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

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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 non-precipitating 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 non-precipitating 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 sub-tropical 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 a striking 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, 7, 8, 9, 10, 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 [12, 3, 13]. Here we show that, despite providing a striking 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, 15, 16, 17].

In non-precipitating Sc with their approximately full cloud cover, cloud response to aerosol perturbations is commonly quantified by the sensitivity [4, 18, 19]

$$S = \frac{dA_c}{dN} = \frac{A_c(1-A_c)}{3N}\left(1 + \frac{5}{2}\frac{d\ln LWP}{d\ln N}\right) \quad (1)$$

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 $d\ln LWP/d\ln N$ of LWP to N. Numerical values for LWP-adjustments $d\ln LWP/d\ln N$ have been derived from detailed modeling and satellite studies [20, 8, 21, 22, 23, 14, 15, 24, 25, 16, 17]. 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 non-precipitating Sc. Morphologically, this regime features an approximately hexagonal arrangement of cloudy (closed) cells, while the precipitation-dominated regime tends to occur as an inverse pattern of open cells [14, 26]. Occurring in 50% to 80% of observations, the non-precipitating is at least as common as the precipitating regime [27, 25]. Non-precipitating 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 due to reduced gravitational settling, which increases the potential for evaporation [7, 8, 9, 28, 10, 11]. 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 non-precipitating Sc, this is the case when $d\ln LWP/d\ln N < -2/5$ 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 [29, 15, 25]. 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, references [15, 30] discuss the effects of N-LWP co-variability that results from large-scale co-variability of aerosol and moisture. 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 [20, 14, 16] 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 [20, 14, 16] provide the best estimate for LWP adjustment. In contrast to this assumption, a recent study of shipping lanes reports significantly negative adjustment values [17].

We show that the current emphasis on satellite studies of ship tracks to estimate LWP adjustments leads to an overestimation of the cooling effect of aerosols in Sc. We furthermore reconcile the broad range of reported adjustment estimates and discuss implications of our results for identifying alternatives to ship-track studies. Our argument is illustrated in Fig. 2 and builds on two key results: Firstly, LWP adjustments become more negative as Sc decks evolve towards a steady-state. Secondly, in ship tracks, this temporal evolution does not proceed long enough to be representative of Sc decks in a polluted climate.

Effect of cloud field evolution towards steady state on adjustment strength.

Climatological satellite studies derive LWP adjustments $d\ln LWP/d\ln N$ as slopes of linear regression lines through median LWP values in N-bins. We apply this methodology to an ensemble of 12h-long LES timeseries that resembles the scope of a satellite dataset (Supplementary Information). To discuss the time-dependence of adjustments, we separately derive LWP-adjustments per time-step. Figs. 3A and S1A illustrate this for $t = 2\mathrm{h}$ (magenta) and $t = 12\mathrm{h}$ (green). Considering all time steps $2 \leq t/h \leq 12$ shows that the LWP adjustment becomes increasingly negative over time (Fig. 3B). This behavior results from the sampling of the N-LWP space by our simulations, which evolves over time. By construction, our dataset initially features an uncorrelated sampling (Supplementary Information). This explains the almost horizontal regression line and corresponding vanishing adjustment observed at $t = 2\mathrm{h}$. An initial co-variability of N and LWP values would have imprinted an initial correlation and corresponding adjustment value between N and LWP.

As our simulations collectively evolve further from the initial state, they approach a steady-state LWP line (blue curve in Figs. 3A, S1) and the sampling of the N-LWP space features an increasingly negative correlation. This relationship replaces the initial relationship because the evolution speed of Sc systems scales with their distance to the steady-state line (Fig. S3). Had we run our simulations beyond 12h, all ensemble members would eventually have reached their steady-state LWP. This means that for $t \to \infty$ only the steady-state line is sampled and the LWP adjustment is quantified by the slope of this line. As the slope of the steady-state LWP line reflects the N-dependence of entrainment [33], the LWP-adjustment at $t \to \infty$, $d\ln LWP_\infty/d\ln N$, is a direct quantification of N– or more generally aerosol– effects on cloud processes.

Employing Gaussian-process emulation, we determine the location of this steady-state line in LWP- N from limited-duration timeseries (Supplementary Information). For non-precipitating Sc, we obtain $d\ln LWP_\infty/d\ln N = -0.64$ (Fig. 3A; uncertainty quantification in Tab. S4). This value constitutes a lower bound; a more negative adjustment value would require a stronger N-dependent entrainment and therefore drier above-cloud conditions than prescribed for our simulations. This is not realistic since our simulations feature very dry conditions already (Tab. S2, Fig. S5). Fig. 1 also supports $-0.64 \leq d\ln LWP/d\ln N$ as a lower bound on previous estimates from the literature. We contrast this value with the positive value of the precipitation-dominated branch, for which we determine a slope of 0.21 (Tab. S4) that lies well within the reported range (Fig. 1).

The equilibration of adjustments to the steady-state value is the collective result of the equilibration of individual Sc systems. This allows us to derive that the observed time-dependence of LWP adjustments is well-described as an exponential decay towards $d\ln LWP_\infty/d\ln N$ (Fig. 3B),

$$
\begin{aligned}
&adj(\Delta t) = \frac{d\ln LWP_\infty}{d\ln N}\left[1 - \exp\left(-\frac{\Delta t}{\tau_{adj}}\right)\right],\\
&\tau_{adj} \approx \tau\left(1 - 1.6\,\frac{d\ln LWP_\infty}{d\ln N}\right) = 2.0\,\tau = 20\ h,
\end{aligned}
\tag{2}
$$

with an adjustment equilibration timescale τ_{adj} that scales with the equilibration timescale of an individual system, $\tau = 9.6h$, and with adjustment strength (Supplementary Information). The time-dependence of LWP adjustments on a timescale of almost a day is in stark contrast to the radiative effect of an increased cloud droplet number, which takes full effect in $5 - 10$ minutes (Supplementary Information).

In summary, 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 co-variability 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 towards 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, Δt, to the characteristic adjustment equilibration timescale, $\tau_{adj} = 20\mathrm{h}$ (Eq. 2). From a Lagrangian perspective, a Sc system is exposed to an aerosol background throughout its lifetime: in the absence of precipitation, the aerosol co-evolves with the boundary layer height [25], i.e. on a multiday-long timescale [35]. Typical Sc trajectories in the subtropics persist on timescales of days, $\Delta t_{clim} > 48\mathrm{h}$, before they transition into the shallow cumulus regime due to advection towards higher sea-surface temperatures [36]. Since $\Delta t_{clim} \gg \tau_{adj}$, the climatological sampling of Sc is dominated by strongly equilibrated LWPs. While not necessarily composed of steady-state LWPs, we can assume that the LWP climatology of non-precipitating Sc is better characterized as a sampling of steady-state LWPs, than as one of highly transient LWPs. Steady-state values as a feasible approximation for Sc properties are in line with previous theoretical studies [37]. A significant probability of Sc being observed close to their steady state is also consistent with relatively narrow climatological distributions of Sc LWPs [15, 38] that indicates dominant sampling of a steady-state line. The scatter around this line corresponds to transient LWPs.

Sc decks being strongly adjusted to the aerosol background in which they evolve has implications for constraining the anthropogenic radiative forcing; LWP adjustments need to compare Sc that are strongly adjusted to an aerosol background typical of an industrial-era aerosol climatology (Fig. 4, cyan circle) to Sc decks that are strongly adjusted to a pre-industrial aerosol background (orange circle). Climatological satellite studies are suitable for this quantification because they predominantly sample strongly-adjusted LWPs close to steady state. As discussed in the previous section, this specifically means that such studies capture cloud processes, and are only weakly confounded by externally-induced N-LWP co-variability.

Ship-track data are obtained throughout the life of the track, with fresh tracks more likely to be sampled due to their better visibility. With a typical lifetime for ship tracks of $6 - 7\mathrm{h}$ [39, 40], this corresponds to an average evolution time until sampling of $\Delta t_{ship} \approx 3\mathrm{h}$. As the characteristic equilibration time exceeds the typical evolution time at sampling, $\Delta t_{ship} \ll \tau_{adj}$, 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, and the other in-track conditions (Fig. 4). As 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 (Fig. 4, purple regression line). After the perturbation, the in-track distribution evolves to an asymptotic LWP value that is different from that of the out-of-track LWP. Due 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

$$adj(\Delta t_{ship} = 3\ h) = -0.1, \tag{3}$$

which matches reported values ranging from -0.2 to 0.0 (Tab. S1). When, in contrast, sampling a climatologically polluted situation, adjustments can evolve to more negative values before being sampled and values of $adj(\Delta t_{clim} = 48\ h) \approx -0.6$, close to the asymptotic value of -0.64 (Fig. 3A), are obtained.

While ship exhaust may at first glance seem 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 due to an anthropogenic shift of the aerosol climatology (Fig. 4, cyan circle vs triangle).

Implications for the effective radiative forcing due to aerosol-cloud interactions.

Ship-track-derived LWP adjustments are less negative than the LWP adjustment that a Sc deck under climatologically-polluted conditions exhibits. As 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 radiative forcing of aerosol-cloud interactions underestimates the absolute effect of LWP adjustments on the radiative forcing. With $-0.64 \leq d\ln LWP/d\ln N$ as lower bound (Fig. 3), this underestimation corresponds to an overestimation of the cooling effect of aerosols on non-precipitating Sc of up to 200% (Supplementary Information). Since non-precipitating Sc occur frequently [27, 25], 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 [25, 15]. Our insight that the effects of external co-variability fade as a Sc system evolves towards its internal steady-state refutes N-LWP co-variability 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 $d\ln LWP/d\ln N = -0.3$ [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 $d\ln LWP_\infty/d\ln N = -0.64$ as a new lower bound for LWP adjustments in non-precipitating 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 to a climatological perturbation. We estimate an effective lifetime of ship tracks in a shipping lane of $\Delta t_{lane} \gtrsim 9$ h (Supplementary Information). With an evolution time that is longer than 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 [25, 15, 16, 17].

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 alternatives 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 [41, 42]. 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 extra-tropical above-cloud moisture may bias towards less-negative values. Under-sampling of the subtropics may also exacerbate the timescale effect and 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) [43], i.e. the advertent 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.

In closing, there is urgent need to quantify the albedo and LWP responses in both precipitating and non-precipitating 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 non-precipitating regime are likely to significantly overestimate climate cooling.

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Supplementary materials

Materials and Methods
Supplementary Text
Figs. S1 to S6
Tables S1 to S4
Reference (*44,45*)

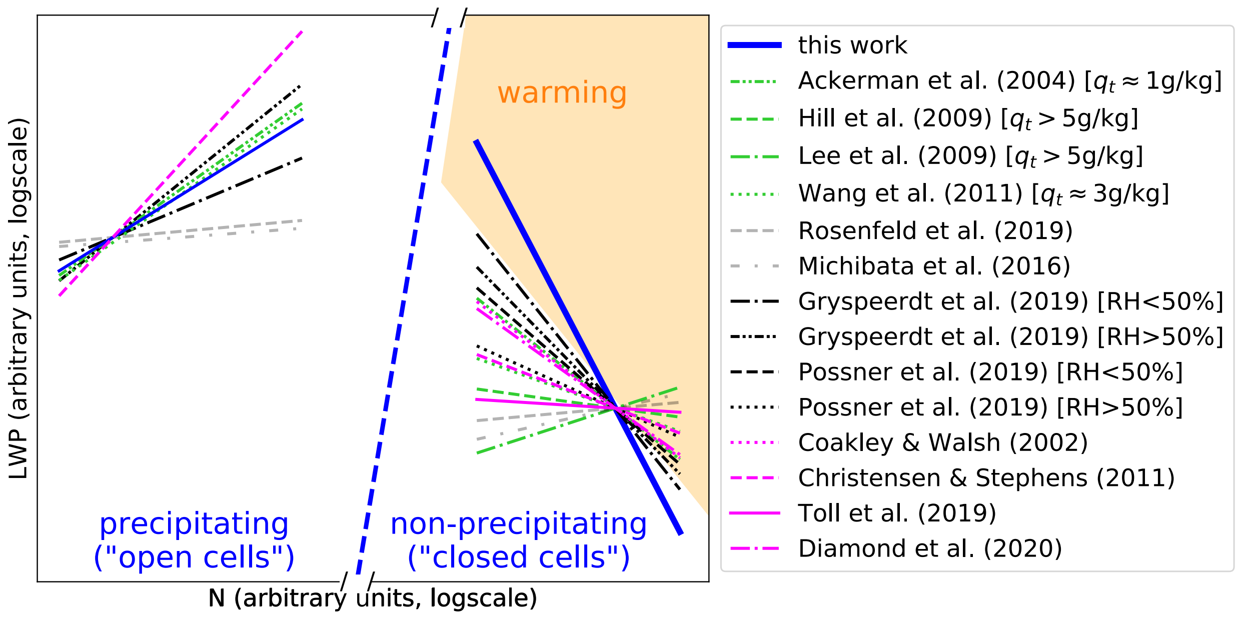

Fig. 1 Reported log-log-linear relationships between liquid-water path, LWP, and cloud droplet number, N, in comparison to this work. Lines are based on reported slopes (Tab. S1), axis intercepts have been added as suitable for illustration. The dashed blue line indicates a critical droplet radius for precipitation formation (based on a mean droplet radius of $12\ \mu$m at cloud top for an adiabatic condensation rate of $2.5 \cdot 10^{-6}\ kg\ m^{-4}$), which separates the precipitation-dominated regime on the left from the non-precipitating, entrainment-dominated regime on the right. The latter is the focus of this study. Colors distinguish results from large-eddy simulations (green), climatological satellite studies (black), and satellite studies of ship tracks (magenta). Results in grey are shown for completeness but are not directly comparable due to differences in methodology. For simulation and climatological studies, above-cloud absolute humidity q_t or ranges of relative humidity RH are indicated. Solid blue lines show values derived in this work (Tab. S1 and S4, in particular $d\ln LWP/d\ln N = -0.64$ in the entrainment regime). The orange shading indicates where LWP adjustments are sufficiently negative to lead to climate warming rather than cooling based on the sign of albedo sensitivity S (Eq. 1).

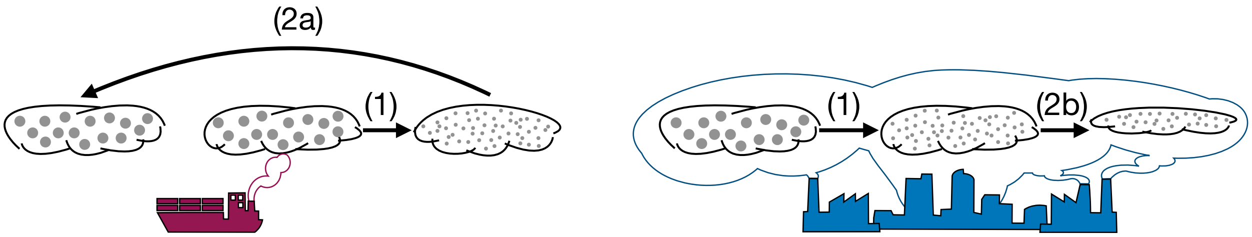

Fig. 2 LWP adjustments in ship tracks, which persist for a few hours, as compared to industrial-era pollution, which perturbs the climatological aerosol background and leads to perturbations that last for days. As an initial response to the aerosol perturbation, both situations feature cloud brightening through more but smaller cloud droplets at constant LWP (step 1). The ship track then returns to its original state because the perturbation ceases (step 2a). An enhanced aerosol background, in contrast, persists and allows for LWP to equilibrate to a new steady state that is characterized by increased entrainment efficiency and a lower LWP (step 2b).

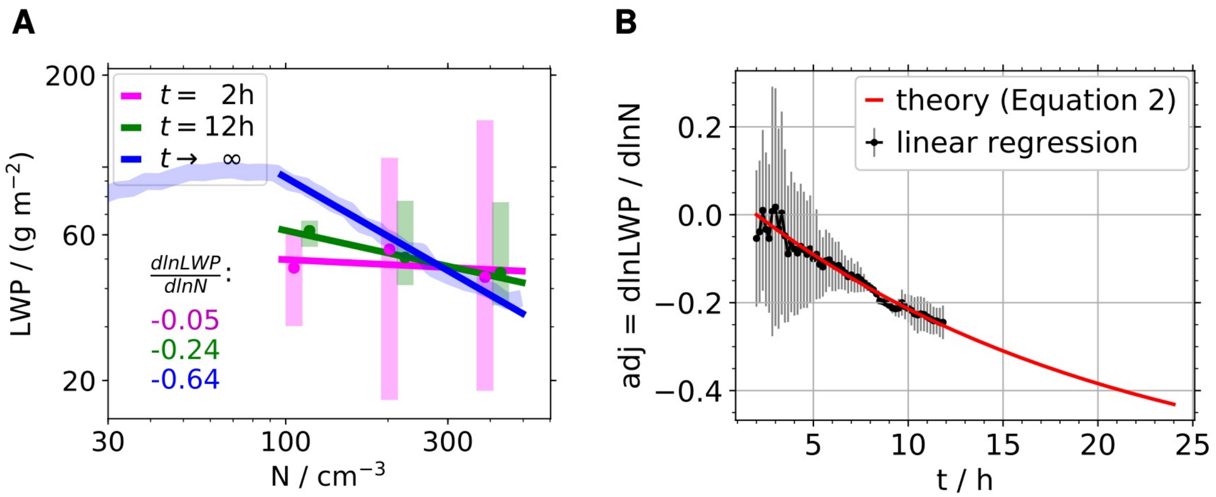

Fig. 3 Time-dependence of LWP-adjustments. (**A**) Data points with error bars show median and 25th/75th percentile of simulated LWP at (magenta) $t = 2$ h and (green) $t = 12$ h for the N-bins indicated in Fig. S1A. The faint blue curve indicates the steady-state LWP as in Fig. S1. Fit slopes $d\ln LWP/d\ln N$ are indicated. (**B**) Each data point indicates an adjustment slope obtained as in (**A**) with error bars for $2\,h \leq t \leq 12$ h. The red line shows the theoretically expected exponential decay (Eq. 2).

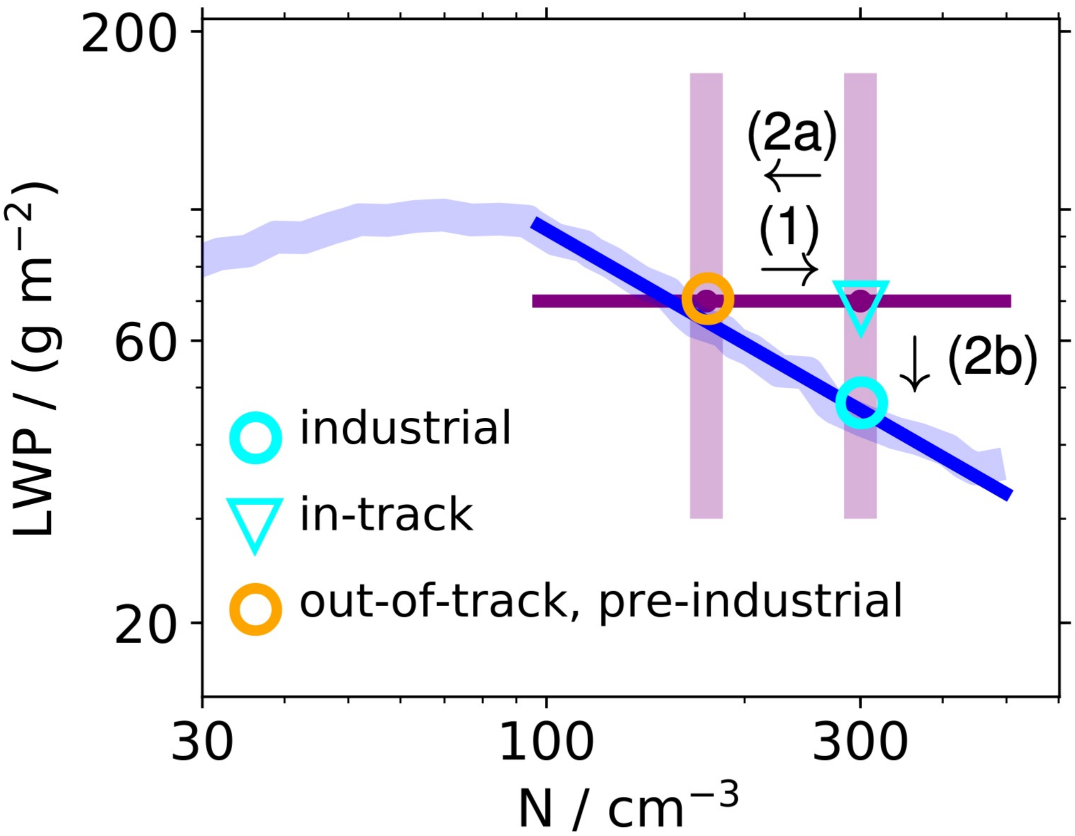

Fig. 4 Conceptual illustration of LWP adjustments as derived from ship tracks in comparison to climatological satellite studies in analogy to Fig. 3A. (Pre-)industrial climatological conditions are strongly-adjusted and represented by circles close to the steady state (blue line). LWPs within ship tracks are weakly adjusted (triangle symbol) such that ship-track studies are based on comparing almost identical LWP distributions (purple; illustration only, no actual data), which implies vanishing LWP adjustments (purple regression line). Labeled arrows correspond to Fig. 2.