Response to Anonymous Referee #2

We wish to thank the referee for their insightful, detailed and helpful comments. We would additionally like to express our gratitude for the care and clarity with which the comments were presented.

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) It is emphasized in the manuscript that the sea salt particles are injected from a point source. There are some descriptions in section 3 on how the injection is done. It is unclear how the sea spray rate of 30 kg s-1 (Salter et al. 2008) was converted to the mass fluxes and number flux used in the model simulations. How large was the perturbation to aerosol number concentration in the model grid cell upon injection?

Here, 'point source' refers to emission of aerosols into a single grid cell, of horizontal dimensions 300 m x 300 m (this follows Wang et al. Wang et al., 2011)). The mass and number fluxes quoted in the paper are emitted over this 300 m x 300 m area. We used the 'point source' term to emphasize that the aerosol emission into a single grid cell is a closer representation of the highly localised and concentrated emissions that would occur with the proposed injection vessels (Salter et al., 2008). This is in contrast to the lower aerosol emission rates associated with uniform emission over the significantly larger grid cells of global climate models for the same total aerosol flux.

The number flux was calculated using an assumed wet droplet diameter of 800nm, as suggested by Salter et al. (2008). The mass flux was calculated using the assumption that the dry particle has one quarter of this diameter (Lewis and Schwartz, 2004) and that the mass of Na and Cl would be distributed according the ratio of atomic masses of the two elements. These calculations are presented below. Thus the aerosol increase into the bottom layer 300 m x 300 m grid cell during the injection time was 1.24×10^{12} m^{-2} s^{-1}.

In order to clarify these assumptions, we add further descriptions to the manuscript to the Introduction (p.2/L.17 and p.3/L14), to the Experimental Design section (p.6/L.28) and add detail on the number and mass flux calculations on p.7/L.4.

Details on the calculation of the mass and number fluxes follow:

Assuming that 30 kg s^{-1} of sea water is injected into the boundary layer:

Number flux:

Assuming a spherical droplet, 800 nm in diameter,

This will have a volume of:

$$V_{each\ droplet} = \frac{4}{3}\pi r^3 = \frac{4}{3}\pi(400 \times 10^{-9})^3 = 2.68 \times 10^{-19}\ m^3 \quad \text{(Eq. 1)}$$

Taking a density of sea water as $1\ m^3 = 1000\ kg$,

$$mass_{each\ droplet} = (2.68 \times 10^{-19}) \times 1000 = 2.68 \times 10^{-16}\ kg \quad \text{(Eq. 2)}$$

Assuming that the $30\ kg\ s^{-1}$ of sea water is injected from a device that is smaller than our 300 m x 300 m grid cell size, and is therefore taken to be emitted uniformly over the grid cell size, the mass flux of sea water becomes:

$$30\ kg\ s^{-1}\ \div (300\ m\ \times 300\ m) = \ 3.3 \times 10^{-4}\ kg\ m^{-2}\ s^{-1} \quad \text{(Eq. 3)}$$

Therefore, the number flux of droplets:

$$(3.3 \times 10^{-4}) kg\ m^{-2} s^{-1} \div 2.68 \times 10^{-16} kg = 1.24 \times 10^{12}\ m^{-2} s^{-1} \quad \text{(Eq. 4)}$$

Mass fluxes:

These were based on the assumption that the dry salt has approximately one quarter of the diameter of the wet droplet (Lewis and Schwartz, 2004), such that a dry salt particle would have a diameter of 200 nm (see below), and is approximately equal to a simple salinity calculation.

The calculations assume a spherical shape of the dry salt particle:

$$V_{each\ dry\ droplet} = \frac{4}{3}\pi r^3 = \frac{4}{3}\pi(100 \times 10^{-9})^3 = 4.1888 \times 10^{-21}\ m^3 \quad \text{(Eq. 5)}$$

Taking a density of sea salt of 2.165 $g\ cm^{-3}$,

$$mass_{each\ dry\ droplet} = (4.1888 \times 10^{-21}) \times 100000 \times 2.165 = 9.068 \times 10^{-15}\ g \quad \text{(Eq. 6)}$$

Each dry droplet is assumed to be composed of Na and Cl. Taking the atomic weight of Na as 23.00 and that of Cl as 35.45,

$$mass\ of\ Na_{each\ dry\ droplet} = (9.07 \times 10^{-15}) \times \frac{23.00}{(23.00+35.45)} = 3.569 \times 10^{-15}\ g \quad \text{(Eq. 7)}$$

$$mass\ of\ Cl_{each\ dry\ droplet} = (9.07 \times 10^{-15}) \times \frac{35.45}{(23.000+35.45)} = 5.501 \times 10^{-15}\ g \quad \text{(Eq. 8)}$$

Using the droplet number of $1.243 \times 10^{12}\ m^{-2}s^{-1}$, the mass flux becomes:

$$mass\ flux_{Na} = (3.569 \times 10^{-15}) \times (1.243 \times 10^{12}) = 4436\ \mu g\ m^{-2}s^{-1} \quad \text{(Eq. 9)}$$

$$mass\ flux_{Cl} = (5.501 \times 10^{-15}) \times (1.243 \times 10^{12}) = 6840\ \mu g\ m^{-2}s^{-1} \quad \text{(Eq. 10)}$$

1 continued) The explanation on how the Salter's full emission rate causes simulation failure seems to be interesting but inadequate to justify the choice of inject rate. It needs more clarification.

Owing to the potential side-effects and complex governance that currently restricts geoengineering field test, the assumptions used in all geoengineering modelling simulations are subject to uncertainties. Thus while the Salter et al. (2008) design proposal for the hardware necessary to implement MCB is comprehensive, some details of the scheme (e.g. sea-water droplet production)

remain in development. Hence, the 30 kg s^{-1} value can only be taken as a possible achievable sea-water injection rate. Additionally, the 30 kg s^{-1} injection rate would be anticipated to be reached at wind speeds of between 6-8 m s^{-1}, with Korhonen et al. (2010) introducing a wind dependent aerosol emission rate. As our upper limit of 0.5 of the Salter suggested maximum flux is within the proposed design range, and produces perturbations to the cloud system that are clearly evident, we suggest that this upper bound is acceptable to use here in identifying patterns of cloud response to the aerosol injection over the diurnal cycle.

In order to detail our justification of this choice of upper bound, we add clarification to the Experimental Design (p.7/L.11).

1 continued) It also sounds odd to use the combination of CAM longwave radition scheme and RRTMG shortwave scheme. The latter is usually used for large-scale model simulations.

The RRTMG scheme is designed to be used across a range of scales. The RRTMG scheme is included rather than the RRTM scheme in the model Version 3 of WRF and is coupled to the aerosol option selected in WRF/Chem.

This combination of long-wave and shortwave radiation schemes produced cloud simulations that were maintained through the night via longwave cloud-top radiative cooling (Fig. 3 c, and Fig. 5 c in the amended manuscript), and that dissipated into the day resulting from shortwave radiative warming of the cloud offsetting the longwave cooling (Fig. 3 c, and Fig. 5 c). The diurnal variation in cloud radiative heating/cooling rate produced mixing (exemplified by vertical velocity variance, Fig. 3 a and Fig. 5 a) that was driven from this cloud top region and that followed the diurnal pattern in weakening during the day, which is typical of marine stratocumulus cloud systems.

As such, the production of diurnal cycles of cloud and dynamical features in both the NP and WP cloud examples accorded us with confidence in the performance of these radiative schemes for use in this study.

We add justification for our scheme selection to the Model Set-up section of the amended manuscript (p.4/L.25).

2) The domain size of 9 km x 9 km is rather small for simulating marine stratocumulus clouds, which usually have organized cloud structures with horizontal scale even larger than this domain size. Moreover, with such a small domain size, there is no way to characterize the mesoscale cloud dynamics and the interactions with cloud microphysical changes caused by the strong local aerosol perturbation. This raises the question of how representative the simulated clouds and their responses to the aerosol inject are for marine stratocumulus.

Owing to computational constraints, these diurnal simulations were limited in domain size. The limited domain size does mean that we are unable to capture the mesoscale cellular cloud features, and feedbacks that have been shown to occur in response to gradients in aerosols (e.g. Sharon et al., 2006; Wang and Feingold, 2009a, b; Wang et al., 2011).

In spite of these omissions, the clouds that are simulated here are able to capture the key features of MSc cloud behaviour over the diurnal cycle, including the importance of radiative cloud-top cooling in 'powering' the turbulence within the cloud system, resulting vertical velocity variance and cloud development, with subsequent break-down into the day as SW warming offsets cloud top cooling. Similarly small domain sizes (or smaller) have previously been used for the investigation of MSc and its sensitivities to conditions (e.g. Ackerman et al., 2009; Chen et al., 2011). Additionally, cloud responses to aerosol injection are as expected, including precipitation suppression, LWP increases and increased entrainment subsequent to N_d increases in the WP case.

Therefore, these simulations represent the rapid response of the clouds at small domain sizes, but do not capture the larger scale feedbacks that can distort the results at larger domain sizes and longer time-scales. In this way, our results are intended to introduce the issue of aerosol injection timing sensitivity, but cannot be fully representative of cloud response at meso-scales.

It is acknowledged that further modelling work is needed to gain a fuller understanding of the sensitivies to aerosol timing. This would include investigation of a larger range of cloud system properties, both at this larger domain size with additional feedbacks, but also incorporating the lifetime of aerosols (over several diurnal cycles). For this, a domain size would be necessary that would capture the horizontal dispersion of aerosols over this time, and also be able to simulate the mesoscale features at scales of tens of kilometres. The computational intensity and difficulties in maintaining a balance of forcings capable of producing reproducible and recovering cloud properties over several diurnal cycles means that these simulations would be challenging.

In response to this comment, we add reasoning for simulation set-up, constraints, usefulness and discussion of future challenges and simulation aims in the amended manuscript on p.6/L.5 to p.6/L.17 and p.19/L.15.

See also response to Referee #1, Comment 3.

3) The total dissipation of clouds in the daytime makes the weakly precipitating case less representative. There are quite a few studies in the literature simulating the same DYCOMS II case using various LES models. None of them seems to have the cloud-free situation. It is unclear why this happened in the present study, which warrants more in-depth explanation than "due to the less turbulent conditions".

The three control cases were intended to represent the possible response to aerosol injection for a range of clouds. The WP case (in which total cloud dissipation occurs) was therefore selected as it simulates a more extreme cloud condition. While this cloud is therefore less representative of typically observed marine stratocumulus, it does embody a bounding cloud case that has been observed previously (Albrecht et al., 1988; Minnis et al., 1992). We discuss this atypical nature of the cloud in the amended manuscript (p.8/L.25 to p.8/L.30). We also acknowledge that the simulations used here are a small sub-set of the vast range of MSc conditions possible (p.16/L.32).

While we used the DYCOMS II RF02 initialisation soundings, other model variables (e.g. wind initialisation, surface fluxes) were different to those used in previous simulations of RF02. Additionally, the background aerosol concentrations were intentionally varied in order to produce a range of base case simulations with different cloud properties. As such, similarity to the RF02 case would not be expected. We clarify this distinction in the text of the amended manuscript (p.5/L.7).

The occurrence of cloud-free conditions in the WP case is the result of the radiative and thermodynamic fluxes associated with the combination of cloud and atmospheric conditions simulated here. Under this combination of conditions, the loss of water through precipitation, combined with the reduced moisture transport to the cloud caused by turbulence decreases resulting from shortwave warming offsetting cloud-top longwave radiative cooling produces a loss in supersaturation and total cloud dissipation. These conditions, and hence the dissipation of the cloud, would be sensitive to changes in variables including large-scale divergence, sea-surface temperature, background aerosol concentration, wind speed and moisture content above the cloud top (cf. Chen et al., 2011). A more thorough description of the processes that lead to the cloud dissipation is added to the amended manuscript on p.8/L.19.

See also response to Referee #2 Comment 2.

3 continued) In addition, the clouds are not totally recovered in any of the cases. If the simulations were extended to a second day, the model boundary layer would have collapsed completely. In other words, the simulated morning, daytime and evening clouds and their responses to aerosol perturbation very likely depend on when the model simulation is started.

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.

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 #1 Comment 1.

4) The use of calculated clear-sky albedo as a measure of direct aerosol effect might be inappropriate. If cloudy columns are excluded from the calculation, should we see stronger direct effect when cloud fraction is lower? If you look at results in Fig 8, it doesn't seem to be the case.

The clear-sky albedo is calculated as the ratio of upward to downward SW radiation at the top of atmosphere in clear-sky conditions for all (including cloudy) columns. i.e. the contribution of cloud to the albedo is omitted from the clear-sky radiation calculation, rather than the cloudy columns in their entirety. As such, the clear-sky albedo increases for injection times 03:00 LT and 13:00 LT in to the weakly precipitating regime are similar (Fig. 9 in the amended manuscript) despite there being a larger cloud fraction at 03:00 LT than at 13:00 LT.

We clarify the description of the clear sky albedo in the amended manuscript (p.11/L.19).

5) Page 24206, line 13: domain or cloud "average"?

The 'average' refers to the domain average. We make the distinction clear in the amended manuscript on p.1/L.21.

6) Page 24206, line 15: change "day" to "daytime" or more exactly "early afternoon". Same for a few other places in the main text.

We agree that this change will clarify the results. We change 'day' to 'early afternoon' in the following places in the amended manuscript: p.1/L.24, p.1/L.28, p.12/L.10, p.12/L.22, p.12/L.32, p.14/L.18, p.16/L.9, p.17/L.26 and p.20/L.5.

7) Page 24206, lines 20-22: The sentence "penetration and accumulation of aerosols.. ...cloud albedo increases" seems to be out of context and incorrect. I suggest remove and clarify more.

As suggested, we remove these sentences from the abstract (p.1/L28) and add clarification and further discussion of the processes involved in the Discussion Section, as detailed in Comment 11.

8) Page 24208, line 22: This statement seems to be just applicable to some particular GCMs.

We agree that this statement may not incorporate the behaviour of all GCMs, and adjust the text in the amended manuscript (p.3/L.21).

9) Page 24212, lines 5-7: how was the 800 nm sea water drops convert to 200 nm dry aerosol particle size.

From Lewis and Schwartz (2004): "The ratio of the radius of a seawater drop at formation r_{98} to the equivalent dry radius r_{dry}... ...$r_{98}/r_{dry} \approx 4.0$". Therefore, we assume that the dry aerosol particle size is one quarter of the diameter of the sea water drop. This assumption is also used by Latham et al. (2012): "Hence, the initially sprayed droplets, drying to a quarter of their initial size, should minimally be of the order of 150-200 nm diameter".

We add this detail, and reference to the amended manuscript (p.7/L.3).

10) Page 24216, lines 21: The defined all-sky planetary albedo might have accounted for ocean surface albedo as well.

We agree that the all-sky planetary albedo will take the ocean surface albedo into account. This ocean surface albedo is constant in the model. We add this point to the manuscript (p.11/L.17).

11) Page 24222, lines 17-18: why are the non-absorbing sea salt particles typically not associated with SW attenuation?

The term "SW attenuation" was incorrectly used here since attenuation describes both the absorption and scattering of SW radiation. This passage was intended to convey the lack of absorption in the ultra-violet/visible wavelength range associated with sea salt particles. In response to this comment, we re-word these passages in order to improve clarity (p.18/L.20 to p.19/L.6).

12) Page 24223, lines 25-27: why should the injection from a point source particularly lead to the penetration of aerosols above cloud top?

The high concentrations of aerosols associated with the point source injection (i.e. emission into one grid cell rather than uniformly over the whole domain) are transported up through the boundary layer by turbulent mixing. These aerosols lead to significant increases in N_d, reductions in cloud droplet size and increases in cloud top radiative cooling. This in turn increases turbulence and the entrainment of overlying warm, dry free tropospheric air at the cloud top. This is evidenced by the increase in cloud top height in injected cases, particularly for into the early and mid mornings (Fig. 6 of the amended manuscript) given that large-scale subsidence remains constant. This entrainment of warm, dry air leads to increased cloud droplet evaporation at the cloud top, and the formation of a layer of interstitial aerosols overlying the cloud top.

This phenomenon may occur more for point source injection than for a more distributed (and hence lower concentration) aerosol emission because the higher local aerosol concentration leads to larger

increases in N_d. For our point source injection, N_d increases by more 1000%. Global climate model simulations, having more uniformly applied aerosol injections, result increases in N_d of up to between 40% (South Pacific; Korhonen et al., 2010) and 80% (South Pacific; Partanen et al., 2012). Thus, the smaller N_d increases may not produce the large increases in entrainment seen for point source injection. We remove reference to the term 'penetration', and add details of this explanation in the amended manuscript (p.18/L.14, p.20/L.6).

ADDITIONAL AMENDMENTS:

1. Removal of erroneous reference on p.6/L.3
2. Update Wang et al. (2011) reference from ACPD to ACP
3. Update Yang et al. (2011) reference from ACPD to ACP
4. Amendments to erroneous contour bars, Figure 6 – Time evolution for...

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