

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

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S1. Aerosol composition and absorption from CAMS EAC4

The composition of the regime aerosol is characterized with the speciated aerosol optical depths of the CAMS EAC4 reanalysis (Inness et al., 2019), sampled over the same domain and period at 12:00 UTC, the archive time closest to the 13:00 UTC analysis hour. Repeating the analysis at 15:00 UTC and on the 13:00 UTC interpolation changes the regime-median carbonaceous fractions by less than 0.3 %.

The composition separates the regimes and the within-regime aerosol gradients (Fig. S1). Carbonaceous aerosol (black carbon plus organic matter) accounts for a median 69 % of the column aerosol optical depth in R3, against 57 % and 60 % in R2 and R1, dust contributes only 2 % in R3, and the total column AOD is also highest in R3 (0.19, against 0.12–0.15). Within R3 the carbonaceous fraction rises from 59 % in the cleanest separated-aerosol quartile to 83 % in the most polluted, with the same increase, from a lower base, in R1 and R2. The AAOD construction of Sect. 2.1 yields column single-scattering albedo medians of 0.941, 0.940, and 0.906 for R1–R3 (Fig. S1c), with the R3 distribution carrying a broad absorbing tail below 0.90. The black-carbon single-scattering albedo is the least certain input to this construction, so the calculation is repeated with values of 0.20 and 0.30. Across this range the R3 AAOD median moves only between 0.017 and 0.019, the R3 SSA median between 0.900 and 0.911, and R3 remains the most absorbing regime under either choice. The days called smoky are therefore days on which an independent reanalysis places a predominantly carbonaceous and absorbing aerosol column over the domain.

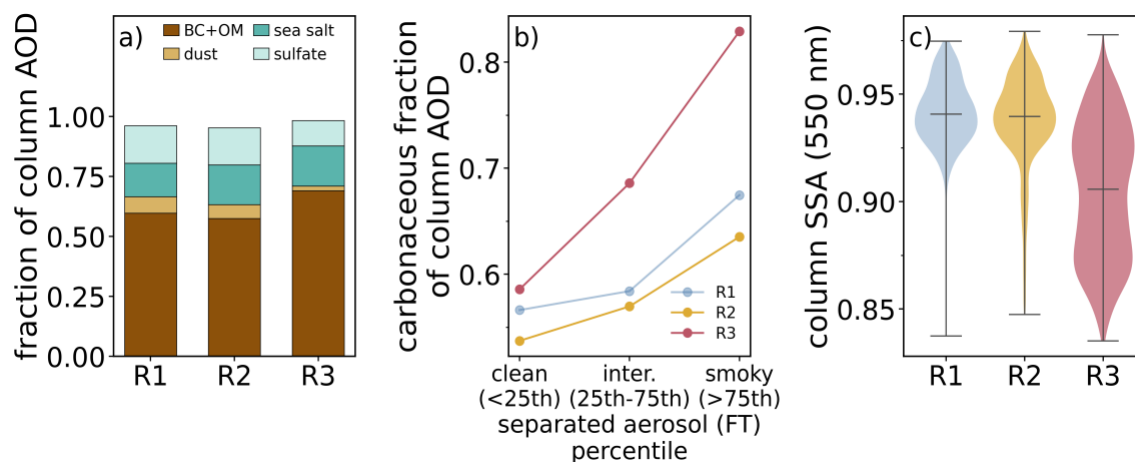


Figure S1. Aerosol composition and absorption from CAMS EAC4. (a) Median composition of the column aerosol optical depth per regime, partitioned into carbonaceous (black carbon plus organic matter), dust, sea salt, and sulfate fractions. (b) Median carbonaceous fraction of the column AOD per within-regime separated-aerosol percentile group. (c) Column single-scattering albedo (550 nm) per regime, derived with the per-species single-scattering albedos (violins; bars denote medians).

S2. Attribution of the above-cloud aerosol

Three independent lines of evidence indicate that biomass-burning smoke reaches the domain most frequently, and dominates the above-cloud aerosol most strongly, in regime R3. First, the depolarization-ratio (DR) method retrieves the optical depth of aerosol layers above opaque water clouds from the layer-integrated attenuated backscatter and depolarization of the underlying cloud, with no assumption on the aerosol lidar ratio (Hu et al., 2007). The global 2006–2022 DR dataset of Jethva et al. (2026) is subset to the analysis domain and the daytime overpasses of 2006–2014, and each retrieval day carries the regime label of Sect. 2.4. Above-cloud aerosol is detected on 3, 6, and 13 of every 100 regime days in R1, R2, and R3 (452, 1619, and 1215 labeled days). Conditional on a detection, the above-cloud optical depth has medians of 0.30, 0.32, and 0.39 (Fig. S2a), and the extinction Ångström exponent medians rise from 0.97 through 1.17 to 1.47 (Fig. S2b), indicating an increasingly fine-particle-dominated above-cloud aerosol from R1 to R3. Second, the CALIPSO–SODA column lidar ratio is type-diagnostic at 532 nm (biomass-burning smoke ~60–70 sr, dust ~40–55 sr, marine aerosol ~20–25 sr; Li et al., 2022). It rises monotonically with the separated-aerosol amount within every regime, from medians of 27–29 sr in each regime’s cleanest quartile, the signature of a marine-dominated column, to 43–46 sr in R1 and R2 and 58 sr in R3’s smokiest quartile (Fig. S2c, d). Because the column lidar ratio is an optical-depth-weighted blend of the marine boundary layer and the elevated layer, a column value near 58–60 sr requires the elevated layer’s intrinsic lidar ratio to lie in the biomass-burning range. Third, the polluted quartiles concentrate in the June–October burning season (Fig. 2b). Overall, the three methods rest on different measurements and assumptions, and they converge on the same result, in which above-cloud aerosol occurs most often, is most fine-particle-dominated, and comes closest to the biomass-burning lidar-ratio range in R3, while the R1 and R2 upper quartiles reach only intermediate values. This attribution is consistent with the CAMS composition of Sect. S1.

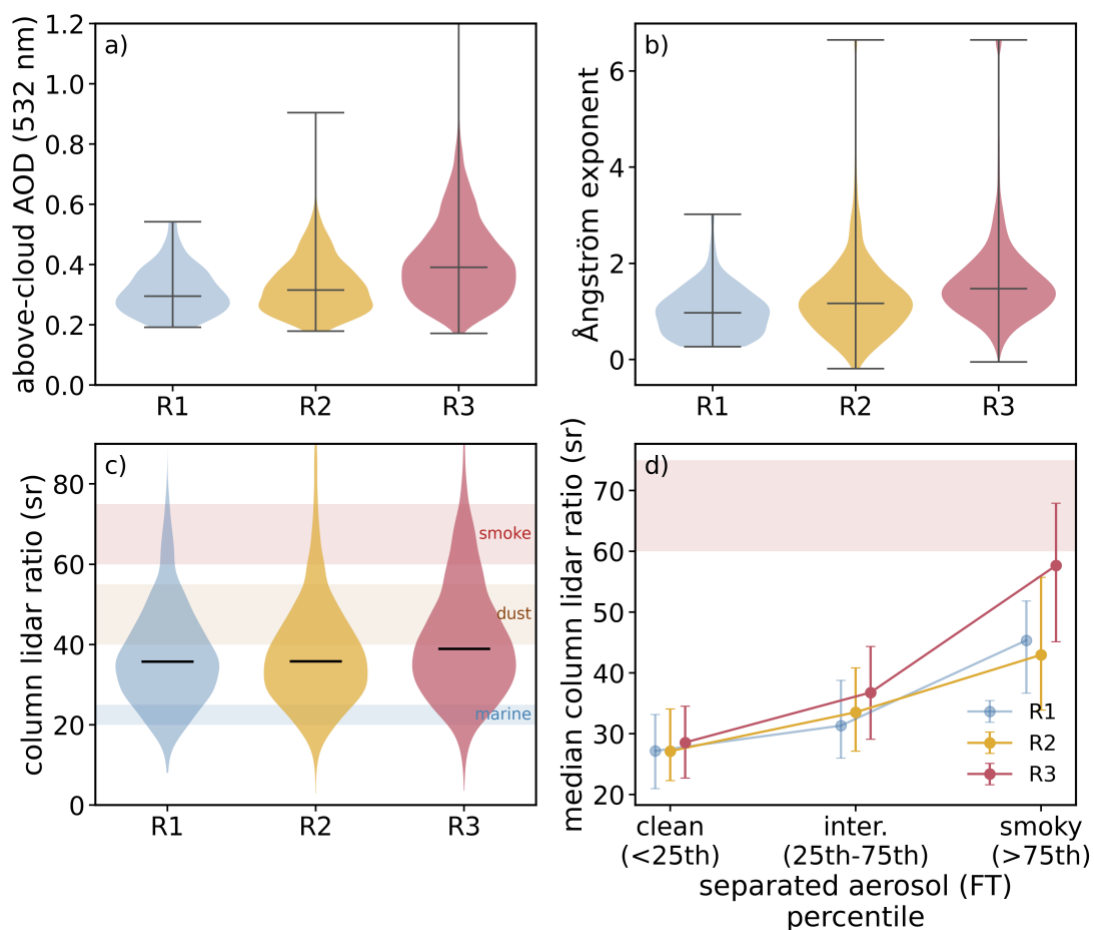


Figure S2. (a) Above-cloud aerosol optical depth (532 nm) per regime from the CALIOP depolarization-ratio retrieval (daytime overpasses, 2006–2014, analysis domain; violins, with bars denoting medians). (b) Extinction Ångström exponent (532–1064 nm) per regime from the same retrieval. (c) Distributions of the CALIPSO–SODA column lidar ratio per regime, with shaded characteristic ranges for marine aerosol, dust, and biomass-burning smoke. (d) Median column lidar ratio per within-regime separated-aerosol percentile group (whiskers denote the interquartile range).

S3. Regime contributions to the all-days aerosol dependence

Figure S3 examines how the all-days aerosol dependence of Sect. 3.3 arises from the regimes. The regime make-up of the all-days sample shifts along the aerosol axis, in which the R3 share of each all-days separated-aerosol percentile bin rises from 0.42 in the cleanest bin to 0.56 in the most polluted (Fig. S3a), so the all-sample statistic compares bins built from different weather. To measure how much of the all-days dependence this shifting make-up explains, each bin is assigned the regime-mean slopes weighted by that bin's regime shares. This mixture prediction would match the all-days slope if the dependence came from the shifting regime shares alone. Instead, it stays nearly flat, between -0.05 and -0.10 , and reproduces only about one fifth of the all-days gradient (Fig. S3b). Most of the all-days signal therefore arises within R3, and the R1 and R2 days mixed into every bin dilute it.

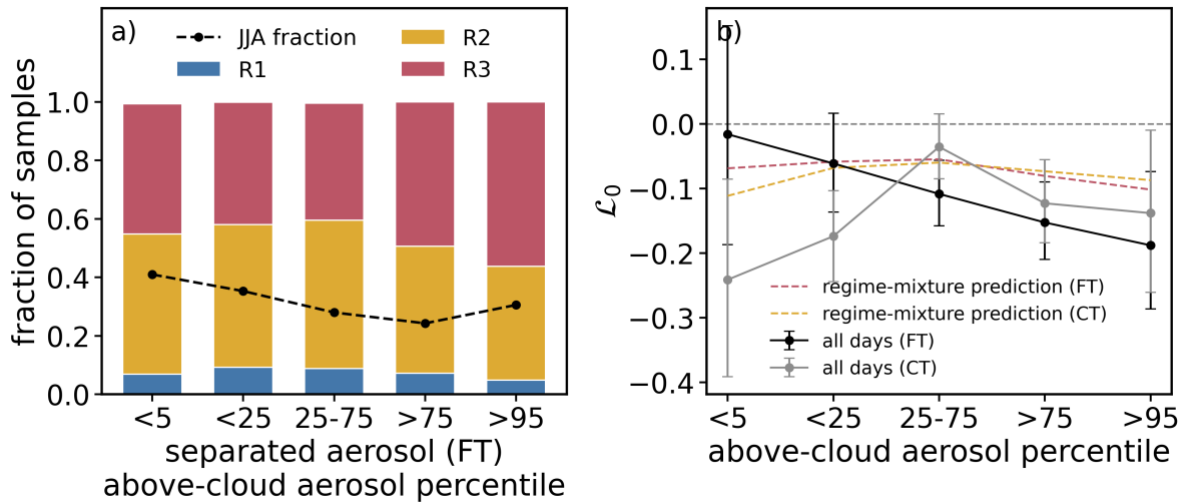


Figure S3. Regime contributions to the all-days aerosol dependence. (a) Regime make-up of the all-days separated-aerosol percentile bins, with the JJA fraction overlaid. (b) All-days slope versus the regime-mixture prediction, for both aerosol–cloud configurations.

S4. The humidity of the near-cloud air

The moisture analyses of Sects. 3.4 and 3.5 are anchored at 700 hPa, the level of the plume. The nearest ERA5 level to the air immediately above the cloud top is 850 hPa, whose surface lies at a median height of 1.52 km against regime-median cloud tops of 1.35–1.51 km (Fig. S4c). Since 36–47 % of cloud columns reach or exceed that surface, all q_{850} statistics use only the columns whose MODIS cloud top lies below it. The percentile groups are defined exactly as in the main text, and no other sample is limited this way.

Both humidity claims of the main text reproduce at this level (Fig. S4a, b). The q_{850} rises from the clean to the most polluted percentile groups in every regime and both configurations, and the contact-air ordering of Sect. 3.5 holds, with R2's cleanest group moister than R3's smokiest (7.80 versus 6.97–7.10 g kg^{-1}). Requiring cloud tops at least 0.2 km below the 850 hPa surface strengthens the contrast (8.40 versus 7.04 g kg^{-1} , with disjoint bootstrap intervals), so the numbers above are conservative. The entrainment-drying control of Fig. 5c also reproduces with $\text{EDI}_{850} = (1 - \text{RH}_{850}/100) \times \text{LTS}$, the R3 clean-to-smoky steepening persisting in its moist (−0.11 to −0.28, $p = 0.022$) and dry (−0.18 to −0.32, $p = 0.034$) thirds.

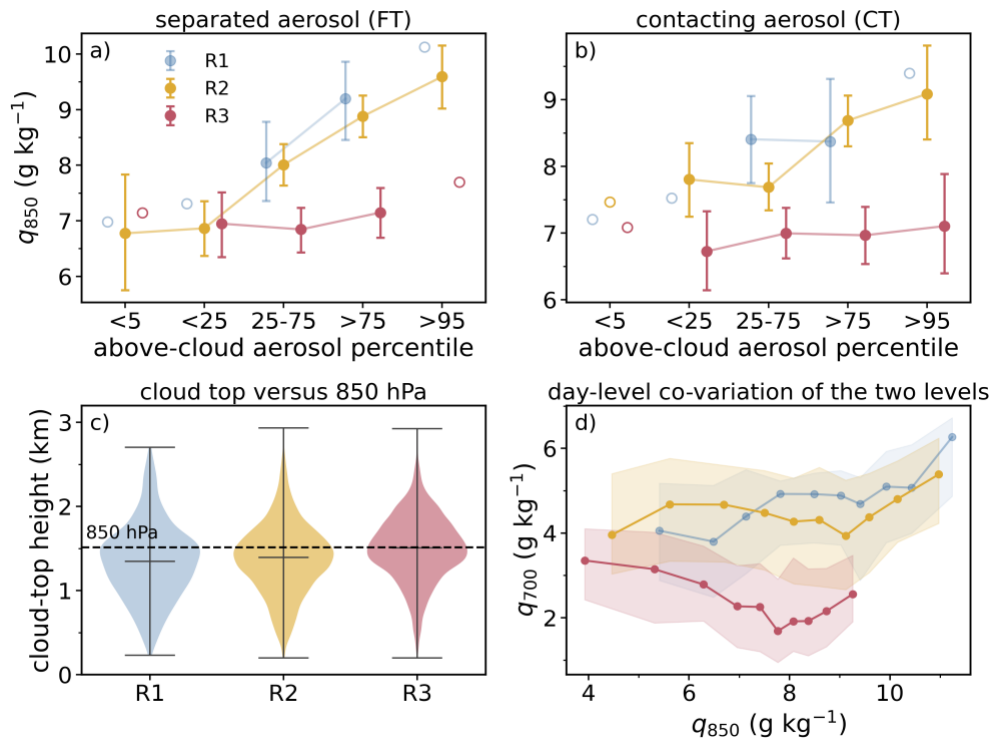


Figure S4. (a, b) The q_{850} per within-regime (a) separated-aerosol and (b) contacting-aerosol percentile group (means with 95 % day-bootstrap intervals; open regime-colored markers denote groups sampled on fewer than 30 days). (c) Cloud-top height distributions per regime (violins; bars denote medians) against the median height of the 850 hPa surface (dashed line). (d) Day-level co-variation of the domain-median q_{700} and q_{850} per regime.

S5. The entrainment-drying index

The entrainment-drying index $EDI = (1 - RH_{700}/100) \times LTS$ is the product of the free-tropospheric subsaturation and the lower-tropospheric stability, so high EDI marks days on which the air available for entrainment is far from saturation beneath a strong cap. Figure S5a shows the clean-versus-polluted separated-aerosol comparison within three equal EDI groups for all regimes, and Fig. S5b the EDI distributions of FT-clean days, the lowest quartile of each regime's own separated-aerosol extinction, against all days. The R3 steepening is present in every group and strongest in the moist, weak-drying group (-0.12 to -0.31 , $p = 0.005$); R2 differences are small and not significant, and R1 is undersampled.

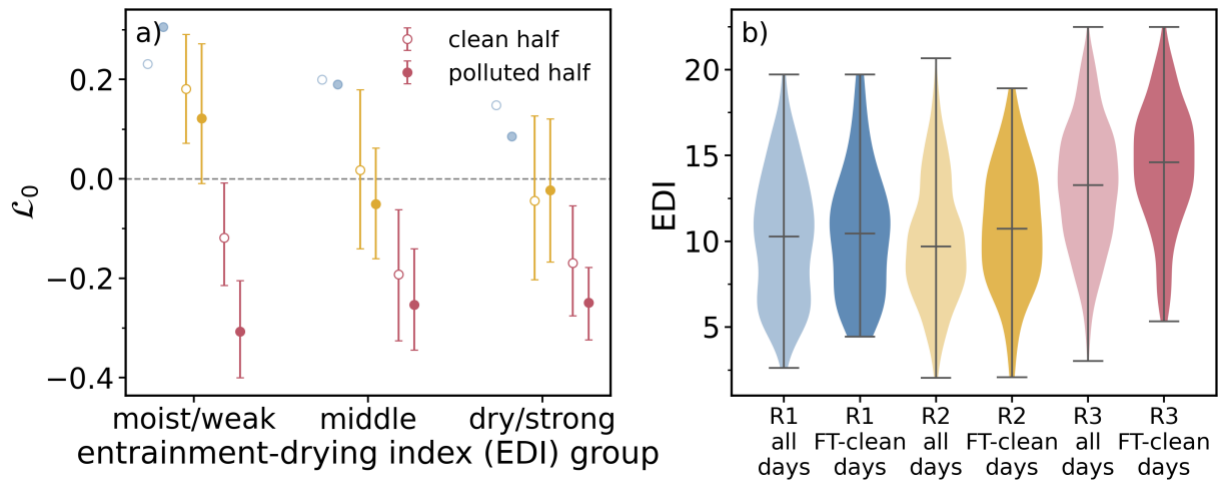


Figure S5. EDI stratification of L_0 and EDI distributions for all regimes. (a) L_0 in the clean and polluted halves of the separated-aerosol distribution within three equal groups of the entrainment-drying index (EDI), per regime, with 95 % day-bootstrap intervals; groups sampled on fewer than 30 days carry no interval. (b) EDI distributions of all days and of FT-clean days, the lowest quartile of each regime's separated-aerosol extinction, per regime (violins; bars denote medians).

S6. E3SMv2 diagnosis of aerosol placement, rain mass, and activation

Figure S6 collects the model diagnostics of Sect. 4. Figure S6a shows the normalized extinction profiles of E3SMv2 against CALIOP per regime. The model places its extinction maximum at the top of the boundary layer and preserves the regime ordering aloft, with a smoother and deeper elevated layer than the discrete CALIOP maximum. Figure S6b shows the distribution of the in-cloud rainwater path, which occurs in all cloud columns at a median of 0.05 g m^{-2} (regime means 1.5 , 1.0 , and 1.0 g m^{-2} for R1–R3). Figure S6c shows the model droplet-number susceptibility to the below-cloud extinction, the same predictor as Fig. 7c. The model values of $\text{dln}N_d/\text{dln}\sigma_{\text{EXT}}(\text{MBL}) = 0.13$, 0.16 , and 0.38 for R1–R3 reproduce the ascending regime ordering of the observed 0.12 , 0.26 , and 0.63 . The rain-stratified adjustment referenced in Sect. 4 comes from the same columns. In the low-aerosol groups of every regime the N_d -based adjustment is more positive in the low-rain than in the high-rain columns, by 0.22 – 0.25 ($p \leq 0.04$). 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.

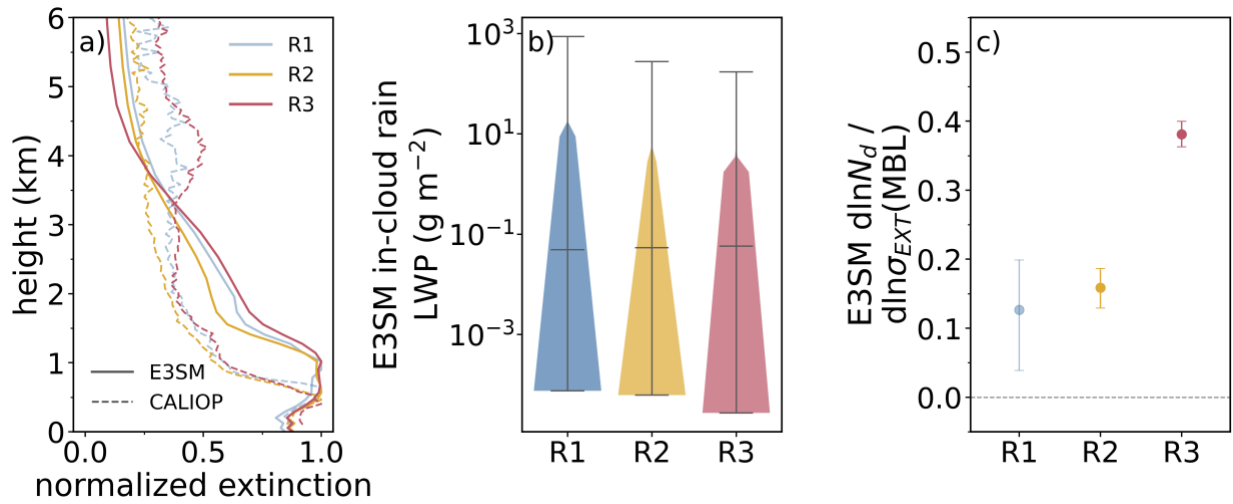


Figure S6. (a) Regime-mean aerosol extinction profiles of E3SMv2 (solid) and CALIOP (dashed), each normalized by its maximum. (b) Distributions of the E3SMv2 in-cloud rainwater path per regime (violins on a logarithmic axis; bars denote medians). (c) The model droplet-number susceptibility to the below-cloud extinction, $\text{dln}N_d/\text{dln}\sigma_{\text{EXT}}(\text{MBL})$, per regime with 95 % day-bootstrap intervals.

S7. Model aerosol sampling and the CCN predictor

The model aerosol predictors of Sect. 4 are layer means of the MAM4 aerosol extinction over the same three bands as the satellite predictors (Sect. 2.3), with the layers anchored to the diagnosed inversion base. The activation-explicit companion predictor is the E3SMv2 CCN concentration diagnosed at 0.2 % supersaturation, averaged below the cloud base. Against this predictor the model droplet-number susceptibility $d\ln N_d/d\ln \text{CCN}$ is 0.34, 0.33, and 0.39 for R1–R3, and the LWP slope $d\ln \text{LWP}/d\ln \text{CCN}$ is -0.20 and -0.12 in R1 and R2 and -0.01 (95 % CI -0.05 to 0.03) in R3 (Fig. S7). Along this axis the LWP response is therefore negative in R1 and R2 and statistically indistinguishable from zero in R3, and in no regime does it reach the positive values of the model’s LWP– N_d relation in Sect. 4.

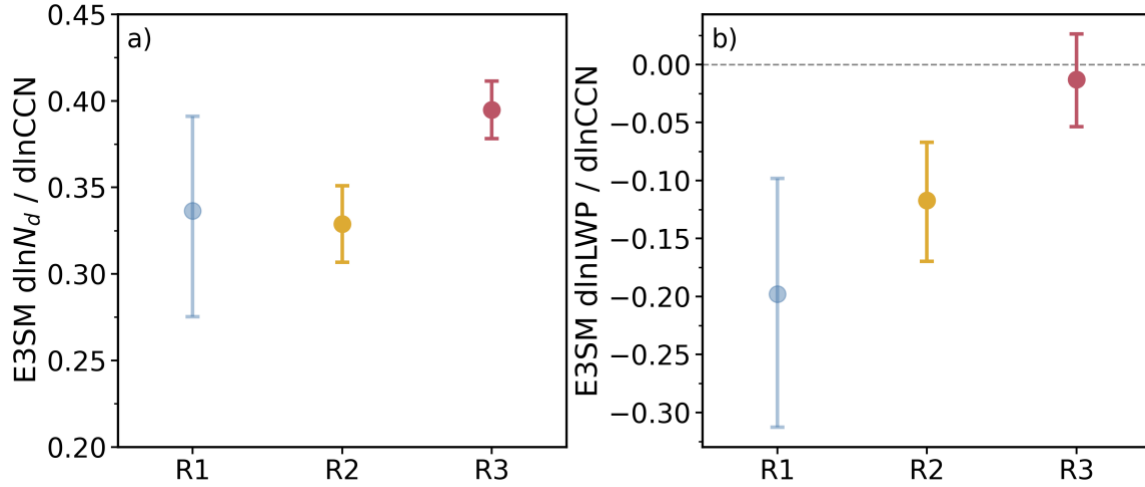


Figure S7. E3SMv2 slopes against the below-cloud CCN concentration (0.2 % supersaturation, averaged below the cloud base), per regime with 95 % day-bootstrap intervals. (a) The droplet-number susceptibility $d\ln N_d/d\ln \text{CCN}$. (b) The LWP slope $d\ln \text{LWP}/d\ln \text{CCN}$.

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