Review Response for "Deconvolution of Boundary Layer Depth and Aerosol Constraints on Cloud Water Path in Subtropical Stratocumulus Decks"

We would like to thank both reviewers for their comments, which greatly helped to improve the clarity of this manuscript. Individual comments and concerns of each reviewer are addressed below. The colour code is as follows: reviewer comments and author response.

Reviewer 1:

Comments on: Deconvolution of Boundary Layer Depth and Aerosol Constraints on Cloud Water Path in Subtropical Stratocumuli By Possner et al. In this paper the authors use 10 years of measurements (primarily from MODIS) to investigate the LWP response to changes in cloud droplet number concentrations and boundary layer depth in subtropical Sc. They show that, in agreement with previous studies, LWP increase (decrease) with Nd for precipitating (non-precipitating) clouds. The rate of decrease (or susceptibility) in LWP with Nd under non-precipitating conditions is shown to increase with the BL depth. The authors further claim that the deep BL conditions are underrepresented in previous studies, hence, previous estimations of LWP susceptibility may be underestimated. The paper is well written and presents important and timely results. Hence, I support its publication after the following comment are addressed:

We thank the reviewer for their comments, which we address individually below.

General comment: One of the main conclusions/messages of this paper is that relatively deep BL clouds are underrepresented in studies of aerosol effect on LWP. However, there were previous LES studies simulating the transition between marine stratocumulus (Sc) to cumulus (Cu) and the aerosol effect on it. These studies include phases of deep BL. In addition, there were also many previous studies examining the aerosol effect on LWP in Cu clouds, with BL depth of 1.5 km and even more. I appreciate the focus on Sc, however, it looks to me as slightly artificial separation, especially if the focus is on relatively deep BL. I would expect that many of the physical processes acting in deep Sc and in Cu would be similar (as warm clouds cover the entire spectrum between Sc to Cu). For example, fig. 1 presents PDF of "disorganised" Sc. Looking on Fig. 1 of Muhlbauer et al., (2014), these disorganised Sc could definitely be (or at least be similar to) Cu. The fact that the data used here don't have any information on the decoupling level in the boundary layer (L.163) only strength the relevancy of the Cu regime.

We would like to thank the reviewer for this comment as it touches on several aspects which were insufficiently addressed in the previously submitted version. Firstly, we entirely agree with the reviewer that in the case of shallow cumulus (Cu) fields detraining into thin cloud decks, such as can be found below a strong inversions, and precipitating stratocumulus (Sc), the distinction between the two regimes is obsolete. However, in case of Cu cloud fields characterised by a low cloud fraction (30-40%), the LWP adjustment may differ to that of Sc decks (CF>80%).
Due to the increased fraction of sub-saturated clear-sky regions, the LWP adjustment seems governed by lateral entrainment and convergence processes (e.g. Jiang et al. (2006); Seifert et al. (2015)) as opposed to cloud thinning through vertical entrainment processes hypothesised to govern the LWP adjustment in marine stratocumuli. We agree that it is not clear at which point the distinction between Sc and Cu becomes somewhat semantic, but believe it is fair to contrast the behaviour of extensive Sc cloud decks of high cloud fraction to that of low-cloud fraction Cu.
To address this concern in the manuscript, we have:

- removed the “disorganised” PDF from Fig. 1 (extension of Fig. 10 in Muehlbauer et al (2014)). This PDF is associated with cloud fields of an average cloud fraction of 40% and is thus likely to be more representative of shallow cumulus fields as opposed to the stratocumulus sheets studied here.
- Included results from the ATEX campaign, which is technically sampling detraining Cu, but there is no a priori reason to believe that the processes governing the LWP adjustment should be substantially different to that of Sc
- Changed the title of the manuscript to highlight the implications for Sc cloud decks to: “Deconvolution of Boundary Layer Depth and Aerosol Constraints on Cloud Water Path in Subtropical Stratocumulus Decks”
- included two paragraphs that summarise this discussion:

“Fig. 1 shows the global distribution of stratocumulus regimes across BL depth which was characterised by Muhlbauer et al. (2014) in terms of cloud-top height (Fig. 10 in Muhlbauer et al. (2014)). The Muhlbauer et al. (2014) PDF is representative of all low-clouds over the oceans (see original paper for further methodology). We find the global PDF to be comparable to the distribution of stratocumuli against BL depth in the subtropics alone (Fig. S1). The PDF for disorganised clouds in Fig. 10 of Muhlbauer et al. (2014) was omitted here. These scenes were governed by broken cloud decks of low CF (CF = 40 %) resembling shallow cumuli rather than stratocumui.
The LW P adjustment within shallow cumuli seems governed by lateral entrainment effects and moisture gradients (e.g. Jiang et al. (2006); Seifert et al. (2015)). This is in stark contrast to stratocumulus cloud decks (CF 80 %) where the LW P adjustment is predominantly governed by vertical gradients in moisture, stability and aerosol. Thus, the LWP adjustment in shallow cumuli may differ from adjustments in stratocumuli, which is the focus of this study. The distinction between detraining shallow cumuli under strong inversions and precipitating stratocumuli becomes semantic in the case of cloud scenes associated with high cloud fraction. For this reason results of the Atlantic Trade Wind Experiment (ATEX) are included in Fig. 1.”

We further appreciate that several field campaigns and modelling studies have focused on Sc to Cu transitions and have explored the potential aerosol influence within these transitions. We have discussed this during the writing of this manuscript and decided to omit these studies for the following reasons:

(I) it is still debated to which extent precipitation and thus aerosol-sensitive cloud processes govern the breakup of the Sc deck into Cu cloud fields (e.g. Sandu & Stevens (2011) and Yamaguchi et al (2017)).

(ii) The LWP adjustment that originates from potentially delayed transitions likely only plays a secondary role to the inherent cloud fraction adjustment, which is not addressed in this study.

(iii) It is not clear how potential adjustments inferred from altered transition time scales relate to LWP adjustments within Sc decks generally.

References:
Sandu, I. and B. Stevens, 2011: On the Factors Modulating the Stratocumulus to Cumulus Transitions. *J. Atmos. Sci.*, **68**, 1865–1881, https://doi.org/10.1175/2011JAS3614.1.

Yamaguchi, T., Feingold, G., & Kazil, J. (2017). Stratocumulus to cumulus transition by drizzle. *Journal of Advances in Modeling Earth Systems*, 9, 2333– 2349, https://doi.org/10.1002/2017MS001104.

Specific comments:
Abstract: I think it is better not to use "susceptibility" in the abstract without defining it as some readers may not know what it is.
This section of the abstract was reworded as:
" An unequivocal attribution of LW P adjustments to changes in aerosol concentration from climatology remains difficult due to the considerable covariance between meteorological conditions alongside changes in aerosol concentrations. We utilise the susceptibility framework to quantify the potential change in LW P adjustment with boundary layer (BL) depth in subtropical marine stratocumuli. We show that the LW P susceptibility, i.e. the relative change in LW P scaled by the relative change in cloud droplet number concentration,
in marine BLs triples in magnitude from −0.1 to −0.33 as the BL deepens."

L27: I think that decreased precipitation rates are a micro-physical effect and not "dynamic or thermodynamic adjustments".
This has been rephrased as: "Through microphysical or thermodynamic adjustments [...]"
Figure 1. The PDFs taken from Muhlbauer et al., (2014) are based on which data? These processes (including the effect of the BL depth) were studied in Cu clouds.
Please see our response to the general comment above. Essentially we agree that these effects were studied in Cu clouds for different BL depths. Yet these regimes may not be identical in response to the Sc cloud decks studied here.
Technical comments:
L15: "due to be". Rephrased as "[…] due to changes […]".
L16: "estimates in". Rephrased ("estimates off").
L168: "the stronger". Rephrased as "a larger".

<u>Reviewer2:</u>

In this study, the authors investigated the dependency of the Liquid Water Path (LWP) susceptibility of stratocumulus deck upon the boundary layer (BL) depth using 10 years (from 2007 to 2016) data. From their analyses, the authors elucidated that the susceptibility increases with deepening BL, and magnitude of susceptibility triples with deepening BL. LWP adjustment is one of the important topics in the climate science. And I agree authors' suggestion that "the discussion based on the knowledge obtained from limited area of stratocumulus below shallow BL" can mislead the scientific community. So, I think this is an important study in the scientific community of the climate science. Most of the discussions in this manuscript is clear, and I agree most of the authors' suggestions. However, some discussions based on the previous process modeling study (slow- and fast- manifold mechanism) need to be modified. In addition, there are some technical problems. Based on the descriptions shown above, my decision is "not-so-major revision", and I encourage the authors to modify the manuscript. Detail comments are shown below.

General Comment:
1: The authors discuss the relationship between LWP-Hc and LWP-HBL in section 3, and try to interpret the difference between LWP-Hc and LWP-HBL relationship, and effects of aerosols on LWP-HBL relationship based on the slow and fast manifold mechanism. I agree that the discussions about slow and fast manifold mechanism are important for reducing the uncertainties of the cloud adjustment process. However, it is difficult for me to connect the results of this study to slow/fast manifold mechanism based on the analyses shown in the body of the manuscript. So, the discussion about the fast/slow manifold mechanism should be modified or removed from the manuscript.

We appreciate the reviewers criticism. As this is not a key aspect and hard to prove from a Eulerian perspective, we decided to remove the fast/slow manifold discussion from the revised manuscript. We thus remove Fig. 4 of the submitted manuscript and revised the text accordingly. The revised text still addresses that changes in H_c and H_{BL} are constrained on different timescales and by different factors.

2: The authors indicate the negative and positive LWP susceptibility in non-precipitating clouds and precipitating clouds, respectively (Table 1). This result supports the results of Gryspeerdt et al. (2019). In contrast, Chen et al. (2014), Michibata et al. (2016), Sato et al. (2018) indicated that the susceptibility is negative and positive or zero over precipitating and non-precipitating cloud areas. The authors should add some discussions about the reasons the inconsistency between the results of this study and the previous studies.

We would like to thank the reviewer for raising this issue. To facilitate the discussion of this issue and also to contextualise our results of our revised Fig. 4 (previously Fig. 5 in original submission) we included an additional figure (Fig. 5) in the revised manuscript. Although we find predominantly -ve values of s_{lwp}, we also find +ve slopes in regions of moderate to high precipitation occurrence (which has been added to Fig. 2).
We agree that this is an important question to be resolved within the community, but that the resolution of this contradiction is beyond the scope of this study. We acknowledge and compare our results to the aforementioned studies. At this stage it would require a targeted effort by all participants to address to which degree the assumptions and methodologies differ and impact s_{lwp} estimates.
In the revised manuscript we explicitly discuss this within a new paragraph added to the conclusions:
"Different remote-sensing-based estimates for slwp have been proposed. Their spatial distribution not only differs in magnitude, but also in sign among one another (e.g. Michibata et al. (2016) and Gryspeerdt et al. (2019)), as well as compared to Fig. 5 of this study. This is likely a result of different methodologies of categorising and processing different retrievals. Different methodologies to distinguish between precipitating and non-precipitating clouds, as well as different methods to retrieve and process Nd may impact slwp estimates. In particular, Nd remains a highly uncertain retrieval from space-born observations. For this study, we chose to limit the uncertainty of the physical retrieval of Nd while capturing as much of the variability in the subtropics as possible. Stricter filtering approaches may yield less retrieval uncertainty, but may imply a loss of some of the variability characteristic to the system. Either approach could influence slwp estimates. Thus our results, like previous studies, are subject to this uncertainty and remain to be verified in independent data sets."

Specific Comment:
Title: This study targets on the stratocumulus "decks", and open cellar stratocumuli are excluded from the analyses (CF > 80 %). So, I think the title with the word "deck" is better. For example, "Deconvolution of Boundary Layer Depth and Aerosol Constraints on Cloud Water Path in Subtropical Stratocumulus decks". This is just an example.

As suggested by the reviewer we changed the title to "Deconvolution of Boundary Layer Depth and Aerosol Constraints on Cloud Water Path in Subtropical Stratocumulus Decks".
It is worth noting though, that open cells may well be associated with cloud fractions of 80% and higher (see e.g. McCoy et al 2017).

Line 63- 64: "In Fig. 1, we show that . . ." should be "Figure 1 shows that. . .". The Figure 1 is originated from Fig. 10 of Muhlbauer et al. (2014), not the authors' work.

This was rephrased.

Figure 1: LES intercomparison studies targeting on stratocumulus like Stevens et al. (2005); Ackerman et al. (2009), which are representative LES studies for DYCOMS and LES studies on VOLCALS case (Berner et al. 2013) should be added in the figures.

We would like to thank the reviewer for the additional references. Ackerman et al (2009) and Berner et al (2013) were added to the figure. Although we agree that Stevens et al (2005) is one of *the* key publications in simulating stratocumuli, it does not explicitly focus on the impact of aerosols on cloud properties. This was a criterion for inclusion in this summary.
During this review we also added Xue et al (2008): "Aerosol Effects on Clouds, Precipitation, and the Organization of Shallow Cumulus Convection" and the ATEX campaign (Stevens et al 2001).

Line 68-69: "merely two campaigns and even fewer LES studies": Some concrete descriptions about the campaigns and LES studies targeting on the deep BL are helpful for readers to identify the previous studies targeting on deep BL.

We have expanded on this as follows:
"In the subtropics merely 30% of stratocumuli reside at the predominant depth range sampled in the field and studied within most LES. Results from merely three campaigns and few LES studies are discussed within the literature that reside within a height range deeper than 1 km where over 70% of marine stratocumuli are found. The campaigns containing measurements of deep stratocumulus cloud decks are ATEX, EPIC (East Pacific Investigation of Climate), and VOCALS-REx (VAMOS –Variability of the American Monsoons – Ocean-Cloud-Atmosphere-Land Study Regional Experiment). Merely 25% of all cloud-resolving modelling studies investigating the influence of aerosol concentrations on cloud properties in marine stratocumulus decks (i.e. Xue et al., 2008; Caldwell and Bretherton, 2009; Mechem et al., 2012; Berner et al., 2013; Possner et al., 2018) are based on deep BL field campaigns."

Line 86: Fig. S2: The authors discuss Reff through the Fig. S2, but no data of Reff in Fig S2. Fig. S2 is same as Fig. S3, so, I think this is just a mistake. The authors should exchange the figure to correct one.

We would like to thank the reviewer for catching this. The figure has been updated.

Figure 3: The data for non-precipitating case like Fig. S3 is useful for the reader, because the authors also discuss non-precipitating case in the body of the manuscript.

An additional figure was included in the supplementary material (Fig. S3) which shows the scaling relationships against BL depth for non-precipitating clouds in comparison to all clouds.

Line 140-157: As I mentioned in the general comment, it is difficult for me to connect the discussion of the LWP-Hc and LWP-HBL relationship to slow and fast manifold mechanism, through the results of this manuscript. I agree that the slow and fast manifold mechanism need to be considered when we discuss about the LWP adjustment. However, it is no evidence in this manuscript to justify that LWP-Hc and LWP-HBL relationship is regard as slow and fast manifold mechanism. The authors tried to justify through Hc-HBL relationship and Clausius Clapeyron, but I think these discussions could not convince readers that the LWP-Hc and LWP-HBL is regarded as the slow and fast manifold mechanism. In my understanding, the discussions about slow and fast manifold mechanism are not the main topic of the manuscript. So, the elimination of this part is one of the options. If the authors want to remain this part, I require the authors to add evidences to justify that LWP-Hc and LWP-HBL relationship can be regarded as slow and fast manifold mechanism.

We have removed our discussion of slow and fast manifolds within the manuscript (see comment above).

Line 158-164: The authors discuss about the effect of the decoupling, but as the authors mentions in Line 162-163, no conclusion about the decoupling is obtained. So, I think this part is not necessary, and can be removed from the manuscript.

This section has been removed from the manuscript.

Table 1: Sample number for each column is helpful for readers. In addition, the regression statistics (e.g., error, residual, and so on) are helpful for readers. The information can be added as a supplemental material.
Following your suggestion additional regression statistics (sample number and statistical error) are put are included in a new table within the supplemental material Table S1.

Line 174-177: In this part, the authors suggest that the anticorrelation between Nd and HBL is attributed to the climatological deepening of BL and increase of the distance to continental sources of anthropogenic pollution. However, there are no results to confirm these two suggestions. The trend of BL height and distance to continental sources are helpful for readers.

Thank you for this suggestion. We now show this explicitly in a new figure in the Supplement (Fig. S4).

Line 178-180: The authors suggested that the anticorrelation between Nd and HBL vanishes in a deregionalised and deseasonalised version as shown in Fig. S3. However, weak anticorrelation, which is shown in black line of Fig. S3, is seen in climatological mean (red) and non-precipitating clouds (green) shown in Fig. S3. Is the word "anticorrelation" is same as "negative correlation"? If so, the authors should add some descriptions about the weak anticorrelation in climatological mean (red) and non-precipitating clouds (green) shown in Fig. S3. The value of slope for each case in Fig. S3 is helpful for readers. If not, please added the definition of anticorrelation more correctly.

We do not find a significant trend in the non-precipitating deregionalised/deseasonalised Nd-HBL relationship shown in Fig. S5 of the revised supplement. A significant slope is only determined for the full deregionalised/deseasonalised dataset (black line – fit to red points). However, this slope originates from the different weighting of the precipitating and non-precipitating curves as BL depth increases and the precipitating fraction of clouds increases. i.e. initially the all-cloud curve (red) is governed by non-precipitating clouds (green) in shallow BLs and increasingly influenced by precipitating clouds (blue) in deeper BLs. This is discussed in lines 203ff ("In addition...").

Line 182-183: In this part, the authors suggest that the Nd and HBL climatology are not impacted by the precipitation, but the negative correlation in precipitating case is small but non-precipitating case is large. I think this means that the negative correlation is impacted.

This section has been rephrased. The main message is that "[...], the N_d climatology of all subtropical stratocumuli is constrained to first order by precipitation and to second order by the

proximity to sources of cloud condensation nuclei". We intended to state here that neither the BL deepening perpendicular to the coast (Fig. S4), nor the proximity to aerosol sources is directly impacted by precipitation. Yet the negative correlation between N_d and H_{BL} vanishes. Therefore, precipitation governs the N_d signal despite the underlying processes which result in a negative correlation otherwise.
This section was rephrased for clarity as:
"The observed negative correlation also disappears in the presence of precipitation (Fig.3e and Table 1). Our two process hypotheses governing the negative correlation between N_d and H_{BL} are not impacted directly by precipitation. Yet the negative correlation vanishes. This also holds for the deseasonalised and deregionalised N_d climatology (Fig. S4). It follows that precipitation is the predominant constraint on climatological N_d in sub-tropical marine stratocumuli at this scale."

Line 220-221: As I mentioned in the comment for Table 1, sample number for each column is helpful for readers.
This information is now included.

Line 273-274: As I mentioned in general comment, it is difficult to regard LWP-Hc and LWP- HBL relationship as the fast and flow manifold mechanism from the results shown in the manuscript. Please do not misunderstanding, I agree the importance of slow and fast manifold mechanism.

Please see our response to your main comment.

Minor or technical Comment:
Figure 2: There are many contour lines around tropics, mid-latitude area, and ITCZ zone, and it is difficult to see the value over these areas. Of course, I understand that these areas are out of the scope of this study, but the figure need to be modified.
This figure has been revised for clarity.

Line 147: "Hc" should be italic form and "c" should be subscript.
Corrected.
Figure 5: Unit of each variable in logarithmic is helpful for readers.
Line 153: Full spelling of SST (sea surface temperature) and FT (may by free troposphere) is helpful for readers.
Was added.
Line 228: "(6b)" should be "(Fig. 6b)".
Corrected.
Line 234: I think the word "LWP adjustment" is used as slwp. Is this right? If so, slwp is easy to be understood.
Changed to slwp.
Figures S1, S2, and S3: The label of Figure 1, 2 and 3 shown in supplemental material should be Figure S1, S2, and S3.
Corrected.

Reference:
Ackerman, A. S., and Coauthors, 2009: Large-Eddy Simulations of a Drizzling, Stratocumulus-Topped Marine Boundary Layer. Mon. Weather Rev., 137, 1083–1110, https://doi.org/10.1175/2008MWR2582.1. Berner, A. H., C. S. Bretherton, R. Wood, and A. Muhlbauer, 2013: Marine boundary layer cloud regimes and POC formation in a CRM coupled to a bulk aerosol scheme. Atmos. Chem. Phys., 13, 12549–12572, https://doi.org/10.5194/acp-13-12549-2013.

Chen, Y.-C., M. W. Christensen, G. L. Stephens, and J. H. Seinfeld, 2014: Satellite-based estimate of global aerosol–cloud radiative forcing by marine warm clouds. Nat. Geosci., 7, 643–646, https://doi.org/10.1038/ngeo2214.
Matsui, T., H. Masunaga, S. M. Kreidenweis, R. a. Pielke, W.-K. Tao, M. Chin, and Y. J. Kaufman, 2006: Satellite-based assessment of marine low cloud variability associated with aerosol, atmospheric stability, and the diurnal cycle. J. Geophys. Res., 111, D17204, https://doi.org/10.1029/2005JD006097.
Michibata, T., K. Suzuki, Y. Sato, and T. Takemura, 2016: The source of discrepancies in aerosol–cloud–precipitation interactions between GCM and A-Train retrievals. Atmos. Chem. Phys., 16, 15413–15424, https://doi.org/10.5194/acp-16-15413-2016.

Sato, Y., D. Goto, T. Michibata, K. Suzuki, T. Takemura, H. Tomita, and T. Nakajima, 2018: Aerosol effects on cloud water amounts were successfully simulated by a global cloud-system resolving model. Nat. Commun., 9, 985, https://doi.org/10.1038/s41467-018-03379-6. Stevens, B., and Coauthors, 2005: Evaluation of Large-Eddy Simulations via Observations of Nocturnal Marine Stratocumulus. Mon. Weather Rev., 133, 1443–1462, https://doi.org/10.1175/MWR2930.1.

Deconvolution of Boundary Layer Depth and Aerosol Constraints on Cloud Water Path in Subtropical Stratocumulus Decks

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Abstract. The liquid water path (LWP) adjustment due to aerosol-cloud interactions in marine stratocumulus remains a considerable source of uncertainty for climate sensitivity estimates. An unequivocal attribution of LWP adjustments to changes in aerosol concentration from climatology remains difficult due to the considerable covariance between meteorological conditions alongside changes in aerosol concentrations. We utilise the susceptibility framework to quantify the potential change in LWP adjustment with boundary layer (BL) depth in subtropical marine stratocumulus. We show that the LWP susceptibility, i.e. the relative change in LWP scaled by the relative change in cloud droplet number concentration, in marine BLs triples in magnitude from -0.1 to -0.31 as the BL deepens from 300 m to 1200 m.

We further find deep BLs to be underrepresented in pollution tracks, process modelling and in-situ studies of aerosol-cloud interactions in marine stratocumulus. Susceptibility estimates based on these approaches are skewed towards shallow BLs of moderate LWP susceptibility. Therefore, extrapolating LWP susceptibility estimates from shallow BLs to the entire cloud climatology, may underestimate the true LWP adjustment within subtropical stratocumulus, and thus overestimate the effective aerosol radiative forcing in this region.

Meanwhile, LWP susceptibility estimates in deep BLs remain poorly constrained. While susceptibility estimates in shallow BLs are found to be consistent with process modelling studies, they overestimate pollution track estimates.

1 Introduction

The aerosol radiative forcing due to changes in cloud reflectivity of low-level marine clouds remains one of the largest sources of physical uncertainty in climate sensitivity estimates. Estimates of total aerosol radiative forcing from the Fifth Assessment Report (AR5) issued by the Intergovernmental Panel on Climate Change (IPCC) range from $-0.1\,\mathrm{W\,m^{-2}}$ to $-1.9\,\mathrm{W\,m^{-2}}$ (Boucher et al., 2013; Zelinka et al., 2014). Based on these estimates, increased cloud reflectivity due to anthropogenic aerosol, may have posed a substantial offset to the greenhouse gas forcing.

However, this cooling term is likely to reduce in coming years as anthropogenic emissions of aerosols decline (Smith and Bond, 2014). Yet, the quantification of aerosol induced changes in cloud scene albedo remains important for reducing the uncertainty in overall forcing. Subtropical marine stratocumulus are of particular relevance; the stratocumulus decks in the subtropics contribute strongly to the cooling of the planet by reflecting $\sim 40\%$ of incoming solar radiation on average, in a region of high solar intensity (Bender et al., 2011).

In particular, cloud adjustments to changes in aerosol concentration remain highly uncertain. As defined in IPCC AR5 (Boucher et al., 2013), adjustments quantify the net response in cloud-radiative properties to external forcing agents such as anthropogenic aerosols. Through microphysical or thermodynamic adjustments, such as decreased precipitation rates (Albrecht, 1989), increased mixing rates at cloud top (Ackerman et al., 2004), or the sedimentation-entrainment feedback (Bretherton et al., 2007), the thermodynamics of the cloud is impacted and the liquid water path (LWP) may be altered. Adjustments in cloud fraction (CF) by changes in aerosol concentration may also increase the overall albedo of the cloud scene (Gryspeerdt et al., 2016; Andersen et al., 2017; Possner et al., 2018). However, these effects cannot be addressed within the framework of this study due to the insufficient accuracy in CF retrievals under polluted conditions (e.g. Twohy et al. (2009)). It is therefore mentioned here for completeness, but will not be discussed further.

In order to constrain the uncertainty range reflected within the wide range of AR5 forcing estimates, numerous studies have since quantified the individual contributions of the Twomey effect (Twomey, 1991) and LWP adjustments in global-scale and long-term satellite records (Sekiguchi et al., 2003; Quaas et al., 2008; Lebsock et al., 2008; Bellouin et al., 2013; Bender et al., 2016; Gryspeerdt et al., 2017; McCoy et al., 2017; Gryspeerdt et al., 2019; Rosenfeld et al., 2019), pollution track data sets (Ackerman et al., 2000; Christensen and Stephens, 2011; Christensen et al., 2014; Chen et al., 2015; Malavelle et al., 2017; Toll et al., 2017; Bender et al., 2019; Toll et al., 2019), and large-eddy simulations (LES) or cloud-resolving simulations in combination with field observations (see Fig. 1 for references). Satellite-based estimates of large data sets provide long-term and near-global constraints for the Twomey effect and the LWP adjustment. However, they are prone to numerous sources of uncertainties. These include, but are not limited to, uncertainties in N_d changes for a given change in aerosol metric, the distortion of the true sensitivity due to relatively coarse retrieval scales (McComiskey and Feingold, 2012), and the covariability between meteorological factors and aerosol indices. Average forcing estimates for the Twomey effect alone range between -0.2 to -1.0 $\mathrm{W\,m^{-2}}$ (Quaas et al., 2008; Lebsock et al., 2008; Bellouin et al., 2013; McCoy et al., 2017). The LWP adjustment may induce a partially compensating positive forcing to the Twomey effect, due to a decrease in cloud field LWP (Gryspeerdt et al., 2019). Meanwhile, the LWP adjustment inside the convective cores of low clouds may be positive (Rosenfeld et al., 2019) which would locally amplify the aerosol-cloud forcing due to Twomey.

In the case of pollution tracks, the issue of covariability between confounding factors is avoided and a clear detection and attribution of the cloud response to the aerosol perturbation itself, or at least to the corresponding change in N_d is possible. Each individual track is associated with a spatially confined cloud response due to aerosol perturbations by ship or volcano plumes for a given set of meteorological conditions. However, these tracks are rare. It is estimated that merely 0.002 % of all ocean-going ships generate a ship track (Campmany et al., 2009). Though a recent estimate suggests that this number might underestimate the true ship track frequency (Yuan et al., 2019). Furthermore, they are only found within a narrow window of meteorological conditions (Durkee et al., 2000). Therefore, while these estimates are prone to fewer uncertainties in detection and attribution of aerosol forcing, the representativeness of such estimates remains unclear.

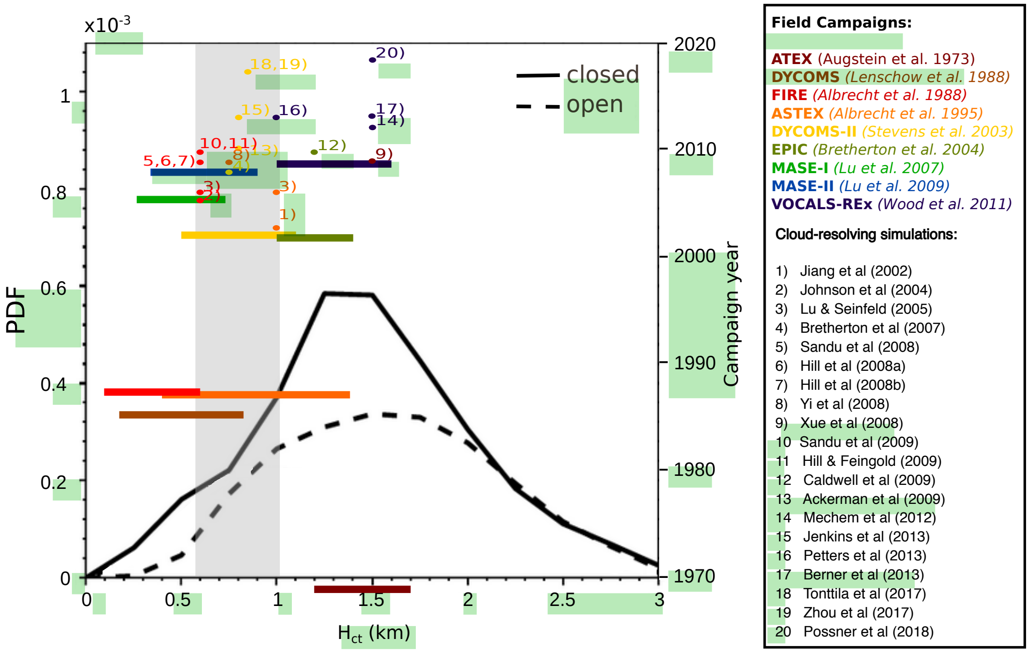

Figure 1. Probability density function (PDF) for closed, open-cell and disorganised stratocumulus layers against cloud top height. This figure is adapted from Fig. 10 in Muhlbauer et al. (2014). Coloured bars denote range of cloud top heights sampled during each campaign listed in the legend. LES and cloud-resolving studies investigating aerosol-cloud-radiative interactions are colour-coded by the campaigns they are based on [with the exception of model study 8 which is based on an idealised profile]. Grey shading denotes narrow BL depth interval within which 75 % of all modelling studies reside. Future analyses of past campaigns summarised in Zuidema et al. (2016) will likely increase the data points sampled in deeper BLs. References: Jiang et al. (2002); Johnson et al. (2004); Lu and Seinfeld (2005); Bretherton et al. (2007); Sandu et al. (2008); Hill et al. (2008); Hill and Dobbie (2008); Yi et al. (2008); Xue et al. (2008); Caldwell and Bretherton (2009); Ackerman et al. (2009); Sandu et al. (2009); Hill and Feingold (2009); Mechem et al. (2012); Jenkins et al. (2013)Petters et al. (2013); Berner et al. (2013); Tonttila et al. (2017); Zhou et al. (2017); Possner et al. (2018); Augstein et al. (1973); Lenschow et al. (1988); Albrecht et al. (1988, 1995); Stevens et al. (2003); Bretherton et al. (2004); Lu et al. (2007, 2009); Wood et al. (2011)

The same holds true for estimates based on LES, cloud-resolving model studies, and field observations. At this resolution insights into the interplay between microphysical, radiative and thermodynamic processes can be obtained. Yet, the estimates are representative for the conditions sampled and may not be valid generally, or at larger spatial scales. The LES community recently started to address these limitations, e.g. through extensive LES ensembles (Glassmeier et al., 2019). Here we would like to draw attention to the fact that previous analyses of LES, cloud-resolving models and field campaigns have predominantly focused on shallow boundary layers. Fig. 1 shows that most field campaigns and high-resolution modelling studies quantifying aerosol-cloud-radiative interactions have been conducted in BLs below 1 km in depth.

Fig. 1 shows the global distribution of stratocumulus regimes across BL depth which was characterised by Muhlbauer et al.

(2014) in terms of cloud-top height (Fig. 10 in Muhlbauer et al. (2014)). The Muhlbauer et al. (2014) PDF is representative of all low-clouds over the oceans (see original paper for further methodology). We find the global PDF to be comparable to the distribution of stratocumulus against BL depth in the subtropics alone (Fig. S1). The PDF for *disorganised* clouds in Fig. 10 of Muhlbauer et al. (2014) was omitted here. These scenes were governed by broken cloud decks of low CF ($\overline{CF} = 40\,\%$) resembling shallow cumulus rather than stratocumulus.

The LWP adjustment within shallow cumulus seems governed by lateral entrainment effects and moisture gradients (e.g. Jiang et al. (2006); Seifert et al. (2015)). This is in stark contrast to stratocumulus cloud decks ($CF > 80\,\%$) where the LWP adjustment is predominantly governed by vertical gradients in moisture, stability and aerosol. Thus, the LWP adjustment in shallow cumulus may differ from adjustments in stratocumulus, which is the focus of this study. The distinction between detraining shallow cumulus under strong inversions and precipitating stratocumulus becomes semantic in the case of cloud scenes associated with high cloud fraction. For this reason results of the Atlantic Trade Wind Experiment (ATEX) are included in Fig. 1.

In the subtropics merely 30% of stratocumulus reside at the predominant depth range sampled in the field and studied within most high-resolution simulations. Results from merely three campaigns and few modelling studies are discussed within the literature that reside within a height range deeper than 1 km where over 70% of marine stratocumulus are found. The campaigns containing measurements of deep stratocumulus cloud decks are ATEX, EPIC (East Pacific Investigation of Climate), and VOCALS-REx (VAMOS –Variability of the American Monsoons – Ocean-Cloud-Atmosphere-Land Study Regional Experiment). Merely 25% (Fig. 1) of all high-resolution modelling studies investigating the influence of aerosol concentrations on cloud properties in marine stratocumulus decks (i.e. Xue et al. (2008); Caldwell and Bretherton (2009); Mechem et al. (2012); Berner et al. (2013); Possner et al. (2018)), are based on deep BL field campaigns.

The lack of process studies in deep boundary layers, despite their prominence, motivates the question of exploring the dependence of cloud adjustments on BL depth. This is further supported by recent findings showing an explicit dependence of the LWP adjustment on BL depth in pollution tracks (Toll et al., 2019). Here, we focus on regions dominated by marine stratocumulus, and explore these relationships within 10-year records in the subtropics. The data set is described in section 2. The change of mean cloud properties with BL depth is presented in section 3, while the impact of BL depth covariance with LWP, and N_d on the LWP adjustment estimate is presented in section 4.

2 Data Description

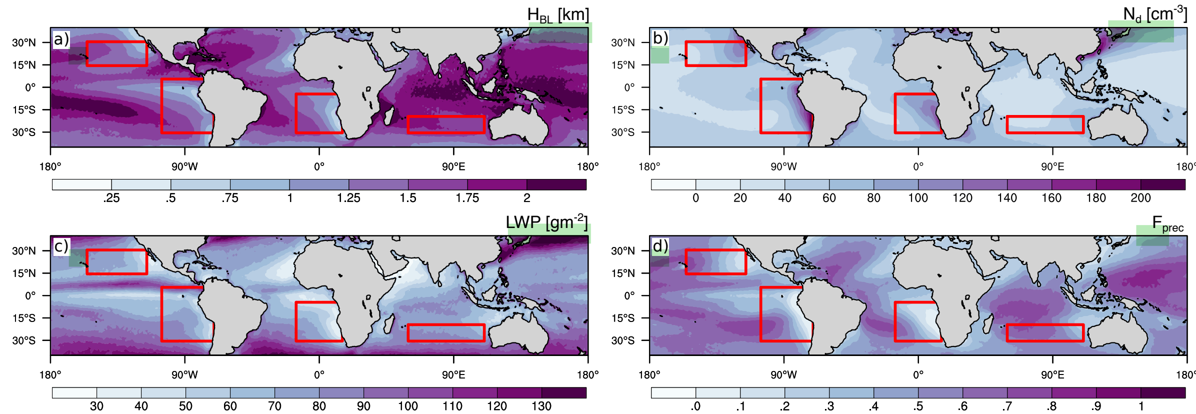

Figure 2. (a) Boundary layer height (H_{BL}), (b) cloud droplet number concentration (N_d), (c) liquid water path (LWP) and (d) low-cloud precipitation probability (F_{prec}) are shown. Regions of subtropical stratocumulus decks are marked in red and were defined in Eastman and Wood (2016) based on surface observations of Hahn and Warren (2007).

The relationship between LWP and N_d at different BL depths is analysed in the semi-permanent stratocumulus regions of the subtropics (Fig. 2). The analysis is based on a 10 - year climatology of daily in-cloud and radiation retrievals between 2007 and 2016, at a spatial resolution of $1 \times 1°$. Day-time in-cloud retrievals for LWP, N_d and effective radius (R_{eff}) are obtained from the level 3 Moderate Resolution Imaging Spectroradiometer (MODIS) collection 6 product (King et al., 2003; Platnick et al., 2017). As in previous collections, independent retrievals of cloud optical depth and R_{eff} are obtained using the visible and near-infrared radiances at 2.1 μm and 0.86 μm (Platnick et al., 2003).

The R_{eff} retrieval is further used to distinguish between precipitating ($R_{eff} \geq 15\,\mu$m) and non-precipitating ($R_{eff} < 15\,\mu$m) cloud scenes. For the year 2007 an independent retrieval of precipitation probability (Eastman et al., 2019) was available (Fig. S2). During this year, the R_{eff} criterion splits the data set into regimes where the precipitation probability remains below 50 % (equivalent to non-precipitating) and above 50 % (equivalent to precipitating).

N_d is estimated based on the relationship established by Boers et al. (2006) and Bennartz (2007) for marine boundary layer clouds:

$$N_d = \sqrt{2}\frac{3}{4k\pi\rho_w}\Gamma_{eff}^{\frac{1}{2}}\frac{LWP^{\frac{1}{2}}}{R^3_{eff|top}} \tag{1}$$

where ρ_w denotes the density of water, $\Gamma_{eff} = f_ad\Gamma_{ad}$ the effective rate of increase in adiabiatic liquid water content with increasing height and $R_{eff|top}$ denotes the effective radius at cloud top. All assumptions regarding the degree of adiabaticity and the proportionality constant k between the true and effective N_d are the same as in Eastman and Wood (2016).

The retrievals are restricted to sensor viewing angles between $0° - 65°$ (Grosvenor and Wood, 2014), which does not pose a strong constraint in the subtropics. The data is further limited to regions with high CFs exceeding 80 %. This restriction permits the best possible accuracy in N_d retrievals, which assumes plane parallel clouds and restricts the analysis to large-scale stratocumulus cloud decks only, which have the largest radiative impact. All cloud properties are in-cloud mean values only, which are not weighted by areal CF within each $1 \times 1°$ grid box.

The retrieval of BL height (H_{BL}) used in this study was first presented in Eastman and Wood (2016) and analysed in Eastman et al. (2017). The retrieval is based on a combination of MODIS and Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) cloud retrievals (Vaughan et al., 2004). The Clouds and Earth's Radiant Energy System (CERES) Single Scanner Footprint One Degree (SSF1deg) retrievals of all-sky (A_{toa}) and clear-sky albedo (A_{clr}) based on the top-of-atmosphere shortwave fluxes (Kato et al., 2013) were used to estimate A_{cld} from:

$$A_{toa} = CF * A_{cld} + (1 - CF) * A_{clr}. \tag{2}$$

It should be noted that the above equation can only provide an estimate of the cloud albedo. This definition, due to the separation of clear and cloudy skies, is highly sensitive to the definition of CF. CF retrievals are afflicted with uncertainty, due to swelling of aerosols in the high relative humidity environment near cloud edges (Twohy et al., 2009), and rather than a dichotomy, clear and cloud skies represent a continuum of albedo values (Charlson et al., 2007). Yet, Eq 3 has been shown to provide useful estimate of cloud albedo in the subtropical stratocumulus regions we focus on here.

Further environmental factors considered in this study, such as the lower tropospheric stability (LTS), and the free troposphere relative humidity (RH_{FT}), are obtained from the Modern-Era Retrospective Analysis for Research and Applications Version 2 (MERRA2) reanalysis (Rienecker et al., 2011; Molod et al., 2015). The LTS is defined as the change in potential temperature between 700 hPa and the surface. Conditions are considered non-stable, if the change in potential temperature between these two pressure levels remains below 15 K. RH_{FT} is diagnosed as the mean RH between the inversion and 700 hPa. Environmental conditions are considered to be dry if RH_{FT} falls below 50% and moist otherwise.

3 Covariance between Cloud Properties and Boundary Layer Depth

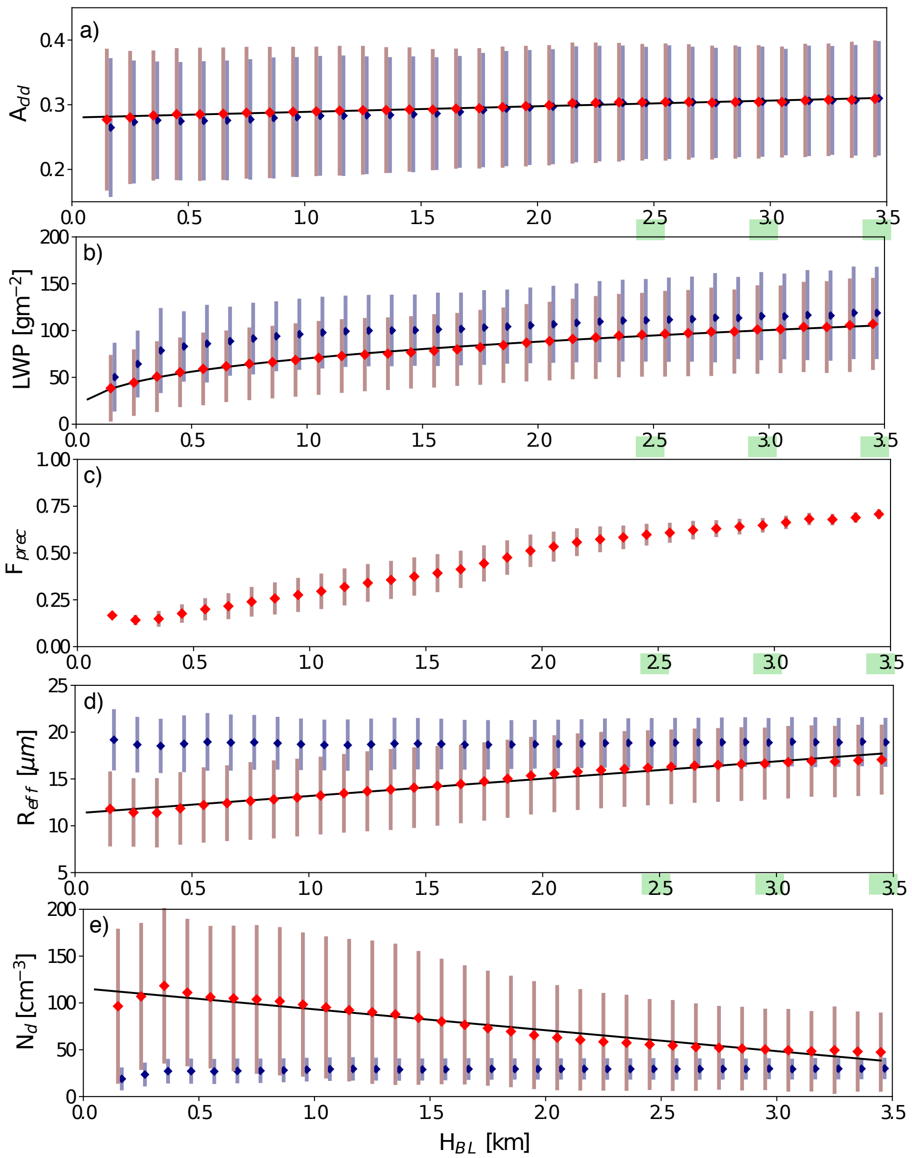

Figure 3. Scaling of (a) A_{cld}, (b) LWP, (c) F_{prec}, (d) R_{eff}, and e) N_d against H_{BL}. Mean and standard deviation of the 10-year subtropical stratocumulus climatology (see Fig. 2) are shown in red within each height bin (100-m intervals). Fits across the climatology are superimposed in black (see table 1 for details on fitting parameters). The climatology of precipitating clouds is shown in blue. For completeness the climatology for non-precipitating clouds is shown in Fig. S3.

Here, we analyse the change in cloud properties of subtropical stratocumulus as a function of H_{BL}. Each cloud property is binned into 100 m H_{BL} intervals within which the 10 – year mean and standard deviation are computed. The resulting relationships, which include data from all four predominant stratocumulus cloud decks, are shown in Fig. 3. All cloud properties change significantly (at the 95 % level) with H_{BL}. The largest relative changes are observed for LWP (Fig. 3b) and N_d (Fig. 3e), while merely moderate and small changes are observed in R_{eff} (Fig. 3d) and A_{cld} (Fig. 3a) respectively.

The adiabatic LWP (LWP_{ad}) scales with cloud depth (H_c) as $LWP_{ad} \propto H_c^2$, where $H_c = H_{BL} - H_b$ and H_b denotes cloud base. Based on this relationship we regress $\ln LWP$ against $\ln H_{BL}$ to identify the exponent of the LWP–H_{BL} relationship (Fig. 3b). Meanwhile, a simple linear regression is obtained for all other cloud climatologies (Fig. 3a,d,e). To understand the relative sensitivity amongst cloud properties to changes in H_{BL}, the slopes obtained by linear regression in the physical, as opposed to logarithmic space, are scaled by the climatological mean (Table 1).

As expected, larger LWPs are associated with deeper BLs (Fig. 3b). In particular we find LWP to scale as $LWP \propto H_{BL}^{0.33}$ (Table 1). Therefore LWP scales considerably weaker with H_{BL} than H_c. Combining these two relationships, it follows that in adiabatic clouds $H_c \propto H_{BL}^{0.17}$. That is, H_c increases on average by merely 2 m for every 100 m increase in H_{BL}. Thus H_c seems largely independent of H_{BL} variations.

Adiabaticity is known to change with cloud depth in marine stratocumulus (Merk et al., 2016; Braun et al., 2018). Furthermore, we find that clouds in deep BLs are more likely to precipitate than clouds in shallow BLs (Fig. 3c). Consequently, one might expect cloud adiabaticity to change as a function of H_{BL} due to the change in likelihood of precipitation (F_{prec}). However, we find the $LWP - H_{BL}$ relationship is hardly impacted by precipitation (Table 1: columns 6 and 7). It also seems unlikely that the functional relationship between adiabaticity and H_c would be sufficient to reduce the quadratic exponent of the LWP-H_c relationship to that of the sub-linear exponent in the LWP-H_{BL} relationship. It therefore follows that LWP scales very differently and seemingly independently with H_{BL} and H_c in marine subtropical stratocumulus.

Thermodynamic adjustments in LWP and H_c occur rapidly (hourly timescale) while adjustments in BL depth and thus LWP occur on longer time scales (multi-day timescale). H_c is predominantly constrained by the vertical displacement of H_b, as has been quantified by LWP budgets (Wood, 2007; van der Dussen et al., 2014; Ghonima et al., 2015; Hoffmann et al., submitted). H_b in turn is governed by Clausius Clapeyron in response to variability in BL humidity and temperature. Meanwhile, the multi-day evolution of H_{BL} and thus LWP, characterises their evolution as a function of external drivers such as gradients in sea surface temperature (SST), FT conditions, and large-scale advection. This is consistent with the weak relationship between H_c and H_{BL} inferred here from climatology.

LWP increases more rapidly with H_{BL} under dry FT and non-stable lower tropospheric conditions (Table 1 columns 2 – 4). This behaviour is consistent with cloud-scale observations (Eastman and Wood, 2018), simulations (Bretherton et al., 2013) and mixed-layer theory (Dal Gesso et al., 2014). Under low RH_{FT} cloud-top cooling and cloud-top generated mixing are more effective. Therefore, a deeper and moister mixed layer associated with larger LWP can be maintained. Thus, the reinforcement of the cloud through stronger radiative cooling has a larger impact on LWP, than the increased drying through entrainment under low RH_{FT} conditions. Similarly, the weaker buoyancy jump across the inversion under non-stable lower troposphere conditions, likely induces less warming in the sub-cloud layer as the BL deepens, which corresponds to a weaker upward shift of the cloud base.

Meanwhile, deeper BLs are characterised by lower N_d (Fig. 3e). As the BL deepens, F_{prec} (Fig. 3c), and thus the N_d sink through collision-coalescence processes, increases. Yet, N_d primarily decreases with H_{BL} in non-precipitating BLs. This suggests that precipitation scavenging is not the only constraint on N_d. In the absence of precipitation we attribute the negative correlation between N_d and H_{BL} to (i) the climatological deepening of the BL away from the cold upwelling zones near the coasts (Fig. 2a), and (ii) the increasing distance to continental sources of anthropogenic pollution, which manifest in a pronounced negative gradient in N_d (Fig. 2b). This is also explicitly illustrated in Fig. S4.

The negative correlation vanishes in a deregionalised and deseasonalised version of this data set (Fig. S5). Following Bender et al. (2016) we remove geographical and seasonal trends. In doing so, the significant negative correlation between N_d and H_{BL} in non-precipitating clouds disappears. This further confirms, that the observed negative correlation between N_d and H_{BL} in non-precipitating clouds is intrinsic to the data, but not a manifestation of a given physical process.

The observed negative correlation disappears in the presence of precipitation (Fig. 3e and Table 1). Our two process hypotheses governing the negative correlation between N_d and H_{BL} are not impacted directly by precipitation. Yet the negative correlation vanishes. This also holds for the deseasonalised and deregionalised N_d climatology (Fig. S5). It follows that precipitation is the predominant constraint on climatological N_d in subtropical marine stratocumulus at this scale. In addition F_{prec} changes with H_{BL}. Thus, a significant negative slope manifests within the whole N_d climatology ($\overline{c_{N_d}} = -0.3$), as the fraction of precipitating to non-precipitating clouds changes. Therefore, the N_d climatology of all subtropical stratocumulus is constrained to first order by precipitation and to second order by the proximity to sources of cloud condensation nuclei.

The weakly positive scaling in R_{eff} against H_{BL} is consistent with the relationship between LWP and N_d, and H_{BL}. The decrease in N_d with H_{BL} is insufficient to offset the increase in LWP. The combined increase in LWP and R_{eff} with BL deepening results in a significant, but inconsequential increase in A_{cld} with H_{BL} (Table 1). Stratocumulus with cloud tops above 1 km are associated with larger LWP, lower N_d, larger R_{eff}, and an elevated A_{cld} of 0.01 as compared to stratocumulus with cloud tops below 1 km.

4 Liquid Water Path Adjustment

The climatological fields of LWP and N_d display a significant correlation and anticorrelation with H_{BL}. The largest climatological values of LWP are found in deep BLs with low N_d (Fig. 4a). The increase in LWP with H_{BL} is consistent with Fig. 3b). The displayed sensitivity of LWP to N_d is potentially attributable to a multitude of competing factors; not all representative of cloud adjustments. The decrease in LWP with increased levels of pollution has been noted multiple times in observations and various process hypotheses have been put forward.

Less polluted clouds could potentially be associated with weaker entrainment drying through the entrainment-sedimentation feedback (Bretherton et al., 2007). Alternatively, increased rates of precipitation in cleaner environments could stabilise the cloud (Wood, 2012), which results in weaker overall cloud-top entrainment of dry sub-saturated air (Ackerman et al., 2004).

Quantity	stability		above cloud RH		cloud-base precipitation			all
	stable	non-stable	dry	moist	no-rain	rain		
$c_{\ln LWP}$	0.4	0.6	0.5	0.2	0.3	0.3		0.42
$\overline{c_{R_{eff}}}$	0.10	0.08	0.13	0.13	0.07	x		0.13
$\overline{c_{N_d}}$	-0.2	x	-0.3	-0.28	-0.13	x		-0.3
$\overline{c_{A_{cld}}}$	0.03	0.04	0.04	x	0.03	0.04		0.03
						fully	intermittently	
s_{lwp} (bivariate)	-0.29	-0.18	-0.29	-0.15	-0.28	0.14	-0.23	-0.28
s_{lwp} (N_d-only)	-0.31	-0.2	-0.32	-0.16	-0.28	0.28	-0.26	-0.33

Table 1. This table summarises the regime dependence of each slope. The relationship between cloud properties and H_{BL} is determined logarithmically ($\ln\Psi \sim c\_\ln\Psi \times \ln H_{BL}$ for $\Psi = LWP$), or as normalised linear slopes ($\overline{c_\Psi} = c\_\Psi/\overline{\Psi}$ where $\Psi \sim c\_\Psi \times H_{BL}$ for $\Psi \in \{R_{eff}, N_d, A_{cld}\}$ and $\overline{\Psi}$ denotes the average). Slopes were determined by linear regression if and only if: (i) a significant fit was obtained at the 95-% confidence level and (ii) the fit explained at least 80% of the variance of the climatological relationship shown in Fig. 3. If no such fit is obtained, "x" is given. The regime dependence of s_{lwp}, which is defined and discussed in section 4, is summarised in the last two rows. Estimates for s_{lwp} were either obtained by simple linear regression or by a bivariate fit taking the covariability between LWP, N_d and H_{BL} into account. All slopes are given to the significant digit which is determined based on the error of the respective fit. Error statistics and sample sizes for each category are provided in Table S1.

Furthermore, recent results show that the strengthening of convective overturning in the sub-cloud layer through precipitation can also have a net positive impact on LWP (Goren et al., 2018). All these, are examples of adjustments to the initial cloud microphysical response caused by an increase in droplet number. However, other factors not representative of cloud adjustments, such as the climatological covariance between H_{BL} and N_d noted in Section 3, may impact s_{lwp} estimates.

Here, we estimate s_{lwp} by either fitting $\ln LWP$ against $\ln N_d$ as in (Gryspeerdt et al., 2019), or by fitting the two-dimensional surface of $\ln LWP$ against $\ln H_{BL}$ and $\ln N_d$. Both fits are simple linear, single- or bivariate regressions across the phase space containing 80 % of all data (Fig. 4a). Both fitting approaches yield similar and negative s_{lwp} estimates. Taking the covariance with H_{BL} into account merely reduces the magnitude of s_{lwp} from -0.33, which is consistent with previous global estimates of marine low-level clouds (Gryspeerdt et al., 2019), to -0.28 (Table 1). Therefore the bivariate fit of the entire climatology is likely subject to the same confounding factors impacting LWP adjustments as in Gryspeerdt et al. (2019). Furthermore, the two predictor variables H_{BL} and N_d of the bivariate fit are not independent (Fig. 3e), which may bias the s_{lwp} estimate.

The entire phase space can be further characterised by F_{prec} (Fig. 4e)): 14% (Table S1) of the climatological phase space is characterised by precipitating cloud scenes (Fig. 4b)), 37% by intermittently precipitating cloud scenes (Fig. 4c)), and 48% by non-precipitating clouds (Fig. 4d)). The analysis shows that $\ln LWP$ increases with $\ln N_d$ in the precipitating fraction of the cloud climatology ($s_{lwp} = 0.14$) and decreases in the intermittently ($s_{lwp} = -0.23$) and non-precipitating climatologies ($s_{lwp} = -0.28$). Thus s_{lwp} inferred from the entire climatology is dominated by the LWP-N_d relationship in non-precipitating clouds.

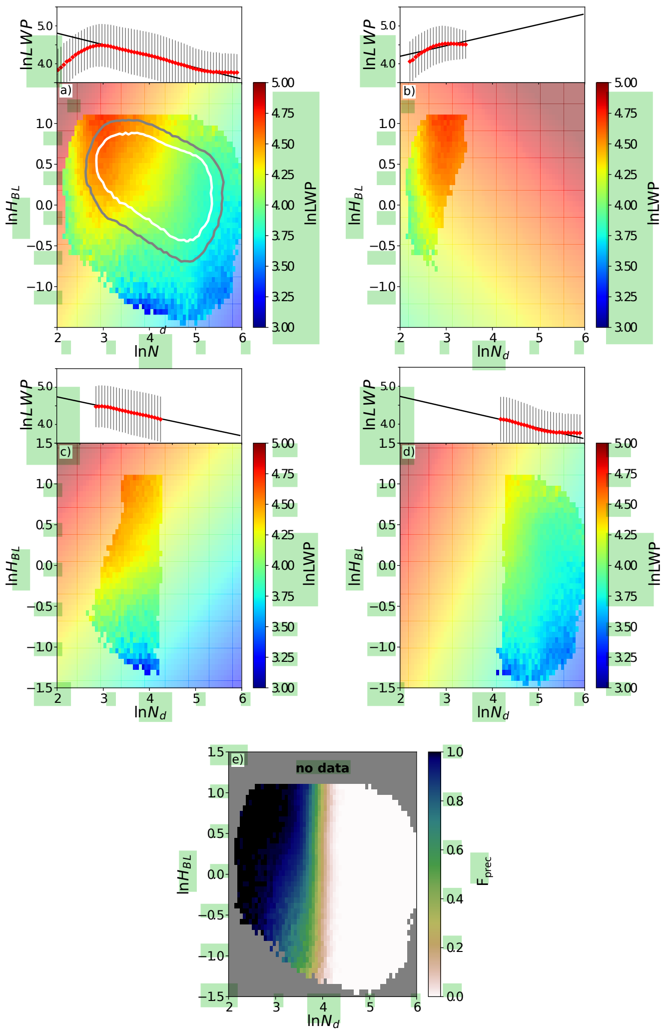

Figure 4. Climatology of $\ln LWP$ against $\ln N_d$ and $\ln H_{BL}$ for (a) all subtropical stratocumulus (see Fig. 2), (b) fully precipitating, (c) intermittently precipitating and (d) non-precipitating stratocumulus. Points are classified as fully precipitating (non-precipitating) if the fraction of precipitating cloud scenes shown in panel (e) exceeds 0.98 (remains below 0.02). All other cloud scenes are classified as intermittently precipitating. At the top of each panel (a) – (d) $\ln LWP$ binned in $\ln N_d$ is shown as in Gryspeerdt et al. (2019). A minimum number of 100 points within each $\ln N_d - \ln H_{BL}$ bin was required to be included in the climatology shown in opaque contours (a – e). The bivariate (simple linear) regression across the 2-dimensional (one-dimensional) climatology is shown in transparent contours (as black line) in panels (a) – (d). White and grey lines in panel (a) denote the region of the phase space containing 80 % and 90 % of all data respectively. The slopes of all fits are summarised in Table 1.

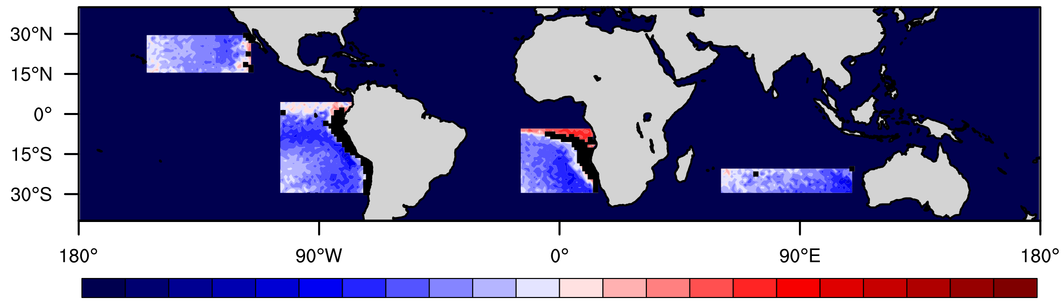

Figure 5. Map s_{lwp} which was obtained from bivariate linear regression of daily $\ln LWP$, $\ln N_d$ and $\ln H_{BL}$ retrievals at a spatial scale of $1^\circ \times 1^\circ$. Regions where s_{lwp} is not statistically significant are masked in black.

The opposing response in precipitating and non-precipitating regions is consistent with numerous previous studies (Albrecht, 1989; Ackerman et al., 2004; Bretherton et al., 2007; Wood, 2007; Wang et al., 2012; Suzuki et al., 2013; Gryspeerdt et al., 2019). While the largest fraction of H_{BL}-N_d phase space is characterised by non-precipitating clouds (Table S2), only a narrow band in close proximity to the coast lines of the Americas and the African continent is characterised by little to no precipitation ($F_{prec} < 0.1$, Fig.2d). Most regions are characterised by intermittend rain occurrence ($0.2 < F_{prec} < 0.8$) and are associated with more moderate susceptibilities of -0.23 or less (Fig. 5). Thus the results of Fig. 4c) are representative for most stratocumulus regions in the subtropics. Overall, few stratocumulus regions are associated with an average positive susceptibility.

Non-precipitating and intermittently precipitating cloud climatologies are not sensitive to the fitting technique applied (Table 1). Yet, s_{lwp} halved when the covariance of LWP with H_{BL} is taken into account in consistently precipitating clouds. In order to gain further insight into the potential variance of s_{lwp} with H_{BL}, we calculated s_{lwp} within constrained BL-depth ranges for the three precipitation regimes characterised in Fig. 4.

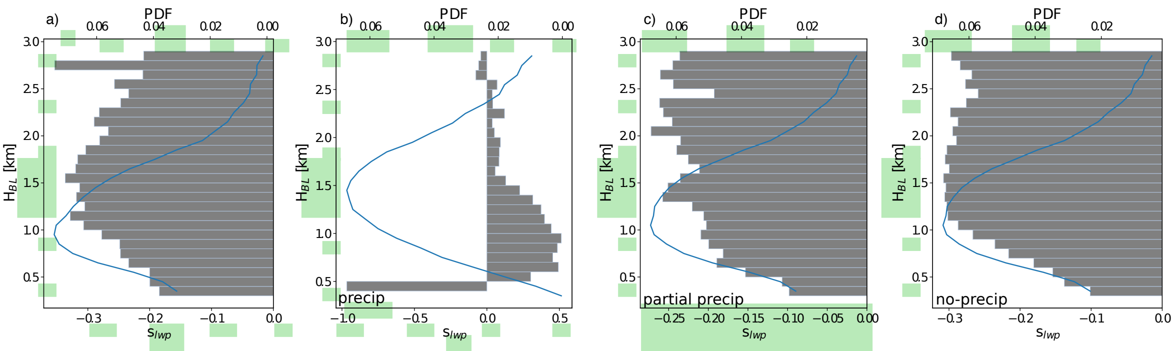

Figure 6. (a) s_{lwp} determined within 100 -m height intervals for subtropical stratocumulus, (b) precipitating clouds only, (c) intermittently precipitating clouds, and (d) non-precipitating clouds. S_{lwp} was obtained from bivariate linear regression of the $\ln LWP$ surfaces shown in Fig. 4(a) – (d) within each height interval respectively. Only statistically significant results at the 95 % confidence level are shown. The probability density function (PDF) of the subtropical cloud climatology across the height bins is superimposed.

Analyses of ship tracks (Christensen and Stephens, 2011) and Lagrangian studies of cloud evolution (Eastman et al., 2017) have shown that H_{BL} may increase under more polluted conditions. This, however, is not manifested within the H_{BL}-N_d climatology (Fig. 3e). Therefore, by constraining s_{lwp} estimates in this manner, we attempt to remove some of the covariance between LWP, N_d and H_{BL} which may impact the estimated strength of the LWP adjustment.

S_{lwp} in precipitating subtropical stratocumulus is considerably larger in BLs below 1.5 km in depth than in deeper BLs (Fig. 6b). While s_{lwp} may be as large as 0.48, which constitutes a tremendous cloud adjustment in shallow BLs, it does not exceed 0.08 in deep BL clouds. It should be noted that the large negative adjustment of $s_{lwp} = -1.0$ within the first height bin in Fig. 6b) is statistically significant, but characterises a very small sub-sample of the total climatology.

Meanwhile, s_{lwp} increases in magnitude from -0.1 in BLs below 500 m in altitude to -0.31 in BLs exceeding 1 km in depth (Fig. 6d). A weaker height dependence is observed for intermittently precipitating cloud scenes (Fig. 6c). Both results of Fig. 6c) and Fig. 6d) are consistent with the increase in LWP susceptibility noted within pollution tracks around the globe (Toll et al., 2019). Moreover, the change in s_{lwp} with H_{BL} shown in Fig. 6c), which characterises the behaviour in most stratocumulus regions, is within a 1σ uncertainty range of s_{lwp} estimates based on pollution tracks. Within pollution tracks s_{lwp} increased in magnitude from less than -0.01 ± 0.13 in shallow BL clouds to -0.1 ± 0.13 for a cloud top height of 2 km (Toll et al., 2019).

5 Conclusions

Isolating the LWP adjustment due to changes in N_d from potentially covarying meteorological factors has remained a significant hindrance in quantifying the radiative forcing of aerosol-cloud interactions. It also is a likely cause between diverging estimates from low-cloud climatology, process-scale models, and pollution track estimates.

Here, we address whether LWP adjustments vary with BL depth, and whether climatological susceptibility estimates are impacted by the covariance of cloud properties with H_{BL}. Like previous studies we find evidence for a positive relationship between LWP and N_d climatologies in precipitating marine stratocumulus (Albrecht, 1989; Christensen and Stephens, 2011; Wang et al., 2012; Suzuki et al., 2013; Rosenfeld et al., 2019) which is consistent with the suppression of precipitation. Particularly in shallow precipitating BLs ($H_{BL} < 1$ km) the estimated susceptibility can become very large ($s_{lwp} > 0.4$, Fig. 6). Such adjustments would correspond to a considerable enhancement of the negative cloud-radiative forcing. However, these shallow precipitating BLs are rare (10 – 25 % of all cloud scenes analysed within the 10 –year climatology). Therefore, such cloud scenes are unlikely to govern the radiative forcing of aerosol-cloud interactions.

The LWP adjustment inferred from the entire climatology of marine subtropical stratocumulus ($s_{lwp} = -0.33$) is driven by non-precipitating cloud climatologies which govern the climatological statistics, but are only representative for a small subset stratocumulus regions in the subtropics. Susceptibility estimates restricted to the phase space of intermittently precipitating climatologies which represent most stratocumulus regions in the subtropics are lower ($s_{lwp} = -0.23$) and are representative for most stratocumulus regions in the subtropics (Fig. 2d). Performing a bivariate fit of the $\ln LWP$ phase space, which removes any potential impact of the LWP-H_{BL} covariance on estimates of s_{lwp}, does not provide substantially different results (Table 1) to previous global estimates of s_{lwp} in marine low clouds (Michibata et al., 2016; Gryspeerdt et al., 2019).

A further division of the entire phase space into BL-depth regimes showed that overall, cloud adjustments are less effective in shallow BLs. The potential increase in LWP adjustment with BL depth has very recently been noted in pollution tracks (Toll et al., 2019). Here, we show that this behaviour may generalise to the whole climatology. The simulated change in s_{lwp} with H_{BL} within clouds of intermittent precipitation is consistent with the lower end of s_{lwp} estimates within the 1σ range inferred from pollution tracks. Stratifying the $\ln LWP - \ln N_d$ surface by BL depth, further closes the gap between s_{lwp} estimates inferred from climatology and cloud-scale modelling. Shallow BLs, such as the ones sampled during ASTEX and DYCOMS-II (Fig. 1) are associated with $-0.22 < s_{lwp} < -0.1$ (Fig. 6c – d). This is consistent with estimates of s_{lwp} obtained in LES experiments of these campaigns (Ackerman et al., 2004; Bretherton et al., 2007).

Different remote-sensing-based estimates for s_{lwp} have been proposed. Their spatial distribution not only differs in magnitude, but also in sign among one another (e.g. Michibata et al. (2016) and Gryspeerdt et al. (2019)), as well as compared to Fig. 5 of this study. This is likely a result of different methodologies of categorising and processing different retrievals. Different methodologies to distinguish between precipitating and non-precipitating clouds, as well as different methods to retrieve and process N_d may impact s_{lwp} estimates. In particular, N_d remains a highly uncertain retrieval from space-born observations. For this study, we chose to limit the uncertainty of the physical retrieval of N_d while capturing as much of the variability in the subtropics as possible. Stricter filtering approaches may yield less retrieval uncertainty, but may imply a loss of some of the variability characteristic to the system. Either approach could influence s_{lwp} estimates. Thus our results, like previous studies, are subject to this uncertainty and remain to be verified in independent data sets.

In summary, our results show that aerosol-cloud interactions may manifest differently in deep precipitating, and non-precipitating, marine BLs as compared to shallow BLs. Furthermore, this work highlights the importance of understanding aerosol-cloud interactions in deep marine stratocumulus, which are underrepresented in currently analysed field data, numerical process models, and pollution tracks.

Data availability. MODIS cloud retrievals were obtained from: https://ladsweb.modaps.eosdis.nasa.gov/archive/allData/61/MYD08_D3 (last accessed January 2019). MERRA-2 data were downloaded from https://disc.gsfc.nasa.gov/datasets (last accessed March 2019). Albedo fields from CERES were downloaded from https://ceres.larc.nasa.gov/compare_products.php (last accessed November 2018)

Author contributions. AP conceived this study and wrote the manuscript. RE compiled the remote sensing retrievals for the study. AP and RE performed the analyses with input from FB and FG. All authors contributed to the discussion of the results, and editing of the manuscript.

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