Response to anonymous referee 2

We thank the reviewer for thorough and thoughtful comments. Our responses to each of the points are given below.

Specific comments
1. The paper discusses the results in terms of their implications for the marine cloud brightening geo-engineering scheme. The results from this study are most certainly relevant but there are some key differences. The plume from a ship stack is highly buoyant and so will reach the cloud even if the boundary layer is stably stratified. The surface generation of sea-salt aerosol will lead to a plume which is if anything slightly negatively buoyant so may well not reach the cloud level in conditions where a reduction in cloud albedo is likely. This point needs to be discussed. This study does have considerable interest for the general topic of cloud aerosol interaction extending beyond any geo-engineering application a point which needs to be made.

>> **Response**: The following has been added: "While the present study has been motivated predominantly by gaining a more thorough understanding of the response of marine stratocumulus to perturbations in aerosol concentrations, it has obvious implication in terms of marine cloud brightening geo-engineering. There are some important differences, however; the buoyancy of ship exhaust will generally cause the plume to reach the overlying clouds, whereas surface generation of sea salt will lead to a plume that is largely neutrally buoyant. The efficiency of vertical transport of a plume of sea salt aerosol, under a variety of conditions, needs to be considered in the design of a geo-engineering strategy. "

2. The physical explanations of the key processes responsible for the range of effects seemed to be clear but somewhat brief. Quantitatively the key to the paper is equation 3 and the explanation of this was less clear. The assumptions in deriving this equation need to be carefully explained along with the limitations in their use. The clouds are clearly not always adiabatic e.g. the effects of entrainment and precipitation are discussed in the paper; but there should be some discussion of how significant this is and whether the assumptions that are made still hold in these cases. The units of each of the quantities need to be stated, since some are stated while others are not.
I do not intend to pick up on all the minor text changes or typos (there are a few but these may change/re-appear after revision anyway). However, as stated above, I would like to see some additional explanation of terms in equations – particularly Eq. 3, with better provision of units for most terms since the few given are not the same and are mixed together.

>> **Response**: Concerning the adiabatic assumption applied in Eq. (2) and (3), we have added more detailed explanation: "The assumption of adiabatic conditions in Eq. (2) may not be appropriate for heavy drizzling clouds and/or partly cloudy (i.e., open cell cloud) conditions (e.g., Hayes et al., 2010). Comparison of cloud albedo susceptibility using Eqs. (1) and (2) will be investigated to evaluate the effect of the adiabatic assumption (Sect. 3.1)."

The effect of the adiabatic assumption has been examined in Section 3.1 and added into Figure 4:
"Comparing the cloud albedo susceptibility estimated from the analytical formulation based on the adiabatic assumption (Eq. (3)) and derived using the droplet size spectrum (Eq. (1)), it is shown that the two derived albedo susceptibilities exhibit only small difference for non-precipitating and light-drizzling clouds (RF18, 19, and 24, as shown in Fig. 4). However, for heavy drizzling clouds with open cell structure (RF20), the cloud albedo susceptibility derived using Eq. (1) is about twice as large as that from Eq. (3), indicating that the assumption of adiabatic conditions is not applicable for heavy drizzling clouds (see also Hayes et al., 2010)."

Concerning the units for each term and additional explanation in Eq. (2) and (3), we have added the followings accordingly: "C_w is the moist adiabatic condensation coefficient, ranging from 1 to 2.5×10^{-3} g m^{-4} for temperatures between 0°C and 40°C (Brenguier et al., 2000); *k* is a parameter inversely proportional to the droplet distribution breadth (unitless), approaching 0 for a very wide droplet spectrum and 1 for a monodisperse droplet population."

Additional explanation of Eq. (3) has also been added: "The three effects represented in Eq. (3) are the major ones governing the response of cloud albedo to a perturbation in cloud droplet number concentration. As an increase in emitted aerosol particles can lead to an increase in N_d (the strength of which depends on background aerosol number concentration, particles acting as cloud condensation nuclei, etc), Eq. (3) can be applied to the ship exhaust observations, expressing the change between the unperturbed clouds, subject only to the marine background N_a (thus background N_d), and those perturbed by ship exhaust."

The definition of cloud drops and drizzle drops seems to be somewhat arbitrary, e.g. P13556, line 14: cloud drops 1.77 µm (e.g. why was this value chosen?) P13556, line 17: the size limit used in the definition of drizzle (20 µm) is somewhat low in comparison to what is often used – but it is defined clearly so at least the reader has been made aware of this.
This leads on to the issue that there is little discussion about measurement details e.g. with respect to the probes used (CIP is said to be used for drizzle but never described at all wrt its size range or type – I assume a CIP-25 is used as part of CAPS – providing the larger end particle size measurements up to 1.6mm from CAPS); how well they were working (e.g. what calibration checks or maintenance checks were undertaken and how well did the probes perform - to inform the reader about how "good" or trustworthy the data are); or how well similar measurements (by different probes) compared (e.g. particle size distributions) particularly at crossover points in that comparison too.
Leading on from this, there is no discussion as to what fraction of which instruments data set comprised the final data set presented (or used). Although I suspect this will not affect the main arguments presented in this paper, there should be some discussion or summary of these instrumentation/data set details so that the reader can have confidence in the data presented (for those that are not measurement experts, and also to allay the concerns of those readers who may be measurement experts!). Basically this is a request to provide an estimate of uncertainty in the measurements used. I suspect the data presented on occasions come from a single probe and that definitions of "cloud drop" and "drizzle drops" are also somewhat tied into the size ranges of the different instruments. There should be some statement also that the range of probes used was capable of capturing the full size range of the particles present – e.g. that the CIP probe did not miss a number of larger precipitation sized particles for example.

>> **Response**: The definition of cloud drops and drizzle drops is the same as that in Lu et al. (2007). The following has been added in Section 2.1: "These values (radius 1.77 and 20 µm) were chosen based on the particle size range of each measuring bin. Cloud droplet is defined as drops with radius larger than ~2 µm (e.g., Chen et al., 2011), and the corresponding size bin in CAPS is between 1.77 and 2.35 µm in radius. Therefore 1.77 µm was chosen as the threshold of cloud droplet size. For drizzle drops, the cutoff radius between cloud and drizzle drops is defined as 25 µm (e.g., Wang and Feingold, 2009). The closest size bin in CIP is that ranging from ~20 to 32 µm, with 25 µm being the geometric mean. Thus 20 µm was chosen as the lowest boundary of drizzle drops."

Concerning the CIP, it is included in the CAPS package, as noted in the footnote of Table 1. We have added the following in Section 2.1 second paragraph: "Drizzle drop size distribution was measured using the Cloud-Imaging Probe (CIP, included in the CAPS package, with size range from ~15 µm to 1.6 mm)."

The instruments used during this field campaign have been applied in previous field studies MASE-I (Lu et al., 2007) and MASE-II (Lu et al., 2009). The data presented here were measured by the instruments introduced in Section 2.1. Though some instruments measure the same parameter (for example, both CAPS and cloud droplet probe (CDP) measure the cloud droplet distribution), during these four flights the CDP were inoperative and thus no direct comparison between two measurements was available. Nevertheless, in some other cases where both instruments functioned, the size spectra match reasonably well. Concerning the uncertainty, the following sentence has been added: “In situ measurements are subject to a variety of uncertainties and limitations. The measurement uncertainty of the probes is documented in several studies (e.g., Baumgardner et al., 2001; Conant et al., 2004; Lance et al., 2010). The probes were repeatedly calibrated during the E-PEACE field mission.”

A similar statement(s) should be made about the quality/limitations of the remote sensing data too (P13558)

>> **Response**: The following has been added in Section 2.2: “Droplet effective radius and cloud optical thickness were derived from the 3.7-µm reflectances and obtained using the MODIS cloud product (MYD06, King et al., 1998). One-kilometer pixels were screened to include only those with full cloud coverage and fitting the requirement of a single layer, low-level (cloud top pressure greater than 600 hPa), and warm phase cloud. The screening criteria are similar to those applied in Christensen and Stephens (2012). Cloud albedo was calculated using BUGSrad (Stephens et al., 2001), a two-stream radiative transfer model. LWP was derived from the effective radius and optical depth through LWP = $(2/3)\rho_w r_e \tau$ (Stephens, 1978), assuming that the cloud contains spherical droplets and that liquid water content follows an adiabatic vertical profile. These assumptions lead to ~30 % error at the pixel scale, as derived from Bennartz et al. (2007). Therefore, numerous pixels, a minimum of 30 for a ship track, were grouped together into segments to reduce the uncertainty, thereby producing a more representative average of the cloud optical properties derived from MODIS.”

P13559, line 22 and Fig, 2: the Reff axes in Fig.2(a) and (b) are not labelled (or described in the legend)

>> **Response**: The x-axes in Fig. 2 are liquid water content, as shown on the Fig. The effective radius has been presented by the size of symbols, as shown in the legend and the caption. To make it clearer, we have added the label “R_e” above the symbols in Fig. 2.

P13564, line 25 Conclusion: change “led” to “leads”/ P13565, line 4: change “deeper” to “higher”

>> **Response**: These have been changed accordingly.

P13565 “Conclusions” and Fig. 8: I do not find the “conceptual figure” provided to be particularly illuminating, especially since it needs the long description in the figure legend to be present (repeating what is shown graphically above) to make sense of it.
This long description should be in the main body of text rather than the legend. Then the requirement for the figure is debateable, so the continued presence of this figure should be justified or it should be removed.

>> **Response**: Figure 8 is intended to illustrate the key processes regulating the cloud albedo response. We would like to retain this figure. The figure legend has been revised as the follows: “Conceptual diagram displaying the interactions among aerosol, cloud, precipitation, and meteorology. The response of each property/phenomenon to increased aerosol (*N*a) is shown as a red plus (signifying positive response), and a blue minus (negative response) sign. Footnotes to figure: (1) Twomey effect (Twomey, 1991). (2) Albrecht effect (Albrecht, 1989). (3) Sedimentation-entrainment effect (Ackerman et al., 2004). (4) Drizzle-entrainment effect (Wood, 2007). (5) Significant meteorological conditions, such as free tropospheric humidity (*q*ft), large scale divergence rate, as well as cloud top height (z_i), can control the MSc structure (Wood, 2007; Chen et al., 2011).”
Also, the following has been added in the main text in conclusions: “The so-called Twomey and Albrecht effects can lead to cloud brightening and thus cooling. On the other hand, in response to an aerosol perturbation, reduced in-cloud sedimentation leads to an increase of cloud water and evaporation in entrainment regions, resulting in stronger entrainment (Ackerman et al., 2004; Bretherton et al., 2007). Besides, less drizzle reduces below-cloud evaporative cooling and in-cloud latent heat release, resulting in higher turbulent kinetic energy and thus stronger entrainment (Wood, 2007).“

Table 2 (and text P13558): The significance of values and changes in "k" (the droplet spectral shape parameter) are not really discussed anywhere in any useful sense.
This is true of many parameters introduced into the main body of text (e.g. dispersion). Please justify their inclusion.

>> **Response**: As optical depth can be written as a function of *k* (as shown in Eq. (2)), the dispersion effect (dlnk/dlnN_a) in Eq. (3) can be significant in governing the response of cloud albedo to a perturbation in cloud droplet number concentration. Based on our calculations, the magnitude of the dispersion effect has the smallest contribution to the albedo susceptibility (Fig. 4), and thus is discussed less compared to the other effects.
The following has been revised/added in Section 3.1 (for RF18 and RF24): "The sign of the dispersion effect is slightly negative in these two cases where non/light drizzle exists. The broadening of the spectrum is caused by the competition for water vapor in the relatively polluted, condensation-dominated regime, offsetting the cooling from the Twomey effect (Feingold and Siebert, 2009). This result is consistent with previous observational studies (e.g., Ackerman et al., 2000; Liu and Daum, 2002)."
And the following has been revised/added for RF20: "The dispersion effect is positive (narrower droplet size spectrum under polluted condition) in this heavy-drizzling case, an opposite trend to the cases with non/light drizzle (RF18, 19, 24). This result agrees with the large eddy simulation (LES) studies in Lu and Seinfeld (2006) and Chen et al. (2011), where a larger value of the dispersion effect occurs for clouds with stronger precipitation. With increased aerosol, smaller droplets suppress collision-coalescence, leading to less spectral broadening. Also, higher updraft velocity (due to stronger turbulence) leads to droplet condensational growth and thus spectral narrowing (Lu and Seinfeld, 2006). In RF20, the dispersion effect acts to enhance the Twomey effect. Among these three major effects, the dispersion effect plays a minor role in the total albedo susceptibility."

Fig 1: there is no indication of ambient wind - is it low and hence unimportant – then say so (quantitatively)

>> **Response**: The ambient wind speed ranges from 2.5 to 10 m s^{-1} in these four cases. The following has been added in Section 3.1: "The wind speed is lower in RF18 and 24 (2.5 and 5.5 m s^{-1}, respectively), where negative albedo responses were observed. Stronger wind speed was measured in RF19 and 20 (9.5 and 10 m s^{-1}, respectively), where positive albedo responses occurred. Weaker wind is associated with smaller latent heat flux from the ocean surface, and thus less moisture supply to the boundary layer. This may partially contribute to the negative cloud albedo response as the cloud gets drier through entrainment, yet does not accumulate moisture through the mixing."

Fig 2: (as above) – label effective radius axes/ Fig. 7: the 5K and 200m described are: "5K wide bins" and "200m wide bins"/Fig. 8: Move long explanation in legend to main body of text – justify the need for the figure.

>> **Response**: These have been revised/addressed.