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### Interactive comment on "Enhancement of marine cloud albedo via controlled sea spray injections: a global model study of the influence of emission rates, microphysics and transport" by H. Korhonen et al.

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We thank the two reviewers for their comments and suggestions. Our point-by-point reply to these comments is below.

In addition to the changes made based on the reviewer comments, we have added to the Introduction the following sentence that discusses the results of Rasch et al. (2009), which was published after the completion of this manuscript:

"Rasch et al. (2009) concluded that cloud seeding could restore global averages of

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temperature, precipitation or sea ice extent to present day values (but not simultaneously) even in the case of doubled CO2 concentration, assuming that it is possible to increase the CDCN to a very high value of 1000 cm-3 over the seeded areas that cover 20-70% of the world's oceans. "

#### Referee 1:

**1.** Page 739, line 13, and throughout the paper. Make it clear whether the values refer to particle radius or diameter.

The values refer to particle diameter throughout the manuscript. We have now clarified this.

2. Page 740, lines 8-14. This paragraph raises a couple of points: - The authors should explain why they do not include precipitation scavenging from low-level clouds. This seems an odd omission, as drizzle from stratocumulus is an important and common part of the boundary-layer processes in these areas. Can the authors speculate on the impact on their results of including this process?

In the current model version, low level clouds and rain formation are diagnosed separately. Whereas sulphate formation in BL clouds is calculated in the GLOMAP microphysics, precipitation formation is parameterized in host model TOMCAT which treats convective rain arising from sub-grid convective events and frontal rain diagnosed from humidity profiles.

We admit that the omission of drizzle formation in BL clouds is a shortcoming in the current model framework and efforts to improve the description of precipitation in the model are ongoing. To clarify that the omission is a model feature (and not an assumption made only for this study), we have reformulated the corresponding sentence in section 2.1 to:

"Precipitation scavenging in these low-level clouds is currently not described in GLOMAP."

We have also added to Conclusions section the following discussion of the impact of neglecting this process:

"Since the current version of GLOMAP includes precipitation scavenging only for convective and frontal clouds (and not for BL clouds), our study is likely to underestimate the wet deposition rate of MBL particles to some extent. As a result, our estimates of the increases in CDNC due to cloud additional spray emissions are probably an upper estimate (i.e., even higher spray fluxes would be needed to enhance CDNC). However, this effect is expected to be relatively small in the presented annual averages and does not change the conclusions of this study."

## Why do they use ECMWF analyses for frontal and convective clouds but ISCCP products for boundary-layer clouds? Do ECMWF analyses not include boundary-layer clouds?

ECMWF analyses do include cloud data for BL clouds but it is not used in this study because they are available only in forecast mode, not in the re-analyses. The ISCCP product provides a realistic, validated climatological product that is well suited for the current study (but not necessarily for case studies involving specific observations).

### 3. Page 741, lines 14 15. This is unclear: how does the clause in parentheses, which mentions either 10% or 100% of spray particles activating to form cloud droplets, relate to the single CDNC figure of 400 cm-3 ?

Latham (2002) based his calculation on the assumption that every individual particle emitted at the surface will be activated and form a new droplet (CDNC) at cloud altitude (i.e., 100% activated). He acknowledged, however, that some of the particles will be lost by deposition before activation or may not be transported to high enough altitude for other reasons – however, he suggested that the lost fraction will not be larger than 90% (i.e., "worst case scenario" is that only 10% are activated). The fraction of emitted particles that are ultimately activated determines the emission flux required to produce 400 cm-3 new particles.

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In order to clarify this, we have reformulated the paragraph and it now reads:

"— presented in Latham (2002) for the flux required to produce 400 cm-3 additional cloud droplets. Figure 1 compares the geoengineering particle emissions used in this study (red lines) and those calculated by Latham (2002) (black dashed lines; the two lines represent the two assumptions made by Latham (2002): the top line assumes that 100% of particles sprayed at the surface activate as droplets at cloud altitude and the bottom line assumes only 10% activation) —"

### 4. Page 741, line 25. A couple of questions: - At what height above the surface were the sea-spray particles emitted in the model?

The particles were emitted into the lowest-altitude grid box, which over the oceans typically covers the lowest  $\sim$ 60 m of the atmosphere. Thus the effective emission altitude was  $\sim$ 30 m. We have added this information to section 2.2.

#### - What is the vertical resolution of the model in the boundary layer?

Over the oceans 7 model levels represent the lowest  ${\sim}2$  km, of which 5 levels are in the lowest  ${\sim}1$  km. We have added this to the Model description section.

# 5. Methods section in general. Somewhere in this section it should be clearly stated that the model used is coupled in one direction only, i.e. that cloud cover meteorology can affect the aerosol microphysics and predicted CDNC, but that the latter has no impact on model clouds, boundary layer structure etc.

We have added to the end of Model description section:

"It should be noted that in GLOMAP (as in all CTMs) changes in aerosol population do not feed back to model meteorology and cloud properties. However, the predicted aerosol fields can still be used together with information about typical marine boundary layer updrafts to calculate changes in CDNC at chosen altitudes."

#### 6. Page 744, lines 7-9. I suggest adding something like "....for the spray rates

#### currently being considered (Ref)." to the end of this sentence. Otherwise it ends up sounding too much like "it can't be done", rather than "it can't be done using these spray rates."

We have reformulated the sentence:

"— we predict significantly lower CDNC for the spray rates considered here than the 375 cm-3 assumed in earlier climate model studies —"

## 7. Page 745, discussion of supersaturation suppression. Is this argument in any way affected by the extra water injected (and then evaporated) into the boundary layer as part of the sea-spray process?

The effect is likely to be small since the maximum supersaturation reached at the cloud base depends on the condensation rate of water into the aerosol population and the change in temperature due to external (e.g. adiabatic cooling) or internal (latent heat release) forcing. If temperature and aerosol population remained unchanged, extra water would mean lowered cloud base, but the supersaturation reached would stay approximately the same. We anticipate much larger effects from changing boundary layer dynamics due to evaporation of water and the cooling it causes. We plan to investigate these processes in a future LEM study.

8. Page 748, line 26. The concept of an "individual stratocumulus cloud" is rather a vague one - arguably an entire sub-tropical Sc cloud deck could be considered as one cloud, and therefore entirely resolvable by a large-scale model. Perhaps the phrase "individual stratocumulus cloud cell" should be used instead.

Changed as suggested.

#### Referee 2

### While these two deficiencies could be easily remedied by additional model runs C2025

# and interpretation that would make this manuscript highly suitable for a very interesting paper in ACP, in its present form it seems to be a better fit for a more geoengineering-focused journal where the projected cooling effect is of greater interest than the scientific understanding of the atmosphere.

We do not present any estimates of the cooling effect in this manuscript; one would need a fully coupled climate-ocean model to do that. Instead, we present for the first time a global scale study that includes explicit aerosol emissions, transport and microphysics. We investigate the local variation in the efficacy of cloud modification as well as the inadvertent effects of cloud seeding (such as suppression of supersaturation and effects on natural sulphur cycle). We think that these findings improve the understanding of the atmospheric physics related to sea spray geoengineering, and thus make the manuscript suitable for ACP.

Furthermore, we are not sure which other journal(s) the reviewer refers to. Since intense research in the field of geoengineering is very recent (sparked by Crutzen's essay in 2006), there aren't established journals yet that focus on geoengineering. All previous papers on the sea spray method have been published in general atmospheric journals (JGR, Atmospheric Science Letters, Atmospheric Research) or in even more general science journals (Nature, Phil. Trans. Royal Soc., Environmental Research Letters). Furthermore, ACP has published geoengineering studies before – even ones that discuss also non-atmospheric methods (Lenton and Vaughan, 2009).

## (1) Sensitivity studies of both the particle distribution scheme and the other key assumptions should be carried out to explore the physical constraints on why the proposed cloud albedo modification is unlikely to work.

We want to stress that we do not make any claims on whether cloud modification as such is likely or unlikely to work. Instead, we conclude that desired increases in CDNC are likely to require higher emission fluxes than previously suggested – these higher fluxes may perfectly well be obtainable and thus this is not a statement against the

spray method.

The proposed sensitivity studies are commented below.

Specifically, the authors should consider: 1. Running different salt distribution scenarios, including those previously published by Latham et al. 2006 and 2002. It strikes me that this is likely to be particularly fruitful because the spray distribution scheme being considered, which is highest when/where natural spray is highest, is particularly ill-suited to target the most susceptible clouds.

We agree that it would be ideal to target the clouds with lowest background drop concentration, and that in remote marine areas such clouds can often be found in regions with low surface level wind speeds.

However, to our knowledge, the only concrete plan to date of a spray vessel design is from Steven Salter and his coworkers – at least, it is the only design published so far in scientific literature (Salter et al., 2008). Therefore, we feel justified to construct our flux scheme around this design, rather than assume e.g. a wind-speed-independent spray rate. Although the latter was implicitly suggested in Latham's fairly conceptual 2002 paper (note that Latham et al. (2008) do not propose any salt distribution scenario but simply impose changes to the cloud drop number), there are no published plans how such constant but high spray rate could be technically achieved in unmanned, wind-powered vessels. Should it be possible to build such a vessel, it of course could potentially be superior to the one proposed by Salter et al. (2008). However, we wanted to concentrate on proposed rather than hypothetical vessel designs. (It is also noteworthy that John Latham himself is a co-author of the Salter et al. (2008) paper.)

## 2. Running different precip schemes (or at least one and one off) would also seem imperative to identify whether any of the schemes is effective at lofting and maintaining aerosol.

Wet removal is obviously an important factor determining the aerosol lifetime and ma-

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jor deficiencies in it could bias the results. However, previous studies using the same host model TOMCAT have not revealed any major discrepancy in precipitation scavenging (Giannakopoulos et al., 1999; Rasch et al., 2000), and earlier comparisons of GLOMAP results to aerosol field measurements in the midlatitude MBL (Spracklen et al., 2007; Korhonen et al, 2008a) show good agreement and thus do not imply significant problems in aerosol wet removal. Based on these earlier validation studies we are confident that the precipitation scheme from convective and frontal clouds gives reliable results over the investigated regions and time scales (annual averages).

However, the current version of GLOMAP does not include precipitation from BL clouds (see also reply to referee 1, comment 2) and to acknowledge this, we have added to Conclusions section:

"Since the current version of GLOMAP includes precipitation scavenging only for convective and frontal clouds (and not for BL clouds), our study is likely to underestimate the wet deposition rate of MBL particles to some extent. As a result, our estimates of the increases in CDNC due to cloud additional spray emissions are probably an upper estimate (i.e., even higher spray fluxes would be needed to enhance CDNC). However, this effect is expected to be relatively small in the presented annual averages and does not change the conclusions of this study."

#### References:

- Giannakopoulos et al. (1999), Validation and intercomparison of wet and dry deposition schemes using 210Pb in a global three-dimensional off-line chemical transport model, JGR, 105, 23761

- Rasch et al. (2000), A comparison of scavenging deposition processes in global models: Results from the WCRP Cambridge workshop of 1995, Tellus 52B, 1025

#### 3. The suppression of supersaturation is noted, but I think this merits further

### discussion, as this is scientifically the most interesting part of the paper. How frequently does this occur? How important is it? Does it yield thermodynamically consistent clouds or is LWP increased? If so, how?

Concerning the frequency of supersaturation suppression: Adding new particles that are large enough to activate as droplets always suppresses (at least a little bit) the supersaturation reached during cloud formation (see Figure 5). This is because each new droplet acts as an additional sink for the water vapour. Whether this effect leads to a reduced CDNC depends on several factors, such as cloud updraught (effect most significant at low updraught velocities) and aerosol size distribution (as explained in the third paragraph of section 3.2. in the manuscript). In this study, the CDNC was calculated offline using the modeled monthly mean aerosol at 1 km altitude, a parameterization of drop formation by Nenes and Seinfeld (2003), and a probability distribution for updraught velocity that is typical in marine stratocumulus conditions.

We have added to Section 3.2:

"Overall, we predict reduced CDNC in more than 20% of the seeded grid boxes for updraughts less than 0.4 m/s in the GEO run. The regional differences are, however, large: while more than half of the seeded area in the North Pacific shows CDNC reduction for updraughts below 0.3 m/s, the same areal extent of reduction is not seen over the South Atlantic even at the lowest studied updraught velocity of 0.05 m/s."

Concerning the importance of supersaturation suppression: We think that the importance is well stated in the paper, and in fact we have highlighted it as one of the most important factors that limit the effectiveness of sea spray geoengineering

Concerning thermodynamic consistency: Changes in aerosol do not feed back to clouds and meteorology in offline/chemical transport models like GLOMAP. Therefore, the effects of supersaturation suppression on LWP or other cloud properties cannot be studied with the current model but require either a cloud-resolving model or a climate model. Now that we have highlighted the issue others can follow up the details.

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To clarify this, we have added to the end of Methods section:

"It should be noted that in GLOMAP (as in all CTMs) changes in aerosol population do not feed back to model meteorology and cloud properties. However, the predicted aerosol fields can still be used together with information about typical marine boundary layer updrafts to calculate changes in CDNC at chosen altitudes"

(2) Comparison to published literature should be quantified explicitly: 1. On the last point [(1)-3.], it seems this is not entirely new, and there is possibly observational evidence to use as constraints from the group of the handling editor (e.g. Russell et al., J. Geophys. Res., 1999) showed reduced Sc for measured conditions and fixed updrafts. How are the reductions predicted in this model compared to what was found there?

Our results are in qualitative agreement with those of Russell et al. A meaningful quantitative comparison is very difficult due to different background aerosol distributions, additional particle concentrations (from ship plume or spray vessel) and updraught velocity distributions.

To better indicate that this is not a completely new finding, we have added to section 3.2:

"Cloud model studies have previously demonstrated this effect [suppression of supersaturation] for ship tracks (Russell et al., 1999) as well as sea spray geoengineering (Bower et al., 2006)"

2. There is a limited discussion of and comparison to Bower et al; it seems more is merited since opposing conclusions were drawn. Is the specific difference in the scaling to the global distribution, or is the CDNC prediction different? I feel very strongly that a quantitative comparison to the existing literature is essential to place this work in context.

We are not sure what the reviewer means by opposing conclusions. The Bower et al.

study concludes that (1) albedo change is very sensitive to cloud droplet change (we do not investigate albedo change in our study, but agree with their conclusion), (2) albedo change is greatest in clean and smallest in polluted environments (we conclude the same for relative cloud droplet number which to a large extent determines the albedo change), (3) calculated albedo changes exceed those necessary to compensate warming from doubling of CO2 (we do not predict albedo change or climate cooling in our study) and (4) their calculations provide quantitative support for the physical viability of the mitigation scheme (we do not discuss the viability of the scheme).

It is difficult to see how a quantitative comparison to Bower et al. could be done in a meaningful way. Bower et al. prescribe an aerosol distribution (background + spray) and use it in cloud model calculations with a specified updraft velocity. Thus, each of the values given in their study for change in cloud drop number is for a specific set of aerosol and cloud properties. On the other hand, we calculate the aerosol emissions and atmospheric processes explicitly. Depending on the emission flux, transport and microphysics, the predicted aerosol distribution from which the CDNC is calculated varies hugely between model grid cells (and can be very different from the prescribed ones in Bower et al.). A further complication is that we calculate our CDNC using a probability distribution for the updraught velocity, whereas Bower et al. use a single value in each of their sets. These factors make a direct comparison of the predicted CDNC in this and Bower's study impossible.

# 3. For global modeling results, we need a more specific quantitative comparison to Rasch and Jones models reported in Latham et al. (PhilTransRoySoc, 2008); this work is cited but they should be added to graphs and quantitative regional differences should be explored and explained.

The previous studies have investigated the radiative forcing and climate effects from sea spray geoengineering. However, there is a big difference between our studies that makes direct comparison meaningless: The other studies have simply assumed that it is possible to produce totally homogeneous cloud drop fields of very high CDNC con-

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centrations (375 cm-3 or even 1000 cm-3) over large areas of the oceans, while we have actually calculated the CDNC mechanistically. The quantities that they do calculate are radiative forcing and cooling potential, neither of which we have calculated. We do compare our CDNC with their fixed values in the text and explain the difference, but this is as far as any comparison can go.

(3) The discussion of the choice of sea spray choice should be more comprehensive, and the degree to which this choice is arbitrary should be made more explicit. The "5xGEO" does not seem to represent the spirit of the Latham proposal, as the particle generation is not to be done by increasing wind speed. In fact, the Rasch et al. work shows that a better approach is to target the areas with low wind speed, providing a disproportionately larger effect in those areas. If the main difference of this model is that the authors use the Salter design...

See reply to comment (1)-1.

To acknowledge that Salter's design is not the only possible one (although the only published one) we have added to section 2.2:

"It should be noted that while in theory it would be ideal to produce the highest spray fluxes in low wind speed conditions, in which the background aerosol concentration is likely to be low and therefore clouds more susceptible to modification, no concrete designs of wind-driven spray vessels capable of such spray generation have been proposed. In fact, the Salter et al. (2008) design is the only one published in scientific literature to date."

We don't think it makes sense studying hypothetical sensitivity experiments. The 5xGEO run is as far as it seems sensible to go at present.

(4) "Personal communications" should be explicitly approved by the person to whom they are attributed, when they cannot be avoided altogether. Can the authors verify that S. Salter has approved this interpretation of information he pro-

#### vided?

Since Salter et al. (2008), which is the only published article on the vessel design, did not contain information on the wind speed dependence of the spray rate, we obtained this information directly from Steven Salter (email communication 18.6.2008). Since submission, we have confirmed directly that he is happy for this information to be used in this study. We are happy to provide the emails to the editor, should he wish to see them.

We now give the date of Salter's original reply in the manuscript.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 735, 2010.

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