

Response to referee #2

We thank the reviewer for the useful comments our response is below, with the reviewer's comments in italics.

This study explores the sensitivity of the activated cloud droplet concentration to different concentrations of artificially-produced sea-salt particles under different assumptions about the updraft strength and distribution and for different background aerosol size distributions produced by three global models. As far as this reviewer can tell, the global models are used only for producing the background aerosol fields (which are not interactive in the sense that activation outcomes do not feed back onto aerosol fields) and only their activation schemes are tested here. Impacts upon cloud optical properties and radiation are not examined, nor is the model used to predict the updraft speeds used in the activation. It seems more a test of the model activation parameterization under a range of background aerosol scenarios than a true test of the efficacy of geoengineering. I think the reader needs to be made aware of what the manuscript is attempting to do from the start, because the current title does not indicate that it is only the efficacy of the activation process that is tested here. There is no attempt to examine the feedbacks of changes in cloud droplet concentration on cloud condensate or cover. It is fine to do this, but a test of the efficacy of this type of geoengineering would need to include much more. As process studies show, such feedbacks are likely to be critical in determining the overall efficacy of sea spray geoengineering. That said, I think the manuscript does contain some interesting findings and I think will be acceptable for publication subject to some revision.

Major points:

1. *The authors need to rename their title to make it clear what has been done and to avoid misleading those thinking that the study provides global model estimates of geoengineering efficacy (i.e. cloud albedo increases driven by deliberate salt particle emissions). I might suggest something like:*

"An assessment of the efficacy of increasing cloud droplet concentration by sea spray geoengineering for different updraft speed assumptions and for background aerosol states derived from three global models"

The title has been changed to: "A multi-model assessment of the impact of sea spray geoengineering on cloud droplet number"

2. *This study replicates the essence of previous parcel model studies (it should be noted that the models used here are essentially using activation schemes derived from the parcel-type approach). The most important finding in this study that differentiates it from previous studies is that the cloud droplet concentration enhancement with seaspray geoengineering is found not to be very sensitive to the assumed width of the injected sea-spray aerosols.*

We replicate parcel model studies and then extend this analysis to the global scale, using aerosol distributions simulated by three different aerosol models and different assumptions about the injected mode (e.g. width, diameter).

3. *The main substantive criticism I have of the paper is that the assumed updraft speeds are somewhat low compared with most real stratocumulus clouds over the oceans. In citing only recent papers ignores the rich history of documenting and understanding the turbulent structure of Sc determined from observations from the 1980s onward. These include Nicholls (1984), Nicholls and Leighton (1986), Nicholls (1989), all QJRMS, Hignett (1991, JAS), deRoode and Duijnkerke 1995 (QJRMS), 1997 (JAS), Wood (2005, JAS) etc. and a multitude of modeling papers from the 1990s. The review paper of Wood (2012, MWR) discusses this history in detail.*

Typical updraft speeds in most marine low clouds are larger than those in Lu et al. (2007) that are used to justify the authors' choice. The boundary layers in Lu et al. are very near the Californian coast and are atypically shallow compared with boundary layer depths over the broader subtropical oceans where the maxima in marine stratocumulus occur.

The observed convective velocity scale in the ASTEX Lagrangian case are consistently above 0.6 m/s throughout (deRoode and Duynkerke, 1997, JAS, Table 2). See also Bretherton et. al. (2010, ACP) for some measurements of the vertical velocity pdf in deeper MBLs more typical of the open ocean stratocumulus regions. These results showed vertical wind standard deviations in the range 0.3-0.8 m s⁻¹.

Thus, I am concerned that the conclusion about not being able to achieve 375 cm⁻³ everywhere might be problematic, since with 0.2 m/s it can be achieved everywhere. I would like to see some more discussion of this. It would be much more robust a statement if the model actually predicted the updraft speed and its geographical variability.

We agree that it would be ideal to predict updraft in the global models but at present updraft velocity is a major uncertainty in general circulation models and although parameterisations exist the authors believe the uncertainties associated with them to be too significant to use them in this work. For example Partanen *et al* (2011) state that the simulated updrafts in ECHAM-HAM2 in the stratocumulus regions were 1.0 – 1.4 m s⁻¹ despite the fact that these velocities are not supported by field measurements. This potentially large bias in simulated updrafts would influence the conclusions reached and for this reason we choose to prescribe updraft velocities offline.

Also, although updraft speeds of 0.3-0.8 m s⁻¹ are observed in marine stratocumulus clouds they generally occur in the centre of the cloud and not at cloud base - the region most important for droplet activation. As stated in the paper, high updrafts in the centre of the cloud tend to grow existing droplets, which is important for LWC, but does not generally activate many additional droplets. The references (and model simulation) we use all give cloud base values. The older literature tends to give cloud centre values, or does not specify the location.

We include results from the ASTEX case in the LEM model results presented in Table 3 (as presented by *deRoode and Duynkerke, 1997*), the advantage in using the LEM model results is that we can isolate the cloud base data and frequency statistics. Even with this approach it is difficult to settle on a “representative” in-cloud updraft velocity as, as the reviewer notes, it is dependent on the meteorological conditions. In this work we aim to draw attention to the finding that updraft may be a potentially important constraint on the efficiency of sea spray geoengineering, but it is uncertain and poorly parameterised in models and thus should be the subject of future investigations. We have tried to clarify this in the text.

4. The authors need to state what is new here that isn't in existing papers (e.g. those focusing upon geoengineering itself: e.g., Bower et al. 2006). Also, there are a number of papers that have explored the competition effects of introducing coarse mode aerosols (papers by Ghan and others come to mind) that surely have provided insights for this study.

The Bower et al 2006 paper explores the change in CDN and albedo to the injection of a range of different injection fluxes and updraft velocities; we build on this work and explore a larger region of parameter space using background aerosol properties from three global models. So we are essentially extending previous studies to a global scale and accounting for the diversity in modelled aerosol. We agree that work by Ghan et al 1998 is relevant and have added in reference to it.

5. Why is the geometric standard deviation of injected particles assumed to be 1.1? Is this just a guess, or is there some physical basis for this number? Are the results sensitive to this choice?

A geometric standard deviation of 1.1 represents a narrow lognormal mode, geoengineering literature (e.g. Latham *et al* 1990, Latham *et al* 2008) suggested that the injection of a monodisperse mode would give the optimum increase in CDN.

We examine the response to wider modes in the 3D models in Figure 7 which shows that increasing the mode width reduces the simulated increase in CDN.

We also examined this in the OD model where we found that increasing the mode width reduced the CDN, except when the injected mode was very small. In this scenario the large mode width was advantageous as it increased the number of particles that were large enough to activate.

We have now included this finding in the text: “We repeated these experiments assuming a geometric standard deviation of 1.3 and 1.6 (not shown). With the wider modes the pattern of the change in CDN is similar but the wider mode reduces the CDN achieved (fewer particles activate) unless the injection diameter is very small, in which case the wide mode increases the fraction of the mode where particles are large enough to activate.”

6. What exactly is a marine "background". Does such a thing exist? It suggests that there's some kind of steady state in the marine boundary layer, but all the studies and observations I have seen suggest quite the opposite: an aerosol that is highly variable in time and space and can sometimes be almost completely wiped out by precipitation.

In the OD plots we use the climatological marine aerosol distribution from Heintzenberg *et al* 2004 as a baseline pre-existing marine aerosol. It is compiled from ship cruise data and thus includes the effects of the variations mentioned above. We used the term “background” to try and distinguish between the pre-existing marine aerosol and that added through geoengineering, but we agree it can be confusing. Where relevant we have replaced it with the term “pre-existing marine aerosol”.

7. Statements such as "geoengineering becomes more efficient" are misleading. All you know is that the increase of CDN becomes more efficient. This is only one part of the geoengineering problem.

Changed to “the increase in CDN becomes greater”

8. The model is just used to give the fields of the unperturbed aerosol size distribution. Doesn't the model need a vertical velocity to produce such fields, since they depend on physical processes such as activation, precipitation scavenging, cloud processing etc.? What updraft is assumed for these and is it consistent with what is used in this study?

The three models take different approaches to simulating the effect of clouds on aerosol; UKCA assumes all accumulation and coarse mode particles can activate, ECHAM-HAM and EMAC have (different) more explicit cloud activation parameterisations. The wet removal schemes also differ significantly between models. These different approaches reflect the level of scientific uncertainty within the field of aerosol modelling. Thus they are indeed inconsistent with each other and with the updrafts used in the paper.

We have added: “This approach means that the updrafts used in the CDN parameterisation are not the same as those used within the aerosol activation and wet removal parameterisations within the models. We take this approach as the models all have different aerosol activation and wet removal parameterisations thus the assumed updraft velocities vary between models.”

9. Does the use of monthly mean aerosols fields average out a lot of potentially important variability? This can be checked by comparing with the results performed on instantaneous model aerosol fields.

Unfortunately only monthly mean aerosol fields are available from the AeroCom intercomparison. We have added an additional caveat to the text (Methods): “It should also be noted that by using monthly mean aerosol fields we cannot capture the day to day variability of the aerosol distribution which could lead to biases in the predicted annual mean change in CDN, this is a limitation of the offline approach used.”

10. Are the authors comfortable with the fact that the ECHAM-HAM has zero accumulation mode aerosol over most of the global ocean and no apparent sign of continental sources showing up over the ocean?

We include all three models as they reflect the diversity that exists within aerosol modelling; the model results as submitted to the AeroCom inter-comparison study, within this exercise the models will be evaluated against observations (Mann *et al*, *in preparation*) thus we do not repeat this here. It is worth noting that (as stated in

the paper) the models give more similar distributions at the surface and it is only at altitude, where observations are scarce, that ECHAM-HAM has low accumulation mode concentrations.

11. It would be great if the authors showed the MODIS observations for comparison. Unless one is familiar with the Bennartz study, it is difficult to assess the relative model skill at getting the correct mean cloud droplet concentrations.

Comparing our predicted CDN concentrations with satellite observations is tricky for three reasons: (i) the in-cloud updraught velocity at the time of the observation must be known; (ii) MODIS gives the CDN at cloud top, thus the rate of collision coalescence within the cloud is also required; (iii) the MODIS retrievals are reliable only during periods of high cloud fraction, so a point by point comparison would be needed if a perfunctory analysis is to be avoided. This will be the subject of future work but is challenging and out with the scope of this paper.

12. Besides needing increases in CDN, one also needs existing clouds with intermediate albedo (50% is ideal) to effectively brighten clouds. The authors might wish to comment on this. Do the models produce realistic cloud fields?

We agree this is a further uncertainty which we have not considered, we have added a clarification in the conclusion:

“This study only examines the response of the number of activated aerosol to the injected particles, in order to assess the climate impact one needs to also consider the change in albedo that arises from the geoengineering. This is dependent on the cloud liquid water response, the initial cloud albedo and the simulated cloud cover. Further work examining the model dependence of these properties is also required for a robust assessment of the efficacy of sea spray geoengineering for albedo modification. “

OTHER COMMENTS

1. At least one of the authors of the Korhonen et al. study is an author on this paper. Yet the assessment of the causes of the differences between the two studies is in places vague and speculative (P7140, line 23-25). The utility of models is the ability to test such hypotheses.

Korhonen has followed up her 2010 paper that used GLOMAP-Bin by repeating the study within the ECHAM-HAM model and a different aerosol activation scheme (Partanen, Korhonen *et al* 2012). They find larger increases in CDN in ECHAM-HAM (than GLOMAP-Bin) but also do not fully explain the differences as they use a different model and a different activation scheme.

From our research we think that the difference largely arises from changes to the activation parameterisation that have been made since the Korhonen publication and thus we have changed the text to:

“Differences in the simulated change in CDN could arise from differences in the simulated background aerosol distribution. However, GLOMAP-mode and GLOMAP-bin simulations were compared in detail in \citep{mann12}, which found that GLOMAP-mode predicts between 25 -- 100 \% more CCN and thus would be expected to produce a smaller increase in CDN than GLOMAP-bin. We therefore conclude that the main reason for the discrepancy is that \citep{korhonen10} used the activation parameterisation of \citep{nenes03} without the additional treatment of giant CCN of \citep{barahona10}, which is used in this work (BN10). We propose that the suppression of supersaturation responsible for the small increase in CDN in \citep{korhonen10} was overestimated without this treatment.”

2. All the results in Fig. 6 look almost identical for the four regions. Why then not just focus on one region and look in more depth?

We have altered the figure and now show just one region and combine Figures 6 and 7.

3. P7144, Lines 1-4: *I don't understand this statement. The updraft speed is not produced by the model, it is assumed. I agree that more attention needs to be paid to the frequency distribution of updrafts in the real world and in attempting to predict this in large scale numerical models. There are ways to do this.*

We have changed this to: "but in all models when the updraught is assumed to be low"

4. P7145, L10: *Do the authors mean the opposite? Why inversely proportional?*

This was a typo, we have changed it to:

"In the absence of geoengineering, the global distribution of CDN is similar to the distribution of accumulation mode ($50 < R_{\text{p}} < 500$ nm) particles"