Response to Anonymous Referee #1

We would like to thank the referee for their pertinent and thought-provoking comments, and also for their valuable suggestions towards improving this work.

Below, we quote the referee's specific comments (in bold), and follow them with responses.

To aid transparency, we also refer to specific manuscript changes that we propose in response to the comments (as a page number/line number). This amended manuscript is uploaded as part of the Interactive Discussion.

1) Aerosol lifetime is longer than one day (typically it is 2+ days) in the MBL. Therefore, many of the particles injected at a particular time will still be present much later. So this renders testing the seeding time somewhat moot because real clouds would experience the high aerosol concentrations whatever the time of the seeding.

We do not believe that the seeding time tests are invalid, although a high aerosol concentration will still be present in the boundary layer much later than is covered in these simulations, the overall albedo perturbation will be dependent both on the effect that the aerosols have on the clouds and cloud system, and on the vertical and horizontal distribution of the aerosols over time. Both of these show sensitivity to the aerosol injection time, the results of which can be seen in the short-term cloud response simulated here.

For example, our results show that aerosols injected into the early morning are activated as cloud droplets and produce rapid changes in the cloud. These changes are also indicative of a change from WP to NP regime. This would result in the cloud being more likely to persist through the day, when the SW radiation can be reflected back to space with associated cooling effect. Thus, injection into the early morning produces immediate and sustained albedo increase. However, aerosols that are injected only 6 hours later (into the day, 13:00 LT) have no initial cloud albedo response. Whilst the high concentration of aerosols may then be taken into the cloud during the subsequent night, thus altering the cloud properties, it would be early the next morning before these perturbed clouds could begin to reflect the SW radiation necessary to produce the desired cooling effect. In this case, this would produce a delay in cloud effects of 16 hours, thus diminishing the fraction of the aerosol lifetime over which the perturbed cloud could be effective in producing a cooling effect.

Changes to the cloud also affect the cloud system behaviour (e.g. vertical velocity variance, TKE and skewness; Figure 6 in the amended manuscript) which can in turn alter the extent of aerosol distribution from the high concentrations emitted at the point source. Wang et al. (2011) show that the distribution of aerosols affects the strength of albedo increase, but that this relationship is dependent on conditions. They show that for their weakly precipitating cloud case, the uniform distribution of added aerosols produces a larger albedo increase than when the aerosols are emitted as a point source. However, the opposite is true for their more heavily precipitating case.

Modelling, at increased time scales, increased domain sizes and covering a wider range of the possible MSc cloud properties (e.g. cloud top height, jumps of temperature and moisture across the inversion, and large-scale subsidence strengths) would be necessary to further explore these sensitivities. However, modelling the whole aerosol lifetime – shown in the global modelling of Jones and Haywood (2012) to be up to 4.8 days – at the resolutions needed to simulate the eddies typical of MSc clouds would be challenging both in terms of model configuration and forcing, and computational expense. For example, these simulations would need to incorporate forcings demonstrating sufficient balance to maintain a recovering cloud system over several diurnal cycles to prevent the drift that is commonly seen in these styles of simulations (see also Comment 1 continued). Despite resolutions needing to be maintained below hundreds of metres for the resolution of smaller eddies, the domain size would need to be extended sufficiently not only to encompass mesoscale circulations (of the order of tens to hundreds of kilometres), but also to encompass the horizontal dispersal of aerosols that would occur over their lifetime. This would require substantial computational commitment.

We add discussions of the limitations of our simulations and the extent of their usefulness in the altered manuscript (p.17/L.10 to p.18/L.2).

1 continued) Also, because the clouds are thinning over the course of the simulation, I am somewhat concerned that the results may not really be telling us much about the physical system but will mainly reflect the fact that there is less cloud to seed as time goes on. The authors should test this by comparing the results of seeding at 03hr on day 2 with seeding at 03hr on day 1. In other words, to comprehensively test their hypothesis, the authors would need to construct clouds that recover on the second night. I would suggest that the authors at least discuss this issue.

We agree that there is thinning of the cloud, but disagree that conclusions cannot be drawn into the pattern of albedo changes with respect to the physical system. The four aerosol injection times used were selected in order to exemplify four different characteristic states of the cloud and cloud system through the diurnal cycle. i.e. during the peak cloud amount with strong vertical mixing (early morning); during increasing uncoupling and decreasing cloud amount of the mid-morning; during the stagnant day; and as the cloud system becomes more turbulent into the evening as the cloud rebuilds. While the cloud properties exhibit drift over the diurnal cycle, these patterns of characteristic cloud physics are evident in both the WP and NP simulations (evidenced in properties shown in Figures 3 and 5).

In considering these four cloud and cloud system states over the diurnal cycle, we use the magnitude of albedo change as an indicator of the pattern of cloud response to the injected aerosol, but relate this pattern of changes to the characteristic state of the cloud and cloud physics at the time of aerosol injection. For example, for aerosol injection into the WP early morning, the presence of precipitation results in comparative LWP gains and albedo increases. For injection into the WP daytime, the cloud has dissipated, as has precipitation, and no albedo increase results. Despite the magnitude of cloud dissipation being sensitive to the drift, the pattern of uncoupling and decrease of turbulence during the day would remain, as would the reduction in cloud albedo increase. For injection into the evening, there is cloud thickening associated with the increasing turbulence, although it is not yet sufficient to produce precipitation. Whilst the drift may alter the strength and timing of this cloud thickening, the general pattern will again remain, with the opportunity for albedo increase in the evening being smaller than it was for the early morning as precipitation is not as well established.

Thus, as suggested, the magnitude of albedo increase for seeding at 03hr on day 2 would likely differ from the magnitude of albedo increase for seeding at 03hr on day 1. However, the conclusion in terms of relation to the physical system would likely remain, with the precipitation rate recovering sufficiently to produce significant LWP increases upon suppression by the injected aerosol.

The construction of a system that supports total cloud recovery into the second night is a challenge, with the drift being caused by imbalances in forcings. Owing to the complexity of the MSc system, these imbalances tend to be common in models aiming to simulate the MSc diurnal cycle (e.g. Wang et al. (2011), Chen et al. (2011)). The development of such a balanced MSc system would be of interest for future work.

This framework of analysis, based on consideration of the physical and dynamical cloud state at the time of injection (rather than cloud properties alone) is outlined in the amended manuscript on p.10/L.27. The challenges for future modelling are presented on p.19/L.15.

See also response to Referee #2 Comment 3.

2) In the simulations practically all the simulated clouds essentially disappear in the middle of the day. Real subtropical stratocumulus clouds do not do this, but instead one sees a drop of perhaps 20-30% during the day, from close to zero at night (e.g. paper by Rozendaal et al. 1994, JCLI). Can the authors put their results in the context of real clouds?

In this paper, we aim to demonstrate the response to aerosol injection for a range of clouds. As such, the WP cloud with total cloud dissipation is intended to represent a more extreme cloud case. Such cloud behaviour, whilst not being typical, has been observed previously (Albrecht et al., 1988; Minnis et al., 1992).

The disparity between this extreme case and the more typically reported cloud behaviour may also be a consequence of the scale of observations. The Albrecht et al. (1988) and Minnis et al. (1992) observations of real cloud total dissipation were made at a single location, whilst the ISCCP data used by Rozendaal et al. (1995) uses a 4 to 7 km pixel size averaged over a 280 km grid resolution. Stubenrauch et al. (2006) similarly do not observe total cloud dissipation, but the diurnal cycle data is produced over larger regions (i.e. five latitude bands), over seasonal time periods.

Previous simulations of the MSc diurnal cycle using similar model configurations to that used here also suggest larger cloud fraction deviation than typically observed. For example, Wang et al. (2011) simulations include decreases by up to 75% in their W100 case, and decreases of 60% in their W50 case, with Chen et al. (2011) also simulating decreases in cloud fraction of up to 100% for their 100 cm^{-3} background aerosol concentration case.

We clarify the atypical nature of the WP cloud fraction diurnal cycle in the amended manuscript (p.8/L.25 to p.8/L.30), and state that there is a wide range of cloud behaviour expected following the large range in possible cloud states (p.16/L.32).

See also response to Referee #2, Comment 3.

3) The domain is very small given the low resolution. 30x30 points at 300 m resolution for stratocumulus is not really cloud resolving because stratocumulus large eddies are typically 100-1000 m in size. The simulations should be termed "convection permitting". The authors should perform a sensitivity test to demonstrate that their results are robust. Otherwise, it's hard to believe their findings. What is the resolution near the MBL inversion? This needs to be better than 10 m and preferably 5 m to even begin to resolve entrainment

We accept that the small size of the domain prohibits the simulation of mesoscale cellular cloud patterns that are characteristic of MSc clouds at scales of tens of kilometres. The restriction of the domain size was necessary owing to computational constraints. In spite of the domain size restrictions, our simulations represent the physics and dynamics of the cloud system adequately for turbulence and the distribution of updrafts and downdrafts to be formed at scales of hundreds of meters. Thus cloud and cloud system properties are simulated that include the radiative effects and subsequent patterns of dynamics and cloud properties that are driven by the cloud-top radiative cooling and are characteristic of MSc clouds over the diurnal cycle. Importantly, entrainment (seen as cloud top height perturbations, given the constant large-scale divergence) also demonstrate expected behaviour. Entrainment decreases into the daytime as SW offsets the LW radiative cloud top cooling and the cloud-top production of turbulent kinetic energy (TKE) decreases. Entrainment increases in response to the increase in N_d associated with aerosol injection into the less polluted WP cloud as the smaller cloud droplets result in increased TKE production.

Sensitivity testing of the model resolution and domain size (carried out using an alternate set of input sounding) indicates that an improved LWP recovery into the second night was achieved for a higher horizontal resolution (100 m compared to 300 m), as shown in Figure C3A. Given the computational restrictions however, increasing the resolution would decrease the domain size and owing to the already limited horizontal domain size (and hence post-injection analysis time prior to the aerosols reaching the simulation boundaries), a concession was made in using the poorer resolution.

Increasing both the horizontal resolution from 300 m to 100 m and the vertical resolution from ~30 m to ~15 m resulted in a poorer recovery of LWP and cloud fraction (Figure C3B) which suggests that the input soundings, large-scale atmospheric features (i.e. subsidence) and radiative response of the cloud in this case did not produce an improved cloud representation, as may be expected. Increasing the vertical resolution similarly did not produce significant improvements in cloud property response in the sensitivity study carried out by Chen et al. (2011) when comparing 20 m, 10 m, and 5 m vertical resolutions. Thus, while the ~30m vertical resolution used here is greater than the 5-10 m vertical resolution suggested by Bretherton et al. (1999) and Stevens and Bretherton (1999), the response of the cloud and cloud system demonstrates characteristic behaviour typical for the cloud. It is also noted that these two investigations used horizontal resolutions of 50 m, which may also affect the sensitivity of vertical resolution.

Doubling the domain size led to a larger number of clouds being simulated (Figure C3C), although there was no change to the diurnal cycle of LWP (Figure C3D). As the domain size was extended to only 18 km (again owing to computational constraints), the production of mesoscale features was again unattainable. The similarity of cloud properties between the two domain sizes was therefore expected given that there was no change in system behaviour.

These sensitivity experiments demonstrate that, within our constraints, the physical characteristics of the simulations are robust. A measure of the robustness of the aerosol injection results must however incorporate broader features, i.e. the processes associated with domain sizes large enough to capture the meso-scale cellular features and feedbacks; a larger sample of cloud system properties (i.e. varying the cloud top height, jumps of temperature and moisture across the inversion, and large-scale subsidence strengths); and a timescale sufficient to incorporate the full life-cycle of the injected aerosols. The challenges involved with this extended modelling are further discussed in the response to Comment 1. As the configuration of our model currently prohibits consideration of these features, we present our findings as an introduction to the sensitivity of albedo increase to aerosol timing, but acknowledge that more extensive modelling work must be undertaken to gain a fuller understanding of the processes involved.

The reasons behind the simulation setup; constraints; usefulness; and future challenges are now stated clearly in the amended manuscript on p.6/L.5 to p.6/L.17 and p.19/L.15. In order to avoid potential confusion regarding the cloud-top driven mechanics of the MSc clouds, we omit inclusion of the 'convection permitting' term, but feel that the connotations of this phrase are captured in the explicit statement of inability to capture the larger cellular/mesoscale patterns of the MSc clouds.

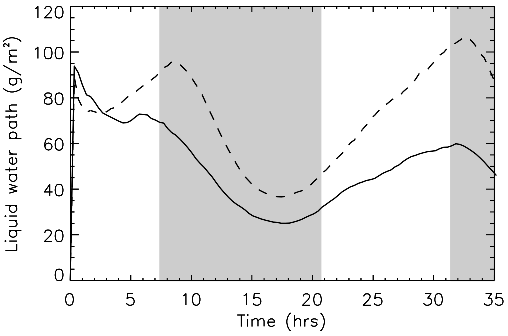

Figure 3CA – Liquid water path 30×30 horizontal cells at 300m horizontal resolution (black line) and for 30×30 horizontal cells at 100m horizontal resolution (dashed line). Shading indicates daytime.

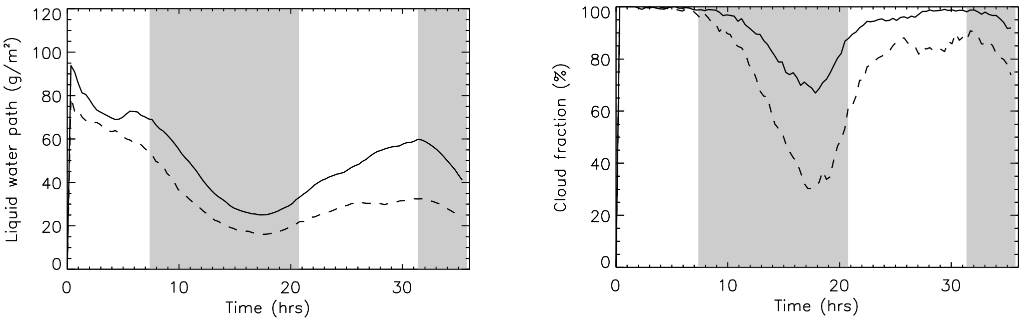

Figure 3CB – (left) liquid water path and (right) cloud fraction for 30x30 horizontal cells at 300 m horizontal resolution and ~30m vertical resolution (solid line); and for 30x30 horizontal cells at 100 m horizontal resolution and ~15 m vertical resolution (dashed line). Shading indicates daytime.

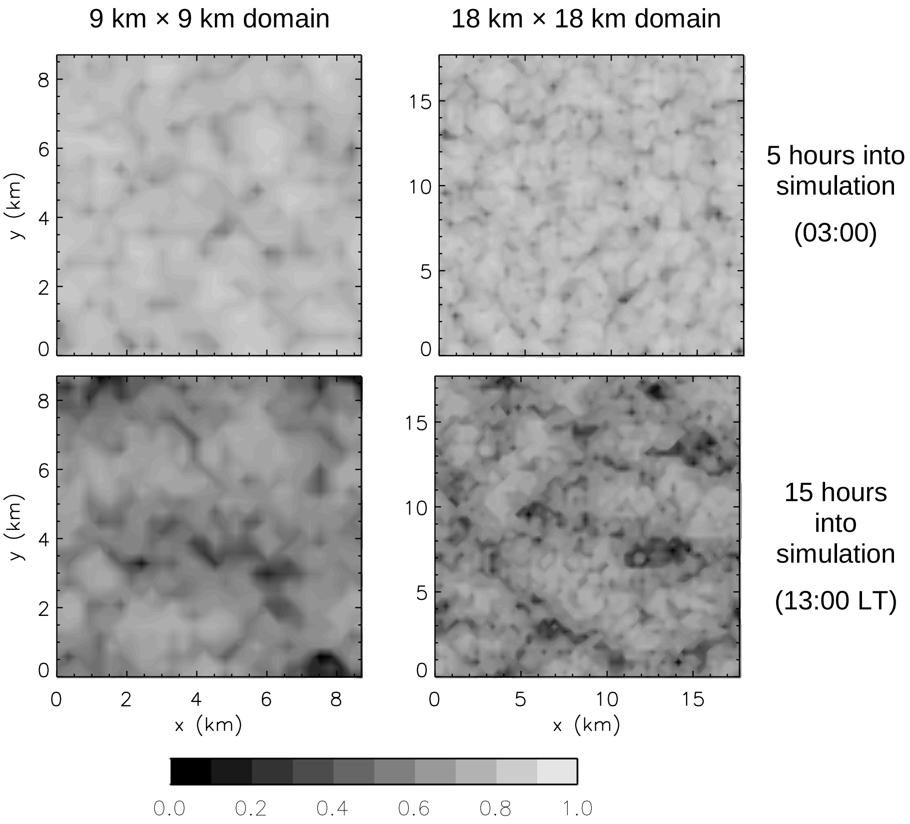

Figure C3C – Cloud albedo snapshots for 9×9 km and 18×18 km simulations

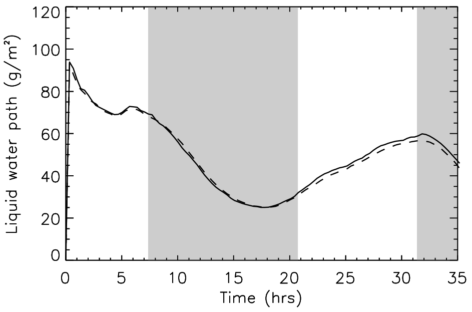

Figure C3D – Liquid water path timeseries for 9×9 km (solid line) and 18×18 km (dashed line) simulations. Shading indicates daytime.

See also response to Referee #2 Comment 3.

4) Is cloud droplet sedimentation included? If so, please say so. If not, then a major feedback that could be working here for the non-precipitating cases is being omitted. See Ackerman et al. (2009) and Bretherton et al. (2007) for details.

Cloud droplet sedimentation is included in the Morrison 2-moment microphysics scheme used. Cloud droplet sedimentation is an important point, the discussion of which is currently omitted. Clarification is added to the amended manuscript (p.4/L.21). The importance of sedimentation is also discussed in Comment V.

Other points:

I) If the wind is zero, then the surface fluxes are zero and there is no moisture transport into the cloud. This doesn't make much sense given that these cloud layers practically always have surface winds 5-10 m/s. What are the surface fluxes of LHF and SHF?

Time series of the domain average surface fluxes of latent and sensible heat are shown in Figure C4A. While our wind is initialised at zero (following Wang and Feingold, 2009a; Wang et al., 2011), our non-zero surface heat fluxes arise from maximum wind speeds of 2 m s^{-1} (Figure C4A) produced by the convergence of downdrafts. As suggested however, these are below the 5-10 m s^{-1} surface wind speeds typical of MSc regions (Fan et al., 2012). Both surface latent and sensible heat fluxes show some diurnal cycle. Daytime latent heat fluxes are around 6 W m^{-2} for the WP case, and 3 W m^{-2} for the NP cases increasing to maxima of around 11 W m^{-2} in both cases by the end of the simulation. The daytime sensible heat flux for the WP case is just over 1 W m^{-2}, increasing to around 4 W m^{-2} by the simulation end. The sensible heat flux in the NP case remains at around 0 W m^{-2}. For comparison, the surface heat fluxes observed in the nocturnal DYCOMS-II RF02 field study were 93 W m^{-2} (latent) and 16 W m^{-2} (sensible), although because of differences in the simulation initialisations, we would not expect to reproduce this case absolutely.

In spite of the lower than typical surface heat fluxes, our simulations include diurnal patterns of cloud properties and system dynamics that are characteristic of MSc. Importantly, these simulations demonstrate uncoupling of the cloud during the day, associated with a lack of moisture transport to the cloud and a less well-mixed vertical moisture profile (Jones et al., 2011), as seen in Figures 3g and 5g. The simulations also show apparent re-coupling during the second night, with the daytime accumulation of total water mixing ratio near to the surface dissipating and the boundary layer again becoming well-mixed (Figures 3g and 5g).

Whilst, as suggested, surface fluxes are an important component in representing the MSc processes, the physical cloud system is sensitive to a range of variables. For example, Chen et al. (2011) investigated the sensitivity of MSc using a similar model configuration to that used here, and found that for their experimental ranges, cloud properties were less sensitive to a change in wind speed than to changes in sea-surface temperature, free-tropospheric moisture, and the strength of large-scale subsidence.

The prescription of surface fluxes was considered, however, in order to simulate the diurnal cloud cycle, these values too would need to be diurnally varying. Making the necessary assumptions regarding the magnitude and diurnal pattern of surface heat fluxes associated with this particular combination of variables would likely lead to further imbalances in the system. It was therefore decided to allow the model to calculate the surface fluxes.

We amend the manuscript by stating the magnitude of the surface fluxes, and discussing the implications for our simulations (p.8/L.6, p.9/L.11 and p.15./L.21 onwards).

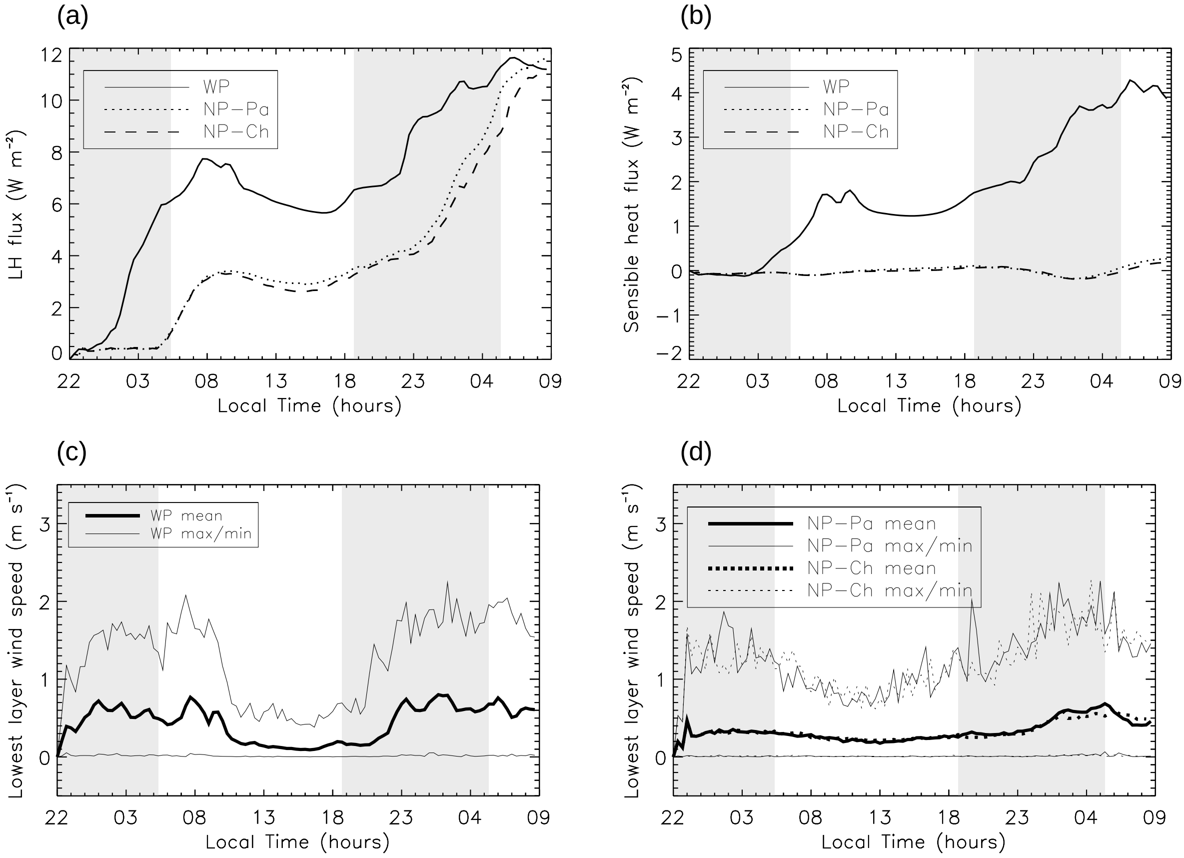

Figure C4A – (a) Surface latent heat flux (W m^{-2}); (b) Surface sensible heat flux (W m^{-2}); and (c) wind speed in the lowest model layer for the WP control case and (d) wind speed in the lowest model layer for the NP control cases

II) P24211, line 1: Fluxes have units of /m2/s. So is this a flux or an increased concentration? Is this consistent with what one would expect from DMS?

This is an increase in SO_2 concentration, and not (as was incorrectly stated in our manuscript) a flux. The increased concentration of 1 pptv h^{-1} is included as a representation of DMS production based on the following assumptions:

- From observations (Yang et al., 2011, p.5087) the production of SO_2 from DMS is estimated at approximately 2 pptv h^{-1}.
- From observations and chemical modelling (Gray et al., 2011), the loss of boundary layer SO_2 associated with dry deposition (which is not simulated in our model) is approximately half of the production rate.
- Therefore, we add 1 pptv h^{-1} in order to represent the DMS processes.

We accept this incorrect usage of terminology, and correct it in the amended manuscript (p.5/L.24).

III) The addition of a table providing details of the unseeded aerosol state in these cases would be extremely helpful. Aerosol number concentrations, CCN concentrations and sizes should be included. Without this it is impossible for a reader to put the seeding in any kind of context.

The unseeded aerosol state conditions are clarified in Tables 2 and 3 (p.31 and p.32) by expressing the absolute values of the unperturbed and perturbed states explicitly (replacing the percentage-change terminology previously used).

While investigation of the sensitivity of albedo response to the relative background and injected aerosol sizes (as considered by Bower et al. (2006) and Partanen et al. (2012)) are outside the scope of this current investigation, as suggested, we add further detail of the unseeded aerosol state through the addition of a new figure (Figure 1 on p.33 and added text on p.5/L.18). This figure elucidates the 8-bin size distribution of the background aerosols on initialisation of the simulation. We also add a discussion outlining this interesting area with a suggestion for future work (p.17/L.1 onwards).

IV) Do any secondary circulations form, or does the aerosol become well-mixed within the domain after some time?

No secondary circulations form and the aerosols tend towards a well-mixed state. The domain size of the simulations prevents circulations at the scales necessary for the secondary feedbacks that are seen in e.g. Wang and Feingold (2009a), Wang and Feingold (2009b), and Wang et al. (2011). The implications of looking at MSc at these smaller domain sizes are discussed in depth in Comment 3. In response the query, and to aid reader clarity, we emphasise the lack of secondary circulations in text added to the amended manuscript (p.6/L.9).

See also response to Referee #2 Comment 2.

V) Can the authors explain the LWP changes? It is not obvious that there SHOULD be increases (see e.g. Ackerman et al. 2004, Wood 2007), so the authors need to do more to provide understanding.

The net change in LWP results from a balance between two processes (Ackerman et al., 2004). The addition of aerosols decreases the cloud droplet size. This reduces coalescence. In the case of a cloud that loses LWP as surface precipitation, reduced coalescence may suppress this surface precipitation sufficiently to produce a comparative gain in LWP. The amount of LWP comparatively gained would be dependent on the amount of precipitation reaching the surface, and hence on several variables including N_d, boundary layer moisture content and temperature, and cloud base height. In making the cloud droplets smaller, cloud top radiative cooling increases, as does turbulence. This increases the entrainment of the drier, warmer overlying free tropospheric air. Therefore, droplet evaporation increases, and LWP is comparatively lost. The amount of relative cloud drying will again be dependent on several factors, including the change in N_d and the free tropospheric moisture content. The net change in LWP is therefore dependent on these two processes, with the magnitude and sign of change being dependent on conditions.

Whilst the inclusion of cloud droplet sedimentation (Comment 4) removes droplets from the cloud-top, thus reducing entrainment of the overlying air and increasing the LWP (Bretherton et al., 2007), sedimentation is reduced for smaller droplets, allowing them to stay in the entrainment zone for longer, and hence increase entrainment drying.

In addition to the investigations of Ackerman et al. (2004), Wood (2007) uses a mixed-layer model to find that cloud base height is an important parameter in deciding the sign of LWP change. As such, they find a threshold cloud base height of 400 m, above which LWP losses are more significant than LWP gains, resulting in cloud thinning. For cloud base heights below 400 m, the LWP gains from precipitation suppression outweigh LWP losses from increased cloud top evaporation and the cloud thickens. The results for our LWP gains in the WP case (with cloud base below 400 m) follow this threshold, leading to increases in LWP. In our non-precipitating cases (with cloud bases above 400m), small LWP are seen (NP-Ch in the morning). Thus, our simulations extend to cover both LWP gains and losses (or both negative and positive second indirect aerosol effects given that these relate to macroscopic changes in cloud, incorporating changes in LWP that are held constant for the first indirect aerosol effect).

This is an interesting addition to the analysis of the cloud responses, and as suggested, we make amendments to the manuscript (p.16/L.1 to p.16/L.33) that now frame our LWP results against understanding of the processes, and sensitivities, discussed in literature.

VI) It is not only the magnitude of the second indirect effect that is uncertain, but also the sign (Ackerman et al. 2004).

Again, we agree that this point was omitted from the original manuscript. As the second indirect effect is related to LWP changes in the cloud (with the first indirect aerosol effect assuming a constant LWP, Twomey, 1974), the large dependence on conditions described in the response to Comment V means that the sign of the second indirect effect is indeed often uncertain. In addition to the added discussions detailed in Comment V, we emphasise the point further by adding the sign of the second indirect effect to the text in the Abstract (p.1/L.17) and in the Conclusions (p.20/L.3).

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