Authors response to: Interactive comment on “The efficacy of aerosol-cloud-radiative perturbations from near-surface emissions in deep open-cell stratocumulus” by Anna Possner et al.

Anonymous Referee #1
Received and published: 11 September 2018

Possner et al. present a modelling study in which the effects of idealised ship emissions on clouds and radiation are examined. Tools and analysis are adequate, and the diligent analysis by Possner et al. allowed for very important insights. A particularly important finding is that a substantial increases in all-sky albedo even relatively far away from the aerosol emission source may be expected, and that there is no simple way of identifying these changes given the large natural variability of the cloud properties. With a known location of the emission source, in turn, detection and attribution seems feasible.

The study is very relevant to the readership of Atmos. Chem. Phys. and should be published after some minor revisions.
We thank the reviewer for their assessment and have addressed all issues raised in the revised manuscript. Please find a repy to each individual comment below.

The most important one is that the authors need to clarify better than they did which quantities are reported as all-sky, and which as cloudy-sky averages. Some further mistakes or unclear aspects that I list below also should be corrected.
Domain means have been clarified with respect to when they were taken over all-sky, cloudy sky or clear sky. To this end the captions of Table 1 and Table 2 were revised and minor edits were undertaken in the main text of the manuscript.

Specific comments

p1l19 – the forcing is for the anthropogenic perturbation of the aerosol
This was corrected in manuscript.
p2l5 – invert the order of the Durkee references
Done.
p2l15 – Toll rather did conclude that the LWP change was small
The increase in LWP was quantified in main text for accuracy. According to Fig.2 of Toll et al 2017 increase in the precipitating regime was between 16 – 24%.
p2l19 – correct “Agency” reference
Done.
p2l31 – unit missing (cm-3?)
Added.
p2l32 – reference to Platnick and Twomey is missing
Added.
Table 1 – interquartile range: of the temporal variability of the domain-mean quantities? should be specified. Are LWP and Nd in-cloud or all-sky in both, observations and model results?
The caption has been revised for clarity. Domain-mean and in-cloud Nd are given in the table (correspondingly for observations and simulations). The interquartile range covers the spatial and temporal variability of the cloud field. This is now clarified in the revised caption.
p3l14 – The quantities in Table 1 are not numerous, and don’t include boundary layer properties
Sentence has been rephrased.
p4l20 – a reference for the Quickscat data source would be good
Added.

p5l10 – for completeness, the symbols τ , h, and z could be explained, too. ρ w presumably is a constant?
A definition of all variables was added.
p6l6 – how do the authors come to the conclusion that these are "realistic"?
The cell sizes of open cells observed furing VOCALS Rex were around 30-40km (p3L15), which is consistent with Fig.1d. Rephrased "realistic" with "observed".
p7l6 – if the authors want to make this point, they should consider listing the observed range in Table 1.
A reference to Fig.S2, where the range of observed Rcb is shown, was added.
Fig. 2 – how is cloud top defined?
Reference to definition of cloud top given in revised caption of Table 1 was added.
Table 2 – the caption should report the definition of "ship-seeded" / "ship-unseeded" as well as "detrained" and "wall"
The definitions were added to the caption. Entries were rephrased as "seeded" and "unseeded" also.
p8 l14 – the plume with Na> 1000 cm-3 seems to me much narrower than 60 km. I would guess, only a few km
The 60km refer to the length and not the width of the plume. This was clarified in the revised manuscript.
p9l4 – were → was
Done.
p10 last line – reduction compared to what? to the ctrl simulation?
The decrease in Nd_top is with respect to the cell wall cores. This has been clarified in the revised manuscript.
p12l7-9 – I have difficulties following this argument. What is meant by "domain-average Acld"? Is it what is labeled "albedo" in Table 2? It is hard to understand that the increase in in-cloud albedo by 5
The revised caption in Table 2 together with the renaming of "albedo" as Aall and Acld as appropriate should clarify this discussion in the revised manuscript.
The calculation is that:
CF_filament_ship*Acld_filament_ctrl+CF_well_ship*Acld_wall_ctrl=0.9* CF_filament_ship*Acld_filament_ship+CF_well_ship*Acld_wall_ship
where e.g. "CF_filament_ship" corresponds to the cloud fraction entry of Table 2 for the cloud filaments of the ship simulation.
p12l17 – this decrease in in-cloud LWP is not documented in the Tables or figures. Or is the area covered by detrained + wall the entire cloudy area?
Yes, the table entries under filament and wall are cloud-mean only and one can see the decrease from detrained ctrl to detrain seeded of the ship simulation. This is now clarified in the revised caption of table 2.
p12l20 – given the relatively small mean increases in Nd, and the fact that albedo is much less sensitive to Nd changes than to L changes, is this plausible? Or is the distribution of the relative changes of both quantities relevant?
The increase in in-cloud Acld of the filaments was attributable to changes in Nd and the Twomey effect. In-cloud mean LWP within the filaments is found to decrease and can therefore not contribute to the brightening. However, it should be noted that 90% of the increase in domain-mean Acld does not come from brighter clouds, but is attributed to the increase in cloud-fraction assuming an equally bright cloud distribution. This should now be clearer in the revised manuscript given the more carefull defintion of all-sky and in-cloud means.
p14l5 – it probably rather is "satellite-based estimates in CRE changes"
Done.
p14l16 – maybe specify "even in the absence"?
We decided to keep the sentence as is.
p16l1 – superfluous comma
Removed.

p16l26 – the
Done.
p16l27 - “annual mean insolation”
Changed.
p16l31 – extent
Changed.

Supplement
Fig. 2a – this seems to me an awkward definition of a CDF. I am used to CDFs that increase monotonically and asymptotically approach 1 (as in Fig. 5, in fact).
This was a mis-noma of the Figure. Indeed the probability density function (PDF) is shown. This has been corrected.

Fig. 3a – it would be good to indicate the ship position here, as well as the past ship track
We have played around with super-imposing the aerosol perturbation in a transparent layer over the cloud fields and decided for clarity of the figures not to do it. The information is instead displayed separately in Fig 3 of the main manuscript (Fig 3a - aerosol plume and Fig 3b - Nd_top field) separately.
Fig. 6 – this figure seems to be wrong. I believe the authors intended to show Nd_top vs. Acld histograms in all rows.
Thank you for pointing this out. The Figure has been corrected in the revised supplement.

Authors response to: "Interactive comment on "The efficacy of aerosol-cloud-radiative perturbations from near-surface emissions in deep open-cell stratocumulus" by Anna Possner et al.

Anonymous Referee #2
Received and published: 18 September 2018

General comments: Possner et al present an interesting modeling study in which the effect of ship emissions on cloud microphysical and macrophysical properties of deep open cells is examined. Based on field campaign measurements, and previous modeling study of Wang et al 2010, Possner et al. show that despite the lack of typical linear ship tracks, the cloud adjustments can be significantly larger than one would expect. The manuscript is well written and the analysis support the authors conclusions. I recommend the manuscript to be published after minor revisions.

We thank the reviewer for their evaluation and have addressed their comments in the revised manuscript. Individual responses to each of the issues raised are inserted below.

Specific comments: 1. The authors cite that 70% of marine Sc form in deep boundary layers (p16l19; citation needed, e.g., Muhlbauer et al., 2014). Do the 70% compose mainly open cells? How much of the 70% are closed cells? This needs to be mentioned in order to asses the global effect of ship emissions in deep open cells.

From Table 4 of Muhlbauer et al 2014 we know that open-cell and disorganised stratocumulus clouds occur far more often than the closed-cell regime. A further height-dependend split of the frequency of occurrence in open, closed and disorganised stratocumuli was not obtained by Muhlbauer et al 2014 and is not available to the authors. While the authors agree that this would be needed for a global assessment of the radiative impact of ship emissions, it is beyond the scope of this study which focused on the feedback mechanisms in such regimes and explored the potential for radiative impacts, which remained previously unexplored and unquantified.

The issue of relative regime occurrence was previously only mentioned in the introduction of the submitted manuscript (P2L26ff):
"Yet, over 70% of stratocumulus clouds are found in deeper boundary layers (Muhlbauer et al., 2014). The potential for albedo changes is particularly high in the open-cell, and disorganised stratocumulus regimes, which occur more frequently in the sub-tropics, than in the closed cells regime (Muhlbauer et al., 2014)." A more quantitative statement has now also been added to the conclusions (including reference to Muhlbauer et al 2014).

2. The domain mean increase in albedo is function of the domain size. For a smaller domain, the increase would be larger, and for larger domain size, smaller. Therefore, domain mean increase in albedo of 0.05 is somewhat arbitrary. If the authors can estimate the density of ship tracks in a given regions with frequently observed deep open cells, a more meaningful value of regional mean increase in albedo can be estimated.

The following paragraph on this, which incorporates the concerns raised under point 6 has been added to the manuscript at P14L20:
"Furthermore, while these simulations are highly idealised in their setup, they do not necessarily reflect unrealistic emission conditions. The prescribed ship is assumed to travel periodically along an identical emission line without any cross-wind, which may reduce the plume size or dilute emissions more effectively. Within the 48h simulation, a total of 5 ships traverse the 180 × 180 km2 domain and the cloud response to their combined emissions is assessed. Throughout most of the North Pacific a shipping density of around 30 ships per 100 km2 per year is observed

(MarineTraffic). Assuming a speed of 10 m s^{-1}, such a density corresponds to an estimated number of 58 ships within the simulation domain on average. Within the North Atlantic, the higher density of ships could even correspond to over 200 ships within a 180 × 180 km^2 domain (MarineTraffic). Therefore, our emission scenario is equivalent to merely □ 10 % (3 %) of these ships contributing to increased CCN concentrations within the seeded domain."

3. The authors claim in the abstract that changes in cloud-radiative properties are masked by the natural variability. What is the meaning of natural variability in this context?
For clarification we rephrased "natural variability" with "naturally occurring variability" (i.e. the variability occurring within the cloud field without an anthropogenic aerosol perturbation).

The abstract further says that the above can be overcome by utilizing the spatio-temporal distribution of the aerosol perturbation. However, in Figure 3 the aerosol plume can be easily seen in Nd, which serves as a tracer to where one can expect cloud adjustment. This can be used in observational studies.
In Figure 3 only small parts of the entire seeded domain are characterised by perturbations in cloud-top Nd of 100 cm-3 or above. Throughout most of the seeded domain, where CCN concentrations are increased by a factor 2, or even in regions were the CCN concentrations exceed 100cm-3, Nd is well within the background variability. Furthermore, the higher concentrations of Nd~100cm-3 are neither unrealistically high for a naturally occuring background, nor spatially coherent to be picked up as an unambiguous marker of an anthropogenic perturbation. Finally, if one were to use only sub-regions where Nd is increased, one would likely miss the full radiative response simulated here for open-cells, where the predominant forcing seems due to an increase in cloud cover of the cloud filaments where Nd is low. For these reasons the authors remain with the statement that our simulations indicate that the spatio-temporal distribution of the aerosol is needed to determine the full extent of the cloud-radiative impact by the ship emissions in this regime.

4. The authors should improve the description of the tables: Table 1: The caption says the simulated values are domain mean. These values are compared with RF06, which seems to be in-clouds values (for LWP at least). Clarification is needed. Table2: The left column is unclear. What is the difference between ship, ship-seeded and ship-unseeded? (ship-unseeded is not mentionedanywhere else in the text). Under the CF column, how CF can be not 100% inside walls? given that walls are defined by ascending air? Is the wall CF is the fraction of walls out of the total CF/domain? If so it means that there is also a dynamical adjustment.

We agree with the reviewers, that the captions provided insufficient information. Both captions have been revised and the LHS column of Table 2 renamed in the revised manuscript.

5. I recommend to elaborate more in the introduction on previous studies that attempted to quantify the regional effect of ship tracks (e.g., Schrier et al. 2006, 2007, Peters et al. 2011).
Missing references were added to the manuscript.

6. The simulation assumes an idealized case with no perpendicular winds. I assume that most ship tracks don't have head/tail winds, rather side winds. Would the wind direction relative to the emission source increase/decrease the regional area that is affected by the emissions? This should discussed.
This is incorporated into the new paragraph included in the revised manuscript, which is presented under point 2.

Technical corrections: Section 2.2: What was the duration of the simulations?

P4L20: Both simulations were run for 48h.
P8l6: Remove "and".
"A comma was introduced to clarify the sentence."
P8l15: Any statistical tests were done to determine the 30km band around the emission line? It is mentioned that inside this region Na_sub are elevated, but by how much? I also would expect the plume to expand and dilute as it gets more mature, and not being fixed.
The seeded domain was conservatively identified as the corridor, where CCN concentrations were increased and the meandering plume, consisting of the super-position of 5 consecutive ships, remained within the bounds of this region. We agree that for an individual ship one would expect a widening of the plume with distance. In previous analyses performed for this study we have quantified the core plume in these simulations where Na was outside 3 standard deviations of the background concentration, but this constrained the analysis to a rather narrow region of highest concentration around the emission line. No further insight was obtained from these results and they were therefore omitted in the manuscript. We therefore felt, that it was more insightfull to distinguish in the analysis between a region where CCN concentrations were elevated and a region where no increase in CCN was detected.

P12l7-10: This paragraph is not clear.
The paragraph has been rephrased for clarity.
P12l28: Observational studies showed ship tracks closing open cells (e.g., Goren and Rosenfled 2012 where at least part of the open cells seems to be deep, based on the cells spatial scale; Christensen and Stephens 2012). While simulations does not show a reverse transition, observational evidences should be provided as well.
The "large open cells" in Goren & Rosenfeld (2012) are estimated to be around 20-25km, which is roughly half the size of the cells simulated here. Furthermore, observational evidence (Durkee et al 2000b, Toll et al 2017, Chen et al 2015, Christensen & Stephens 2012) suggests that ship tracks in boundary layers deeper than 1km are extremely unlikely. Therefore, we would not expect to simulate a track-like structure in these simulations. However, it seems that a similar process occurs in deep boundary layers, where the large open cells partially fill in, but the filaments never stretch across the entire cell, which would then allow it to recover (given sufficient mixing generated through cloud-top cooling to overcome sub-cloud stability). The Goren and Rosenfeld reference was added to the manuscript (P12L33).

P16l17: Consider changing "such tracks" to "linear shaped tracks", and to add that they are rare in deep boundary layers in comparison to shallow boundary layers (reference is needed).
Rephrasing was done and references from the introduction repeated here.
P16l28-29: How do the fractional percentage calculated? From which table?
The paragraph was rephrased slightly such that now all numerical results are merely summarised here without reference, but are referenced to to corresponding figures and tables in section 3.
P16l26 the → the.
Done.
P16l27 annular → annual.
Done.
Figure 5: In the caption the boundary layer depth for each of the simulations should be provided (i.e., shallower in Wang et al 2011).
The information was added.
Figure 6: In order to cover also the night time in Figure 6d, consider replacing (or adding) cloud optical thickness with cloud albedo?
As only day-time values in Aall and Acld are considered throughout the paper, the night-time values are not shown here for consistency. Although they are diagnosed, we ommitted them in the averaging process, as they have no physical meaning but affect the temporal mean due to the diurnal cycle (clouds are optically thicker at night).

Supplementary: Caption 3: What is “ship_open”?
This typo was removed.
Caption 6: remove “and” in line 3. Caption is not clear. X axis label is not consistent.
This was changed.

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The efficacy of aerosol-cloud-radiative perturbations from near-surface emissions in deep open-cell stratocumulus

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Abstract. Aerosol-cloud-radiative effects are determined and quantified in simulations of deep open-cell stratocumuli observed during the VOCALS-REx campaign off the West coast of Chile. The cloud deck forms in a 1.5 km-deep boundary layer with cell sizes reaching 50 km in diameter. Global data bases of ship tracks suggest that these linear structures are seldom found in boundary layers this deep. Here, we quantify the changes in cloud-radiative properties to a continuous aerosol point source moving along a fixed emission line releasing 10^{17} particles per second. We show that a spatially coherent cloud perturbation is not evident along the emission line. Yet our model simulates an increase in domain-mean all-sky albedo of 0.05, corresponding to a diurnally-averaged cloud-radiative effect of 20 W m^{-2}, given the annual mean solar insolation at the VOCALS-REx site. Therefore, marked changes in cloud-radiative properties in precipitating deep open cells may be driven by anthropogenic near-surface aerosol perturbations, such as those generated by ships.
Furthermore, we demonstrate that these changes in cloud-radiative properties are masked by the naturally occurring variability within the organised cloud field. A clear detection and attribution of cloud-radiative effects to a perturbation in aerosol concentrations becomes possible when sub-filtering of the cloud field is applied, using the spatio-temporal distribution of the aerosol perturbation. Therefore, this work has implications for the detection and attribution of effective cloud-radiative forcing in marine stratocumuli, which constitutes one of the major physical uncertainties within the climate system. Our results suggest that ships may sometimes have a substantial radiative effect on marine clouds and albedo, even when ship tracks are not readily visible.

1 Introduction

Aerosol-cloud interactions (aci) in low-level clouds, which span just over a fifth of the Earth's ocean surface (Wood, 2012), contribute the largest uncertainty to estimates of global mean effective radiative forcing (ERF) of anthropogenic aerosols (Myhre et al., 2013). Current estimates of ERF$_{aci}$ range from -1.2 W m^{-2}, which would constitute a strong global cooling which would partially offset the effects of warming due to anthropogenic greenhouse gas emissions, to 0.0 W m^{-2}, which would render these effects negligible at the global scale (Boucher et al., 2013).

To reduce this uncertainty substantially through use of satellite retrievals and global climate models (GCMs) remains challenging. These challenges include issues of collocation in retrievals of aerosol and cloud properties from space (Koren et al., 2007; Charlson et al., 2007), and the inadequate representation of small-scale dynamical processes that contribute to the cloud response in coarse-scale models (Nam et al., 2012; Schneider et al., 2017). Valuable insights on the involved processes and plausible ranges of cloud-radiative perturbations through aerosols have been obtained by the study of ship tracks. These *anomalous cloud lines* (Conover, 1966) are a phenomenon associated with a characteristic spatial structure, which occurs in low-level stratocumulus clouds. The changed cloud-radiative properties within these tracks can be attributed to localised aerosol perturbations (Durkee et al., 2000a, c; Schreier et al., 2006).

Data bases obtained from satellite retrievals (Coakley and Walsh, 2002; Christensen and Stephens, 2012; Chen et al., 2015), as well as high-resolution modelling studies (Wang and Feingold, 2009; Wang et al., 2011; Berner et al., 2015), show that the net cloud-radiative effect (CRE) in individual ship tracks does not depend only on cloud droplet number increases and size decreases, which occur in almost all cases (Chen et al., 2012), but also on induced changes in cloud morphology, cloud fraction and liquid water path (LWP). Cloud albedo (A_{cld}) may not always increase with increased levels of pollution, but may also decrease (Christensen and Stephens, 2012; Berner et al., 2015). Furthermore, localised gradients in aerosol concentration have been shown to induce self-sustaining mesoscale circulations (Chen et al., 2015; Wang et al., 2011), by the local suppression of precipitation in the polluted cloud and the convergence of cold pools transporting moisture into the polluted cloud from the surrounding precipitating clouds. In the global mean, LWP increases of between 16 % and 24 % (depending on above-cloud moisture content) were found in ship tracks that formed within the precipitating cloud regime (Toll et al., 2017).

However, ship tracks are rare in comparison to the number of ocean-going ships that criss-cross the world's oceans (Schreier et al., 2007). Merely 1924 ship tracks were detected over two years worldwide (Campmany et al., 2009), while the total ocean-going fleet consists of over 50,000 ships that exceed 500 gross tons in weight (European Maritime Safety Agency, 2014). Understanding what constrains their occurrence in terms of background pollution, boundary layer dynamics and large-scale stability, may facilitate constraining regimes and magnitudes of global effective radiative forcing estimates. In particular, studies of ship tracks in high-resolution models and satellite retrievals have been mostly limited to extremely shallow boundary layers, that range in depth from 300 m to 600 m (Christensen and Stephens, 2012; Berner et al., 2015; Chen et al., 2015). Ship track formation within one slightly deeper boundary layer of 800 m was investigated in high-resolution simulations by Wang and Feingold (2009) and Wang et al. (2011). Indeed ship tracks are very rarely detected in satellite retrievals of boundary layers deeper than 800 m (Durkee et al., 2000b; Toll et al., 2017). Yet over 70 % of stratocumulus clouds are found in deeper boundary layers (Muhlbauer et al., 2014).

The potential for albedo changes is particularly high in the open-cell, and disorganised stratocumulus regimes, which occur more frequently in the sub-tropics, than in the closed cell regime (Muhlbauer et al., 2014). Both of these regimes are characterised by shallow convective cloud structures that detrain laterally at cloud top. The detrained cloud sheets that span the regions between the convective structures are optically thin (cloud optical thickness $\tau < 3$), are often associated with low droplet number concentrations ($N_d \sim 5\,\mathrm{cm}^{-3}$), and may contribute substantially to the overall cloud fraction (Wood et al., 2018). Thus, their albedo is highly susceptible to aerosol perturbations from the perspective of Platnick and Twomey's (1994) albedo susceptibility definition. Yet, the efficacy of aerosol-cloud-radiative interactions within these detrained cloud segments remains unclear. In general, the processes governing aerosol-cloud interactions in deep stratocumulus boundary layers remain weakly constrained, with only few process-level studies (Wang et al., 2010; Kazil et al., 2011; Wood et al., 2011b; Zuidema et al., 2016) that quantify effects on cloud characteristics through aerosol pollution.

Within this study we quantify changes in cloud-radiative properties due to aerosol perturbations in deep (boundary layer depth of $\sim 1.5\,\mathrm{km}$) open-cell stratocumulus clouds and discuss dominant mechanisms that constrain the cloud-albedo response.

2 Methodology

2.1 Case description

This study is based on a well-documented case of open-cell stratocumulus clouds embedded within a $\sim 1.5\,\mathrm{km}$ deep boundary layer, which was observed during research flight 6 of the VAMOS Ocean-Cloud-Atmosphere-Land Study Regional Experiment (VOCALS-REx) campaign. Detailed information on the particular case and measurement techniques can be found in Wood et al. (2011a) and Wood et al. (2011b) respectively. Here we give an overview of the two cloud regimes and their characteristics relevant to this study.

Table 1. Spatio-temporal averages of liquid water path (LWP), surface precipitation (R_{sfc}), cloud base precipitation (R_{cb}), cloud fraction (CF), sub-cloud mean boundary layer aerosol concentration (N_{a_sub}) and cloud-top droplet number concentration (N_{d_top}) are presented. The first row containing data shows the observations of the open-cell stratocumulus deck obtained during research flight RF06 of the VOCALS-Rex campaign on October, 28^{th}, 2008 between 08:00 – 13:30 UTC. The numerical results, shown in the last two rows, were averaged over the identical time periods over both simulated days. Domain-mean values were computed for LWP, CF, and N_{a_sub}. R_{sfc} was averaged only over values exceeding $0.1\,\mathrm{mm\,day^{-1}}$ and R_{cb} was averaged for all $R_{\mathrm{cb}} > 0\,\mathrm{mm\,day^{-1}}$ (consistent with observations). N_{d_top} was diagnosed at the highest model level where cloud water content exceeded $0.01\,\mathrm{g\,m^{-3}}$ and averaged horizontally (cloudy-points only). Numbers in brackets denote the interquartile range of each variable, which covers the spatial and temporal variability of the cloud field. Numerical results are shown for the control simulation (*ctrl*) and the aerosol-perturbed simulation (*ship*). Further details on simulations can be obtained in text.

08:00 – 13:30 UTC averages						
Sim/obs	LWP [g m^{-2}]	R$_{sfc}$ [mm day^{-1}]	R$_{\mathrm{cb}}$ [mm day^{-1}]	CF [%]	N$_{a_sub}$ [cm^{-3}]	N$_{d_top}$ [cm^{-3}]
RF06	141	1	4 – 5	56	30	10
ctrl	75 [6, 70]	4.2 [0.2, 2.4]	9.1 [0.1, 2.9]	50	34 [32, 37]	8 [3, 11]
ship	82 [10, 81]	4.1 [0.2, 2.2]	8.6 [0.1, 2.9]	57	45 [32, 42]	12 [3, 13]

The cloud regime was sampled during the early morning hours (03 am to 08:30 am local time) on October 28^{th} in 2008. A summary of cloud properties measured during the campaign is given in Table 1. The characteristic cell size was found to be between 30 – 40 km, which is detected frequently in Southeast Pacific stratocumulus clouds (Wood and Hartmann, 2006). A cloud fraction of 56 % was measured in the open-cell regime, which is consistent with the observed high level of detrained cloudy air masses, which spread from the updraft cores into inner regions of the cell. Furthermore, a cloud cover of this extent is typical for marine open-cell stratocumulus (Muhlbauer et al., 2014; Terai et al., 2014).

The open-cell clouds coincided with moister sub-cloud layer air masses, as compared to the neighbouring closed-cell regime, and were characterised by low sub-cloud layer aerosol concentrations ($30\,\mathrm{cm}^{-3}$). A strong vertical gradient in aerosol concentration was observed within the open cells near cloud base where concentrations decreased rapidly. A strong horizontal gradient in cloud-top droplet number concentration (N_{d_top}) was observed (Wood et al., 2011a) between the updraft cores ($N_{d\_top} \sim 30\,\mathrm{cm}^{-3}$) and the detrained cloud filaments ($N_{d\_top} \sim 1-10\,\mathrm{cm}^{-3}$). Substantial rates of drizzle were observed at cloud base. Yet over 50 % evaporated before reaching the surface.

2.2 Simulation Setup

Two simulations were performed with the weather research forecast (WRF) model at the convection-resolving scale with a horizontal grid resolution of $300 \times 300\,\mathrm{m}^2$, a vertical resolution of 30 m and a time step of 3 s following Wang et al. (2010). The idealised simulations with periodic boundary conditions at the domain edges were initialised with meteorological profiles obtained during research flight 6 of the VOCALS-REx field campaign (Wang et al., 2011; Wood et al., 2011a). A brief overview of the research flight is given in the previous section 2.1. Given the large characteristic spatial scales of the cellular organisation of the cloud field with cells sizes ranging from 30 km to 40 km, simulations were performed on a large domain of $180 \times 180\,\mathrm{km}^2$. The domain was centered on 78°W and 15°S, which is off the west coast of Chile. The model top was specified at an altitude of 2 km, which is 600 m above the boundary layer top. Above this height a standard clear-sky atmosphere profile is assumed for the computation of the radiative fluxes until the top of atmosphere.

Both simulations were run for 48 h with a fixed divergence rate of $1.67 \times 10^{-6}\,\mathrm{s}^{-1}$, which was estimated from QuickSCAT surface winds (NASA, 2012), and prescribed surface fluxes. Surface latent heat and sensible heat fluxes were specified, according to field measurements, as $120\,\mathrm{W\,m}^{-2}$ and $15\,\mathrm{W\,m}^{-2}$ (defined as +ve upward) respectively. The surface pressure was specified as 1018 hPa. For simplicity, mean advective tendencies in the wind field were removed from the soundings.

The simulations were performed with the two-moment Morrison et al. (2009) microphysics scheme with a prognostic treatment of number and mass concentrations for cloud water and rain. The exponents of the cloud liquid water content and N_d in the Khairoutdinov and Kogan (2000) autoconversion rate were adjusted to values obtained from the VOCALS-REx field campaign as 3.19 (cloud water exponent) and -1.49 (N_d exponent) respectively. These exponents were obtained for the VOCALS-REx field data using the approach described in Wood (2005). Precipitation formation was artificially suppressed in the first 2 h of simulation to facilitate a thermodynamic adjustment to the initialisation sounding before including moisture sinks. Cloud condensation nuclei (CCN) were treated as in Wang et al. (2011) with a prognostic log-normal sea-salt mode centered at a mean diameter of 500 nm and variance of 1.5. Aerosols were advected according to grid-scale and subgrid-scale transport tendencies and aerosol-cloud interactions were included by removing aerosols upon activation, which was treated as in Kravitz et al. (2014). The release of aerosol upon complete evaporation of cloud droplets and rain drops was also simulated. A surface sea-salt emission flux of 20 $\mathrm{m^{-2}\,h^{-1}}$ was specified in line with estimates from previous simulations (Wang et al., 2010; Kazil et al., 2011).

In addition to the control simulation, from here on named *ctrl* simulation, an aerosol-perturbation experiment was designed. The simulation, named *ship* simulation, followed the setup of Wang et al. (2011) for direct comparison between the deep boundary layer case and shallow boundary layer case in terms of aerosol-cloud-radiative perturbations. A ship moving at 5 $\mathrm{m\,s^{-1}}$ through the center of the domain was allowed to continuously emit sea-salt at a rate of $10^{17}\ \mathrm{s^{-1}}$ and a mean dry radius of 300 nm. This flux was chosen to match emissions within a previously studied case within a shallow open-cell regime (Wang et al., 2011) (see section 3.2.1 for an in-depth discussion). Furthermore, these emissions were consistent with estimates proposed by Salter et al. (2008) for marine cloud brightening applications. The CAM radiation scheme was used in the simulations and A_{cld} was estimated as $A_{cld} = \tau/(\tau + 6.8)$, where τ denotes the cloud optical depth, which in turn was diagnosed as:

$$\tau = \int_{z=0}^{\infty} \frac{3q_l}{2\rho_w R_{eff}} dz, \qquad \rho_w = 997.0\ \mathrm{kg\,m^{-3}}, q_l = \text{liquid water content}, R_{eff} = \text{effective cloud droplet radius}, z = \text{height}. \tag{1}$$

The cloud base precipitation rate (R_{cb}) was computed as the mean precipitation flux across the lowest third of the cloud vertical extent, which is consistent with its estimation from observations.

3 Results

3.1 Evaluation of open-cell characteristics

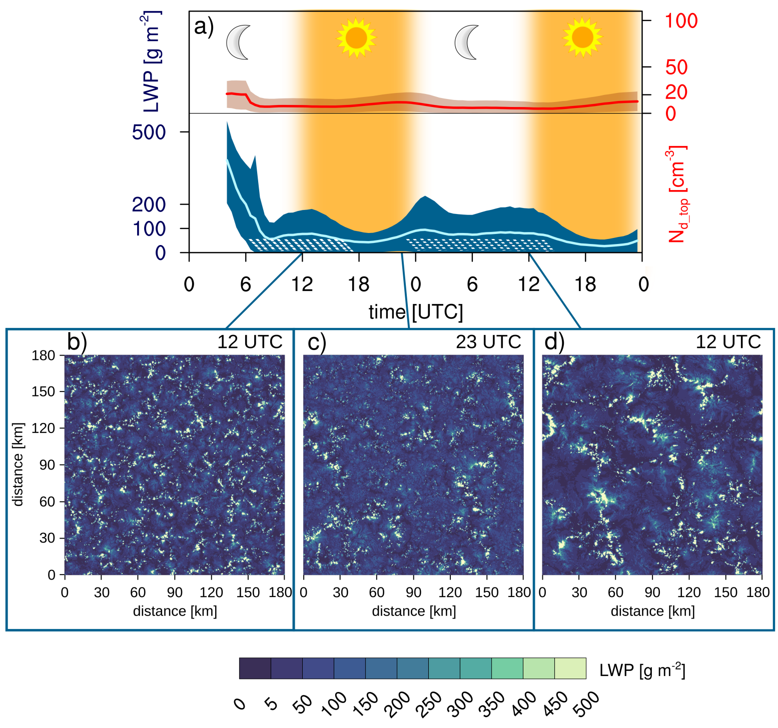

Figure 1. (a) Time series of domain-mean liquid water path, LWP (green), and cloud-top droplet number concentration, N_{d_top} (red),for the *ctrl* simulation. Shading (blue for LWP, black for N_{d_top}) denotes interdecile percentile range. Snapshots of LWP are shown in (b) after initial organised structures developed, (c) after the solar maximum and (d) for the second day organised state.

In order to assess the radiative effect of concentrated and localised aerosol pollution on deep open-cell clouds, the simulations need to demonstrate sufficient skill in capturing the characteristics and dynamics of the open-cell regime. Following initialisation, an unorganised stratiform cloud deck formed in the *ctrl* simulation. Initial organised structures appeared 6 h after initialisation following the onset of precipitation (Fig. 1). Following the second night, observed length scales of organisation (see section 2.1) were simulated.

The diurnal evolution of LWP and N_{d_top} is shown in Fig. 1. Periods, when R_{cb} exceeds 3 mm day^{-1} are marked in white. The simulation showed a pronounced diurnal cycle during both days in LWP and R_{cb}. As in Wang et al. (2010), solar heating was found to break up the cell walls, which led to a reduction of LWP in the upper percentiles, a reduction in cloud-base precipitation rates to $R_{cb} < 2$ mm day^{-1}, and consequently the loss of cloud-field organisation in the late afternoon. During the night the cloud deck recovered and organisation was re-established.

The *ctrl* simulation was characterised by a well-mixed cloud layer and stably stratified sub-cloud layer (see Fig. S1), which is characteristic for deep boundary layers. This structure developed rapidly following initialisation from the well-mixed state. Within the first 3.5 h the boundary layer deepened by 180 m before stabilising at 1.5 km and the sub-cloud layer became stratified. A mean R_{cb} of 9.1 mm day^{-1} (Table 1) was simulated in the early morning hours of the VOCALS-REx field campaign. Although simulated mean R_{cb} was within the spread of observed precipitation rates (Fig. S2), it was roughly twice as high as the mean R_{cb} rates inferred from observations (Table 1). Meanwhile, the mean LWP was underestimated by a factor 2 in the open-cell regime, which is consistent with an over-estimation in precipitation. However, the simulated cumulative precipitation distribution shown in the supporting material (Fig. S2a) showed that the overall distribution of R_{cb} was well captured in the *ctrl* simulation and that the bias in the mean originates from the slight overestimation of intense precipitation events exceeding 20 mm day^{-1}. These events are likely to be found within the walls of the open-cells, which are characterised by strong updrafts (Fig. S2b).

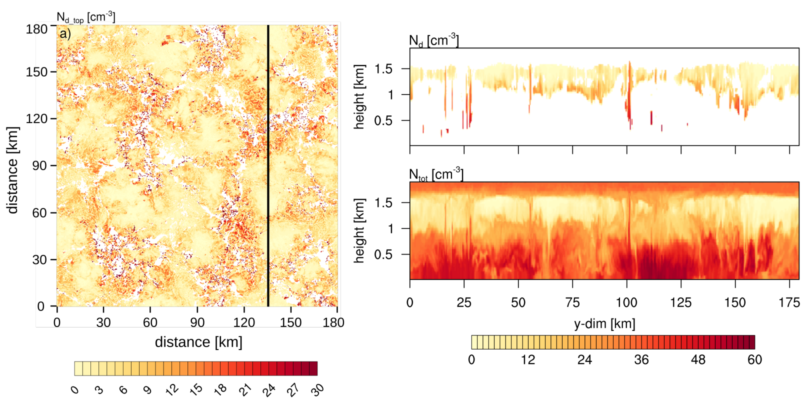

Figure 2. (a) instantaneous cloud-top droplet number concentration, N_{d_top} (corresponding snapshot to LWP field shown in Fig. 1d). Black line denotes location of cross-section of (b) cloud droplet number concentration (N_d) and (c) total number concentration ($N_{tot} = N_a + N_d$, where N_a denotes the aerosol number concentration). Cloud top is defined as in Table 1.

The microphysical quantities, such as the mean sub-cloud layer aerosol concentration ($N_{a\_sub} = 34\,\mathrm{cm}^{-3}$) and $N_{d\_top} = 8\,\mathrm{cm}^{-3}$ were in good agreement with the observations. In the simulations the aerosol particles are lifted into the cloud layer within the cell walls, where they activated and N_d was relatively high. Cloud filaments, many of which are quite optically thin, were detrained horizontally, and are characterised by low N_{d_top} (Fig. 2a) due to the efficient removal through precipitation. While cell-wall N_d may reach up to 40 cm^{-3} in the *ctrl* simulation (Fig. 2b), characteristic N_d in detrained cloud filaments, sometimes reffered to as "veil clouds" (Wood et al., 2018), were as low as 2–3 cm^{-3}. The efficient removal of aerosol particles through cloud processing combined with the stable stratification in the sub-cloud layer induced strong vertical gradients in the combined particle number concentration N_{tot} defined as $N_{tot} = N_a + N_d$, where N_a denotes the aerosol number concentration. Sub-cloud layer N_{tot} ranged between $30-60\,\mathrm{cm}^{-3}$, while values below $10\,\mathrm{cm}^{-3}$ above cloud base height were simulated frequently (Fig.2c).

In summary, despite remaining biases in the mean LWP and R_{cb}, the simulation overall captured a realistic evolution of the open-cell cloud deck with a pronounced diurnal cycle. Since the overall cloud-cell statistics (Table. 1) and the horizontal cloud cover are consistent with observations, it gives us confidence that the underlying cloud dynamics were captured in the *ctrl* simulation. Regions of detrained cloud spanned 36% of the domain, and were characterised by low in-cloud LWP and N_{d_top}, which makes them particularly susceptible to aerosol-induced cloud-radiative perturbations. Yet any near-surface source of pollution will predominantly be transported into the cloud layer through the cell walls given the pronounced vertical stratification in the sub-cloud layer, where wet aerosol removal processes are efficient. It therefore remains to be seen whether substantial changes in cloud-radiative properties can be induced by near-surface aerosol perturbations.

3.2 Efficacy of aerosol perturbation

Table 2. Same as Table 1 but for mean values of the last 24 h period. The following additional variables were added to the table: cloud albedo (A_{cld}), all-sky albedo $A_{all} = CF * A_{cld} + (1 - CF) * A_{clr}$ and cloud-top effective cloud droplet radius (R_{eff_top}). A_{clr} denotes the clear-sky albedo which was determined as $A_{clr} = 0.06$ in both simulations. R_{eff_top} was diagnosed similarly to N_{d_top} and averaged over cloudy regions only. For A_{all} and A_{cld} domain averages (i.e. including clear-sky and cloudy-sky) are given. All entries for "wall" (cloud with updraft $> 0.5\,\mathrm{m\,s}^{-1}$) and "detrained" (non-wall cloud) regions, denote in-cloud averages only. CF for wall and detrained cloud denotes the domain area fraction covered by each category. Inside cell walls and cloud filaments CF is 100 %. The "seeded" region is defined as ±30 km around the emission line and the remainder of the domain is classified as "unseeded".

2nd day mean									
Sim/obs	LWP [g m^{-2}]	R_{sfc} [mm day^{-1}]	R_{cb} [mm day^{-1}]	A_{cld}	A_{all}	CF [%]	N_{a_sub} [cm^{-3}]	N_{d_top} [cm^{-3}]	R_{eff_top}
ctrl	64 [2, 57]	4.6 [0.2, 2.4]	8.4 [0.1, 2.3]	0.18 [<0.01, 0.30]	0.22	44	36 [32, 39]	9 [3, 12]	21 [17, 26]
detrained	92 [38, 101]	4.5 [0.2, 2.6]	9.2 [0.3, 4.6]	0.37 [0.28, 0.43]	0.13	36	–	8 [3, 12]	23 [19, 27]
wall	331 [71, 449]	10.0 [0.4, 7.8]	33.7 [0.4, 18.5]	0.61 [0.45, 0.77]	0.05	8	–	13 [4, 18]	21 [17, 25]
ship	75 [10, 73]	4.5 [0.2, 2.1]	7.7 [0.1, 2.2]	0.26 [0.07, 0.41]	0.27	58	50 [33, 50]	14 [4, 17]	21 [16, 26]
seeded	87 [18, 88]	4.8 [0.2, 2.2]	7.6 [0.1, 2.0]	0.33 [0.16, 0.48]	0.35	72	77 [47, 81]	24 [6, 31]	18 [14, 22]
detrained	87 [38, 99]	4.1 [0.2, 2.1]	5.9 [0.1, 2.4]	0.43 [0.32, 0.52]	0.25	59	–	25 [7, 32]	19 [14, 22]
wall	254 [54, 307]	9.6 [0.3, 6.8]	24.6 [0.2, 8.1]	0.59 [0.43, 0.75]	0.08	13	–	36 [10, 46]	18 [14, 21]
unseeded	67 [5, 67]	4.3 [0.2, 2.1]	7.7 [0.1, 2.3]	0.21 [0.04, 0.36]	0.24	51	35 [31, 38]	9 [3, 11]	22 [18, 27]
detrained	92 [41, 104]	4.1 [0.2, 2.2]	8.1 [0.4, 4.1]	0.37 [0.29, 0.44]	0.16	43	–	8 [3, 11]	24 [19, 28]
wall	299 [68, 414]	9.6 [0.3, 7.1]	30.2 [0.4, 15.2]	0.57 [0.41, 0.73]	0.05	8	–	12 [4, 17]	22 [18, 26]

The sea-salt perturbed simulation displayed a spatially constrained aerosol plume meandering around the emission line (Fig. 3a). The highest values of N_{a_sub} exceeding $1000\,\mathrm{cm}^{-3}$ were found within a narrow plume extending in length up to 60 km behind the point source. Overall, the aerosol perturbation remained spatially constrained within the boundary layer in a region of $\pm 30\,\mathrm{km}$ around the emission line. This strip of the domain (spanning $60 – 120\,\mathrm{km}$ in the y-direction) is characterised by increased levels of N_{a_sub} and will be from here on referred to as "seeded", whereas the domain outside this region will be referred to as "unseeded".

Inside the seeded region the emitted aerosol was predominantly transported into the cloud within the updrafts of the cell walls (Fig. S3). Despite efficient wet-removal processes within the cell walls, the largest absolute changes in N_d were as large as $600\,\mathrm{cm}^{-3}$. At cloud-top, increases in N_{d_top} of up to $150\,\mathrm{cm}^{-3}$ were found (Fig. 3b). From the cell walls, the increased levels of N_d persisted to the detrained cloud regions (Fig. S3), where the largest relative increases in N_{d_top} were found. On average N_{d_top} increased by 177 % within the cell walls and by 213 % within the stratified detrained cloud (Table 2). In this analysis cell walls were diagnosed as cloud-covered regions with updraft speeds exceeding $0.5\,\mathrm{m\,s^{-1}}$. All remaining, non-wall cloudy grid points were classified as "detrained cloud".

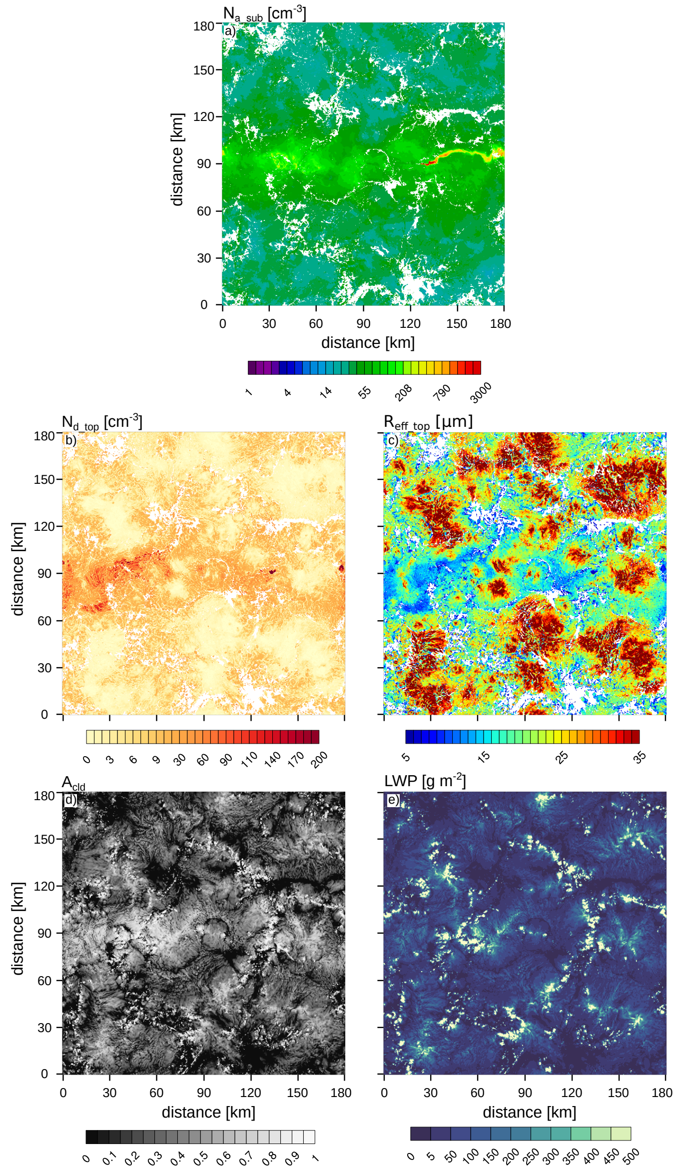

Figure 3. Snapshots of a) vertically averaged sub-cloud layer aerosol concentration (N_{a_sub}), b) cloud-top cloud droplet number concentration (N_{d_top}), c) cloud-top mean effective cloud droplet radius (R_{eff_top}), d) cloud albedo (A_{cld}) and e) liquid water path (LWP). Instantaneous fields are shown at 12 UTC for the *ship* simulation. Fields for the *ctrl* simulation are shown in the supporting material (Fig. S4).

The largest decreases in the cloud droplet effective radius at cloud top (R_{eff_top}) were found to coincide with regions of large increases in N_{d_top} (Fig. 3c and Fig. 3b respectively). R_{eff_top} may be reduced by up to 10 μm locally. The largest decreases in R_{eff_top} were found within the vicinity of strong updrafts. Here, many aerosols were carried into the cloud layer and were activated. Efficient in-cloud scavenging led to a reduction in N_{d_top} and an increase in R_{eff_top} going radially outward from the center of the updraft cores. Averaged over the seeded domain, a mean reduction in R_{eff_top} between 3 – 4 μm was simulated in the cell walls and detrained cloud regions (Table 2).

The changes in cloud-microphysical properties led to an increase in domain-averaged LWP (Table 2) within the seeded (36 %) and unseeded regions (5 %). Yet lower mean-values of in-cloud LWP were found within the detrained cloud and cell-wall regimes in both domains of the *ship* simulation (Table 2). R_{cb} was found to decrease by 0.7 mm day^{-1}, while R_{sfc} remained largely unaffected by the aerosol perturbation (Table 2).

Due to the reduction in R_{cb}, more cloud water was retained within the updraft and detrained horizontally into the stratified cloud filaments, which penetrated deeper into the open cells. The increase in areal extent of the detrained cloud sheets was accompanied by a shift (Fig. S5) in in-cloud LWP distribution towards lower LWP between 50 – 150 g m^{-2}. Therefore, the increase in domain-mean LWP, despite the decrease in in-cloud LWP, was attributed to the 14 % increase in cloud fraction from 44 % in the *ctrl* simulation to 58 % in the *ship* simulation (Table 2). Yet the open cells remain partially uncovered, which prevents a potential transition from the open-cell state to the closed-cell regime.

Mean A_{cld} is increased by 0.15 from 0.18 in the *ctrl* simulation to 0.33 inside the seeded region of the *ship* simulation (Table 2). This translates to a change in all-sky albedo (A_{all}) of 0.11 inside the seeded region, which corresponds to a shortwave cloud-radiative effect (SW CRE) of 44 W m^{-2} at an annual mean solar insolation of 404 W m^{-2} at the VOCALS-REx field site. Although the strongest increase in A_{all} was confined to the seeded domain, A_{all} was found to be increased throughout the simulation domain (Fig. S6). Averaged over the entire domain A_{all} increased by 0.05, which is equivalent to a SW CRE of 20 W m^{-2} exerted over an area of 180 × 180 km^2.

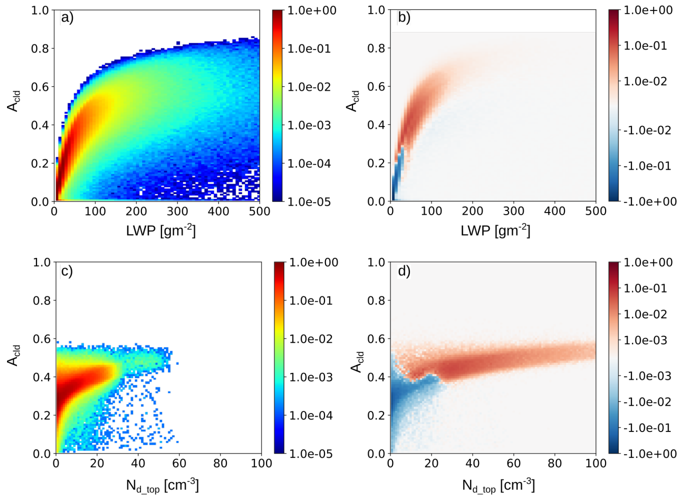

Figure 4. Occurrence rate F [%] for the a) liquid water path (LWP) versus cloud albedo (A_{cld}) phase space and c) the cloud-top droplet number concentration (N_{d_top}) versus A_{cld} phase space. The N_{d_top}-A_{cld} space was sub-filtered for LWP within the range of $40-60\,\mathrm{g\,m^{-2}}$. Results are shown in a) and c) for the last 24 h of the *ctrl* simulation. Absolute changes in F for the *ship* simulation with respect to the *ctrl* simulation are shown in b) and d) respectively. F is normalised to 100 % across the shown phase space. The bin widths for each of which F is defined are $\Delta LWP : 7\,\mathrm{g\,m^{-2}}$, $\Delta N_{d\_top} : 1\,\mathrm{cm^{-3}}$, and $\Delta A_{cld} : 0.01$.

The changes in domain-mean A_{all} were attributed to albedo changes of the detrained cloud sheets spanning the domain between the cell walls. Both the areal coverage and reflectivity of the detrained cloud sheets increased in the *ship* simulation as compared to the *ctrl* simulation (Table 2). Meanwhile, the cell-wall albedo of 0.6 remained unaffected by the aerosol perturbation. Furthermore, we attributed the simulated change in A_{all} predominantly to adjustments in macrophysical cloud properties of the detrained cloud regions. Changes in cloud microphysical properties and the associated Twomey (1991) effect were found to be of secondary importance to the change in all-sky albedo.

The increase in cloud fraction alone, while assuming no further changes in in-cloud A_{cld} (i.e. assuming A_{cld} as in the *ctrl* detrained and cell-wall regions in Table 2 and multiplying these values by the areal coverage of wall and detrained cloud regions of the *ship* simulation), accounted for 90% (100%) of the increase in domain-averaged A_{cld} inside the seeded (unseeded) domain. The additional increase in domain-mean A_{cld} within the seeded domain was attributed to the increase in A_{cld} from 0.37 to 0.43 within the cloud filaments of the *ship* simulation (Table 2).

Fig. 4 shows the normalised occurrence rate (F) within the detrained cloud regions. F, and the change in F due to the aerosol perturbation, is shown for each bin within the LWP-A_{cld} phase space (Fig. 4a and Fig. 4b respectively). The behaviour of F within the N_{d_top}-A_{cld} space, which was sub-filtered to only include points where in-cloud LWP ranged between $40-60\,\mathrm{g\,m^{-2}}$, is shown in Fig. 4c and Fig. 4d. The behaviour of F for other LWP sub-ranges was found to be qualitatively similar (Fig. S7).

The increased occurrence of moderate LWP values ($50 \leq LWP < 150\,\mathrm{g\,m^{-2}}$), may locally coincide with an increase in A_{cld} (Fig. 4b). Yet the overall decrease in in-cloud LWP by $5-23\,\%$ (Table 2) implied that the increase in A_{cld} within the cloud filaments could not be attributed to LWP adjustments. If anything, A_{cld} would be expected to decrease given the reduction in in-cloud LWP. Meanwhile, Fig. 4d displayed a clear shift in F towards higher N_{d_top} associated with locally increased A_{cld}. Hence, the increase in in-cloud A_{cld} was attributed to the Twomey (1991) effect within the stratified cloud.

3.2.1 Contrasting the cloud response in deep and shallow open cells

Although the areal coverage of the detrained cloud amount between the cell walls of the open cells increased, which contributed to the brightening of the cloud deck, the highly concentrated aerosol perturbation was insufficient to induce a transition from open to closed cells in these simulations. Aerosols may impact this transition via aerosol-precipitation interactions. Decreases in N_a from $90\,\mathrm{cm}^{-3}$ to $10\,\mathrm{cm}^{-3}$ facilitated a rapid transition from the closed to the open-cell state (Feingold et al., 2015) in previous simulations within the 800 m deep boundary layer observed during DYCOMS-II. Yet, the reverse transition from the open-cell state to the closed-cell state occurred over far longer time scales, if at all (Wang and Feingold, 2009; Feingold et al., 2015). Nonetheless, strongly concentrated sea salt emissions of 10^{17} particles s^{-1} within the same boundary layer, induced a transition from the open-cell to a filled-in cloud-cell state along the seeding line. Along the seeding line a secondary circulation maintained the cloud layer within the track while depleting the surrounding cloud (Wang et al., 2011). Such transitions from open cells to a closed-cell state along ship tracks have also been observed using remote sensing (Goren and Rosenfeld, 2012).

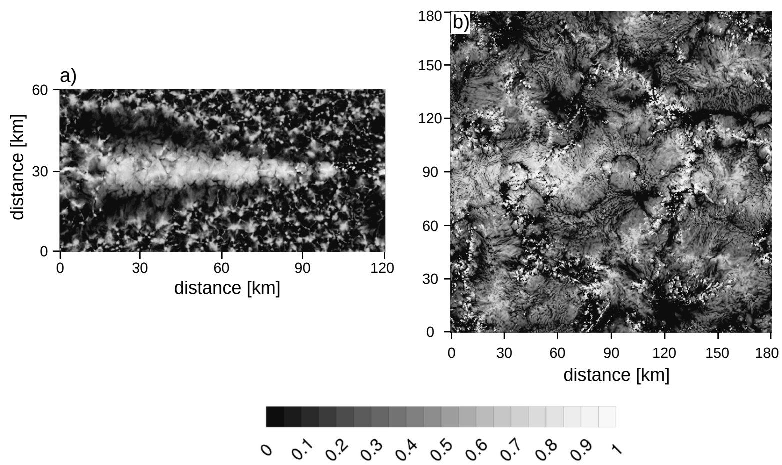

Figure 5. a) Cloud albedo field from Wang et al. (2011) Fig. 1a for an 800 m deep boundary layer. b) Cloud albedo at 12 UTC on second day of the *ship* simulation for a 1.5 km deep boundary layer. Both simulations were subject to an equal seeding source of 10^{17} particles s^{-1}.

While a ship track formed in the shallow boundary layer with open-cells between 10 – 15 km in size (Fig. 5a), a well-defined track does not form in the deep boundary layer with characteristic cloud cell sizes of 30 – 40 km (Fig. 5b). A ship track is also not detected in N_{d_top} or R_{reff_top} (Fig.3b and Fig.3c, respectively). The absence of a track in the deep boundary layer is largely attributed to: (i) the large spatial scales of variability within the background cloud state which is determined by the cloud dynamics and cloud-field organisation, and (ii) the incomplete filling in of the detrained cloud amount between cell walls, which prevents the transition to a 100% cloud-covered state.

A change in A_{cld} of 0.15, which is of the same magnitude as previously identified in ship tracks (Christensen and Stephens, 2011; Goren and Rosenfeld, 2012; Wang et al., 2011) is found embedded in deep open cells of the *ship* simulation. N_{d_top} was increased by 167 % and R_{eff_top} decreased by 14 % (Table 2). Yet, these effects remain seemingly hidden in the large variability of the cloud properties governed by the dynamics of the cloud cells. Furthermore, these effects may not easily be attributed to aerosol perturbations via remote sensing techniques of cloud properties as most of the changes in local cloud properties remain within the variability of the system.

Knowing the position and extent of the aerosol perturbation allows one to remove a sufficient amount of variability within the *ship* simulation to obtain a spatially constrained, detectable and attributable response within the cloud properties. As one averages along the spatial dimension of the aerosol perturbation (coinciding with the x-direction of the simulation domain) the pronounced shift in cloud properties between the seeded and unseeded regions of the *ship* simulation (Fig. 6) is highlighted. However, while changes in total albedo induced within the seeded region may be identified in this manner, the change in total albedo of 0.03 (Table 2) within the detrained cloud regions of the surrounding unseeded domain would still not be accounted for. Furthermore, changes in A_{cld} within the detrained cloud sheets were found up to 60 km from the emission line, which has implications for the definition of the truly unperturbed albedo within satellite retrievals of such scenes.

4 Implications for aerosol radiative forcing estimates in marine stratocumuli

Estimating the aerosol-induced radiative forcing in low-level marine clouds constitutes a considerable uncertainty in the overall cloud-radiative forcing of anthropogenic aerosols. Satellite-based estimates of CRE changes due to ship exhaust have remained inconclusive due to the high degree of variability within the natural cloud scene (Peters et al., 2011). GCM estimates provide a wide range of CRE changes between -0.6 Wm^{-2} and -0.07 Wm^{-2} (Lauer et al., 2007; Righi et al., 2011; Peters et al., 2012; Partanen et al., 2013) due to open-ocean shipping. Furthermore, it remains unclear whether GCMs represent the relevant scales of variability to provide reliable CRE estimates. The analysis of global datasets of ship tracks (Chen et al., 2015) and volcano plumes (Toll et al., 2017), which have been used as analogues to study the cloud response to anthropogenic emissions, have shown that in the global mean, the cloud response within the tracks largely follows the brightening expected by Twomey (1991). In the global mean, increases and decreases in LWP within the different cloud regimes seem to offset one another, while many GCMs predict a positive LWP response only (Wang et al., 2012; Ghan et al., 2016; Malavelle et al., 2017; Toll et al., 2017).

In this study we demonstrate that non-negligible amounts of brightening due to anthropogenic shipping emissions may persist in the absence of a clear ship track. In deep open cells, perturbations in A_{cld} were found to be as large as 0.15 in regions where ΔN_{a_sub} is high and as large as 0.08 when integrated over the whole simulation domain of $180 \times 180\,\mathrm{km}^2$. Furthermore, the induced brightening, which is almost as high as in simulations displaying a pronounced ship track ($\Delta A_{cld} = 0.1$ in Wang et al. (2011)) remains obscured by the variability of the unpolluted cloud, where LWP and N_{d_top} in itself may differ by an order of magnitude between convective cell walls and stratified regions of detrained cloud (Fig. 2).

Furthermore, while these simulations are highly idealised in their setup, they do not necessarily reflect unrealistic emission conditions. The prescribed ship is assumed to travel periodically along an identical emission line without any crosswind, which may alter the plume size or dilute emissions more effectively. Within the 48 h simulation, a total of 5 ships traverse the $180 \times 180\,\mathrm{km}^2$ domain repeatedly at a constant sailing speed of $5\,\mathrm{m\,s}^{-1}$, and the cloud response to their combined emissions is assessed. Throughout most of the North Pacific a shipping density of around 30 ships per $100\,\mathrm{km}^2$ per year is observed (MarineTraffic, 2018). Assuming a speed of $5\,\mathrm{m\,s}^{-1}$ (or even $10\,\mathrm{m\,s}^{-1}$), such a density corresponds to an estimated number of 116 (58) ships within the simulation domain on average. Within the North Atlantic, the higher density of ships could even correspond to over 200 (400) ships within a $180 \times 180\,\mathrm{km}^2$ domain (MarineTraffic, 2018). Therefore, our emission scenario is equivalent to merely $1-9\,\%$ of these ships contributing to increased CCN concentrations within the *seeded* domain.

Increases in cloud-scene albedo were attributed to changes in brightness within the stratified, detrained cloud regions covering the boundary layer between convective cell walls. These detrained cloud regions are optically thin ($\overline{\tau} = 2.8$) and are often referred to as veil clouds. They are connected to the sub-cloud layer aerosol through the convective cell walls feeding into the detrained cloud regions. In our simulations, these detrained cloud filaments contributed 82 % to the overall cloud fraction.

In summary, our results suggest that although detectable ship tracks are extremely rare in deep boundary layers, an increase in A_{cld} of the order of 0.1 may persist in deeper boundary layers of open-cell stratocumuli. Furthermore, our simulations suggest that the albedo increase within this regime, which is currently not picked up in ship track analyses, could be driven predominantly by increases in cloud fraction, as opposed to the Twomey (1991) effect.

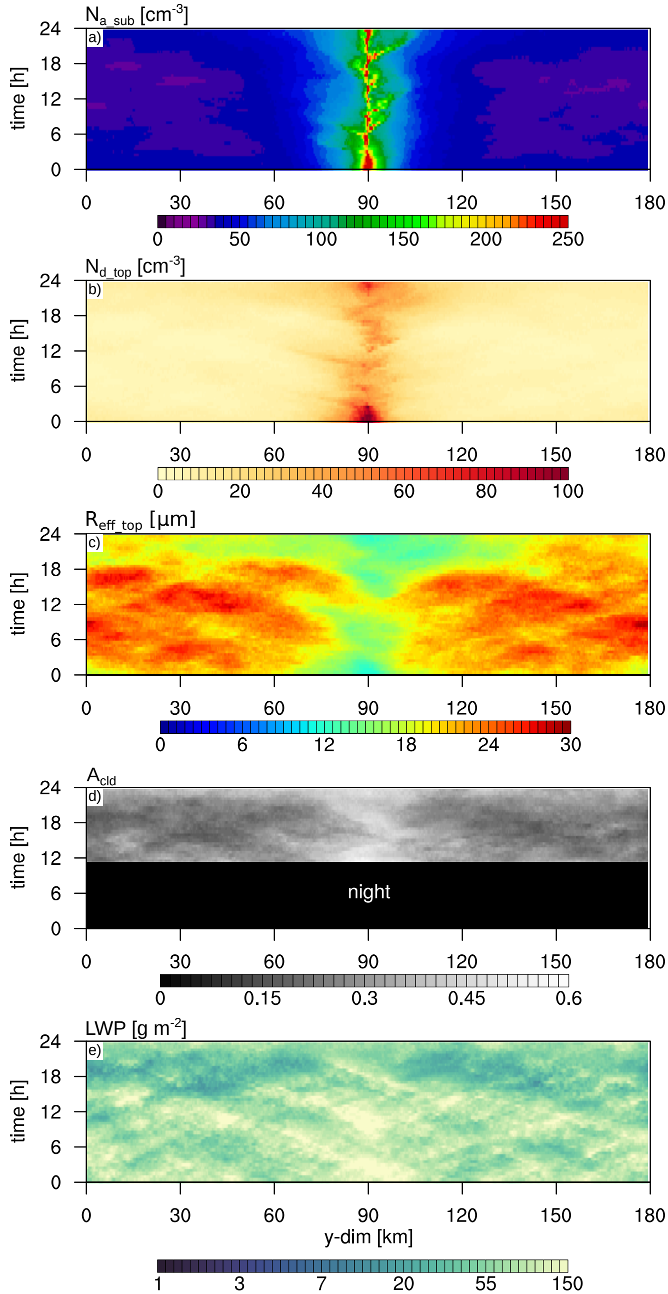

Figure 6. Hovmoeller diagrams of a) sub-cloud layer mean aerosol concentration (N_{a_sub}), b) cloud-top cloud droplet number concentration (N_{d_top}), c) cloud-top mean effective cloud droplet radius (R_{eff_top}), d) cloud albedo (A_{cld}) and e) liquid water path (LWP). Spatial averages were obtained along the emission line dimension (coinciding with x-dimension of simulation domain). Hovmoeller diagrams for the *ctrl* simulation are shown in the supporting material (Fig. S8).

While these simulations are limited in their generality, they do demonstrate that substantial changes in A_{cld} may occur in optically thin veil clouds and that the aerosol-induced changes in cloud scene albedo may prove extremely difficult to attribute without knowing the spatio-temporal distribution of the aerosol perturbation. Despite significant changes in cloud-scene albedo, an attribution of these changes to an aerosol perturbation using satellite retrievals of cloud-properties and vertically integrated aerosol metrics alone could prove to be extremely difficult in this cloud regime (Fig. 3). Yet such open-cell cloud scenes with substantial cloud-fraction and a high percentage of veil clouds, occur often (McCoy et al., 2017) and occur in regions of high solar insolation. Therefore, aerosol induced cloud-radiative perturbations within these clouds may be relevant to global estimates of aerosol-cloud-radiative forcing.

Our results strongly motivate further research into the efficacy of aerosol perturbations in deep open-cell stratocumulus. Here we demonstrate, that the aerosol forcing in this regime could be substantial. Yet for a clear assessment the occurrence rate and magnitude of A_{cld} changes in stratified detrained cloud remnants need to be known. One approach to constrain these aerosol induced perturbations could be field measurements around known aerosol perturbations. Such measurements would allow the detection and attribution of cloud-radiative effects to aerosol perturbations.

5 Conclusions

The analysis of ship tracks and changes in cloud-radiative properties within them has arguably provided an extremely useful framework to develop a mechanistic understanding of aerosol-cloud-radiative interactions, and to constrain the effective cloud-radiative forcing within marine low-level clouds. However, linear shaped tracks are extremely rare and tend to form in shallow boundary layers with a top below 800 m (Durkee et al., 2000b; Christensen and Stephens, 2012; Chen et al., 2015; Toll et al., 2017).

At least 70 % of marine stratocumuli form in deeper boundary layers, where distinct ship tracks due to ship emissions are very rarely detected. Furthermore, 73 % of all stratocumuli globally are likely to occur within the open-cell or disorganised regime (Muhlbauer et al., 2014). Here, we assessed in idealised cloud-resolving simulations, whether significant cloud-radiative perturbations persist in a field of deep (boundary layer top at 1.5 km) open-cell stratocumulus, which was observed during RF06 of the VOCALS-REx campaign. Our key-findings are summarised as follows:

1. Albedo changes equivalent to albedo increases in previously observed ship tracks within shallow open-cell stratocumuli were embedded within a stratocumulus deck of deep open cells, despite the absence of a spatially coherent structure such as a ship track. The domain-mean all-sky albedo increased by 0.05 due to a prescribed seeding source (sea salt emission moving at 5 m s^{-1}, which released particles of 300 nm in size at a rate of 10^{17} particles s^{-1}). This translates to a change in the SW CRE of 20 W m^{-2}, for an annual solar mean insolation of 404 W m^{-2} at this site.

2. Regional changes in A_{cld} (increase by 0.15), cloud microphysical (167 % increase in N_{d_top} and 14 % decrease in R_{eff_top}), and macrophysical properties (14 % absolute increase in CF and a 5 – 23 % decrease in in-cloud LWP) within the seeded domain (±30 km around the emission line) remain obscured by the naturally occurring variability

of cloud field. Reducing the variability of the clouds by averaging along the spatial extent of the aerosol perturbation permitted the detection and attribution of these cloud-radiative effects to the aerosol perturbation. Hence, knowledge of the spatio-temporal distribution of the aerosol perturbation was found to be necessary for the remote attribution of aerosol effects on cloud-radiative properties within this regime.

3. The simulated cloud brightening was attributed to the brightening of the detrained cloud filaments that spanned the regions between the convective cell walls of the open cells. These so-called veil clouds occur frequently in low-level cloud layers and are connected to sub-cloud aerosol sources through the convective cloud cores within the cell walls. Within these clouds the brightening was largely attributed to increases in cloud fraction, with a secondary contribution to brightening due to changes in cloud microphysical properties.

Competing interests. The authors are not aware of any competing interests.

Acknowledgements. We acknowledge the Fund for Innovative Climate and Energy Research grant for the financial support of this research and the high-performance computing support from Yellowstone (ark:/85065/d7wd3xhc). This support was provided by NCAR's Computational and Information Systems Laboratory, sponsored by the National Science Foundation. H. Wang acknowledges support from the U.S. Department of Energy (DOE) Office of Science, Biological and Environmental Research. The Pacific Northwest National Laboratory (PNNL) is operated for DOE by Battelle Memorial Institute under contract DE-AC05-76RLO1830.

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