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Delayed cloud response reshapes aerosol radiative effects on marine low clouds

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Abstract

Aerosol perturbations modify marine low clouds and their radiative effects, yet the magnitude and sign of this impact remain uncertain. Most existing studies have examined this impact from a fixed time and place, implicitly assuming an instantaneous response. Tracking marine low clouds along three-day trajectories across three subtropical ocean regions, we find that the cloud-fraction adjustment to aerosol perturbations, the dominant control on cloud radiative effect, peaks one day and several hundreds kilometers downstream of the perturbation rather than where it is largest, biasing any fixed-time, fixed-location view of this coupling. This one-day lag is consistent across regions and seasons, with its magnitude of the cloud adjustment largely constrained by droplet effective radius and lower-tropospheric stability, and further modulated within individual regimes by the co-evolving meteorological state. These results show that aerosol–cloud radiative coupling is intrinsically time-dependent, helping explain the spread in satellite-based estimates of aerosol indirect effects and motivating a cloud-evolution perspective on aerosol perturbations.

Keywords: aerosol–cloud interaction, marine low clouds, cloud radiative effect, Lagrangian framework, stratocumulus-to-cumulus transition

Introduction

Marine low clouds strongly influence Earth’s energy budget, and their response to a changing climate remains one of the largest sources of uncertainty in climate sensitivity

[1, 2]. Aerosols perturb these clouds primarily by increasing cloud droplet number concentration (N_c), which can suppress precipitation and alter cloud fraction [3, 4], brighten clouds through the Twomey effect [5], and thereby modify the cloud radiative effect (CRE). Observational estimates of the sign and magnitude of this aerosol indirect effect nonetheless vary widely [1, 2, 6], largely because aerosol-driven adjustments are difficult to separate from co-varying meteorological influences [7–9].

Part of this difficulty is that marine low clouds are not a fixed cloud state but a continuously evolving one. As they are advected by the trade winds from cool eastern ocean basins toward warmer downstream regions, they undergo the stratocumulus-to-cumulus transition (SCT), a multi-day evolution from extensive, optically thick stratocumulus decks to broken, shallow cumulus, with cloud fraction and CRE declining systematically along the way [10–15]. Aerosols can accelerate or decelerate this transition by modifying precipitation suppression and entrainment efficiency [3, 16–18], and the transition itself is paced by cloud-controlling factors (CCFs) – inversion strength, free-tropospheric humidity, sea surface temperature, and surface wind speed among them – that vary across regions and seasons and act on their own, often multi-day, timescales [8, 19–30]. A cloud’s radiative response to an aerosol perturbation is therefore not sealed at the instant the perturbation occurs – it continues to be reshaped by the cloud’s subsequent evolution [31].

Yet almost all observational constraints on aerosol sensitivity relate cloud properties and radiative effects to aerosol proxies at a fixed place and time – an approach that implicitly assumes the radiative impact is largest exactly where and when the perturbation in N_c is largest. A handful of studies have instead followed clouds forward in time along their trajectories [16, 32], but none has directly tested whether the aerosol-driven radiative response itself is delayed relative to the perturbation that produced it, even though marine low clouds continuously evolve under CCFs operating across a range of distinct timescales [33–37]. If the response is in fact delayed, fixed-time observations would systematically underestimate, the true aerosol-driven signal [31].

Here, we test this possibility directly by tracking marine low clouds along three-day trajectories across three subtropical marine regions [33]. We show that the aerosol sensitivity of the cloud radiative effect does not peak at the time of the largest aerosol perturbation, but roughly one day (several hundred kilometers) later, once the initial droplet-number anomaly has already substantially decayed – a delay we trace to the aerosol sensitivity of optically thick cloud fraction, the dominant driver of CRE variability in these clouds. This one-day delay is a robust, previously underappreciated feature of the aerosol–cloud system, with direct implications for how satellite-based aerosol–cloud relationships are interpreted and how low-cloud feedbacks are constrained.

Results

Delayed peak in cloud radiative effect sensitivity

To examine how aerosol perturbations influence cloud radiative effects during cloud evolution, we analyze satellite and reanalysis observations along three-day Lagrangian trajectories over three subtropical marine regions – the Northeast Pacific (NEP),

Southeast Pacific (SEP), and Southeast Atlantic (SEA). Figure 1 tracks the aerosol sensitivity of cloud radiative effect anomalies, $\partial \text{CRE}' / \partial \ln(N'_{c,0})$, along composite trajectories for each region: marker size shows the fraction of the initial $N'_{c,0}$ perturbation still remaining, marker shading encodes the magnitude of $\partial \text{CRE}' / \partial \ln(N'_{c,0})$ itself, with lighter (white) shading denoting a stronger sensitivity, and trajectory color denotes season (primes denote anomalies relative to the local monthly climatology; Day 0 is the initial time). This combined encoding allows the full seasonal and regional range of composite trajectories to be compared directly against their radiative sensitivity as cloud evolution proceeds.

The aerosol perturbation itself decays steadily and monotonically along the trajectory: on average, only $\sim 20\%$ of the initial $N'_{c,0}$ anomaly remains by Day 3, consistent with previously reported N_c e-folding timescales [34]. The radiative response tracks a different, non-monotonic course: in all three regions, CRE' sensitivity is weak at Day 0, rises to a maximum on Day 1 (roughly several hundred kilometers downstream), and then gradually weakens over Days 2–3 as the underlying perturbation continues to fade. These two time series are therefore decoupled – the perturbation is decaying at every point along the trajectory, yet the radiative response it produces rises before it falls – demonstrating that the cloud’s radiative response to a given aerosol perturbation is not sealed at the moment the perturbation occurs, but continues to build, peak, and only then decline over the following days. A purely Eulerian, snapshot-based analysis, which by construction cannot follow an air mass forward in time, would place the aerosol radiative signal where it is weak and miss its actual peak [31].

This delayed peak is not confined to a single region or season: the same qualitative pattern – weak sensitivity at Day 0, a maximum on Day 1, gradual weakening thereafter – recurs across NEP, SEP, and SEA and across all seasons sampled within each region, though its magnitude differs systematically between them, with the SEP consistently the most sensitive region and the SEA the least (Fig. 1). This regional ordering recurs throughout our results and is examined mechanistically in the sections that follow.

Dominant contribution of cloud fraction adjustment on CRE

To understand why the CRE signal in Fig. 1 emerges downstream rather than at Day 0, we first identify which component of the cloud radiative effect (CRE) is responsible for its variability. Figure 2a summarizes the same regional and seasonal sensitivity, decomposing the total anomaly as

$$\text{CRE}' = \text{SWCRE}' + \text{LWCRE}', \quad (1)$$

with the shortwave term further separated into optically thick and thin cloud contributions,

$$\text{SWCRE}' = \text{SWCRE}'_{\text{thick}} + \text{SWCRE}'_{\text{thin}}. \quad (2)$$

We separate thick and thin clouds because N_c retrievals are more reliable for optically thick scenes [38], and because optically thin clouds play a disproportionate radiative role during the SCT [39]; the full derivation is given in Methods.

$\partial \text{CRE}' / \partial N'_{c,0}$ shows a broadly consistent seasonal cycle across regions, strongest when boundary layers are shallow and EIS is low (Fig. S1), with the SEP consistently the most sensitive region and the SEA the least – likely reflecting the influence of overlying biomass-burning smoke on cloud-circulation coupling in the SEA [40, 41]. Across nearly all region-season combinations, this variability is dominated by the shortwave contribution from optically thick clouds [39]; contributions from thin clouds and the longwave term are secondary, typically 5–15% of the total and generally below 20%, and often partly offsetting. The thin-cloud contribution in particular tends to oppose that of thick clouds, consistent with a trade-off between cloud regimes [42]. The longwave term, meanwhile, can introduce a compensating warming when cloud fraction increases are accompanied by a large temperature contrast between the sea surface and a systematically cooler cloud top, which enhances longwave trapping [40].

Figure 2b decomposes the aerosol sensitivity of $\text{SWCRE}'_{\text{thick}}$ into contributions from optically thick cloud fraction, $f'_{c,\text{thick}}$, and cloud scene albedo, A_{thick} (Methods). Across all regions and seasons, the variability is overwhelmingly controlled by $f'_{c,\text{thick}}$, while the albedo contribution remains small: an approximate further decomposition (Fig. S2) shows that the underlying Twomey and liquid-water-path adjustment terms are each comparable in size to the fraction contribution but largely cancel one another. This contrasts with several previous studies that identify cloud albedo change as the primary shortwave driver [4, 42, 43], and instead points to the spatial and temporal coverage of optically thick clouds as the leading channel – a discrepancy that may partly reflect our Lagrangian framework, which follows cloud evolution, relative to Eulerian analyses within fixed regions.

Together, Fig. 2 establishes a simple hierarchy: total CRE' is governed by $\text{SWCRE}'_{\text{thick}}$, which is in turn governed by $f'_{c,\text{thick}}$. Because cloud fraction is the dominant driver of total CRE' over a cloud field’s lifetime, we ask whether the radiative delay in Fig. 1 traces to a delay in the fraction response itself. Figure S3 shows that it does: the sensitivity of thick-cloud fraction to aerosol, $\partial f'_{c,\text{thick}} / \partial \ln(N'_{c,0})$, likewise does *not* peak at Day 0, but consistently reaches its maximum on Day 1 in all three regions – the mechanism identified here as the dominant driver of the radiative response, emerging only after a finite adjustment time of roughly one day, and providing a direct physical explanation for the delayed CRE signal in Fig. 1.

The magnitude of this delayed fraction response also differs systematically by region: peak sensitivities on Day 1 reach ~25% in the SEP, ~15% in the NEP, and ~8% in the SEA (Fig. S3) – the same ordering already evident for CRE sensitivity in Fig. 1, confirming that the regional contrast originates in the fraction response itself rather than in a separate radiative pathway. The inset panels of Fig. S3 show that, at Day 1, trajectories with stronger sensitivity tend to sit in regimes of higher EIS and larger droplet effective radius (r_e) – the key meteorological and microphysical controls quantified in the next section.

Physically, a one-day delay is consistent with the time needed for an aerosol-driven reduction in precipitation efficiency to translate into measurable changes in cloud liquid water path, and ultimately cloud fraction and breakup [3, 16, 17, 44]. Rather than the radiative footprint of an aerosol perturbation peaking at the location and time of maximum N_c enhancement, our results show that it peaks downstream, after

cloud microphysical adjustment has had time to act – implying an aerosol impact on the pace of the SCT itself.

Constraints from droplet size and meteorology

Figure 3a evaluates the Day-1 sensitivity $\partial f'_{c,\text{thick}}/\partial \ln(N'_{c,0})$ – the time at which the delayed response is largest – against droplet effective radius, using composite subgroups stratified by five cloud-controlling factors (CCFs; Methods) across all regions and seasons. This choice is motivated by r_e on Day 1 itself being primarily set by $N_{c,0}$ and the initial optically thick cloud liquid water path (Fig. S4), linking the timing of peak sensitivity to the pace of microphysical adjustment. The two show a clear linear relationship ($R^2 = 0.67$) that holds both between regions and across seasons within a region, consistent with larger droplets being more susceptible to precipitation suppression and associated increases in liquid water path and cloud fraction [3, 45].

Within this relationship, the color shading of the scatter points in Fig. 3a denotes EIS and reveals an additional, systematic structure: relative to the fitted line, states with higher EIS tend to lie above it, while those with lower EIS fall below – indicating that lower-tropospheric stability modulates the magnitude of the delayed response beyond its covariance with r_e alone.

Figure 3b isolates this effect directly. After removing the r_e dependence, the residual sensitivity is systematically related to EIS: higher EIS is associated with a stronger delayed response, and lower EIS with a weaker one, robustly across all regions and seasons. The color shading in Fig. 3b denotes SST, showing that EIS and SST strongly covary – weaker inversions occur over warmer SSTs, stronger inversions over cooler ones – so this modulation should be understood as reflecting the broader, coupled thermodynamic–dynamical state of these regimes rather than EIS in isolation.

Figure 3c compares the skill of an r_e -only model against one that also incorporates the CCFs: combining r_e and the CCFs in a single linear model raises the explained variance from $R^2 = 0.67$ to $R^2 = 0.86$, showing that meteorological context resolves systematic deviations that microphysics alone cannot. The same improvement holds on Days 2–3, when the delayed signal has weakened but not disappeared (Figs. S5, S6). The magnitude of the one-day delayed cloud-fraction response is therefore jointly, and predictably, constrained by cloud microphysics and the large-scale meteorological environment.

Aerosol–meteorology coupling

The analysis above stratifies trajectories by fixed background conditions; we next ask whether aerosol susceptibility itself depends on how meteorology and aerosol perturbations co-evolve. We express variations in CRE' as the sum of three terms (Eq. (8) in Methods): a first-order term capturing the direct sensitivity to the aerosol perturbation, a first-order term summing the direct sensitivities to each cloud-controlling factor (CCF), and a second-order term capturing the coupling between the aerosol perturbation and each CCF.

Figure 4a and Table 1 show that the first-order terms recover established physical relationships. Higher RH_{700} consistently contributes to cooling across all three

regions, consistent with reduced entrainment-driven drying in moister environments, which favors cloud persistence and larger cloud fraction [21]. Stronger WS_{surf} similarly enhances cooling, likely reflecting increased turbulence and surface fluxes that support thicker, more extensive cloud layers [26, 27]. Higher SST instead contributes to warming, consistent with warmer sea-surface conditions favoring cloud breakup and reduced low-cloud coverage [23], and higher cloud-top height (CTH) is likewise associated with warming, reflecting both more rapid cloud breakup via decoupling in deeper boundary layers [11, 33, 46–48] and enhanced longwave warming from a larger sea-surface-to-cloud-top temperature contrast [49, 50].

The EIS term shows a more complex, region-dependent pattern: in the NEP and SEP, higher EIS enhances cooling, consistent with stronger lower-tropospheric stability favoring more extensive stratocumulus cover [19], but this sign reverses in the SEA. The reversal traces to region-specific covariability between EIS and the residual aerosol perturbation within our anomaly-based framework: in the SEA, high-EIS conditions coincide with a more negative N'_c anomaly that strengthens over Days 1–2, whereas the opposite holds in the NEP (Fig. S7). The apparent sign reversal therefore reflects regional aerosol–meteorology covariability rather than a breakdown of the underlying stability–cloud relationship: evaluated against the absolute (rather than anomaly) value of EIS, higher EIS is associated with larger $f_{c,\text{thick}}$ and stronger cooling on climatological timescales (Fig. S8).

The second-order (coupling) terms in Fig. 4b show that this aerosol susceptibility is not fixed, but modulated by the evolving meteorological state – and not always in the direction the first-order terms alone would suggest. For EIS, the coupling term is generally negative (aerosol-induced cooling is enhanced under more stable conditions), though its regional magnitude covaries with the SST term, since EIS and SST are themselves coupled; considered individually rather than within the multilinear framework, the EIS coupling remains robustly negative across all three regions (Fig. S9). SST shows a robust positive second-order contribution, consistent with its first-order warming role: higher SST is associated with reduced aerosol-induced cooling, suggesting that cloud regimes prone to breakup under warm surface conditions are less responsive to aerosol perturbations [24]. The RH_{700} coupling term is negative, though smaller in magnitude, in all three regions, consistent with reduced entrainment drying enhancing the sensitivity of cloud fraction to microphysical perturbations in moister environments [7, 51].

Surface wind speed and cloud-top height stand out because each shows a sign reversal between its first- and second-order contribution: stronger winds and shallower clouds both favor cloudiness and cooling in a linear sense, yet both *reduce* aerosol susceptibility in the coupling term. A parsimonious explanation is that these variables govern the pace of the cloud’s own evolution, which the aerosol perturbation acts to delay or accelerate rather than override – for example, stronger turbulent mixing under high winds can transiently thicken clouds while simultaneously accelerating precipitation loss and the decay of the N_c anomaly [52], weakening the aerosol signal by the time it would otherwise appear; similarly, deeper cloud layers break up more readily [33, 53] while also suppressing more precipitation in response to a given aerosol perturbation, so the two effects compete rather than acting independently.

This behavior provides an independent line of evidence, beyond the timing results in Figs. 1 and S3, that aerosol-driven cloud responses are shaped by the pace of ongoing meteorological processes rather than occurring instantaneously, and that aerosol sensitivity is best understood as an emergent property of cloud evolution rather than a fixed characteristic of a given cloud regime.

Discussion

This study shows that the radiative response of marine low clouds to aerosol perturbations is not instantaneous: the aerosol sensitivity of optically thick cloud fraction, the dominant control on shortwave CRE variability in these clouds, peaks roughly one day and several hundred kilometers downstream of the perturbation rather than at the time and place where the perturbation itself is largest. This delay is consistent across three subtropical ocean basins and all seasons, and is large enough – comparable in magnitude to the peak sensitivity itself – that it cannot be dismissed as noise around an effectively instantaneous response.

This finding builds on, and helps resolve some ambiguity in, an established hierarchy of controls on aerosol-driven CRE variability: total CRE' is governed by $SWCRE'_{\text{thick}}$, which is in turn governed by variability in thick-cloud fraction rather than cloud albedo [4]. In contrast to earlier work emphasizing the Twomey effect and liquid-water-path adjustments as the dominant shortwave pathway [4, 43], our decomposition shows that Twomey and LWP-adjustment contributions to albedo are each substantial but largely cancel, leaving cloud-fraction variability as the leading term. Because cloud fraction is the dominant driver of total CRE' over a cloud field’s lifetime, this hierarchy is precisely what allows the time-dependence identified here to be expressed at all – and offers a physical explanation for why studies emphasizing albedo-driven pathways, evaluated at a single time, may have underestimated the role of cloud evolution.

The one-day timescale is physically consistent with the time required for an aerosol-driven reduction in precipitation efficiency to alter cloud liquid water path and, ultimately, cloud breakup [3, 16, 17, 44], and is quantitatively linked to droplet effective radius and lower-tropospheric stability, which together explain most of the regional and seasonal variation in the delayed response’s magnitude ($R^2 = 0.86$, up from $R^2 = 0.67$ for droplet size alone). The same r_e –EIS relationship holds not only between regions but across seasons within a single region, pointing to a common organizing framework rather than region-specific behavior. A second, independent line of evidence for time-dependence comes from the aerosol–meteorology coupling terms: higher wind speed and lower cloud-top height both promote cloudiness in a linear sense while suppressing aerosol susceptibility once coupling is considered, consistent with these variables governing the pace of cloud evolution rather than acting instantaneously in their own right. [52, 53]. Taken together, these results indicate that aerosol sensitivity of cloud radiative effects is an emergent, time-dependent property of cloud evolution, not a fixed attribute of a cloud regime – helping to reconcile why estimates of aerosol indirect effects vary so widely, in magnitude and sometimes sign, across studies that sample cloud fields at different stages of evolution [7, 43].

This time-dependence has practical consequences for how aerosol perturbations are assessed, both as a source of natural variability and as a deliberate or inadvertent forcing. Because cloud-controlling factors set both the pace of cloud evolution and its radiative response, low-frequency climate variability such as the Pacific Decadal Oscillation [54] or El Niño–Southern Oscillation [55] can shift the baseline meteorological state and thereby change how strongly a given aerosol perturbation is expressed from one year, or decade, to the next – a source of apparent inconsistency across observational studies conducted in different periods that is distinct from, but compounds, the timing issue identified here.

The same logic applies to engineered or regulatory aerosol perturbations. Proposed marine cloud brightening strategies aim to enhance cloud reflectivity at the point of aerosol injection [56], yet our results indicate that the dominant radiative response may instead emerge roughly a day downstream, as the perturbed air mass evolves into a more receptive meteorological regime [17, 57–59] – implying that the radiative efficacy of such interventions cannot be judged solely from conditions at the injection site. An analogous argument applies to interpreting the cloud and radiative response to sharp, large-scale aerosol emission reductions such as the IMO 2020 marine-fuel sulfur regulation, where purely Eulerian detection strategies may miss a response that is displaced in time or space from the emission change itself [60–63].

Several limitations should be kept in mind when interpreting these results. First, the trajectory-aligned composite approach emphasizes mean structural differences between predefined high and low CCF groups, and therefore captures conditional, population-level tendencies rather than fully causal, trajectory-specific responses; residual confounding from covarying dynamical or thermodynamic factors not explicitly controlled for may remain, and compositing necessarily averages over substantial within-group variability, so episodic extremes may be underrepresented. Second, while the linear and selective second-order formulation used here is tractable and interpretable, cloud evolution and aerosol responses may involve genuinely nonlinear transitions that this framework does not fully capture [64]. Third, because trajectories are grouped using bulk thermodynamic and microphysical contrasts, the framework does not distinguish between distinct physical pathways of the SCT – for example, precipitation-driven depletion [17, 44] versus boundary-layer deepening associated with radiative or dynamical warming [13] – nor does it explicitly resolve mesoscale cloud organization, such as transitions between closed- and open-cell structures, which are known to strongly shape aerosol–cloud interactions and cloud radiative properties [39, 42, 65–67]: closed-to-disorganized transitions are considerably more common than closed-to-open transitions in this region [65], so the latter likely represent a minority of our trajectories. Averaging across these structurally distinct pathways may contribute to the regional and seasonal spread in the first- and second-order contributions in Fig. 4, since different regions preferentially sample different cloud morphologies and transition mechanisms; incorporating explicit measures of mesoscale organization and SCT pathway would provide a more process-resolving perspective in future work.

Overall, by demonstrating that aerosol impacts on marine low clouds unfold over a finite, physically interpretable timescale rather than instantaneously, this work identifies a mechanism that snapshot-based, Eulerian analyses are structurally unable to

capture, and points toward a Lagrangian, cloud-evolution perspective as necessary for constraining aerosol indirect effects and the low-cloud feedbacks that depend on them.

Methods

Datasets

We analyze satellite observations over three subtropical Eastern ocean regions – the Northeast Pacific (NEP), Southeast Pacific (SEP), and Southeast Atlantic (SEA) – for 2010–2018, focusing on marine low-cloud regimes.

Cloud properties are obtained from the MODIS Aqua Level-2 Collection 6.1 cloud product [68] at native $1\text{ km} \times 1\text{ km}$ resolution, including cloud fraction (f_c), cloud optical thickness (τ), liquid water path (LWP), cloud-top temperature (CTT), cloud-top height (CTH), and cloud effective radius (r_e). We restrict the analysis to single-layer, liquid-phase clouds (MODIS multilayer flag and cloud-phase metric together with $\text{CTT} > 263\text{ K}$), and exclude pixels with sensor zenith angle $> 55^\circ$ or solar zenith angle $> 65^\circ$ [69]. Optically thick and thin clouds are separated using $\tau = 3$ as threshold [39, 66], and all filtered properties are aggregated to a $1^\circ \times 1^\circ$ grid.

Cloud albedo is approximated from optical thickness using a two-stream formulation [70], $\alpha = \tau/(\tau + 7.7)$, applied separately to thick and thin clouds. The scene (top-of-atmosphere) albedo A accounts for multiple reflection between cloud, atmosphere, and surface [71, 72]:

$$\alpha^* = \alpha_s \frac{(1 - \alpha_{\text{cld}})^2}{1 - \alpha_{\text{cld}}\alpha_s}, \quad A = \alpha_{\text{atm}} + \frac{(1 - \alpha_{\text{atm}})^2(\alpha_{\text{cld}} + \alpha^*)}{1 - \alpha_{\text{atm}}(\alpha_{\text{cld}} + \alpha^*)}, \quad (3)$$

with ocean surface albedo $\alpha_s = 0.07$ and clear-atmosphere albedo $\alpha_{\text{atm}}(\theta) = 0.219/(1 + 0.816 \cos \theta)$ as a function of solar zenith angle θ [70]. Cloud-free scene albedo ($\alpha_{\text{cld}} = 0$) gives the clear-sky ocean albedo, A_{ocean} . All albedo quantities are computed at the pixel level before spatial aggregation.

Cloud droplet number concentration is estimated from MODIS τ and r_e under an adiabatic approximation [73],

$$N_c = \sqrt{C^2 \tau (10^{-4} r_e)^{-5}}, \quad C = 1.4067 \times 10^{-6} \text{ cm}^{-1/2}, \quad (4)$$

with r_e in micrometers.

Top-of-atmosphere radiative fluxes and albedo (all-sky and clear-sky) are obtained from the CERES SYN1deg Edition 4A product [74, 75] at $1^\circ \times 1^\circ$, hourly resolution; shortwave and longwave CRE are the all-sky minus clear-sky flux difference. Meteorological fields are obtained from ERA-Interim [76, 77], from which we derive the five cloud-controlling factors used to stratify trajectories: EIS [19], RH_{700} , WS_{surf} , SST, and CTH.

All cloud, radiative, and meteorological variables are analyzed as anomalies relative to local monthly climatology, $X' = X - \bar{X}_{\text{month}}$, to remove covariability associated with fixed regional differences in background cloudiness, radiation, and thermodynamic

state, isolating variability tied to cloud evolution and aerosol perturbations along trajectories.

Decomposition of the shortwave cloud radiative effect

SWCRE is partitioned into thick- and thin-cloud contributions assuming their relative shares scale with the product of cloud fraction and albedo contrast with the clear-sky ocean:

$$\text{SWCRE}'_{\text{thick}} = \text{SWCRE}' \frac{f'_{c,\text{thick}}(A'_{\text{thick}} - A_{\text{ocean}})}{f'_{c,\text{thick}}(A'_{\text{thick}} - A_{\text{ocean}}) + f'_{c,\text{thin}}(A'_{\text{thin}} - A_{\text{ocean}})}, \quad (5)$$

with $\text{SWCRE}_{\text{thin}} = \text{SWCRE} - \text{SWCRE}_{\text{thick}}$ obtained as the residual. Approximating $\text{SWCRE}'_{\text{thick}}$ as proportional to insolation, cloud fraction, and albedo contrast,

$$\text{SWCRE}'_{\text{thick}} \approx SW_{\text{in}} f'_{c,\text{thick}} (A'_{\text{thick}} - A_{\text{ocean}}), \quad (6)$$

a logarithmic derivative with respect to $\ln(N_{c,0})$ gives an additive sensitivity decomposition,

$$\frac{\partial \ln(\text{SWCRE}'_{\text{thick}})}{\partial \ln(N_{c,0})} = \frac{\partial \ln(f'_{c,\text{thick}})}{\partial \ln(N_{c,0})} + \frac{\partial \ln(A'_{\text{thick}} - A_{\text{ocean}})}{\partial \ln(N_{c,0})}, \quad (7)$$

used in Fig. 2b to separate fraction- and albedo-driven contributions. This decomposition is validated in Fig. S10 (slope = 0.98, $R^2 = 0.94$). Figure S2 further splits the albedo term of Eq. (7) into approximate Twomey and LWP-adjustment components using $A \propto N_c^{1/3} \mathcal{L}^{5/6}$; because this term is based on the sensitivity of $(A'_{\text{thick}} - A_{\text{ocean}})$ rather than cloud albedo alone, the split is approximate and intended only to illustrate relative magnitudes.

Trajectory construction and Lagrangian framework

Trajectories are constructed for each $1^\circ \times 1^\circ$ grid cell over the NEP, SEP, and SEA using ERA-Interim 925-hPa winds [33], spanning 74 hours and anchored to the MODIS Aqua overpass ($\sim 13:30$ local time, Day 0; Fig. S11). The first ~ 2 hours around the overpass define Day 0; subsequent 24-hour windows define Days 1 (2–26 h), 2 (26–50 h), and 3 (50–74 h). MODIS cloud properties are sampled once per day at the overpass time; CERES and ERA5 fields are sampled continuously along the trajectory.

Trajectories are retained only if cloud fraction $\geq 50\%$ at Day 0, motion is westward and equatorward (consistent with trade-wind advection), and valid MODIS retrievals exist at Day 0, yielding 88,361 trajectories (examples in Fig. S12).

Composite analysis and stratification

Trajectories are grouped by region, season, and then by five cloud-controlling factors (CCFs), each split into high/low groups by within-subset median anomaly ($2^5 = 32$ CCF subgroups per region–season; sample sizes in Fig. S13). $N_{c,0}$ and CTH_0 are evaluated at Day 0; EIS, RH_{700} , WS_{surf} , and SST are evaluated as trajectory means (Fig. S1). Within each CCF subgroup, trajectories are further divided

into low/mid/high $N_{c,0}$ tertiles (Fig. S14), so that aerosol effects (differences between adjacent $N_{c,0}$ groups) are evaluated under comparable meteorological conditions, and CCF effects (high–low group differences) are evaluated with $\ln(N_{c,0})$ -referenced sensitivities.

Regression framework for first- and second-order contributions

We express variations in CRE' using a selective second-order Taylor expansion,

$$\begin{aligned} \Delta \text{CRE}' \approx & \underbrace{\Delta \ln N'_{c,0} \left. \frac{\partial \text{CRE}'}{\partial \ln N'_{c,0}} \right|_{\text{CCFs}}}_{\text{first-order } N'_{c,0} \text{ response}} + \underbrace{\sum_{x_i \in \text{CCFs}} \Delta x'_i \left. \frac{\partial \text{CRE}'}{\partial x'_i} \right|_{X'_j}}_{\text{sum of first-order CCF responses}} \\ & + \underbrace{\sum_{x_i \in \text{CCFs}} \Delta x'_i \Delta \ln N'_{c,0} \left. \frac{\partial^2 \text{CRE}'}{\partial x'_i \partial \ln N'_{c,0}} \right|_{X'_j}}_{\text{sum of second-order aerosol–meteorology coupling}}, \end{aligned} \quad (8)$$

where x'_i is the anomaly of the i -th CCF, X'_j denotes the remaining CCFs, and Δ denotes a finite perturbation.

To estimate the terms in Eq. (8), we first regress CRE' against $\ln(N'_{c,0})$ across all stratified composites to obtain the first-order aerosol term; the fitted contribution is removed, and the residual is regressed simultaneously against the five CCFs (multilinear regression) to obtain the first-order meteorological terms. This two-step, hierarchical procedure avoids conflating N_c variability with CCF terms, since CCF groups are not independently stratified in N_c . Second, $\partial \text{CRE}' / \partial \ln(N'_{c,0})$ is evaluated within each region–season–CCF subgroup and regressed against the five CCFs to obtain the second-order coupling terms. Both steps use the same stratified composites (Fig. 4, Table 1). This framework, together with a single-variable stratification cross-check (Fig. S9), reconstructs the observed $\Delta \text{CRE}'$ ($R^2 = 0.85$, improving modestly but significantly to $R^2 = 0.87$ with coupling terms included) and the observed aerosol sensitivity itself (Fig. S15), confirming that meteorological covariability among CCFs does not materially bias the inferred hierarchy of controls.

Supplementary information. Supplementary Figures S1–S15.

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Consent for publication. Not applicable.

Data availability. MODIS Level-2 cloud property data [78] are available from the NASA LAADS DAAC (<https://ladsweb.modaps.eosdis.nasa.gov/>). CERES SYN1deg data are available from the NASA CERES data archive (<https://ceres.larc.nasa.gov/data/>). ERA5 data [76, 77] are available from the Copernicus Climate Change Service Climate Data Store (<https://cds.climate.copernicus.eu/>).

Materials availability. Not applicable.

Code availability. Not applicable.

Author contributions. JC conceived the study, designed the analysis, performed the data analysis, and wrote the manuscript. RE provided the trajectory data and contributed to interpretation and editing. RW contributed to interpretation and editing.

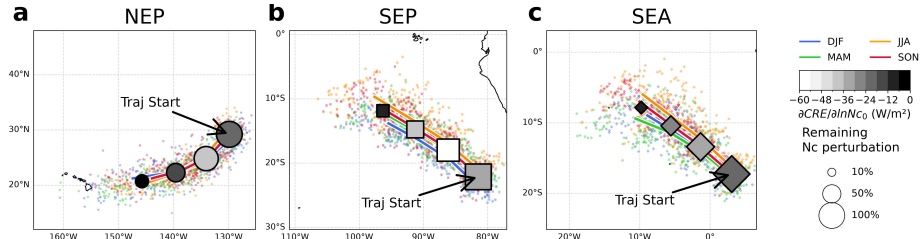


Fig. 1 The aerosol radiative effect peaks downstream of the initial perturbation, not at it. Spatial distribution and temporal evolution of the aerosol sensitivity of cloud radiative effect anomalies, $\partial \text{CRE}' / \partial \ln(N'_{c,0})$, along composite trajectories for (a) NEP, (b) SEP, and (c) SEA. Colored lines show seasonal-mean trajectories (DJF, MAM, JJA, SON); small points show individual daily trajectory locations. Marker size indicates the remaining fraction of the initial $N'_{c,0}$ anomaly relative to Day 0 (largest at trajectory start); marker shading indicates the magnitude of $\partial \text{CRE}' / \partial \ln(N'_{c,0})$, with lighter (white) shading denoting a stronger sensitivity. In all three regions, radiative sensitivity is weak at Day 0, where the aerosol perturbation itself is largest, and strengthens as the air mass evolves downstream.

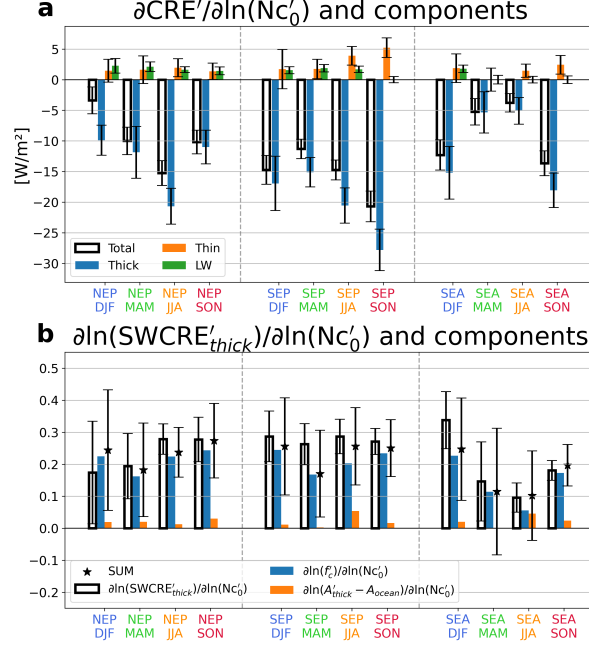


Fig. 2 Regional and seasonal variability of cloud radiative effects and their components. (a) Regional and seasonal mean aerosol sensitivity of cloud radiative effect anomalies, $\partial \text{CRE}' / \partial \ln(N'_{c,0})$, along three-day Lagrangian trajectories over the NEP, SEP, and SEA. Total CRE' is decomposed into shortwave and longwave components, with the shortwave contribution further separated into optically thick and thin cloud components. Bars are composite means for each region–season combination; error bars are 95% confidence intervals. (b) Decomposition of the aerosol sensitivity of the optically thick shortwave cloud radiative effect into contributions from thick-cloud fraction and cloud scene-albedo contrast. Positive values indicate enhanced shortwave cooling.

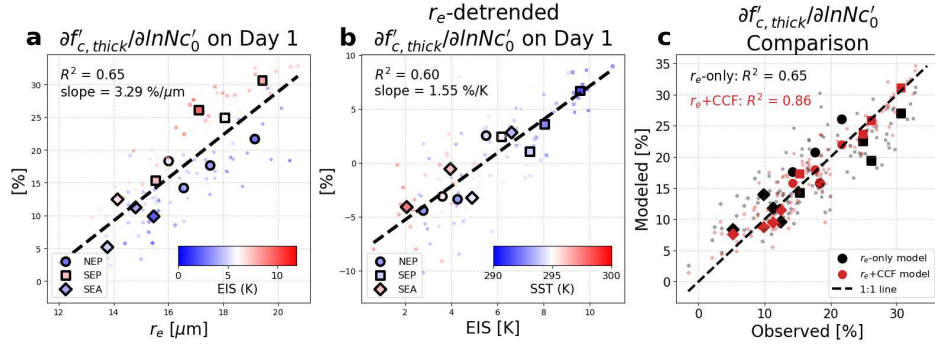


Fig. 3 Thermodynamic and meteorological constraints on the magnitude of the delayed cloud-fraction response. (a) Relationship between the Day-1 optically thick cloud fraction sensitivity and droplet effective radius (r_e) across all regions and seasons. Small points are individual composite subgroups; large symbols are region–season means; color indicates EIS. (b) Residual sensitivity after removing the linear r_e dependence, as a function of EIS; marker color indicates SST. (c) Observed sensitivity versus modeled sensitivity using r_e alone (black) or r_e plus cloud-controlling factors (red).

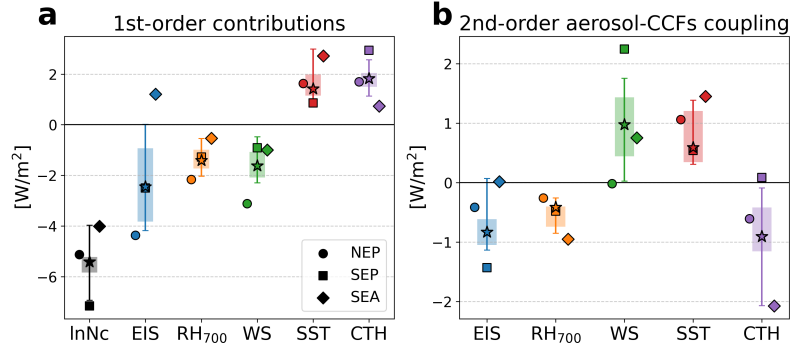


Fig. 4 First- and second-order contributions to $\Delta CRE'$. (a) First-order contributions associated with aerosol loading and individual cloud-controlling factors. Markers are regional medians, stars are regional means; thin (thick) error bars are 95% (66%) bootstrap confidence intervals. Each contribution is the product of the corresponding sensitivity (Table 1) and the regional standard deviation of the associated variable. (b) Second-order (coupling) contributions between aerosol perturbations and cloud-controlling factors, pooled across regions. Because the first-order aerosol contribution is strongly negative, positive second-order values indicate reduced cooling (weaker aerosol susceptibility), not absolute warming.

Table 1 First- and second-order CRE sensitivities. For each predictor (columns), the regression sensitivity of CRE to a perturbation in that predictor. Row “1st” lists first-order sensitivities; row “2nd” lists second-order aerosol-CCF coupling sensitivities. Top number in each cell is the median estimate; bottom line is the 95% bootstrap confidence interval. $\ln N_c$ has no second-order entry because the second-order terms are defined as couplings between $\ln N_c$ and the meteorological/cloud variables.

	$\ln N_c$ (W m ⁻²)	EIS (W m ⁻² K ⁻¹)	RH ₇₀₀ (W m ⁻² % ⁻¹)	WS _{surf} (W m ⁻² (m s ⁻¹) ⁻¹)	SST (W m ⁻² K ⁻¹)	CTH (W m ⁻² km ⁻¹)
1st	-12 (-15, -8)	-2.2 (-3.4, 0.0)	-0.23 (-0.34, -0.09)	-1.7 (-2.6, -0.6)	3.8 (1.6, 6.0)	7.9 (4.7, 15)
2nd		-0.8 (-1.1, 0.0)	-0.08 (-0.16, -0.02)	1.2 (0.2, 1.8)	1.6 (0.7, 5.3)	-4.5 (-10, -0.7)

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**Supplementary Information for
Delayed cloud response reshapes aerosol radiative
effects on marine low clouds**

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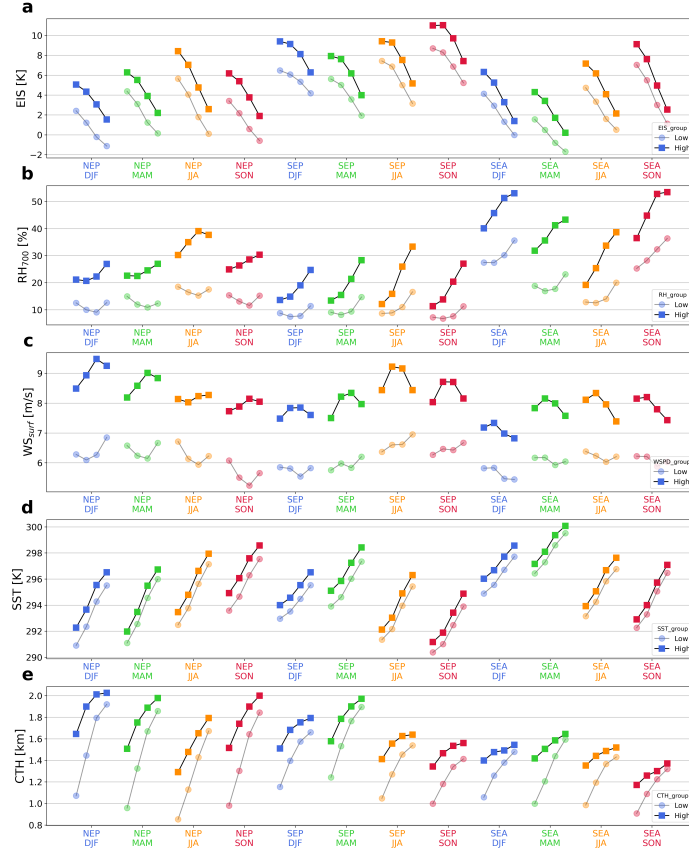


Fig. S1 Evolution of cloud-controlling factors in high and low composite groups along trajectories. Panels (a)–(e): daily evolution of EIS, RH₇₀₀, WS_{surf}, SST, and CTH for the high (dark) and low (light) groups defined in the composite stratification, by region and season, Days 0–3.

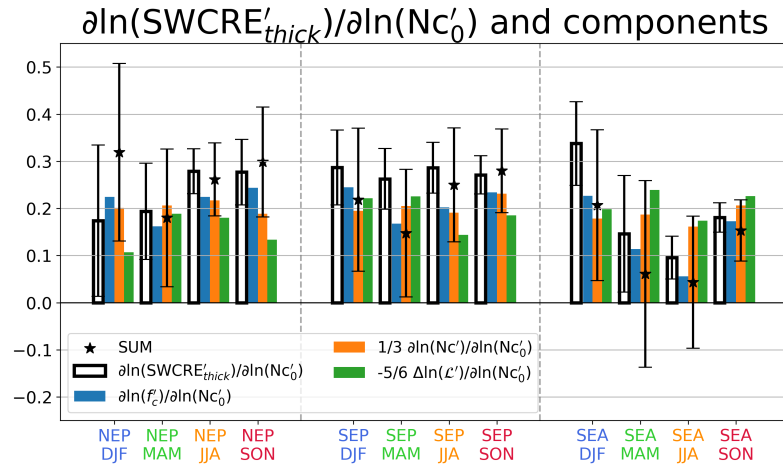


Fig. S2 Approximate decomposition of the aerosol sensitivity of optically thick short-wave cloud radiative effect. Same as Fig. 2b, but with the scene-albedo contribution further decomposed into approximate Twomey and liquid water path ($\mathcal{L}_{\text{thick}}$) adjustment components using $A \propto N_c^{1/3} \mathcal{L}^{5/6}$. This decomposition is approximate and intended to illustrate relative magnitudes only.

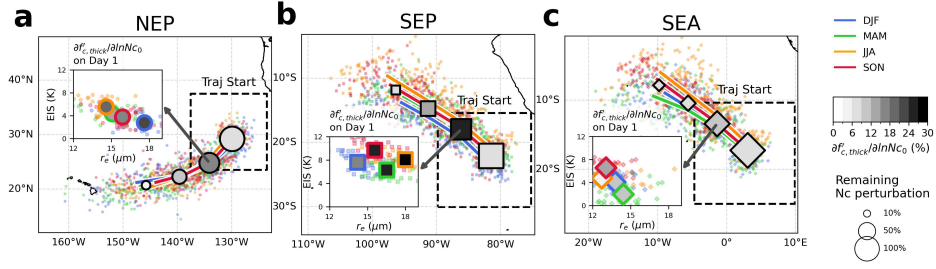


Fig. S3 The underlying cloud-fraction response peaks one day after the perturbation. Spatial distribution and temporal evolution of aerosol sensitivity of optically thick cloud fraction, $\partial f'_{c,thick}/\partial \ln(N'_{c,0})$, along composite trajectories for (a) NEP, (b) SEP, and (c) SEA. Small colored points show daily trajectory locations; solid lines show seasonal-mean trajectories. Large markers show regional daily means, sized by the remaining fraction of the initial $N'_{c,0}$ anomaly relative to Day 0, and shaded by the magnitude of $\partial f'_{c,thick}/\partial \ln(N'_{c,0})$; the response is largest on Day 1 in all three regions, not Day 0. Insets show the distribution of composite trajectories in EIS- r_e space at Day 1, with large symbols indicating seasonal means. This is the cloud-fraction-level counterpart to the CRE-level delay shown in Fig. 1, and the mechanistic basis for it (see Results).

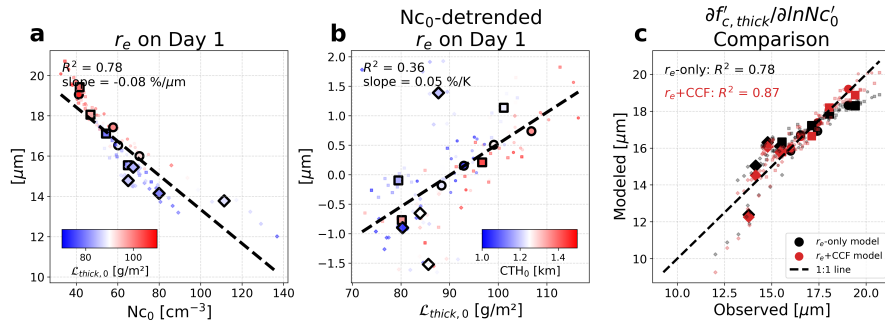


Fig. S4 Determinants of droplet effective radius on Day 1. Same as Fig. 3, but for r_e on Day 1. Panel (a): x-axis is $N_{c,0}$; panel (b): x-axis is initial optically thick cloud liquid water path, $\mathcal{L}_{thick,0}$. Panel (c): observed versus modeled r_e on Day 1.

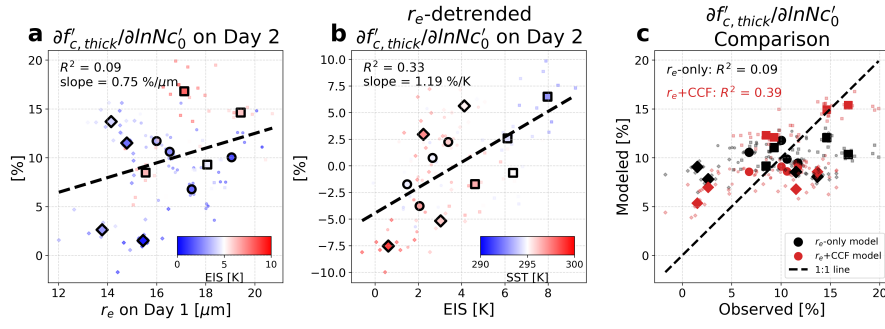


Fig. S5 Thermodynamic and meteorological constraints on cloud fraction sensitivity on Day 2. Same as Fig. 3, but for $\partial f'_{c,thick}/\partial \ln(N'_{c,0})$ on Day 2. (a) Relationship between the Day-2 sensitivity and droplet effective radius (r_e) measured on Day 1, across all regions and seasons; color indicates EIS. (b) Residual sensitivity after removing the linear r_e dependence, as a function of EIS; marker color indicates SST. (c) Observed sensitivity versus modeled sensitivity using r_e alone (black) or r_e plus cloud-controlling factors (red). Both relationships are markedly weaker than on Day 1 ($R^2 = 0.09$ and 0.39 , respectively), consistent with the delayed response beginning to fade by Day 2.

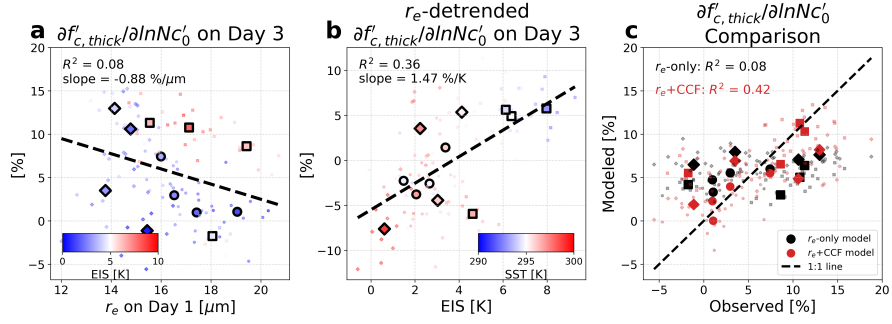


Fig. S6 Thermodynamic and meteorological constraints on cloud fraction sensitivity on Day 3. Same as Fig. 3, but for $\partial f'_{c,thick}/\partial \ln(N'_{c,0})$ on Day 3. (a) Relationship between the Day-3 sensitivity and droplet effective radius (r_e) measured on Day 1, across all regions and seasons; color indicates EIS. (b) Residual sensitivity after removing the linear r_e dependence, as a function of EIS; marker color indicates SST. (c) Observed sensitivity versus modeled sensitivity using r_e alone (black) or r_e plus cloud-controlling factors (red). As on Day 2, both relationships remain markedly weaker than on Day 1 ($R^2 = 0.08$ and 0.42 , respectively), showing that the delayed signal continues to fade but has not fully vanished by Day 3.

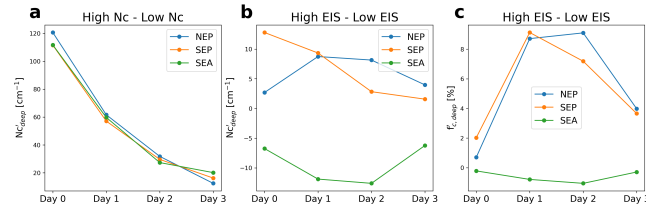


Fig. S7 Evolution of aerosol perturbations and their covariability with EIS. (a) High–low N'_c differences showing decay of the initial perturbation along trajectories. (b) EIS-associated N'_c anomalies, showing regional contrasts (negative in SEA, positive in NEP on Days 1–2). (c) Corresponding changes in thick-cloud fraction. These results indicate that regional EIS–aerosol covariability contributes to the apparent sign reversal in the EIS-related term in Fig. 4a.

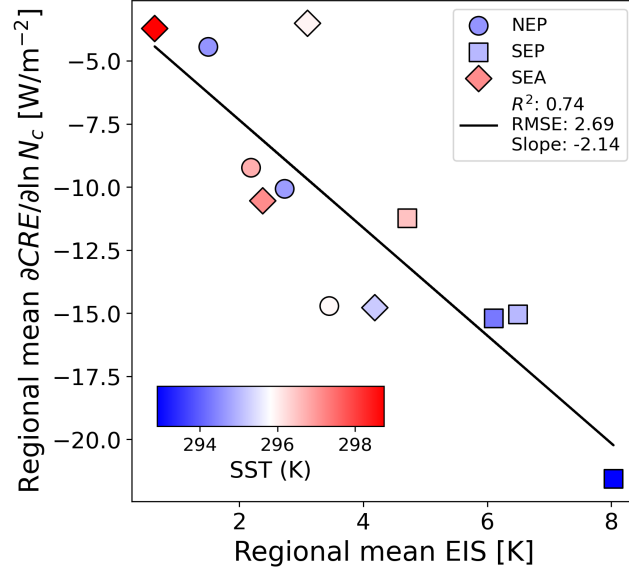


Fig. S8 Relationship between regional-seasonal mean aerosol sensitivity ($\partial CRE' / \partial \ln N_{c,0}$) and EIS. Analogous to Fig. 3b, but averaged over each region-season combination. Marker shape denotes region; marker color indicates seasonal mean SST. Black line: least-squares fit across all region-season points.

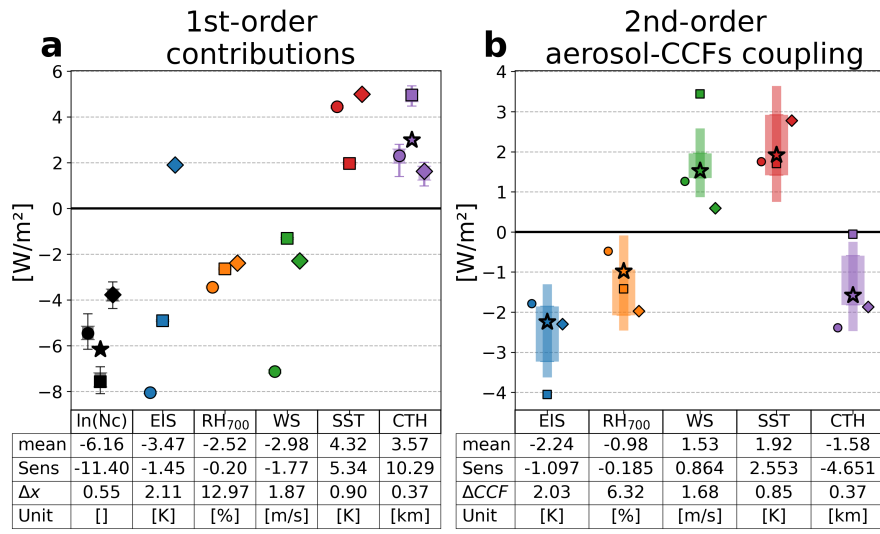


Fig. S9 First- and second-order contributions to $\Delta\text{CRE}'$ from single-variable regressions within stratified composite groups. Analogous to Fig. 4, but sensitivities are derived from single-variable rather than multilinear regressions.

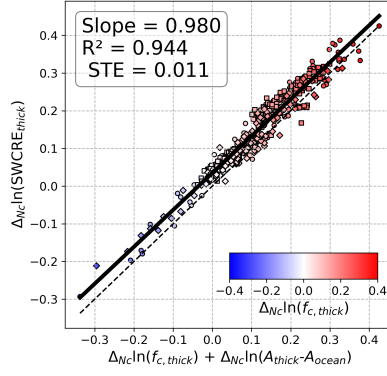


Fig. S10 Validation of the decomposition of optically thick shortwave cloud radiative effect. Scatter plot comparing the decomposed and directly computed changes in $\ln(\text{SWCRE}'_{\text{thick}})$. Δ_{N_c} denotes differences between adjacent N_c groups. The x-axis shows $\Delta_{N_c} \ln(f'_{c,\text{thick}}) + \Delta_{N_c} \ln(A'_{\text{thick}} - A_{\text{ocean}})$; the y-axis shows the corresponding directly computed $\Delta_{N_c} \ln(\text{SWCRE}'_{\text{thick}})$. Close alignment with the 1:1 line (slope 0.98, $R^2 = 0.94$) indicates the decomposition captures variability in $\text{SWCRE}'_{\text{thick}}$ with high fidelity.

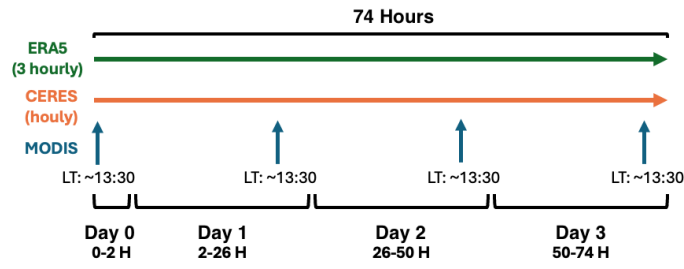


Fig. S11 Schematic illustration of the trajectory-based Lagrangian framework. Forward trajectories span 74 hours, anchored at the MODIS Aqua overpass time (Day 0; ~13:30 local time). MODIS cloud properties are sampled once per day at overpass time; ERA5 and CERES fields are sampled continuously (3-hourly and hourly, respectively). Day 0 covers 0–2 hours after overpass; Days 1–3 are successive 24-hour periods (2–26, 26–50, 50–74 hours).

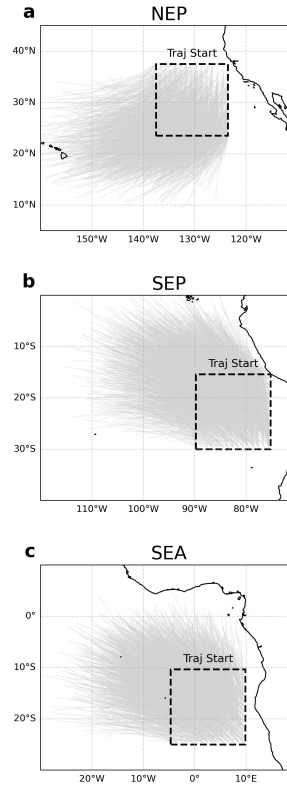


Fig. S12 Examples of filtered Lagrangian trajectories for each study region. Panels (a–c): representative valid trajectories for NEP, SEP, and SEA after applying the selection criteria described in Methods. Thin gray lines are individual 74-hour trajectories; dashed boxes denote the Day-0 starting regions.

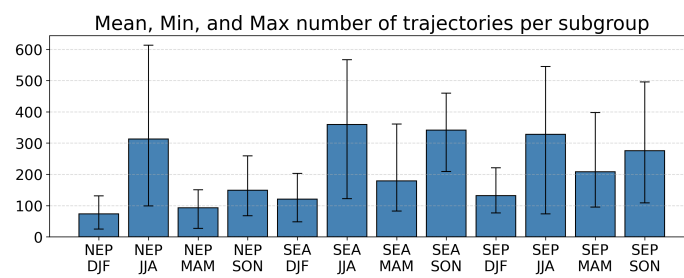


Fig. S13 Distribution of trajectory counts across CCF-stratified subgroups by region and season. Bars: mean number of valid trajectories per subgroup across the 2^5 CCF groups; error bars: minimum and maximum across subgroups.

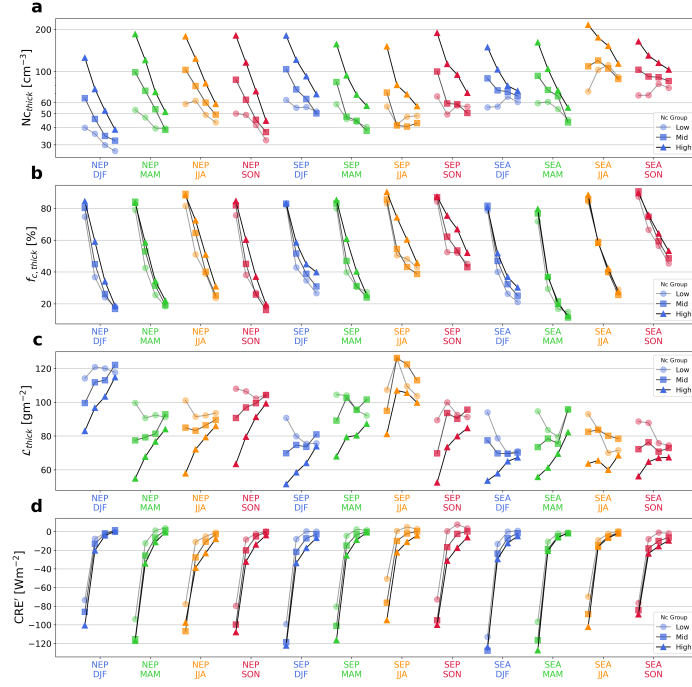


Fig. S14 Same as **Fig. S1**, but for N_c -stratified composite groups. Panels (a)–(d): daily evolution of $N_{c,thick}$, $f_{c,thick}$ and L_{thick} , and CRE' for the low, mid, and high N_c groups defined at Day 0.

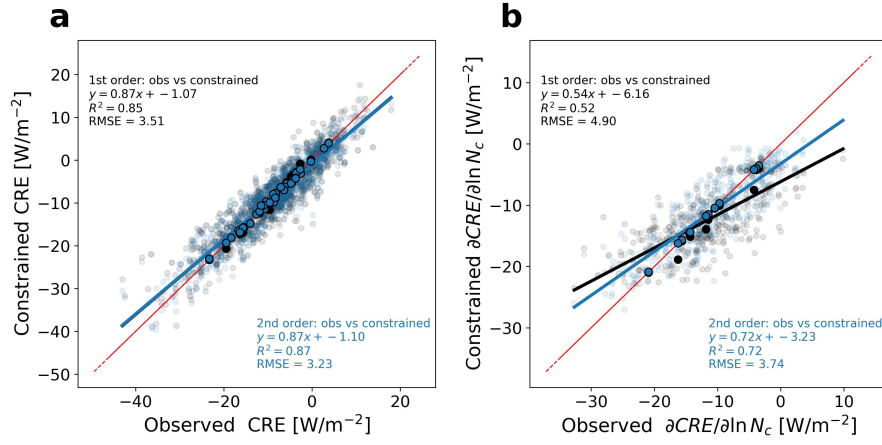


Fig. S15 Validation of constrained CRE and aerosol sensitivity estimates. (a) Observed versus constrained CRE from the first-order approximation (black) and the combined first- and second-order formulation (blue); large markers are averages of the 12 region–season combinations; red dashed line is 1:1. (b) Same as (a), but for aerosol sensitivity $\partial \text{CRE} / \partial \ln N_{c,0}$.