The effect of anthropogenic aerosol on the reflectivity of stratocumulus cloud decks through changes in cloud amount is a major uncertainty in climate projections. In frequently occurring nonprecipitating stratocumulus, cloud amount can decrease through aerosol-enhanced cloud-top mixing. The climatological relevance of this effect is debated because ship exhaust only marginally reduces stratocumulus amount. By comparing detailed numerical simulations with satellite analyses, we show that ship-track studies cannot be generalized to estimate the climatological forcing of anthropogenic aerosol. The ship track–derived sensitivity of the radiative effect of nonprecipitating stratocumulus to aerosol overestimates their cooling effect by up to 200%. The offsetting warming effect of decreasing stratocumulus amount needs to be taken into account if we are to constrain the cloud-mediated radiative forcing of anthropogenic aerosol.

Clouds interact with atmospheric radiation and therefore play an important role in the planetary energy balance. Their net effect is to cool the planet by reflecting incoming solar radiation (1). Covering large parts of the subtropical oceans, stratocumulus (Sc) clouds are by far the largest contributor to this cooling (2). Effects on cloud reflectivity caused by the production of atmospheric aerosol particles are the most uncertain anthropogenic forcing of the climate system (3, 4). As an illustration of this effect, exhaust from ships can create “ship tracks” that manifest as bright linear features in Sc decks. This brightening arises because exhaust-aerosol particles act as nuclei of cloud droplets. A greater abundance of particles means that a cloud consists of more, but smaller, droplets, which enhances the radiant energy reflected to space (5). Changes in the number and size of cloud droplets also influence cloud physical processes (6–11); for the example of ship tracks, this means that the amount of cloud water inside and outside of a track may evolve differently. Globally, the large uncertainty in the cloud-mediated aerosol forcing arises from the unknown magnitude of such adjustments of cloud water in response to aerosol-induced perturbations (3, 12, 13). Here, we show that despite providing an illustration of aerosol–cloud interactions, ship tracks do not provide suitable data to estimate the magnitude of cloud liquid-water adjustments in a polluted climate, in contrast with the common assumption that ship-track data can quantify those adjustments (14–17).

In nonprecipitating Sc with their approximately full cloud cover, cloud response to aerosol perturbations is commonly quantified by the sensitivity (4, 18, 19)

$$S = \frac{dA}{dN} = A_c \left( 1 - A_c \right) \frac{1}{3N} \left( 1 + \frac{5}{2} \frac{d \ln \text{LWP}}{d \ln N} \right)$$  \hspace{1cm} \text{(1)}

of cloud albedo $A_c$ to cloud droplet number $N$. The first term on the right-hand side of Eq. 1 quantifies the albedo effect of changing droplet number when keeping the vertically integrated amount of liquid water, or liquid-water path (LWP), constant; the second term accounts for cloud water adjustments as quantified by the relative sensitivity $d \ln \text{LWP}/d \ln N$ of LWP to $N$. Numerical values for LWP adjustments $d \ln \text{LWP}/d \ln N$ have been derived from detailed modeling and satellite studies (8, 14–17, 20–25). Both approaches have recently converged on the insight that the sign of LWP adjustments is regime-dependent (Fig. 1). Adjustments tend to be positive under precipitating conditions where the addition of particles decreases drop size, increases colloidal stability, and allows for an accumulation of liquid water (6). A positive LWP adjustment thus implies thicker, more reflective clouds that have a stronger cooling effect. In the current work, we focus on nonprecipitating Sc. Morphologically, this regime features an approximately hexagonal arrangement of cloudy (closed) cells, whereas the precipitation-dominated regime tends to occur as an inverse pattern of open cells (14, 26). Occurring in 50 to 80% of observations, the nonprecipitating regime is at least as common as the precipitating regime (25, 27). Nonprecipitating Sc feature negative adjustments, indicating a decrease in LWP for higher aerosol concentrations. The decrease in LWP stems from the accelerated and stronger evaporation of cloud liquid in higher aerosol conditions as the Sc mixes with dry air from above the cloud (entrainment). Smaller droplets evaporate more efficiently because they provide a larger surface (for a given total amount of liquid) and reside closer to the entrainment interface than larger droplets owing to reduced gravitational settling, which increases the potential for evaporation (7–11, 28). Negative LWP adjustment values indicate thinner, less reflective clouds and a weaker cooling effect. When the darkening effect of cloud thinning is stronger than the brightening of increased $N$, negative LWP adjustments can even imply a warming effect. In nonprecipitating Sc, this is the case when $d \ln \text{LWP}/d \ln N < -2/5$ such that Eq. 1 becomes negative (orange shading in Fig. 1).

In addition to the distinction between the entrainment- and precipitation-dominated regimes, satellite studies have identified above-cloud moisture as an important control on the magnitude of LWP adjustments in Sc (15, 25, 29). This is consistent with process understanding from detailed cloud modeling studies [large-eddy simulation (LES)], where drier above-cloud conditions correspond to a stronger aerosol effect on entrainment (Fig. 1). As another factor behind the variability of adjustment estimates, the effects of $N$-LWP covariability that results from large-scale co-variability of aerosol and moisture are discussed in (15, 30). As an example of this confounding effect, compare a maritime situation with a clean and moist atmosphere to a polluted and drier continental case. Observations from these two cases will likely show that higher $N$ is correlated with lower LWP, suggesting a negative LWP adjustment. Clearly, the “adjustment” quantified here is not related to the effect of aerosol on cloud properties driven by entrainment or precipitation formation that we seek to capture but rather to large-scale conditions.

A special appeal of ship tracks has been that they are not affected by external co-variability because the large-scale meteorological conditions are the same inside and outside of the track. Accordingly, results from targeted satellite analyses of ship tracks (14, 16, 20) have been assigned higher credibility than climatological satellite studies, for which external co-variability cannot be ruled out. In particular, the comparably large absolute adjustment values found in the latter studies have been attributed to aerosol-moisture co-variability, assuming that weak-to-almost absent LWP adjustments identified by ship-track studies...
This allows us to derive that the observed time dependence of LWP adjustments is well described as an exponential decay toward \( \frac{d\ln LWP}{\ln N} \) (Fig. 3B)
\[
\text{adj}(\Delta t) = \frac{d\ln LWP}{\ln N} \left[ 1 - \exp\left( -\frac{\Delta t}{\tau_{\text{adj}}} \right) \right],
\]
\[
\tau_{\text{adj}} = 2.0 \; \tau = 20 \; \text{hours}
\]
with an adjustment equilibration time scale \( \tau_{\text{adj}} \) that scales with the equilibration time scale of an individual system, \( \tau = 9.6 \) hours, and with adjustment strength (supplementary materials). The time dependence of LWP adjustments on a time scale of almost a day is in marked contrast to the radiative effect of an increased cloud droplet number, which takes full effect in 5 to 10 min (supplementary materials).

The extent and interpretation of LWP adjustments in a Sc field depends on the proximity of the system’s LWP to its steady-state LWP. Adjustments based on sampling transient LWP, far from steady state, reflect \( N \)-LWP covariability that is externally prescribed on the system, i.e., a mere association; LWP adjustments diagnosed from steady systems reflect aerosol-dependent cloud processes, i.e., a causal relationship; intermediate degrees of proximity result in a mixture of both.

**Insufficient time for evolution of ship tracks toward steady state**

The degree of proximity of an ensemble, or sampling, of Sc systems to its steady-state LWP adjustment can be estimated by comparing the duration of its evolution under an aerosol perturbation, \( \Delta t \), to the characteristic adjustment equilibration time scale, \( \tau_{\text{adj}} = 20 \) hours (Eq. 2).

From a Lagrangian perspective, a Sc system is exposed to an aerosol background throughout its lifetime: In the absence of precipitation, the lifetime of the track, with fresh tracks more likely to be sampled owing to their better visibility. With a typical lifetime for ship tracks of 6 to 7 hours (36, 37), this corresponds to an average evolution time until sampling of \( \Delta t_{\text{adj}} = 3 \) hours.

Because the characteristic equilibration time exceeds the typical evolution time at sampling, \( \Delta t_{\text{adj}} \ll \Delta t_{\text{ship}} \), we conclude that LWPs sampled from ship tracks are not representative of the aerosol-cloud interaction processes, specifically entrainment, that manifest as a Sc system approaches a steady-state LWP. Instead, their sampling of transient LWPs carries a strong imprint of their specific initial conditions. To characterize these conditions, we describe ship-track studies as a sampling within two different \( N \)-bins, one representing out-of-track conditions and the other in-track conditions (Fig. 4).

Because LWP adjustments are not instantaneous, the LWP distributions within these two bins are identical when the ship exhaust first makes contact with the cloud. As for the idealized initial conditions in our dataset, this corresponds to an initial adjustment of zero (purple regression line in Fig. 4). After the perturbation, the in-track distribution evolves toward an asymptotic LWP value that is different from that of the out-of-track LWP. Owing to the short duration of this evolution until sampling, adjustment values diagnosed from ship tracks remain small. Indeed, evolution according to Eq. 2 corresponds to an adjustment value of
\[
\text{adj}(\Delta t_{\text{ship}} = 3 \; \text{hours}) = -0.1
\]
which matches reported values ranging from -0.2 to 0.0 (table S1). By contrast, when sampling a climatologically polluted situation, adjustments can evolve toward more
negative values before being sampled and values of $\text{adj}(\Delta_{\text{clim}} = 48 \text{ hours}) = -0.6$, close to the asymptotic value of $-0.64$ (Fig. 3A), are obtained.

Although ship exhaust may, at first glance, seem to be an intriguing proxy for aerosol conditions typical of the industrial-era aerosol climatology, it does not perturb the pristine background for a sufficiently long time (Fig. 2). In other words, typical LWPs in ship tracks are not comparable to LWPs in Sc that experience a higher aerosol background owing to an anthropogenic shift of the aerosol climatology (cyan circle versus triangle in Fig. 4).

Implications for the cloud-mediated radiative forcing of anthropogenic aerosol

Ship track–derived LWP adjustments are less negative than the LWP adjustment exhibited by a Sc deck under climatologically polluted conditions. Because negative adjustments mean that an increased aerosol load leads to cloud thinning and reduced reflectivity, they imply a warming effect that offsets the cooling associated with cloud brightening (Eq. 1). Ship-track studies underestimate this offsetting warming effect of LWP adjustments (Fig. 2). We contend, therefore, that using ship track–derived adjustment values to estimate the cloud-mediated radiative forcing of anthropogenic aerosol underestimates the absolute effect of LWP adjustments on the radiative forcing. With $-0.64 < \text{dlnLWP}/\text{dlnN} < -0.4$ (as a lower bound (Fig. 3), this underestimation corresponds to an overestimation of the cooling effect of aerosols on nonprecipitating Sc of up to 200% (supplementary materials). Because nonprecipitating Sc occur frequently (25, 27), this warming effect may offset the cooling effect of positive LWP adjustments in precipitating Sc in the overall climate effect of Sc.

Our results are consistent with recent satellite estimates of LWP adjustments in Sc (15, 25). Our insight that the effects of external covariability fade as a Sc system evolves toward its internal steady state refutes $N$-LWP covariability as the likely explanation for the strongly negative adjustment values reported. At the same time, our modeling results show that strongly negative adjustment values are consistent with process understanding. In combination with the limitations of ship track–derived adjustment values discussed above, we therefore conclude that climatological satellite studies should be assigned more weight for estimating LWP adjustments than ship-track studies. Specifically, values of $\text{dlnLWP}/\text{dlnN} = -0.3$ (25) to $-0.4$ (15) should be considered possible central values rather than lower bounds, as in a recent review (4). Our analysis establishes the steady-state adjustment $\text{dlnLWP}/\text{dlnN} = -0.64$ as a new lower bound for LWP adjustments in nonprecipitating Sc.

Our results are moreover consistent with a recent study that derived LWP adjustments from climatological observations of a heavily frequented shipping lane (17). This setup provides more persistent pollution than an individual ship track while still suffering from a certain intermittency of pollution as compared with a climatological perturbation. We estimate an effective lifetime of ship tracks in a shipping lane of $\Delta_{\text{lane}} \gtrsim 9 \text{ hours}$ (supplementary materials). With an evolution time that is longer than that for individual ship tracks but shorter than Sc lifetime, it is not surprising that the shipping lane provides a numerical adjustment value that lies in between those derived from single-ship-track studies and fully climatological studies (Fig. 1). Our results therefore reconcile and explain the differing LWP adjustments that have recently been reported (15–17, 25).

Satellite remote sensing of thin and broken clouds remains a challenge, with large uncertainties in retrieved values. Despite the support for climatological satellite studies that our results provide, it therefore seems desirable to identify alternative to ship-track studies that allow for a direct observation of aerosol effects. Our analysis shows that suitable natural experiments should feature temporally continuous pollution. Spatial continuity of pollution is another criterion, which excludes biases from boundary effects as described for ship tracks (38, 39). Effusive volcanic emission and oceanic outflows of continental air are examples of such continuously polluting natural experiments. Existing datasets do not sample the subtropical Sc regions, however (16). In addition to adjustments being cloud-regime specific, higher extratropical above-cloud moisture may bias toward less-negative values. Undersampling of the sub tropics may also exacerbate the time-scale effect, leading to underestimation of negative anthropogenic LWP adjustments by ship-track data.

Deliberate experiments could, by design, provide suitable aerosol perturbations. Such experiments have been suggested to assess the feasibility of marine cloud brightening (MCB) (40), i.e., the inadvertent mitigation of climate forcing by injecting aerosol into extensive Sc decks. In contrast to a setup with persistent pollution as needed to estimate the climatological aerosol effect, MCB rests on the notion of weak LWP adjustments as observed in ship tracks. Our results indicate that an intermittent aerosol perturbation may maximize the cooling effect by limiting the magnitude of compensating adjustments. To be feasible, MCB strategies will therefore have to balance the potential for LWP adjustments discussed here with the total aerosol perturbation obtainable with a given installation.

There is an urgent need to quantify the albedo and LWP responses in both precipitating and nonprecipitating Sc cloud systems to successfully quantify the cloud-mediated effect of anthropogenic aerosol on the climate system. This will require careful assessment of the frequency of occurrence and areal coverage of these regimes, with attendant consideration of the temporal nature of the LWP responses. Estimates of aerosol-cloud forcing that ignore the nonprecipitating regime are likely to substantially overestimate climate cooling.

REFERENCES AND NOTES


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SUPPLEMENTARY MATERIALS
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Materials and Methods
Supplementary Text
Figs. S1 to S6
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Aerosol-cloud-climate cooling overestimated by ship-track data
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Making tracks
The magnitude of the effect of anthropogenic aerosols on the formation of clouds is an important unknown about how humans are affecting climate. Studies of stratocumulus cloud tracks that are formed by ship exhaust have been used to estimate the radiative impact of this process, but Glassmeier et al. now show that this approach overestimates the cooling effect of aerosol addition by up to 200%. These findings underscore the need to quantify stratocumulus cloud responses to anthropogenic aerosols to understand the climate system.
Science, this issue p. 485
Supplementary Materials for

Aerosol-cloud-climate cooling overestimated by ship-track data

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This PDF file includes:

Materials and Methods
Supplementary Text
Figs. S1 to S6
Tables S1 to S4
References
Materials and Methods

Approach

Our analysis builds on relating satellite datasets to LES. We make these two data sources comparable by creating an ensemble of 144 LES runs that resembles the scope of a satellite dataset in that it samples a broad range of LWP and \( N \) conditions (see Dataset, Fig. S1). In contrast to satellite data, we prevent externally-induced \( N \)-LWP co-variability by sampling initial conditions of ensemble members in a statistically independent way (see Dataset). We furthermore limit externally-induced variability in LWP adjustments by fixing external control parameters of Sc (Table S2). We specifically restrict above-cloud moisture to values \( q_t < 2.8 \text{ g/kg} \) with a median value of \( 0.5 \text{ g/kg} \). This choice of very dry above-cloud conditions allows us to derive a lower bound for LWP adjustments.

We approach our investigation of LWP adjustments to \( N \) perturbations by analyzing the temporal co-evolution of LWP and \( N \) collectively for all members of the Sc ensemble (Fig. S1). In the LWP direction, individual ensemble members collectively evolve towards similar LWPs (along an approximately horizontal line in Fig. S1). For these steady-state LWPs, there exists a balance in the contributions of different processes that are source and sink terms for LWP - in particular radiative cooling (source), and entrainment and precipitation drying (sink) \([31]\). The collective evolution in the \( N \) direction is structured around the critical radius for precipitation formation \([43]\). Precipitating systems (above the dashed line in Fig. S1) contain sufficient drops with radii larger than the critical radius, are colloidally unstable, and feature a rapid reduction in \( N \). For systems with smaller radii (below the dashed line), which are our focus, rain is scant and entrainment dominates.

The individual evolution of Sc cloud fields in the ensemble can be represented as flow vectors \( \vec{v} = (d \ln N/dt, d \ln \text{LWP}/dt) \) in \( N \)-LWP space. Gaussian-process emulation allows us to interpolate such flow vectors from our limited number of simulations (Fig. S1A) to obtain the full flow field illustrated in Fig. S1B (see Derivation of the flow field). This interpolation of the flow field enables us to infer cloud behavior beyond the 12h-duration of our simulations, including the behavior when an LWP steady state (\( d \ln \text{LWP}/dt = 0 \)) is reached (blue curve in Fig. S1B). This enables us to systematically quantify the time-dependence of LWP adjustments over timescales longer than the duration of our simulations. The flow field representation also allows us to determine that individual Sc systems equilibrate to their steady-state with a characteristic equilibration time scale of \( \tau = 9.6 \text{ h} \) (see Characteristic equilibration timescale of individual Sc systems), in excellent agreement with a theoretical estimate \([32]\), and in line with a recent observational study \([45]\). This timescale informs us about the proximity of an observed Sc system to its steady state.
**Dataset**

This study is based on the ensemble of LESs described in reference [41] and references therein. As verbatim indicated in [41], simulations are performed with the System for Atmospheric Modeling (SAM). The domain measures 48 km × 48 km at a horizontal resolution of 200 m and a vertical resolution of 10 m. The time step is 1 s. Simulations are nocturnal and of 12 h duration. The model activates aerosol particles based on the prognosed supersaturation and simulates condensation and/or evaporation and collision-coalescence using a bin-emulating 2-moment approach. Particles are removed by collision-coalescence, scavenging and wet deposition. We assume a surface source of aerosol particles of 70 cm$^{-2}$s$^{-1}$.

In comparison to the original dataset, we have excluded outliers in terms of above-cloud humidity, which results in a dataset of 144 LES runs. Large-scale horizontal wind divergence (subsidence) and surface fluxes are the same across the LES ensemble and are summarized in Table S2. In reality, subsidence and surface fluxes vary. And although they do not directly depend on $N$, they may move the steady state LWP [31] into a region in which precipitation and entrainment have different susceptibilities to $N$, resulting in commensurate changes in the $\frac{d \ln \text{LWP}}{d \ln N}$ slope. Since these changes seem to saturate for larger LWPs [31], we are confident that the $\frac{d \ln \text{LWP}}{d \ln N}$ slope discussed in this study is representative for a large range of natural stratocumulus, especially subtropical stratocumulus [35].

Variability of LWP within the ensemble is achieved by varying the initial profiles of temperature and moisture; individual simulations vary in $N$ because they have been initialized with varying aerosol backgrounds (Table S3). Following reference [42], we prevent co-variability among the initial conditions by means of a 6D latin-hypercube sampling of the internal factors listed in Table S3. For the derivation of the flow field, additional simulations have been added to achieve better coverage of the $N$-LWP space. The dataset and its variants are illustrated in Fig. S2.

**Derivation of the flow field $\vec{v}(N, \text{LWP})$**

The flow field $\vec{v} = (d \ln N/dt, d \ln \text{LWP}/dt)^T = (v_N, v_{\text{LWP}})^T$ shown in Fig. S1B is based on separately deriving the components in the $N$-direction, $v_N$, and the LWP-direction, $v_{\text{LWP}}$ (Fig. S3). To derive these component fields, we first extract tendencies $d \ln N/dt$ and $d \ln \text{LWP}/dt$ from the data and then interpolate the extracted tendencies by means of Gaussian-process emulation to obtain emulators for the tendency surfaces.

To derive tendencies, we split each simulated time-series into six intervals of 100 min duration, each of which contains 10 consecutive data points at a 10 min output frequency. For each of these, we determine tendencies by fitting trend lines. We only consider significant trends (p-value < 0.05) and assign a value of zero otherwise. To account for oscillatory
behavior that occurs because some of our simulations remain influenced by spin-up processes, we assume that the last 100 min-segment of each time-series provides the correct sign of the evolution. The previous segments are then only considered if they feature the same sign. With these restrictions, we obtain a dataset of 828 LWP-tendencies and 783 $N$-tendencies.

We process these datasets largely in the same way as described in detail in Section 3 of reference [41]. Here we only mention adaptations to the technical parameters mentioned therein. Instead of a 50%-50% split of the dataset into training and validation data, we use a smaller fraction of training data (33%) to ensure good validation. To obtain an ensemble of 5 emulated surfaces for $v_{\text{LWP}}$ and $v_{N}$, we do not restrict the fraction of the training data to be used for individual ensemble members.

Quantification and uncertainty of LWP adjustment values in steady state

As a result of our interpolation technique, we obtain a mean emulator surface $v_{\text{LWP}}(N, \text{LWP})$, to which individual emulator ensemble members contribute according to their root-mean-square error (RMSE) in predicting the validation data (Eq. 3 of reference [41]). The central value for the LWP adjustment is based on the zero-contour of this mean surface. For the uncertainty percentiles, we determine LWP adjustments from the zero-contours of a specific sampling of the emulator ensemble. For this sampling, we take 100 samples of each of the 5 ensemble members and then select a subset of these 500 samples, such that each ensemble member contributes proportionally to its RMSE-based weight, i.e. for the ensemble member with the lowest RMSE, all 100 samples are considered, and for the ensemble member with the highest RMSE, no samples are considered. Table S4 summarizes the uncertainty ranges obtained in this way for the LWP adjustment value.

Supplementary Text

Timescale for the radiative effect of an increased cloud droplet number

The timescale for the radiative effect of an increased cloud droplet number is determined by the time it takes for a droplet to travel from cloud base, where an aerosol perturbation first takes effect through cloud droplet activation, to the top of the cloud. Assuming an updraft of 0.5–1 m s$^{-1}$ and a cloud thickness of 300 m results in the 5–10 min mentioned in the main text.
Characteristic equilibration timescale of individual Sc systems

The characteristic equilibration timescale $\tau$ for the equilibration of the LWP of an individual Sc system to its steady-state value $\text{LWP}_\infty$ can be determined by linear stability analysis about $\text{LWP}_\infty$. It is given by

$$\tau^{-1} = \left| \frac{\partial v_{\text{LWP}}}{\partial \ln \text{LWP}} \right|_{\text{LWP}_\infty}, \quad (S1)$$

where the derivative (color contours in Fig. S4) is evaluated at $\text{LWP}_\infty$. Practically, we average Eq. S1 over a steady-state region (white contour in Fig. S4), bounded by the 25th and 75th uncertainty percentile of the location of $v_{\text{LWP}} = 0$ (dashed blue lines as in Fig. S3A) within the entrainment dominated regime ($N > 100 \, \text{cm}^{-3}$). The characteristic equilibration timescale determined in this manner yields $\tau = 9.6 \, \text{h}$, as mentioned in the main text.

While the characteristic equilibration timescale is similar to the autocorrelation timescale of LWP in Lagrangian trajectories of stratocumulus ($15 - 17 \, \text{h} \, [45]$), it is much shorter than the timescale characterizing changes in the boundary layer height ($3 - 6 \, \text{d} \, [32]$). These two very different timescales must be understood as different manifolds of the stratocumulus evolution [9]: The background conditions characterized by the boundary-layer height vary so slowly (the slow manifold) that fast-changing stratocumulus properties (the LWP and other thermodynamic quantities; the fast manifold) are able to adapt to the slowly varying boundary-layer height. Accordingly, the LWP may be in a steady state, while the boundary layer height is not.

Derivation of the adjustment equilibration timescale

To derive the adjustment equilibration timescale, we determine the time required by the entire ensemble of LWPs to reach their respective steady states. We assume that the LWP for any $N$ approaches its steady state $\text{LWP}_\infty(N)$ with approximately the same velocity $[\text{LWP}_{\text{ini}} - \text{LWP}_\infty(N_0)]/\tau$ controlled by the characteristic equilibration timescale $\tau = 9.6 \, \text{h}$ (Fig. S4), where $\text{LWP}_{\text{ini}}$ is an initial non-steady-state LWP. Hence, the linearized exponential change in LWP yields

$$\text{LWP}(t, N) = \text{LWP}_{\text{ini}} - \frac{t}{\tau} [\text{LWP}_{\text{ini}} - \text{LWP}_\infty(N_0)], \quad (S2)$$

where $\text{LWP}_\infty(N_0)$ is the steady-state LWP for the smallest $N$ in the non-precipitating regime, $N_0$. Note that we focus on the LWPs that approach the steady state from larger values since only those require a longer time to reach the steady state for larger $N$.

The steady-state LWP as a function of $N$ is obtained by integrating and linearizing the
adjustment \( \frac{d \ln \text{LWP}}{d \ln N} \),

\[
\text{LWP}_\infty(N) = \text{LWP}_\infty(N_0) \left[ 1 + \frac{\frac{d \ln \text{LWP}_\infty}{d \ln N}}{\ln \left( \frac{N}{N_0} \right)} \right], \tag{S3}
\]

using the same constants of integration as above. Combining Eq. S2 and Eq. S3, and solving for \( t \equiv \tau_{\text{adj}} \), gives the adjustment equilibration timescale necessary to equilibrate the entire system,

\[
\tau_{\text{adj}} = \tau \left[ 1 - \frac{\frac{d \ln \text{LWP}_\infty}{d \ln N}}{\ln \left( \frac{N_2}{N_0} \right)} \frac{\text{LWP}_\infty(N_0)}{\text{LWP}_{\text{ini}} - \text{LWP}_\infty(N_0)} \right], \tag{S4}
\]

where \( N = N_2 \) is the largest droplet concentration in the non-precipitating regime, resulting in the longest time to equilibrate the LWP. With \( N_0 = 107 \text{ cm}^{-3} \), \( N_2 = 390 \text{ cm}^{-3} \) and \( \text{LWP}_\infty(N_0) = 89 \text{ g m}^{-2} \) in the smallest (index 0) and largest (index 2) \( N \)-bin (see Fig. S1A for bin specifications; note the logarithmic scale), and \( \frac{d \ln \text{LWP}_\infty}{d \ln N} = -0.64 \), we obtain best fitting results for \( \tau_{\text{adj}} \) when assuming \( \text{LWP}_{\text{ini}} = 159 \text{ g m}^{-2} \), which amounts to the 78th percentile of LWP in the \( N_2 \)-bin. These numerical values provide the adjustment equilibration timescale of 20 h stated in Eq. 2.

**Effective evolution time of ship tracks in a shipping lane**

Based on the lower-bound steady-state adjustment value \( \frac{d \ln \text{LWP}_\infty}{d \ln N} = -0.64 \), the effective evolution time \( \Delta t_{\text{lane}} \) of a ship track in a shipping lane can be estimated based on Eq. 2,

\[
\text{adj}(\Delta t_{\text{lane}}) = -0.24 \Rightarrow \Delta t_{\text{lane}} \gtrsim 9 \text{ h}, \tag{S5}
\]

where we have used the observed adjustment value \( \frac{d \ln \text{LWP}}{d \ln N} = -0.24 \) from reference [17].

**Cloud-mediated aerosol forcing and cloud radiative effect**

The cloud-mediated aerosol forcing depends on the aerosol sensitivity of the relative cloud radiative effect, \( \text{rCRE} \), which relates downwelling short-wave radiative fluxes at the surface, \( F \), under clear-sky (index clr) and all-sky (index all) conditions [44] and references therein,

\[
\text{rCRE} = \frac{F_{\text{clr}} - F_{\text{all}}}{F_{\text{clr}}} \approx \text{CF} \cdot A_c \approx A_c \tag{S6}
\]

and amounts to cloud albedo \( A_c \) in fully overcast Sc with cloud fraction \( \text{CF} \approx 1 \). We assume that climatological cloud properties can be approximated by steady-state values. With a steady-state cloud albedo of \( A_c = 0.5 \) based on Fig. S6, Eq. 1 for the sensitivity of \( A_c \), or
rCRE, respectively, results in

\[ S = \frac{1}{N} \left( \frac{1}{12} + \frac{5}{24} \frac{d \ln \text{LWP}}{d \ln N} \right). \]  \tag{S7}

With \( \frac{d \ln \text{LWP}}{d \ln N} \approx -0.1 \) (Eq. 3), ship-track studies imply \( S_{\text{ship}} \approx 0.06/N > 0 \) and thus a cooling effect of anthropogenic aerosol via increased cloud brightness at almost constant LWP. In contrast, the steady-state adjustment value of \( \frac{d \ln \text{LWP}_\infty}{d \ln N} = -0.64 \) derived here as a lower bound results in \( S_{\text{clim}} = -0.05/N < 0 \), which indicates that aerosol-induced cloud thinning overcompensates the brightening effect at constant LWP. Ship-track studies thus overestimate the cooling effect of aerosol on Sc by up to \(|(-0.05 - 0.06)/(-0.05)| = 220\% \approx 200\%\).
Fig. S1 (A).

A

(See next page for continuation of figure) LES ensemble dataset and corresponding temporal co-evolution of in-cloud liquid-water path LWP and cloud-droplet number $N$. We focus on the non-precipitating regime below the dashed blue line indicating the critical radius for precipitation formation as in Fig. 1. (A) Temporal evolution of a sample of 144 LES runs with varying initial conditions. Individual simulation runs are indicated by gray lines connecting gray circles. Start (2 h into the simulation to allow for model spin-up, magenta) and end (12 h, green) of a trajectory are color-highlighted. The solid blue line shows the steady-state LWP from (B). Thin vertical black lines indicate the boundaries of $N$-bins used in Fig. 3.
Fig. S1 (B).

(B) Flow-field representation of $N$-LWP co-evolution and location of steady-state LWP (blue line and 25th/75th percentile uncertainty shading, Table S4), which is characterized by $\frac{\text{d} \ln \text{LWP}}{\text{d} t} = 0$. 

$\tau_{\text{cr}} = 12 \mu m$
Dataset illustrated similar to Fig. S1A. Trajectories in faint coloring, whose start is indicated by a cross rather than a circle, indicate runs that were excluded for this study in comparison to the dataset described in reference [41] due to their above-cloud moisture being an outlier. Open magenta-colored circles indicate additional simulations only considered for deriving the flow field \( \vec{v} \). The coloring of trajectories indicates the fractional contribution of in-cloud rain water path RWP to total in-cloud liquid water path LWP.
Fig. S3 (A).

A

Emulated surfaces (average surface from an ensemble of emulators) of (A) $v_{LWP} = \frac{d \ln LWP}{d t}$ and (B) $v_N = \frac{d \ln N}{d t}$ of the flow field $\vec{v}$ as a function of in-cloud LWP and droplet number $N$. The dark gray area confines the convex hull of data points, within which interpolation is possible. The goodness of fit of the emulated surfaces as compared to the validation data set is illustrated by color-filled circles and by the one-to-one scatter plots in the insets, which also indicate a correlation coefficient $r$ and a $p$-value for a linear relationship.
(Continuation from previous page) Hatching indicates the number of training data points $n_{\text{trn}}$ per bin, for a $10 \times 12$ binning of the $N$-LWP space. As discussed in reference [41], insufficient sampling is the largest source of uncertainty. Blue contour lines in (A) indicate $v_{\text{LWP}} = 0$, i.e., LWP = $LWP_\infty$, for the (dashed) 25th, (solid) 50th and (dashed) 75th percentile of a RMSE-weighted sampling from the emulator ensemble (see text). Blue curves in Fig. 3 and S1 correspond to the median sampling (solid blue contour).
Determination of the characteristic equilibration timescale $\tau$ according to Eq. S1 (color contours). The white contour indicates the steady-state region, bounded by the 25th and 75th uncertainty percentile of the location of $v_{LWP} = 0$ (dashed blue lines in Fig. S3A) and restricted to the non-precipitating regime ($N > 100\, \text{cm}^{-3}$). An average value of $\partial v_{LWP}/\partial \ln \text{LWP} = -2.49\, \text{day}^{-1}$ within the white contour indicates a characteristic equilibration timescale of $\tau = 9.6\, \text{h}$. Sampling density as in Fig. S3.
Climatology (1973-2020) of above-cloud mixing ratio at San Clemente Island (Station NSI 72291) in the Sc region off the coast of California. Mixing ratios are obtained at a height of $1.05z_i$, where $z_i$ denotes the inversion height as determined from the maximum gradient in the equivalent potential temperature. Only soundings that reach saturation in the lowest 2000 m were considered. Radiosonde data kindly provided by University of Wyoming (http://www.weather.uwyo.edu/upperair/sounding.html).
Emulated values for cloud albedo $A_c$ as function of cloud droplet number $N$ and in-cloud liquid-water path LWP (re-plot of results from reference [41]). The solid blue line indicates the location of the steady-state LWP as in Fig. 3. The white contour indicates the steady-state region as in Fig. S4. The average cloud albedo within this white contour amounts to $A_c = 0.46$. 
Literature values used in Fig. 1 and details about their derivation. Abbreviations “entr” and “prcp” refer to the entrainment-dominated/non-precipitating and precipitation-dominated regimes of Sc, respectively. Data source “satellite” refers to climatological satellite studies, while “ship track” refers to ship track studies from satellite. The “data type” column indicates whether adjustment information was derived as difference quotient $\Delta \ln \text{LWP}/\Delta \ln \text{N}$ from value pairs, obtained as gradient $d \ln \text{LWP}/d \ln \text{N}$ of linear regression lines through a provided list of data points, or directly provided in one of these two forms. In one case, $\text{N}$ was inferred from cloud optical thickness $\tau$ and effective radius $r_{\text{eff}}$.

<table>
<thead>
<tr>
<th>Table S1.</th>
<th>entr</th>
<th>prcp</th>
<th>reference details</th>
<th>data source</th>
<th>data type</th>
<th>comments</th>
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</thead>
<tbody>
<tr>
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<td>0.21</td>
<td>Fig. 3, Table S4</td>
<td>LES ensemble</td>
<td>regression</td>
<td>steady-state value</td>
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<td>[8]</td>
<td>-0.27</td>
<td>0.24</td>
<td>Supplementary Table 1, prcp: ASTEX, FIRE1, entr: DYCOMS-RF1</td>
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<td>difference quotient</td>
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<tr>
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<td>-</td>
<td>Table 3, HOM_FULL</td>
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<td>LWP, $N$ (lists)</td>
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<td>[22]</td>
<td>0.11</td>
<td>-</td>
<td>Figs. 4 and 8, MID-WET</td>
<td>LES</td>
<td>LWP, $N$ (pairs)</td>
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<td>[15]</td>
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<td>0.24</td>
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<td>LES</td>
<td>LWP, $N$ (pairs)</td>
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<td>[24]</td>
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<td>regression</td>
<td>-</td>
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<tr>
<td>[25]</td>
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<td>Table 1, LTS &gt; 18</td>
<td>satellite</td>
<td>regression</td>
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<td>[16]</td>
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<td>0.14</td>
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<td>regression</td>
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<td>[44]</td>
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<td>Fig. 4, LTS &gt; 15 K, prcp threshold: dBZe &lt; -20</td>
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<td>regression</td>
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<td>[14]</td>
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<td>[17]</td>
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Table S2.

<table>
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<tr>
<th>Parameter</th>
<th>Value</th>
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<tr>
<td>horizontal wind divergence</td>
<td>(3.75 \times 10^{-6}) s(^{-1})</td>
</tr>
<tr>
<td>sensible heat flux</td>
<td>16 W m(^{-2})</td>
</tr>
<tr>
<td>latent heat flux</td>
<td>93 W m(^{-2})</td>
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<tr>
<td>aerosol surface flux</td>
<td>70 cm(^{-2}) s(^{-1})</td>
</tr>
<tr>
<td>above-cloud moisture</td>
<td>0.5 [0.2, 2.8] g kg(^{-1})</td>
</tr>
<tr>
<td>radiation</td>
<td>nocturnal</td>
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</tbody>
</table>

External simulation parameters given by large-scale conditions following reference [41] and references therein. Values of above-cloud moisture refer to the median and, in brackets, the minimum and maximum value within the distribution.
<table>
<thead>
<tr>
<th>Variable</th>
<th>Range</th>
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<td>mixed-layer height $h$ (in m)</td>
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<td>mixed-layer aerosol concentration $N_a$ (in cm$^{-3}$)</td>
<td>[30, 500]</td>
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<tr>
<td>mixed-layer liquid-water potential temperature $\theta_l$ (in K)</td>
<td>[284, 294]</td>
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<tr>
<td>mixed-layer moisture $q_t$ (in g kg$^{-1}$)</td>
<td>[6.5, 10.5]</td>
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<tr>
<td>liquid-water potential temperature inversion strength at $h$, $\Delta\theta_l$ (in K)</td>
<td>[6, 10]</td>
</tr>
<tr>
<td>moisture inversion strength at $h$, $\Delta q_t$ (in g kg$^{-1}$)</td>
<td>[−10, −6]</td>
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</table>

Ranges of values that span the initial conditions of internal variables for the ensemble of LES runs used in this study, following reference [42] and assuming well-mixed initial profiles.
### Table S4.

<table>
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<th>75th</th>
<th>95th</th>
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<td>non-precipitating</td>
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<td>-0.64</td>
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<td>-0.34</td>
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<tr>
<td>precipitating</td>
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<td>0.22</td>
<td>0.21</td>
<td>0.39</td>
<td>0.68</td>
</tr>
</tbody>
</table>

Uncertainty quantification for LWP adjustment values in steady state. See [Quantification and uncertainty of LWP adjustment values in steady state](#) for details.
References and Notes


