We thank Dr. Robert Wood and the anonymous referee for their valuable comments to the manuscript and their helpful suggestions for changes. Below, we explain how the comments and suggestions are addressed and make note of the changes we have made to the manuscript.

Referee #1: Robert Wood, University of Washington

Review of "Manipulating marine stratocumulus cloud amount and albedo: a process-modeling study of aerosol-cloud-precipitation interactions in response to injection of cloud condensation nuclei", by Wang, Rasch, and Feingold

Recommendation: Accept subject to some revision

Overview:
This paper uses large-domain large eddy simulations to explore the sensitivity of marine stratocumulus clouds, under two sets of meteorological conditions and three different background microphysical states, to injections of cloud condensation nuclei (CCN) from ships. The experiments are designed to test a widely-known geoengineering proposal to increase the planetary albedo by increasing the cloud shortwave reflectivity through injections of artificially generated sea-salt aerosol.

The study is very interesting and finds that only for some of the background cloud states (meteorological/microphysical) do the injections significantly increase the albedo. Clouds with the higher concentrations of background CCN typical of moderately polluted marine stratocumulus are barely susceptible to CCN injection. Clouds with low background CCN are typically more susceptible but only when the background state is precipitating. Indeed precipitation suppression appears to be a necessary condition for the CCN injection to increase cloud albedo, but too much drizzle is found to reduce the susceptibility somewhat. This dependence of albedo increase upon precipitation suppression is not solely because precipitating clouds are also those with low CCN and therefore are more susceptible in the Platnick and Twomey sense, since the dry meteorological conditions case has relatively low CCN (55 cm-3 in the unperturbed case) that one would expect to be susceptible.
The results are intriguing and certainly worthy of publication in Atmospheric Chemistry and Physics. The manuscript is well-written and the figures very clear and instructive. The study left me with more questions than answers which is not a criticism in this case. I hope the authors can continue their work in this area. I have some specific suggestions and thoughts that the authors might wish to consider, as detailed below.

Main points:

1. I think Table 2 should include a column with the mean cloud cover as well as the other albedo controlling variables LWP and Nd. Given the rather large increases of Nd in the dry case (55 to 85 cm-3), and given that there is very little LWP reduction, I am surprised that the albedo change is only 0.01, so can one assume that the cloud cover change is somehow compensating for the Nd change?
It would also be useful to break down the overall albedo increase into a fractional part due to changing Nd, a part due to changing cloud LWP and a part due to changes in cloud fractional coverage. Simple formulae could be used to estimate these contributions.

LWPs in the table are domain-averages, which implicitly reflect both in-cloud liquid water path and cloud fractional coverage; however, to facilitate more in-depth offline comparison by readers, cloud fraction has been added to Table 2 as suggested.

In the W200 and D100 cases, CCN injection didn't change the domain cloud fraction. The enhancement in overall cloud albedo due to an increase in drop number was weakened by a reduction in cloud water path. In this study cloud albedo is calculated offline as a function of cloud optical depth τ using $\alpha \approx \frac{(1-g)\tau}{2+(1-g)\tau}$, where the asymmetry parameter g is approximately 0.85 for warm clouds. Given the log-normal drop spectrum defined by *n(r)* for drop radius *r* in a range of (r_{min}, r_{max}), visible-band τ in between z_{min} and z_{max} was explicitly calculated in the model using $\tau = \int_{z_{min}}^{z_{max}} \int_{r_{min}}^{r_{max}} 2\pi r^2 n(r) dr dz$ for each grid. Assuming a mono-disperse cloud spectrum and constant liquid water path, the conventional theory (i.e., Twomey effect) gives an enhancement of cloud albedo $\Delta\alpha = \frac{\alpha(1-\alpha)}{3N_d}\Delta N_d$, which is about 0.025 in the D100 seeding experiments and 0.03 in the W200 experiments. These values are larger than the directly calculated albedo enhancements (0.01 and 0.02) which did not require the assumption of constant LWP and size distribution. Compared to the base cases, although the domain- and temporal average LWP in the seeding experiments decreased only a little, the localized decrease of LWP with increasing N_d was much stronger. For example, in the W200 case (Fig.7), the increase in N_d slows down after t=12h, but LWP keeps decreasing to about 15 g m^{-2} at t=30h. Therefore, the overall average change in cloud albedo, drop number and LWP does not reflect the nonlinear relationships among the three quantities in instant cloud fields. Further investigation may warrant a more in-depth physical explanation.

Cloud drop number, cloud fraction and liquid water path increase in the W50 and W100 cases, which causes a significant increase in cloud albedo. There are a number of studies in the literature that have focused on breaking down the contribution to cloud albedo change by drop number, LWP and other factors. In fact, we are working on a companion paper focusing on this subject using the concept of cloud albedo susceptibility.

2. Given that Ackerman et al. (2009) found that entrainment (and thus presumably feedbacks on LWP through exchange with the free-troposphere) was more sensitive to cloud droplet sedimentation than to drizzle in the RF02 case, why have the authors not mentioned sedimentation or its potential role?

Cloud water sedimentation and drizzle are the focus of the intercomparison study by Ackerman et al. (2009). They found that LWP responds more strongly to cloud water sedimentation than to drizzle through the entrainment process. In our original development of this parameterization (Feingold et al. 1998) we already showed that neglect of cloud droplet sedimentation degraded the simulations compared to the bin treatment that included sedimentation. Therefore cloud droplet sedimentation is explicitly represented in our model. In the present study, drizzle initiates significant changes in cloud cellular structures, mesoscale dynamics and cloud fraction, which is one of the major differences compared to that in Ackerman et al. (2009). We believe that these changes in cloud morphology have a much stronger impact on entrainment and LWP than does cloud droplet sedimentation.

3. Does exchange with the FT matter for the evolution of the clouds in any of the simulations?
That is, is the FT moisture a relevant variable controlling the sensitivity of albedo to CCN injection?

Previous studies (e.g., Ackerman et al. 2004) did find that cloud water and its response to changes in aerosol are sensitive to the humidity of free-tropospheric air. In the two cases considered in this study, the free-tropospheric air is much drier and warmer than the boundary-layer air, and one of the soundings is drier/warmer than the other. The exchange of air does impact the moisture and temperature in the upper boundary-layer where clouds usually form. And because the soundings differ, there is some variability in the effects. However, we didn't modify each of the observed soundings (i.e., changing the FT moisture) to systematically explore this effect. The focus of this study is on aerosol number concentration, precipitation and seeding strategy. Nonetheless, this is a good point to be tested in future process-modeling studies.

4. What justifies the classification of the two meteorological regimes as DRY and WET? Can the authors state what it is about the simulations that warrants this?

As briefly described in the paper, the WET and DRY cases were observed in the precipitating, more moist atmosphere and non-precipitating, drier atmosphere, respectively, in the same geographical regime. In this sense, WET and DRY are relative to each other. We took the measured initial profiles to drive the precipitating and non-precipitating cases. On the other hand, clouds observed/simulated in the former case (WET) have larger cloud water contend that in the latter (DRY) case. We now make it clearer in the manuscript.

5. Fig 1: The simulation W50P3 has a strange wraparound effect whereby close to the injection location on the left of the panels, the CCN appears to be high in the cloud-free regions. Is this an artifact of having the ship essentially perturb the clouds there twice? Does this affect the results in terms of albedo increases?

This is a very interesting point. As briefly mentioned in the manuscript, it's the result of dynamical interactions between the plumes and not because of a repeat in seeding. After precipitation developed sufficiently behind the plume source due to a substantial increase of cloud water caused by mesoscale circulations (Wang and Feingold 2009b), outflows from the two adjacent plumes collide to form new convergent lines as indicated by the bright clouds in-between plumes. However, the injected high-concentration CCN are now in the cloud-free regions where lower-level water vapor source has been cut-off by the circulations. Hence, the shift of cloud lines is not because of perturbing the clouds twice which could have prevented the plume-tail precipitation from developing in the first place (like in the W100P3 case) if the timing were right and/or perturbation were strong enough. We believe that for this snapshot the overall cloud albedo would be higher if the high-concentration CCN were co-located with the bright clouds. However, we do see that in the next seeding cycle the CCN perturbation is right in the lines of clouds, again, because of the changing dynamics. It's unlikely that one would be able to predict the optimum location to inject aerosols to achieve the best albedo enhancement after a few cycles of interactions.

6. Are the Nd and LWP values in Table 1 and discussed in the manuscript means for cloudy regions only or over the entire domain? If so, wouldn't it be more physically instructive to discuss LWP/Nd for the cloudy regions only and cloud fraction separately?

Yes, they are domain averages. We agree that it's more instructive to discuss cloud properties in cloudy regions only; however, the purpose of this study is to assess domain-average changes in albedo and therefore we focus on domain-average N_d and LWP. We feel that adding too much focus on cloud-average properties might be a bit confusing. For example, in the strongly precipitating cases (e.g., the W50 series), cloud-average LWP in the unseeded base case is even larger than that in the seeded cases, as is the cloud-average albedo. Nonetheless, in response to this comment as well as comment #1, we do add an additional column of cloud fraction to Table 2 and add some discussion in the manuscript.

7. Does the fact that the unperturbed states are changing so rapidly (i.e. are far from equilibrium) have any impact on the results? This is especially the case in the RF02 set-up where the observed state was rapidly transitioning from closed to open cells. Figure 7 shows this rapid decrease.

None of the cases simulated here reaches an equilibrium state. First of all, the diurnally varying solar radiation drives a diurnal cycle in cloud properties. Secondly, radiative cooling and entrainment mixing are sensitive to LWP. In the precipitating cases, the change in cellular structure and cloud fraction (i.e., transition from closed to open cells) largely determines the change in LWP and therefore the radiative process as well as consequent feedbacks. In the non-precipitating cases, the continuous entrainment of dry air steadily reduces LWP. Therefore we doubt that there is an equilibrium state for the semi-idealized simulations (i.e., with fixed large-scale forcing but interactive resolved processes). Nonetheless, even without reaching equilibrium, compared to the corresponding base case, the response of the cloud systems to different CCN injections is temporally consistent, indicating that the results are unambiguously robust.

8. Can clouds which have precipitation at cloud base but not at the surface be successfully perturbed this way? The simulations performed here seem rather extreme ends of the spectrum (1.27 and 1.87 mm/day at the surface for the W50 and W100 cases, and zero for the other two cases). What about for clouds somewhere in the middle?

This is a very good question. The numbers provided in Table 2 are spatial averages over the entire domain and temporal averages over a diurnal cycle (24 hours). Local surface rain rates range virtually from 0 to 40 mm day^{-1} in the W50 case. During the course of 30-h simulations, domain-averages rain rates are in the range of 0~2.2 mm day^{-1} (see Figures 5d and 6d). As we argued in the response to comment #7, the basic message is clear and consistent at all values in the range, not just the two mean values.

Anonymous Referee #2

Review of "Manipulating marine stratocumulus cloud amount and albedo: a process modeling study of aerosol-cloud-precipitation interactions in response to injection of cloud condensation nuclei" by Wang et al.

General comments

This manuscript presents cloud resolving model simulations that are designed to test the efficacy of a proposed geoengineering solution, which aims to use surface based sprayers to modulate cloud albedo through the injection of sea-salt aerosol into marine stratocumulus. Sensitivity studies are performed with different sprayer configurations, background aerosol concentrations and meteorological regimes. The results illustrate that significant changes to the cloud albedo only occur under certain conditions, namely in weakly precipitating boundary layers where the injection of additional cloud condensation nuclei (CCN) aerosol are able to reduce drizzle production in the cloud, and in CCN limited environments (for example after washout from heavy precipitation), where additional CCN are required to sustain the cloud layer. The paper is interesting, well structured and certainly addresses scientific questions that are relevant to the journal. I would therefore recommend publication in ACP once the authors have considered the following comments.

Specific comments

1. In the introduction can the authors highlight the key differences (advances) between the aims of this study and the ship-track simulations of Wang and Feingold (2009b).

The basic modeling tools are the same, but the scientific goals of the two papers are different, so there are some differences between the CCN-injection simulations in this study and ship-track simulations of Wang and Feingold (2009b; hereafter WF09b). To name a few:

1) Based on the findings of WF09b, we designed the various simulations with different injection methods (single vs. multiple sprayers), meteorological scenarios (WET vs. DRY; precipitating vs. non-precipitating), and aerosol background (clean vs. polluted) in the present study.
2) The injection rate in this study is based on the rate proposed by Salter et al. (2008) to implement the geoengineering idea, which is larger than ship emission rate.
3) The diurnal variation of solar radiation is included.

Thanks for raising this good point. We have highlighted them in the introduction in the revised manuscript.

2. Additional information on the model set-up would be useful in section 2. Mean thermodynamic and wind profiles would be of interest for example. I noted that the sprayer tracks don't look like they are advected with the wind in the simulations (Fig. 1). Is this because the domain is orientated with the wind direction or that the boundary layer winds are too weak to advect the aerosol? It is also worth mentioning that the diurnal cycle of solar radiation is included in the simulations - I didn't realise this until I got to section 3.2.

Those soundings are from the standard GEWEX Cloud System Study (GCSS) Boundary-layer Cloud stratocumulus case studies. They are available online at http://www.knmi.nl/~siebesma/BLCWG/, and also in the model intercomparison studies

(Stevens et al. 2005; Ackerman et al. 2008) that are referred to in the manuscript. Essentials of the profiles that might be needed to interpret results have been described in the paper. We decided not to add an additional figure. Nonetheless, we make this clearer in the paper now.

Yes, the mean winds are subtracted from the initial soundings so that the tracks are not significantly advected in the domain. Or, one can imagine that the domain itself is drifting with the mean winds. By doing so, numerical error associated with advection is removed. It also helps identification of the sprayer tracks.

The diurnal cycle of solar radiation is now mentioned in the introduction.

3. It is mentioned that the injection rate of particles from the single surface based sprayer is 10 times lower than that proposed by Salter et al. (2008). The justification for this is that the model domain is a factor of 10 times smaller than the horizontal scales that Salter et al. (2008) propose a single sprayer would target. Does this have any implications for the aerosol transport into cloud i.e. if the modelled surface based sprayers had a much higher injection rate, how different would the results of the simulations be, particularly for the precipitating cases? Would the change in albedo be much larger for example?

Although the one-sprayer cases have a 10 times lower injection rate, the domain-average injection rate used in the uniformly seeded cases is the same as that proposed by Salter et al. (2008) who assumed that the injection can be effectively distributed in the domain. We believe that the much higher local injection rate will enhance local cloud albedo but that it won't impact our main point, which is that seeding may have unpredictable consequences that depend on the meteorological and background CCN conditions. In the precipitating cases, the current local CCN injection (with one sprayer at a 10 times lower rate) has already stopped precipitation effectively, so more CCN would not help in terms of reducing local rain. The circulations are still going to develop and transport the injected CCN. For the non-precipitating cases, the domain-wide impact may be even smaller if LWP still keep the decreasing trend with increasing CCN number concentration. As has been done already in the paper a 3-time stronger injection (W200-P3x3, compared to W200-P3), LWP is further reduced. The additional increase in cloud albedo averaged over a 3-time larger domain is negligibly small.

4. How do you assign a "wet" and "dry" definition to each simulation? For example, the W200 series is non-precipitating but is assigned as "wet".

The "wet" and "dry" refer to humidity (or water vapor mixing ratio) of air characterized by the initial profiles observed on the two research flights, RF02 and RF01, respectively. Also, the LWP observed/modeled for RF02 cloud is much higher than for RF01 (170 vs. 60 g m^{-2}). Even clouds in the W200 series do not precipitate, the simulations started with the same sounding and LWP.

Please also see the response to comment #4 of Referee #1. We now make this clearer in the manuscript.

5. When introducing the simulations in section 3 it would be useful if you can refer to tables 1 and 2.

Done.

6. The vertical distribution of injected particles in the W50-P3 case looks less uniformly mixed than in the W50-P1 case close to the sprayers (0-20 km in the x direction if Fig 1 c). This is presumably driven by the complex mesoscale circulations that exist between the plumes. Does this have implications for the optimum separation distance between sprayers?

This is an excellent point. Yes, it is due to the different mesoscale circulations in the two cases. In the W50-P3 case, interactions between adjacent plumes have changed the circulations when sprayers reach that location. The convergent flow that imposes an organized strong lifting on the injected CCN is no longer along the sprayer track. Otherwise, vertical transport has to rely on random large eddies. This does imply that the separation distance between sprayers matters for vertical transport in the precipitating regime; however, because the marine boundary layer is rather shallow, horizontal transport is more of a challenge.

Please also see the response to comment #5 of Referee #1.

7. Is figure 2 representative of one point in the x-direction or averaged over all grid points in the x-direction?

It's averaged along the x-direction; same for Figures 3 and 4. It is now clarified in the figure caption.

8. In the caption for Fig. 3 I suggest changing "in-cloud CCN number" to "in-cloud unactivated CCN number".

Done.

9. The importance of the diurnal cycle is discussed in section 3.2. Does the time of the simulations correspond to local time, such that the simulations begin at night-time when a more well-mixed sub-cloud layer and higher cloud LWP and precipitation rate would be expected? Would there be any significant changes to the simulations if the sprayers were instead initiated in the day-time?

Yes, the simulations started at local mid-night (00 h) so that cloud water can build up to the observed values in a reasonable length of spin up. About the timing of seeding, we briefly discussed this in the manuscript. For W100, seeding prior to nighttime thickening of Sc clouds may be the most effective strategy because it could prevent an overnight shift to open-cellular structure and help sustain solid Sc through the subsequent daytime. After precipitation starts and LWP is significantly reduced (see Fig. 6), seeding would not be as effective. For W50, early morning seeding to recharge the ultra-clean boundary layer would appear to be most effective in terms of albedo enhancement per unit of injection. The most effective timing will clearly be case-dependent, and evaluation of this aspect will be deferred to later study. To answer the second question from a different angle, if the total amount of injected CCN is not limited, the earlier and longer duration of seeding, the more effective it would be at preventing the clouds from raining out. As seen in the figures, cloud drop number concentration is higher in all seeded cases from the very beginning.

10. In the control simulations e.g. green line in Fig. 5 there is a rapid decrease in cloud fraction and LWP in the first 6h of the simulation. Is this because the simulations are not in equilibrium? Note that the

cloud cover and LWP does not recover the following night in the control run. Does this have any implications for the results and would it have been better to initiate the sprayers after this initial period?

The rapid decrease in cloud fraction and LWP is due to precipitation (see Fig. 6d) and the consequent transition from closed-cell structure to open-cell structure. Please also see the response to comment #7 of Referee #1 about why the simulations did not reach equilibrium, and see the response to comment #9 (right above) about the effective timing of seeding.

11. The boundary layer depth in the simulations is shallower than that observed in some other marine Sc regions (see observations in Abel et al. (2010) and Bretherton et al. (2010) in the ACP VOCALS-REx special issue for example). Do the authors expect a deeper marine boundary layer to impact the efficiency of surface based sprayers?

This is an excellent point. The two soundings used in this study are just examples from the Pacific northeast Sc regime, and by no means do we claim that they are typical. Neither do we claim generality of our results. We expect that the decoupling of precipitating boundary layer could have a more profound impact on the transport of CCN sprayed from the surface. The evaluation of this aspect will be deferred to later study, but we now briefly mention it in closing paragraph of the paper.