 2021.
- Choi, K. O., Rasch, P. J., Wood, R., Doherty, S. J., Wan, H., Wang, H., Zhang, S., Zhang, K.,
 and Wu, M.: Evaluation of marine boundary layer clouds over the Northeast Pacific



- 1045 during the CSET campaign in E3SM version 2, *J. Geophys. Res.-Atmos.*, 130, e2024JD042673, <https://doi.org/10.1029/2024JD042673>, 2025.
- Copernicus Atmosphere Monitoring Service: CAMS global reanalysis (EAC4), ECMWF Atmosphere Data Store [data set], <https://doi.org/10.24381/d58bbf47>, 2020.
- 1050 Copernicus Climate Change Service: ERA5 hourly data on pressure levels from 1940 to present, Copernicus Climate Change Service (C3S) Climate Data Store (CDS) [data set], <https://doi.org/10.24381/cds.bd0915c6>, 2018a.
- Copernicus Climate Change Service: ERA5 hourly data on single levels from 1940 to present, Copernicus Climate Change Service (C3S) Climate Data Store (CDS) [data set], <https://doi.org/10.24381/cds.adbb2d47>, 2018b.
- 1055 Costantino, L., and Bréon, F.-M.: Aerosol indirect effect on warm clouds over South-East Atlantic, from co-located MODIS and CALIPSO observations, *Atmos. Chem. Phys.*, 13, 69–88, <https://doi.org/10.5194/acp-13-69-2013>, 2013.
- Das, S., Colarco, P. R., and Harshvardhan, H.: The influence of elevated smoke layers on stratocumulus clouds over the SE Atlantic in the NASA Goddard Earth Observing System (GEOS) model, *J. Geophys. Res.-Atmos.*, 125, e2019JD031209, <https://doi.org/10.1029/2019JD031209>, 2020.
- 1060 de Graaf, M., Sarna, K., Brown, J., Tenner, E. V., Schenkels, M., and Donovan, D. P.: Aerosol first indirect effect of African smoke at the cloud base of marine cumulus clouds over Ascension Island, southern Atlantic Ocean, *Atmos. Chem. Phys.*, 23, 5373–5391, <https://doi.org/10.5194/acp-23-5373-2023>, 2023.
- 1065 de Lozar, A., and Mellado, J. P.: Evaporative cooling amplification of the entrainment velocity in radiatively driven stratocumulus, *Geophys. Res. Lett.*, 42, 7223–7229, <https://doi.org/10.1002/2015GL065529>, 2015.
- 1070 Dedrick, J. L., Russell, L. M., Sedlacek, A. J., Kuang, C., Zawadowicz, M. A., and Lubin, D.: Aerosol-correlated cloud activation for clean conditions in the tropical Atlantic boundary layer during LASIC, *Geophys. Res. Lett.*, 51, e2023GL105798, <https://doi.org/10.1029/2023GL105798>, 2024.
- 1075 Dedrick, J. L., Pelayo, C. N., Russell, L. M., Lubin, D., Mülmenstädt, J., and Miller, M.: Competition response of cloud supersaturation explains diminished Twomey effect for smoky aerosol in the tropical Atlantic, *P. Natl. Acad. Sci. USA*, 122, e2412247122, <https://doi.org/10.1073/pnas.2412247122>, 2025.
- Diamond, M. S., Dobracki, A., Freitag, S., Small Griswold, J. D., Heikkilä, A., Howell, S. G., Kacarab, M. E., Podolske, J. R., Saide, P. E., and Wood, R.: Time-dependent entrainment of smoke presents an observational challenge for assessing aerosol–cloud interactions



- 1080 over the southeast Atlantic Ocean, *Atmos. Chem. Phys.*, 18, 14623–14636,
<https://doi.org/10.5194/acp-18-14623-2018>, 2018.
- Diamond, M. S., Saide, P. E., Zuidema, P., Ackerman, A. S., Doherty, S. J., Fridlind, A. M.,
 Gordon, H., Howes, C., Kazil, J., Yamaguchi, T., Zhang, J., Feingold, G., and Wood, R.:
 Cloud adjustments from large-scale smoke–circulation interactions strongly modulate the
 1085 southeastern Atlantic stratocumulus-to-cumulus transition, *Atmos. Chem. Phys.*, 22,
 12113–12151, <https://doi.org/10.5194/acp-22-12113-2022>, 2022.
- Fernald, F. G.: Analysis of atmospheric lidar observations: some comments, *Appl. Optics*, 23,
 652–653, <https://doi.org/10.1364/AO.23.000652>, 1984.
- Gettelman, A., Gagne, D. J., Chen, C.-C., Christensen, M. W., Lebo, Z. J., Morrison, H., and
 1090 Gantos, G.: Machine learning the warm rain process, *J. Adv. Model. Earth Syst.*, 13,
 e2020MS002268, <https://doi.org/10.1029/2020MS002268>, 2021.
- Gettelman, A., and Morrison, H.: Advanced two-moment bulk microphysics for global models.
 Part I: Off-line tests and comparison with other schemes, *J. Climate*, 28, 1268–1287,
<https://doi.org/10.1175/JCLI-D-14-00102.1>, 2015.
- 1095 Glassmeier, F., Hoffmann, F., Johnson, J. S., Yamaguchi, T., Carslaw, K. S., and Feingold, G.:
 Aerosol-cloud-climate cooling overestimated by ship-track data, *Science*, 371, 485–489,
<https://doi.org/10.1126/science.abd3980>, 2021.
- Golaz, J.-C., Van Roekel, L. P., Zheng, X., Roberts, A. F., Wolfe, J. D., Lin, W., Bradley, A. M.,
 Tang, Q., Maltrud, M. E., Forsyth, R. M., Zhang, C., Zhou, T., Zhang, K., Zender, C. S.,
 1100 Wu, M., Wang, H., Turner, A. K., Singh, B., Richter, J. H., Qin, Y., Petersen, M. R.,
 Mامتjanov, A., Ma, P.-L., Larson, V. E., Krishna, J., Keen, N. D., Jeffery, N., Hunke,
 E. C., Hannah, W. M., Guba, O., Griffin, B. M., Feng, Y., Engwirda, D., Di Vittorio, A.
 V., Dang, C., Conlon, L. M., Chen, C.-C.-J., Brunke, M. A., Bisht, G., Benedict, J. J.,
 Asay-Davis, X. S., Zhang, Y., Zhang, M., Zeng, X., Xie, S., Wolfram, P. J., Vo, T.,
 1105 Veneziani, M., Tesfa, T. K., Sreepathi, S., Salinger, A. G., Reeves Eyre, J. E. J., Prather,
 M. J., Mahajan, S., Li, Q., Jones, P. W., Jacob, R. L., Huebler, G. W., Huang, X.,
 Hillman, B. R., Harrop, B. E., Foucar, J. G., Fang, Y., Comeau, D. S., Caldwell, P. M.,
 Bartoletti, T., Balaguru, K., Taylor, M. A., McCoy, R. B., Leung, L. R., and Bader, D. C.:
 The DOE E3SM Model Version 2: overview of the physical model and initial model
 1110 evaluation, *J. Adv. Model. Earth Sy.*, 14, e2022MS003156,
<https://doi.org/10.1029/2022MS003156>, 2022.
- Gryspeerdt, E., Goren, T., Sourdeval, O., Quaas, J., Mülmenstädt, J., Dipu, S., Unglaub, C.,
 Gettelman, A., and Christensen, M.: Constraining the aerosol influence on cloud liquid
 water path, *Atmos. Chem. Phys.*, 19, 5331–5347, [https://doi.org/10.5194/acp-19-5331-](https://doi.org/10.5194/acp-19-5331-2019)
 1115 2019, 2019.



- Gryspeerd, E., McCoy, D. T., Crosbie, E., Moore, R. H., Nott, G. J., Painemal, D., Small-Griswold, J., Sorooshian, A., and Ziemba, L.: The impact of sampling strategy on the cloud droplet number concentration estimated from satellite data, *Atmos. Meas. Tech.*, 15, 3875–3892, <https://doi.org/10.5194/amt-15-3875-2022>, 2022.
- 1120 Gryspeerd, E., Stier, P., White, B. A., and Kipling, Z.: Wet scavenging limits the detection of aerosol effects on precipitation, *Atmos. Chem. Phys.*, 15, 7557–7570, <https://doi.org/10.5194/acp-15-7557-2015>, 2015.
- Guo, X., Gao, L., Liu, X., and Yin, J.: Improved deep embedded clustering with local structure preservation, in: *Proceedings of the 26th International Joint Conference on Artificial Intelligence (IJCAI-17)*, Melbourne, Australia, 19–25 August 2017, 1753–1759, <https://doi.org/10.24963/ijcai.2017/243>, 2017.
- 1125
- Gupta, S., McFarquhar, G. M., O’Brien, J. R., Delene, D. J., Poellot, M. R., Dobracki, A., Podolske, J. R., Redemann, J., LeBlanc, S. E., Segal-Rozenhaimer, M., and Pistone, K.: Impact of the variability in vertical separation between biomass burning aerosols and marine stratocumulus on cloud microphysical properties over the southeast Atlantic, *Atmos. Chem. Phys.*, 21, 4615–4635, <https://doi.org/10.5194/acp-21-4615-2021>, 2021.
- 1130
- Gupta, S., McFarquhar, G. M., O’Brien, J. R., Poellot, M. R., Delene, D. J., Miller, R. M., and Small Griswold, J. D.: Factors affecting precipitation formation and precipitation susceptibility of marine stratocumulus with variable above- and below-cloud aerosol concentrations over the Southeast Atlantic, *Atmos. Chem. Phys.*, 22, 2769–2793, <https://doi.org/10.5194/acp-22-2769-2022>, 2022.
- 1135
- Haynes, J. M., L’Ecuyer, T. S., Stephens, G. L., Miller, S. D., Mitrescu, C., Wood, N. B., and Tanelli, S.: Rainfall retrieval over the ocean with spaceborne W-band radar, *J. Geophys. Res.-Atmos.*, 114, D00A22, <https://doi.org/10.1029/2008JD009973>, 2009.
- Herbert, R. J., Bellouin, N., Highwood, E. J., and Hill, A. A.: Diurnal cycle of the semi-direct effect from a persistent absorbing aerosol layer over marine stratocumulus in large-eddy simulations, *Atmos. Chem. Phys.*, 20, 1317–1340, <https://doi.org/10.5194/acp-20-1317-2020>, 2020.
- 1140
- Hersbach, H., Bell, B., Berrisford, P., Hirahara, S., Horányi, A., Muñoz-Sabater, J., Nicolas, J., Peubey, C., Radu, R., Schepers, D., Simmons, A., Soci, C., Abdalla, S., Abellan, X., Balsamo, G., Bechtold, P., Biavati, G., Bidlot, J., Bonavita, M., De Chiara, G., Dahlgren, P., Dee, D., Diamantakis, M., Dragani, R., Flemming, J., Forbes, R., Fuentes, M., Geer, A., Haimberger, L., Healy, S., Hogan, R. J., Hólm, E., Janisková, M., Keeley, S., Laloyaux, P., Lopez, P., Lupu, C., Radnoti, G., de Rosnay, P., Rozum, I., Vamborg, F., Villaume, S., and Thépaut, J.-N.: The ERA5 global reanalysis, *Q. J. Roy. Meteor. Soc.*, 146, 1999–2049, <https://doi.org/10.1002/qj.3803>, 2020.
- 1145
- 1150



- 1155 Howes, C., Saide, P. E., Coe, H., Dobracki, A., Freitag, S., Haywood, J. M., Howell, S. G., Gupta, S., Uin, J., Kacarab, M., Kuang, C., Leung, L. R., Nenes, A., McFarquhar, G. M., Podolske, J., Redemann, J., Sedlacek, A. J., Thornhill, K. L., Wong, J. P. S., Wood, R., Wu, H., Zhang, Y., Zhang, J., and Zuidema, P.: Biomass-burning smoke's properties and its interactions with marine stratocumulus clouds in WRF-CAM5 and southeastern Atlantic field campaigns, *Atmos. Chem. Phys.*, 23, 13911–13940, <https://doi.org/10.5194/acp-23-13911-2023>, 2023.
- 1160 Howes, C., Saide, P. E., Barth, M., Feng, Y., Redemann, J., Sedlacek, A. J., Shrivastava, M., Tang, W., Wood, R., Zawadowicz, M., and Zuidema, P.: Model sensitivities of biomass-burning aerosol chemical aging, sulfate formation, and cloud droplet activation in the southeastern Atlantic using CESM and E3SM, *J. Geophys. Res.-Atmos.*, 131, e2025JD043547, <https://doi.org/10.1029/2025JD043547>, 2026.
- 1165 Hu, Y., Vaughan, M., Liu, Z., Powell, K., and Rodier, S.: Retrieving optical depths and lidar ratios for transparent layers above opaque water clouds from CALIPSO lidar measurements, *IEEE Geosci. Remote Sens. Lett.*, 4, 523–526, <https://doi.org/10.1109/LGRS.2007.901085>, 2007.
- 1170 Inness, A., Ades, M., Agustí-Panareda, A., Barré, J., Benedictow, A., Blechschmidt, A.-M., Dominguez, J. J., Engelen, R., Eskes, H., Flemming, J., Huijnen, V., Jones, L., Kipling, Z., Massart, S., Parrington, M., Peuch, V.-H., Razinger, M., Remy, S., Schulz, M., and Suttie, M.: The CAMS reanalysis of atmospheric composition, *Atmos. Chem. Phys.*, 19, 3515–3556, <https://doi.org/10.5194/acp-19-3515-2019>, 2019.
- 1175 Jethva, H., Kayetha, V., Torres, O., and Hu, Y.: Global retrieval dataset of UV-VIS extinction and absorption of above-cloud aerosols from synergy of A-train active-passive measurements: Part I, above-cloud aerosol optical depth from the CALIOP depolarization ratio method, *Front. Environ. Sci.*, <https://doi.org/10.3389/fenvs.2026.1796667>, 2026.
- Johnson, B. T., Shine, K. P., and Forster, P. M.: The semi-direct aerosol effect: impact of absorbing aerosols on marine stratocumulus, *Q. J. Roy. Meteor. Soc.*, 130, 1407–1422, <https://doi.org/10.1256/qj.03.61>, 2004.
- 1180 Kacarab, M., Thornhill, K. L., Dobracki, A., Howell, S. G., O'Brien, J. R., Freitag, S., Poellot, M. R., Wood, R., Zuidema, P., Redemann, J., and Nenes, A.: Biomass burning aerosol as a modulator of the droplet number in the southeast Atlantic region, *Atmos. Chem. Phys.*, 20, 3029–3040, <https://doi.org/10.5194/acp-20-3029-2020>, 2020.
- 1185 Khairoutdinov, M., and Kogan, Y.: A new cloud physics parameterization in a large-eddy simulation model of marine stratocumulus, *Mon. Weather Rev.*, 128, 229–243, [https://doi.org/10.1175/1520-0493\(2000\)128<0229:ANCPPI>2.0.CO;2](https://doi.org/10.1175/1520-0493(2000)128<0229:ANCPPI>2.0.CO;2), 2000.
- Klett, J. D.: Stable analytical inversion solution for processing lidar returns, *Appl. Optics*, 20, 211–220, <https://doi.org/10.1364/AO.20.000211>, 1981.



- 1190 Larson, V. E.: CLUBB-SILHS: a parameterization of subgrid variability in the atmosphere, arXiv [preprint], <https://doi.org/10.48550/arXiv.1711.03675>, 2017.
- Lee, H.-H., Bogenschütz, P., and Yamaguchi, T.: Resolving away stratocumulus biases in modern global climate models, *Geophys. Res. Lett.*, 49, e2022GL099422, <https://doi.org/10.1029/2022GL099422>, 2022.
- 1195 Lenhardt, E. D., Gao, L., Gupta, S., McFarquhar, G. M., Xu, F., Ferrare, R. A., Hostetler, C. A., and Redemann, J.: Evidence of cloud sensitivity to above-cloud CCN as a function of environmental stability in the Southeast Atlantic based on remote sensing observations, *Atmos. Chem. Phys.*, 26, 11709–11732, <https://doi.org/10.5194/acp-26-11709-2026>, 2026.
- 1200 Li, Z., Painemal, D., Feng, Y., and Zheng, X.: Advancing the quantification of aerosol–cloud interactions with the CALIPSO–CloudSat–Aqua/MODIS record, *Atmos. Chem. Phys.*, 26, 7705–7720, <https://doi.org/10.5194/acp-26-7705-2026>, 2026.
- 1205 Li, Z., Painemal, D., Schuster, G., Clayton, M., Ferrare, R., Vaughan, M., Josset, D., Kar, J., and Trepte, C.: Assessment of tropospheric CALIPSO Version 4.2 aerosol types over the ocean using independent CALIPSO–SODA lidar ratios, *Atmos. Meas. Tech.*, 15, 2745–2766, <https://doi.org/10.5194/amt-15-2745-2022>, 2022.
- Liu, X., Ma, P.-L., Wang, H., Tilmes, S., Singh, B., Easter, R. C., Ghan, S. J., and Rasch, P. J.: Description and evaluation of a new four-mode version of the Modal Aerosol Module (MAM4) within version 5.3 of the Community Atmosphere Model, *Geosci. Model Dev.*, 9, 505–522, <https://doi.org/10.5194/gmd-9-505-2016>, 2016.
- 1210 Logan, T., Dong, X., Xi, B., Zheng, X., Wu, L., Abramowitz, A., Goluszka, A., and Harper, M.: Assessing radiative impacts of African smoke aerosols over the southeastern Atlantic Ocean, *Earth Space Sci.*, 11, e2023EA003138, <https://doi.org/10.1029/2023EA003138>, 2024.
- 1215 Martin, G. M., Johnson, D. W., and Spice, A.: The measurement and parameterization of effective radius of droplets in warm stratocumulus clouds, *J. Atmos. Sci.*, 51, 1823–1842, [https://doi.org/10.1175/1520-0469\(1994\)051<1823:TMAPOE>2.0.CO;2](https://doi.org/10.1175/1520-0469(1994)051<1823:TMAPOE>2.0.CO;2), 1994.
- Mellado, J. P.: Cloud-top entrainment in stratocumulus clouds, *Annu. Rev. Fluid Mech.*, 49, 145–169, <https://doi.org/10.1146/annurev-fluid-010816-060231>, 2017.
- 1220 Minnis, P., Sun-Mack, S., Chen, Y., Chang, F.-L., Yost, C. R., Smith, W. L., Jr., Heck, P. W., Arduini, R. F., Bedka, S. T., Yi, Y., Hong, G., Jin, Z., Painemal, D., Palikonda, R., Scarino, B. R., Spangenberg, D. A., Smith, R. A., Trepte, Q. Z., Yang, P., and Xie, Y.: CERES MODIS cloud product retrievals for Edition 4 – Part I: algorithm changes, *IEEE T. Geosci. Remote*, 59, 2744–2780, <https://doi.org/10.1109/TGRS.2020.3008866>, 2021.



- 1225 Mülmenstädt, J., Ackerman, A. S., Fridlind, A. M., Huang, M., Ma, P.-L., Mahfouz, N., Bauer, S. E., Burrows, S. M., Christensen, M. W., Dipu, S., Gettelman, A., Leung, L. R., Tornow, F., Quaas, J., Varble, A. C., Wang, H., Zhang, K., and Zheng, Y.: Can general circulation models (GCMs) represent cloud liquid water path adjustments to aerosol–cloud interactions?, *Atmos. Chem. Phys.*, 24, 13633–13652, <https://doi.org/10.5194/acp-24-13633-2024>, 2024.
- 1230 Mülmenstädt, J., Nam, C., Salzmänn, M., Kretzschmar, J., L’Ecuyer, T. S., Lohmann, U., Ma, P.-L., Myhre, G., Neubauer, D., Stier, P., Suzuki, K., Wang, M., and Quaas, J.: Reducing the aerosol forcing uncertainty using observational constraints on warm rain processes, *Sci. Adv.*, 6, eaaz6433, <https://doi.org/10.1126/sciadv.aaz6433>, 2020.
- 1235 Neale, R. B., Richter, J. H., and Jochum, M.: The impact of convection on ENSO: From a delayed oscillator to a series of events, *J. Climate*, 21, 5904–5924, <https://doi.org/10.1175/2008JCLI2244.1>, 2008.
- 1240 Painemal, D.: CALIPSO IIR Lidar Level 3 Global Energy and Water Cycle Experiment (GEWEX) Cloud, Standard V2-00, NASA Langley Atmospheric Science Data Center Distributed Active Archive Center [data set], https://doi.org/10.5067/SATCORPS/CALIOP_CLOUDSAT_MODIS_25KM_L3.1.0, 2026 (last access: 26 September 2026).
- 1245 Painemal, D., Chang, F.-L., Ferrare, R., Burton, S., Li, Z., Smith Jr., W. L., Minnis, P., Feng, Y., and Clayton, M.: Reducing uncertainties in satellite estimates of aerosol–cloud interactions over the subtropical ocean by integrating vertically resolved aerosol observations, *Atmos. Chem. Phys.*, 20, 7167–7177, <https://doi.org/10.5194/acp-20-7167-2020>, 2020.
- 1250 Painemal, D., Clayton, M., Ferrare, R., Burton, S., Josset, D., and Vaughan, M.: Novel aerosol extinction coefficients and lidar ratios over the ocean from CALIPSO–CloudSat: evaluation and global statistics, *Atmos. Meas. Tech.*, 12, 2201–2217, <https://doi.org/10.5194/amt-12-2201-2019>, 2019.
- Painemal, D., Greenwald, T., Cadeddu, M., and Minnis, P.: First extended validation of satellite microwave liquid water path with ship-based observations of marine low clouds, *Geophys. Res. Lett.*, 43, 6563–6570, <https://doi.org/10.1002/2016GL069061>, 2016.
- 1255 Painemal, D., Minnis, P., and Sun-Mack, S.: The impact of horizontal heterogeneities, cloud fraction, and liquid water path on warm cloud effective radii from CERES-like Aqua MODIS retrievals, *Atmos. Chem. Phys.*, 13, 9997–10003, <https://doi.org/10.5194/acp-13-9997-2013>, 2013.
- 1260 Pandey, S. K., and Adebisi, A. A.: Dust and smoke layers over the Atlantic Ocean weaken the underlying low-level cloud-top radiative cooling through different pathways, *Commun. Earth Environ.*, 7, 160, <https://doi.org/10.1038/s43247-026-03183-x>, 2026.



- 1265 Rasch, P. J., Xie, S., Ma, P.-L., Lin, W., Wang, H., Tang, Q., Burrows, S. M., Caldwell, P., Zhang, K., Easter, R. C., Cameron-Smith, P., Singh, B., Wan, H., Golaz, J.-C., Harrop, B. E., Roesler, E., Bacmeister, J., Larson, V. E., Evans, K. J., Qian, Y., Taylor, M., Leung, L. R., Zhang, Y., Brent, L., Branstetter, M., Hannay, C., Mahajan, S., Mametjanov, A., Neale, R., Richter, J. H., Yoon, J.-H., Zender, C. S., Bader, D., Flanner, M., Foucar, J. G., Jacob, R., Keen, N., Klein, S. A., Liu, X., Salinger, A. G., Shrivastava, M., and Yang, Y.: An overview of the atmospheric component of the Energy Exascale Earth System Model, *J. Adv. Model. Earth Sy.*, 11, 2377–2411, <https://doi.org/10.1029/2019MS001629>, 2019.
- 1270 Redemann, J., Wood, R., Zuidema, P., Doherty, S. J., Luna, B., LeBlanc, S. E., Diamond, M. S., Shinozuka, Y., Chang, I. Y., Ueyama, R., Pfister, L., Ryoo, J.-M., Dobracki, A. N., da Silva, A. M., Longo, K. M., Kacenelenbogen, M. S., Flynn, C. J., Pistone, K., Knox, N. M., Piketh, S. J., Haywood, J. M., Formenti, P., Mallet, M., Stier, P., Ackerman, A. S., Bauer, S. E., Fridlind, A. M., Carmichael, G. R., Saide, P. E., Ferrada, G. A., Howell, S. G., Freitag, S., Cairns, B., Holben, B. N., Knobelspiesse, K. D., Tanelli, S., L’Ecuyer, T. S., Dzambo, A. M., Sy, O. O., McFarquhar, G. M., Poellot, M. R., Gupta, S., O’Brien, J. R., Nenes, A., Kacarab, M., Wong, J. P. S., Small-Griswold, J. D., Thornhill, K. L., Noone, D., Podolske, J. R., Schmidt, K. S., Pilewskie, P., Chen, H., Cochrane, S. P., Sedlacek, A. J., Lang, T. J., Stith, E., Segal-Rozenhaimer, M., Ferrare, R. A., Burton, S. P., Hostetler, C. A., Diner, D. J., Seidel, F. C., Platnick, S. E., Myers, J. S., Meyer, K. G., Spangenberg, D. A., Maring, H., and Gao, L.: An overview of the ORACLES (ObseRvations of Aerosols above CLouds and their intEractionS) project: aerosol–cloud–radiation interactions in the southeast Atlantic basin, *Atmos. Chem. Phys.*, 21, 1507–1563, <https://doi.org/10.5194/acp-21-1507-2021>, 2021.
- 1280 Rousseeuw, P. J.: Silhouettes: a graphical aid to the interpretation and validation of cluster analysis, *J. Comput. Appl. Math.*, 20, 53–65, [https://doi.org/10.1016/0377-0427\(87\)90125-7](https://doi.org/10.1016/0377-0427(87)90125-7), 1987.
- 1290 Ryoo, J.-M., Pfister, L., Ueyama, R., Zuidema, P., Wood, R., Chang, I., and Redemann, J.: A meteorological overview of the ORACLES (ObseRvations of Aerosols above CLouds and their intEractionS) campaign over the southeastern Atlantic during 2016–2018: Part 1 – Climatology, *Atmos. Chem. Phys.*, 21, 16689–16707, <https://doi.org/10.5194/acp-21-16689-2021>, 2021.
- 1295 Ryoo, J.-M., Pfister, L., Ueyama, R., Zuidema, P., Wood, R., Chang, I., and Redemann, J.: A meteorological overview of the ORACLES (ObseRvations of Aerosols above CLouds and their intEractionS) campaign over the southeastern Atlantic during 2016–2018: Part 2 – Daily and synoptic characteristics, *Atmos. Chem. Phys.*, 22, 14209–14241, <https://doi.org/10.5194/acp-22-14209-2022>, 2022.
- Santos, S. P., Caldwell, P. M., and Bretherton, C. S.: Cloud process coupling and time integration in the E3SM atmosphere model, *J. Adv. Model. Earth Sy.*, 13, e2020MS002359, <https://doi.org/10.1029/2020MS002359>, 2021.



- 1300 Shinozuka, Y., Saide, P. E., Ferrada, G. A., Burton, S. P., Ferrare, R., Doherty, S. J., Gordon, H., Longo, K., Mallet, M., Feng, Y., Wang, Q., Cheng, Y., Dobracki, A., Freitag, S., Howell, S. G., LeBlanc, S., Flynn, C., Segal-Rosenhaimer, M., Pistone, K., Podolske, J. R., Stith, E. J., Bennett, J. R., Carmichael, G. R., da Silva, A., Govindaraju, R., Leung, R., Zhang, Y., Pfister, L., Ryoo, J.-M., Redemann, J., Wood, R., and Zuidema, P.: Modeling the smoky troposphere of the southeast Atlantic: a comparison to ORACLES airborne observations from September of 2016, *Atmos. Chem. Phys.*, 20, 11491–11526, <https://doi.org/10.5194/acp-20-11491-2020>, 2020.
- 1305 Stier, P.: Limitations of passive remote sensing to constrain global cloud condensation nuclei, *Atmos. Chem. Phys.*, 16, 6595–6607, <https://doi.org/10.5194/acp-16-6595-2016>, 2016.
- 1310 Sun, J., Zhang, K., Wan, H., Ma, P.-L., Tang, Q., and Zhang, S.: Impact of nudging strategy on the climate representativeness and hindcast skill of constrained EAMv1 simulations, *J. Adv. Model. Earth Sy.*, 11, 3911–3933, <https://doi.org/10.1029/2019MS001831>, 2019.
- Tatro, T., and Zuidema, P.: Weak, shallow, dry convection over Angola increases offshore stratocumulus cloud droplet number concentrations, *Commun. Earth Environ.*, <https://doi.org/10.1038/s43247-026-03995-x>, 2026.
- 1315 Twomey, S.: The influence of pollution on the shortwave albedo of clouds, *J. Atmos. Sci.*, 34, 1149–1152, [https://doi.org/10.1175/1520-0469\(1977\)034<1149:TIOPOT>2.0.CO;2](https://doi.org/10.1175/1520-0469(1977)034<1149:TIOPOT>2.0.CO;2), 1977.
- 1320 Wang, H., Easter, R. C., Zhang, R., Ma, P.-L., Singh, B., Zhang, K., Ganguly, D., Rasch, P. J., Burrows, S. M., Ghan, S. J., Lou, S., Qian, Y., Yang, Y., Feng, Y., Flanner, M., Leung, L. R., Liu, X., Shrivastava, M., Sun, J., Tang, Q., Xie, S., and Yoon, J.-H.: Aerosols in the E3SM Version 1: New developments and their impacts on radiative forcing, *J. Adv. Model. Earth Sy.*, 12, e2019MS001851, <https://doi.org/10.1029/2019MS001851>, 2020.
- 1325 Wang, Y., Zheng, X., Dong, X., Xi, B., Wu, P., Logan, T., and Yung, Y. L.: Impacts of long-range transport of aerosols on marine-boundary-layer clouds in the eastern North Atlantic, *Atmos. Chem. Phys.*, 20, 14741–14755, <https://doi.org/10.5194/acp-20-14741-2020>, 2020.
- 1330 Wang, Y., Zheng, X., Dong, X., Xi, B., and Yung, Y. L.: Insights of warm-cloud biases in Community Atmospheric Model 5 and 6 from the single-column modeling framework and Aerosol and Cloud Experiments in the Eastern North Atlantic, *Atmos. Chem. Phys.*, 23, 8591–8605, <https://doi.org/10.5194/acp-23-8591-2023>, 2023.
- Wilcox, E. M.: Direct and semi-direct radiative forcing of smoke aerosols over clouds, *Atmos. Chem. Phys.*, 12, 139–149, <https://doi.org/10.5194/acp-12-139-2012>, 2012.
- 1335 Winker, D. M., Vaughan, M. A., Omar, A., Hu, Y., Powell, K. A., Liu, Z., Hunt, W. H., and Young, S. A.: Overview of the CALIPSO mission and CALIOP data processing



- algorithms, J. Atmos. Ocean. Tech., 26, 2310–2323, <https://doi.org/10.1175/2009JTECHA1281.1>, 2009.
- 1340 Xi, B., Dong, X., Minnis, P., and Sun-Mack, S.: Comparison of marine boundary layer cloud properties from CERES-MODIS Edition 4 and DOE ARM AMF measurements at the Azores, J. Geophys. Res.-Atmos., 119, 9509–9529, <https://doi.org/10.1002/2014JD021813>, 2014.
- Xie, J., Girshick, R., and Farhadi, A.: Unsupervised deep embedding for clustering analysis, in: Proceedings of the 33rd International Conference on Machine Learning, PMLR, 48, 478–487, 2016.
- 1345 Xie, S., Lin, W., Rasch, P. J., Ma, P.-L., Neale, R., Larson, V. E., Qian, Y., Bogenschutz, P. A., Caldwell, P., Cameron-Smith, P., Golaz, J.-C., Mahajan, S., Singh, B., Tang, Q., Wang, H., Yoon, J.-H., Zhang, K., and Zhang, Y.: Understanding cloud and convective characteristics in version 1 of the E3SM atmosphere model, J. Adv. Model. Earth Sy., 10, 2618–2644, <https://doi.org/10.1029/2018MS001350>, 2018.
- 1350 Xie, S., Wang, Y.-C., Lin, W., Ma, H.-Y., Tang, Q., Tang, S., Zheng, X., Golaz, J.-C., Zhang, G. J., and Zhang, M.: Improved diurnal cycle of precipitation in E3SM with a revised convective triggering function, J. Adv. Model. Earth Sy., 11, 2290–2310, <https://doi.org/10.1029/2019MS001702>, 2019.
- 1355 Yamaguchi, T., Feingold, G., Kazil, J., and McComiskey, A.: Stratocumulus to cumulus transition in the presence of elevated smoke layers, Geophys. Res. Lett., 42, 10478–10485, <https://doi.org/10.1002/2015GL066544>, 2015.
- Zhang, G. J., and McFarlane, N. A.: Sensitivity of climate simulations to the parameterization of cumulus convection in the Canadian Climate Centre general circulation model, Atmos.-Ocean, 33, 407–446, <https://doi.org/10.1080/07055900.1995.9649539>, 1995.
- 1360 Zhang, J., and Zuidema, P.: Sunlight-absorbing aerosol amplifies the seasonal cycle in low-cloud fraction over the southeast Atlantic, Atmos. Chem. Phys., 21, 11179–11199, <https://doi.org/10.5194/acp-21-11179-2021>, 2021.
- Zheng, X.: CLIC: CNN–LSTM–IDEC classifier for meteorological regime identification (v1.0.0), Zenodo [code], <https://doi.org/10.5281/zenodo.23008635>, 2026.
- 1365 Zheng, X., Dong, X., Ward, D. M., Xi, B., Wu, P., and Wang, Y.: Aerosol–cloud–precipitation interactions in a closed-cell and non-homogenous MBL stratocumulus cloud, Adv. Atmos. Sci., 39, 2107–2123, <https://doi.org/10.1007/s00376-022-2013-6>, 2022a.
- 1370 Zheng, X., Dong, X., Xi, B., Logan, T., and Wang, Y.: Distinctive aerosol–cloud–precipitation interactions in marine boundary layer clouds from the ACE-ENA and SOCRATES aircraft field campaigns, Atmos. Chem. Phys., 24, 10323–10347, <https://doi.org/10.5194/acp-24-10323-2024>, 2024.



- 1375 Zheng, X., Feng, Y., Painemal, D., Zhang, M., Xie, S., Li, Z., Jacob, R., and Lusch, B.: Regime-based aerosol–cloud interactions from CALIPSO-MODIS and the Energy Exascale Earth System Model version 2 (E3SMv2) over the Eastern North Atlantic, *Atmos. Chem. Phys.*, 25, 17473–17499, <https://doi.org/10.5194/acp-25-17473-2025>, 2025.
- Zheng, X., Feng, Y., and Xie, S.: E3SMv2 Nudged Model Simulation over the Southeastern Atlantic Region ($10^{\circ} \times 10^{\circ}$, 2005–2014) (v1.0), Zenodo [data set], <https://doi.org/10.5281/zenodo.22972647>, 2026.
- 1380 Zheng, X., Xi, B., Dong, X., Wu, P., Logan, T., and Wang, Y.: Environmental effects on aerosol–cloud interaction in non-precipitating marine boundary layer (MBL) clouds over the eastern North Atlantic, *Atmos. Chem. Phys.*, 22, 335–354, <https://doi.org/10.5194/acp-22-335-2022>, 2022b.
- 1385 Zhou, X., Ackerman, A. S., Fridlind, A. M., Wood, R., and Kollias, P.: Impacts of solar-absorbing aerosol layers on the transition of stratocumulus to trade cumulus clouds, *Atmos. Chem. Phys.*, 17, 12725–12742, <https://doi.org/10.5194/acp-17-12725-2017>, 2017.
- 1390 Zuidema, P., Sedlacek III, A. J., Flynn, C., Springston, S., Delgadillo, R., Zhang, J., Aiken, A. C., Koontz, A., and Muradyan, P.: The Ascension Island boundary layer in the remote southeast Atlantic is often smoky, *Geophys. Res. Lett.*, 45, 4456–4465, <https://doi.org/10.1002/2017GL076926>, 2018.