Response to reviewer #3

We thank the reviewer for their helpful comments. We begin by responding to the general comments, then we address each detailed remark in turn below. Reviewer comments are in bold, and changes made to the manuscript in italics.

General comments:

The paper gives the impression that the general influence of absorbing aerosol on the cloud evolution has been investigated. However, it is only the radiative effect of the absorbing aerosol layer that enters in the discussion on the SDE, not the effect of aerosols "polluting" and thus modifying additionally the thermodynamic and hydrological cycle of the cloud and the BL. This should be highlighted in abstract and introduction, and should not only be mentioned at the end of the conclusion. The title of the paper should already include the key parameter of this study, i.e. " a persistent absorbing aerosol layer".

As suggested by the reviewer, we have highlighted the focus of our study in the title, abstract, and introduction.

The title has been changed to "Diurnal cycle of the semi-direct effect from a persistent absorbing aerosol layer over marine stratocumulus in large-eddy simulations"

Our experiment description in the abstract has been extended and now reads: "Here we use large eddy simulations to investigate the sensitivity of stratocumulus clouds to the properties of an absorbing aerosol layer located above the inversion layer, with a focus on the location, timing, and strength of the radiative heat perturbation"

At the end of the introduction, when the outline of the study is summarised, we include the following: *"In this study the UK Met Office Large Eddy Model (LEM) is used to investigate and quantify the impact that the properties of an elevated absorbing aerosol layer have on the cloud and radiative response of marine stratocumulus, with a focus on the role that the location, timing, and strength of the heat perturbation has on the underlying cloud and boundary layer."*

It is the LES model and its turbulence closure, which primarily determine the simulation results. Also the cloud description, even in simplified way, as it is in this study, affects the finding. Using other LES and cloud models (as done by other studies) will lead to a different result for the semi direct effect. This should be also highlighted in the conclusions of this paper.

We agree that past literature has shown a large amount of diversity when different LES models are used to study semi-direct effects, although we expect that the fundamental causal chain explaining the response in our model would also be involved in the response simulated by other models. The following has been included in the conclusions section:

"Inconsistent responses between LES models can also arise through differences in the representation of processes, including unresolved sub-grid scale turbulence (e.g., Stevens et al. 2005) and microphysics (van der Dussen et al., 2013). Our results show that the heat perturbation above the cloud layer impacts all aspects of the BL profile, therefore it would be beneficial to repeat this study using other LES models to test our conclusions."

123 is the radiation code applied for all 130x130 columns individually or only for one mean profiles of T, Qvap and Qliq?

The radiation scheme is applied to all columns. We have clarified this in Section 2.2 with the following text:

"Radiation calculations are performed for all grid points within the domain every 30 seconds."

212-220 makes it almost impossible to understand the calculation of the SDE. A reference would be helpful (Hill and Dobbie, 1980?). I guess DRE uses the results of the simulations with the aerosol layer, actually not explained in the paper.

As suggested, we have included a reference to Johnson et al. 2004 which provides a description of the SDE calculation.

We state how the DRE is calculated in lines 250 to 251, but have added a sentence to clarify:

"DRE is calculated as the difference between F_{TOA} and that obtained in a second, diagnostic, call to the radiation scheme with the same profiles of liquid water, water vapour, and atmospheric gases, but without aerosol. This second call is only performed for the simulations with aerosol present."

233 in caption of Fig. 3: w'w'w' is named the perturbation in mean vertical velocity. No, it is the perturbation of w to the power of 3, but it has another physical meaning. Why was it selected to illustrate the BL turbulence characteristics?

We agree that a more appropriate variable to illustrate BL turbulence is the mean variance in vertical velocity perturbation (w'w'). We have updated figure 3 and relevant text in Section 3.1, which is the only instance of use throughout the manuscript.

239-240 "During the daytime, ... through weakened surface to atmosphere gradients". The total water profiles (which are dominated by the presence Q_vap) for t = 13h and t = 5.30h in Fig.3c illustrate the contrary. Only the surface gradient in the first 10-20 m above the sea at 5.30h is stronger than the daytime conditions. Also for THETA_liq in Fig. 3b a weak gradient exists during daytime but none at 5.30h. This explanation of the decoupled state of the BL during daytime is not really convincing. Fig.3d and e better indicate the daytime / nighttime differences in the BL which are controlled by the vertical turbulent transport of tke and thus by the turbulence simulated in the LES model.

We agree with the reviewer that our explanation did not provide a convincing description of the day / night differences in BL turbulence. As suggested, we have used figures 3d and 3e to illustrate the different turbulent structures and the decoupling during daytime. The sentence now reads:

"During the daytime, solar heating reduces the buoyancy flux (Fig. 3d) through an offset in the longwave cooling and reduces turbulence throughout the BL (Fig. 3e). This weakens the BL circulation and prevents mixing throughout the BL and promotes a decoupled state in which the flux of moisture from the surface to the cloud is insufficient to maintain the cloud base height, as evident from the non–constant BL profiles of θ_l (Fig. 3b) and q_t (Fig. 3c) at 1300 hours."

241 SST is kept constant, how are surface heat and moisture fluxes calculated? Give the key parameters.

The surface fluxes are calculated using Monin–Obukhov similarity theory, which is reported in section 2.1 (Description of model; line 117). As suggested by the reviewer, we have expanded on this to provide more information. The sentence now reads:

"Surface fluxes of moisture and heat are calculated using Monin–Obukhov similarity theory (Monin and Obukhov, 1954) which predicts the surface frictional stresses and heat fluxes using the local gradients between the surface and the overlying model level. For these experiments a prescribed constant sea surface temperature is used."

274 Terminology in the caption of Fig.4: instead of "response" a more explicit description like "differences between no-aerosol simulation and simulation with an elevated aerosol layer" would make the illustration DCloud, DLWP and DAlbedo and the reference to equation (1) more understandable.

The figure caption has been amended as suggested.

298 ... allows the cloud layer to maintain a higher RH. This is difficult to understand and to believe, as explained in 2.2 the cloud model excess supersaturation is converted in liquid water.

The reviewer is correct. We erroneously referred to the cloud layer, rather than the sub-cloud layer. This has been corrected to read:

"The increase in RH occurs due to the weakened w_e which reduces the amount of warm dry FT air that is mixed into the BL and allows the sub–cloud layer to maintain a higher RH."

301 -302 ... enhanced RH below cloud (caused by an increase in water vapour) ... by the decrease in latent heat flux. This is not credible. Q_total, i.e. mainly Q_vap, continuously decreases; also LHF mainly decreases but RH increases. RH is a function of two independent state variables T and Q_vap. What happened to the temperature T during the "aerosol" simulations in the BL.

The RH below cloud increases (Fig. 5f) due to the increase in qt (total water specific humidity) below cloud, which occurs despite a decrease in the total water path (Fig. 5f). We believe we introduced some confusion with the variable name 'total BL qt' which, as pointed out by reviewer #2, should be correctly termed total water path.

We chose not to show the qt below the cloud in these plots, and instead use the decrease in LHF from the surface and increase in mean RH below cloud to demonstrate this response,

which occurs alongside a small decrease in BL liquid-water potential temperature of ~0.1 K (discussed in more detail below). The increase in mean qt can be seen further on in the manuscript in Fig. 10e.

We have addressed this by replacing 'total BL qt' with total water path (TWP) throughout the manuscript, and as discussed below we have introduced the mean BL liquid water potential temperature into Figures 5 and 6 to support our analysis.

The paper completely omits a discussion on changes of the T profile in the BL after section 3.1. Temperature perturbations above the BL are the key parameter for the different SDE scenario in this study but a discussion of subsequent temperature changes in the BL is completely ignored - why?

In our simulations the temperature only plays a minor role in the BL response, yet as correctly pointed out by the reviewer, we do not discuss this at any point. To address this, we now include the BL liquid-water potential temperature (θ_l) response in Figures 5 and 6, which are shown below as Fig. R1 and R2, respectively.

The figures show that the mean θ_l decreases by ~0.1 K over the initial three days of the simulation (Fig. R1) and ~0.2 K after 10 days (Fig. R2). The sources for changes to the θ_l occur through exchanges across the inversion layer at the top of the BL, changes in LW and SW fluxes, surface fluxes, and precipitation. In our simulations the enhanced inversion strength reduces the flux of warm air into the BL, however, this may be partially offset by the heat perturbation produced by the absorbing aerosol layer. Small decreases in precipitation are offset by reduced latent heat flux at the surface, and due to the high cloud fraction in the simulations, changes to LW and SW fluxes within the BL are small. The pattern of the θ_l response in Fig. R1 is similar to the entrainment rate (Fig. 5b) which makes it likely that the process controlling the simulated response in θ_l is the change to the entrainment rate.

The magnitude of the response in both the initial and the steady-state response is small (up to 0.2 K in the steady-state) which demonstrates that the dominant driver of changes to RH below cloud is from the water vapour, rather than the temperature.

In addition to the new subplots in figures 5 and 6, we have included text in section 3.2.1:

"The increase in RH occurs due to the weakened w_e which reduces the amount of warm dry FT air that is mixed into the BL and allows the sub–cloud layer to maintain a higher RH. The relatively small decrease in potential temperature of ~ 0.1 K (Fig. 5g) suggests that the RH response is driven by an increase in water vapour."

in section 3.2.2:

"The small response in mean BL potential temperature of -0.2 K (Fig. 6f) strengthens the hypothesis that the RH response below-cloud is driven by changes in available water vapour, rather than the decrease in temperature, although it is worth noting that this decrease in temperature will act to slightly increase the RH."

in section 4:

"The reduction in the entrainment of warm and dry air from the FT reduces the amount of mixing, reducing the sink of \bar{q}_t in the cloud layer and allowing the BL to maintain a greater RH. The result is an increase in \bar{q}_t , a small decrease in BL temperature, and an increase in RH."



Figure R1. 'Initial' domain averaged cloud response of BL liquid-water potential temperature – subplot taken from Figure 5 in the revised manuscript.



Figure R2. Domain averaged 'steady-state' cloud response of BL liquid-water potential temperature – subplot taken from Figure 6 in the revised manuscript.

308 where can we see thicker clouds in Fig.5a in the afternoon of day 3? I can't.

The paragraph in question refers to the increase in LWP of ~ 2 g kg⁻¹ that is evident in Fig. 5c from just before midday and continuing through the afternoon on day 3. There is also a corresponding increase in the geometric thickness of the cloud at midday evident in Fig. 5a which shows that the cloud base has decreased more than the cloud top.

We have clarified the paragraph by starting the paragraph with the following:

"The thicker cloud (enhanced LWP; Fig. 5a) on the afternoon of the third day..."

A similar change has been made to the previous paragraph which discusses the thinner cloud on the morning of the third day.

335-336 "The decrease in cloud layer height allows better mixing beneath the cloud base, which enhances the evaporation of moisture from the surface between 0900 and 1500". This is in any case a speculation. It is not coherent with the daytime turbulence profile of Fig. 3e for the non-aerosol simulation.

A given eddy generated in the cloud layer will be able to penetrate a certain distance below the cloud. If the cloud layer is reduced in altitude then the eddy will be able to influence layers closer to the surface. We see evidence of this occurring in our simulations. Figure R3a shows that although the maximum turbulence in the cloud layer is slightly weaker in the aerosol simulation, the decrease in cloud altitude by ~ 200 m systematically shifts the w'w' profile downwards, so that turbulence below cloud is greater in the simulations with aerosol. The buoyancy flux profile (Fig. R3b) shows that this results in positive buoyancy flux below cloud, as opposed to a negative value in the no-aerosol simulation, resulting in the response that we observe in Figure 6.

Figure R3 has been included in the supplementary information (as Figure S2) and is now referred to in the manuscript in section 3.2.2:

"This modification to the diurnal cycle of the cloud is driven by an increased coupling between surface moisture flux and cloud base during the daytime (see Fig. S2 in the supplementary information) ..."



Figure R3. Domain-mean vertical profiles of a) variance in vertical velocity perturbation w'w', and b) buoyancy flux on day 13 of the simulation at 1300 local time for the no-aerosol simulation (black dashed line) and following the introduction of a layer of absorbing aerosol (blue solid line) in the base experiment (0 m cloud–aerosol gap, 250 m thick layer, and AOD of 0.2).

346 rephrase "reduction in evaporation and associated cooling of entrained air". What do you mean with cooling of entrained air?

The air that is entrained into the cloud layer from the free troposphere is dry. The subsequent mixing of the dry and cloudy air results in net evaporation which acts to cool the parcel of air, generating (negative) buoyancy.

As suggested, we have rephrased the sentence, which now reads:

"The weakened BL circulation is therefore due to a reduction in entrainment. The mixing of dry air into the cloud layer results in evaporation and a cooling which generates buoyancy; a reduction in entrainment therefore weakens cloud–top buoyancy production."

347 the reduced vertical motions reduce surface evaporation. Same question as above for 241.

As per the previous question the text concerning the surface fluxes in section 2.1 has been expanded to provide more information.

394 Fig. caption 7: same remark as in 274.

The caption has been changed as suggested.

416 ... quicker than the cloud base. Where or how to detect?

The current manuscript does not provide evidence of this, as also highlighted by Reviewer #2. We have therefore included additional plots in the supplementary information and have rewritten the first paragraph of section 3.3.1 to discuss this in more detail. The paragraph reads:

"The majority of experiments show a positive spike in SDE (Fig. 7d, i and n) just before midday on the first day. This occurs due to the lag–time in response between the direct impact to the cloud from changes to we, and the increase in sub–cloud RH. Figure S1 in the supplementary information focuses on the response in the initial 24 hours. The positive SDE is driven by the decrease in LWP caused by an increase in cloud base height (Fig. 5a and Fig. S1a) without a corresponding change in cloud top height. The decrease in w_e weakens buoyancy production throughout the cloud layer (Fig. S1c), which drives a reduced moisture flux within the cloud and to the cloud base (Fig. S1d). As the day progresses the continued reduction of w_e results in an increase in mean below–cloud RH and a recovery, or increase, of the LWP. This explains why stronger perturbations to the entrainment rate on the first day (such as when the layer is close to the cloud) results in a quicker recovery of the LWP (Fig. 7c, h, and m)."

520 the norain simulation is not a model setup modification, but a change in the modeling physics.

Granted, although for the sake of simplicity we refer to those sensitivity experiments as experimenting with "model setup".

522 the strengthening of the SDE is +1 W/m2 (or +3) from -7 to -8 W/m2 (or -9 to -12) why not -1 W/m2 (and -3)?

We agree that the current text is confusing. We have rewritten the sentence to read:

"Compared to the control setup the noRain setup is characterised by a consistent increase in the magnitude of the SDE by 1 Wm^{-2} when a cloud–aerosol gap is present and up to 3 Wm^{-2} when there is no gap"

527 – 535 This discussion or interpretation of the results cannot be understood with the given information on the 05cool simulations. Thus, this part should be omitted.

As discussed in Section 3.4 the large-scale advective heat tendency is a large-scale forcing term that represents a degree of variability in LES experiments. We therefore do not want to omit this result, and have instead rewritten the paragraph to improve our interpretation of the results. The paragraph now reads:

"When compared to the control setup, increasing the cooling rate of the large–scale advective heat tendency results in stronger BL dynamics, enhanced cloud–top entrainment

of warm dry air, and enhanced surface LHF (which acts as a feedback to enhanced entrainment). As the processes maintaining the cloud layer become more important, they become more sensitive to perturbations. Therefore, when the aerosol layer is present in the 05cool setup, the responses of w_e , LHF, and below–cloud moisture flux are stronger than in the control setup and the simulations are characterised by a quicker decrease in the TWP of the BL. However, this only becomes prominent on the third day and results in little difference from the control setup over the first two days."

537 and 547 Are the SST and wetFT simulations really with no-aerosol? This is probably a typo.

This is not typo. In each set of sensitivity experiments a simulation was run with and without aerosols. This allows us to investigate the response of each setup to the same aerosol perturbation. For each setup we briefly describe what is changing in the 'without aerosol' simulations before discussing the response to the aerosol layers.

We have made this clearer in the revised manuscript by adding a sentence in Sect. 3.4 pointing out that a no–aerosol simulation is run for each setup and referring to the no–aerosol simulations within our interpretation.

549 allowing the BL to maintain below the cloud layer a greater RH?

This has been changed to:

"...allowing the BL to maintain a greater mean RH"

564-565 ... a redistribution of water from the cloud layer to the surface layer (Fig.9b). What do you mean with redistribution? Does it mean that rain is responsible for the significant Qvap increase in the first 300 m? This is most unlikely. Water vapor is accumulated in the lowest levels due to surface evaporation in the decoupled BL.

Yes, the reduced coupling between surface and cloud layers results in an accumulation of water vapor towards the surface, which can be viewed as a redistribution of the water. This does not occur due to precipitation processes.

The sentence has been amended to clarify our point:

"This reduces the flux of water vapour from the surface layer to the cloud, resulting in an accumulation of water vapour close to the surface (Fig. 9b)."

630 3b. The conclusion that RH increase as Q_total increase, implicates that the BL temperature remains constant or decreases. This study, however, withholds this fundamental information.

As per our previous discussion on the temperature response, this line has been amended to read:

"The reduction in the entrainment of warm and dry air from the FT reduces the amount of mixing, reducing the sink of $\overline{q_t}$ in the cloud layer and allowing the BL to maintain a greater

RH. The result is an increase in $\overline{q_t}$, a small decrease in BL temperature, and an increase in RH."

721 regional climate models as HadGEM certainly treat surface fluxes

We have amended the sentence as follows:

"The lack of BL adjustment may be due to processes that are not explicitly treated in HadGEM, such as BL turbulence and subsequent missing feedbacks on surface fluxes..."

References

Stevens, B., and Coauthors, 2005: Evaluation of large-eddy simulations via observations of nocturnal marine stratocumulus. Mon. Wea. Rev., 133, 1443–1462, https://doi.org/10.1175/MWR2930.1.

van der Dussen, J. J., and Coauthors, 2013: The GASS/EUCLIPSE model intercomparison of the stratocumulus transition as observed during ASTEX: LES results, J. Adv. Model. Earth Syst., 5, 483–499, doi:10.1002/jame.20033.