Response to referees

We thank both referees for their thoughtful comments on our manuscript. Please see our comments below. To guide the reader, we have italicised the comments by the referees.

Referee #1:
This manuscript investigates aerosol coagulation in the very early plume development and discusses the resulting implications on geoengineering by marine cloud brightening. The analysis is based on model simulations. The paper is well organized and scientifically sound. The drawbacks of the paper are that it is rather technical and probably not of very broad scientific interest. On the other hand, the results from the analysis are important for marine geoengineering applications and therefore worth to be published. The paper would benefit from discussing shortly whether the obtained results have any implications beyond geoengineering. For example, are there other potential applications for the derived simple parameterization of in-plume coagulation? Other than that, I cannot pinpoint any issues that would require further actions in the paper.

Response: We note that the derived parameterization can be applied generally to any point-source emission of aerosol, such as small combustion sources, given that the assumptions of the Gaussian plume model remain valid. Most notably, the concentrations of aerosol within the plume must be much greater than the background aerosol concentrations and the concentrations of condensing species must be low enough that condensational growth does not significantly affect coagulation rates. We have added comments throughout the manuscript to reflect the general applicability of the parameterization.

Referee #2:

The authors report their work exploring the role of aerosol Brownian coagulation in reducing the efficacy of marine cloud brightening geoengineering by significantly reducing number concentration of sea salt particles in the intentional sea-spray injections, based on a Gaussian plume model and a fine-resolution large-eddy simulation model with detailed aerosol treatments. They also developed a parameterization scheme to account for this effect in global aerosol-climate models. The results should be of interest to ACP readers, particularly, those from the geoengineering community. The manuscript is well written and in general the scientific methods with assumptions/limitations are clearly outlined. I recommend for publication in ACP after the following specific comments be addressed upon revision.

1) The particles were injected to an arbitrarily chosen 10-m deep box. How sensitive is the reduction to this assumption? It would be nice to also plot the size/volume evolution of the plume on Fig.2

Response: There is no gradient in the aerosol concentrations across the box depth, and width and height of the box do not vary across its depth. The coagulation rates, and thus the reduction in particle number, are insensitive to the assumption of box depth. The box depth is used only to calculate total particle number from the aerosol concentrations. This is now stated more clearly in the manuscript. We have added an extra panel to Fig. 2 (Fig. 3 in the updated manuscript, shown as Fig. A below) as suggested by the referee.

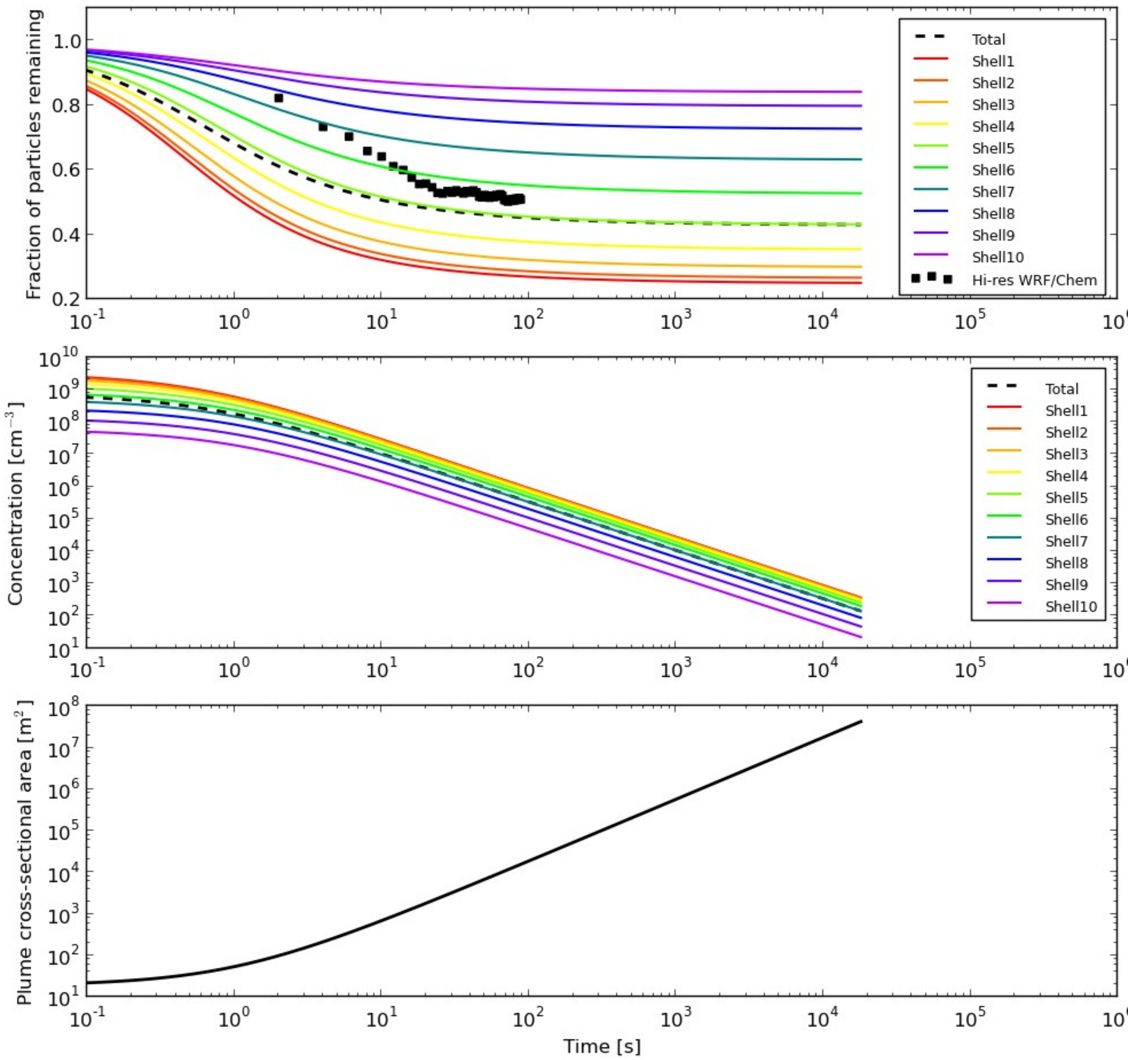

Figure A: The fraction of particles remaining, the concentrations, and the plume cross-sectional area are shown over time for each shell for the base case. The black dashed line shows the average across the plume, and the black square points show high resolution results from the WRF/Chem large-eddy simulation model.

Referee #2:
2) It is not so clear to me how realistically the injection was done to the LES domain and in the plume. In the LES model, particles were introduced to the occupied grid boxes continuously (between model time steps) at a rate equivalent to the 30 kg/s mass flux. The crosswind was assumed to be 6m/s. Have you considered the moving speed of the ship? With the max domain dimension of 120 m, the source would move out of the model domain in a few seconds. Were the particles introduced to the plume model continuously too?

Response: The crosswind speed would indeed be a combination of both the wind speed and the vessel speed. This is indicated in Figure B, where it is noted that as suggested in Salter et al. (2008), vessel motion may be perpendicular to the prevailing wind direction. Given that the wind speed would vary, and that both the direction and speed of the vessel in response to this wind is currently not clearly defined, the 6 m/s crosswind speed was chosen as a representative speed. A sensitivity simulation with crosswind speed of 9 m/s was also performed. While coagulation reduced the aerosol number concentration by 47% in the 6 m/s crosswind simulation, it reduced the aerosol number concentration by 39% in the 9 m/s crosswind simulation as the aerosols were more dispersed under this higher crosswind.

We agree that the current wording ("The crosswind was initialized with a 10m wind speed of 6 ms^{-1}") is confusing and we changed it to "The 10 m crosswind (assumed to comprise of both ambient wind and ship velocity components) was initialized at 6 ms^{-1}".

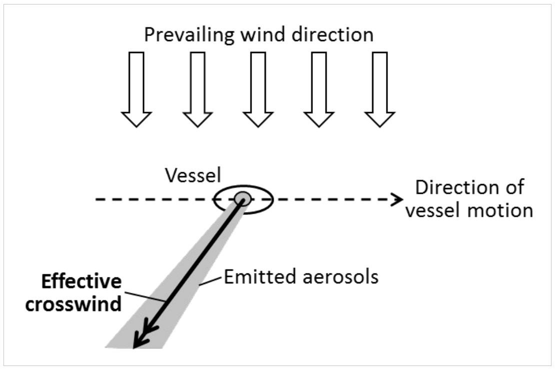

Figure B: Effective crosswind as a combination of the wind and vessel motion

It is also correct that the aerosols move out of the LES domain over a number of seconds. Figure C shows the development of the plume over time. During the first ~16 seconds, the crosswind causes the aerosol plume to extend towards the end of the domain (upper panels), increasing both the domain average aerosol number concentration and aerosol mass (lower panels). After this time, the plume now reaches the end of the domain, and an approximate steady-state condition is reached whereby the mass of aerosols being emitted into the domain approximately equals the mass of aerosols leaving the domain.

Within the Gaussian-plume model, the particles are not introduced continuously. All of the particles are instantaneously added to the shells at the beginning of the simulation. We agree with the referee that the current wording is unclear ("Particles are added to the plume at the beginning of the simulation corresponding to the number of particles that would be emitted into a 10-meter-deep box (and the width of the stack diameter) if the box is traveling at the same speed as the wind.") and we have changed it to "The plume is initialized at the beginning of the simulation with the total number of particles that would be emitted into a box that is 10 meters deep (and the width of the stack diameter) if the box is traveling at the same speed as the wind."

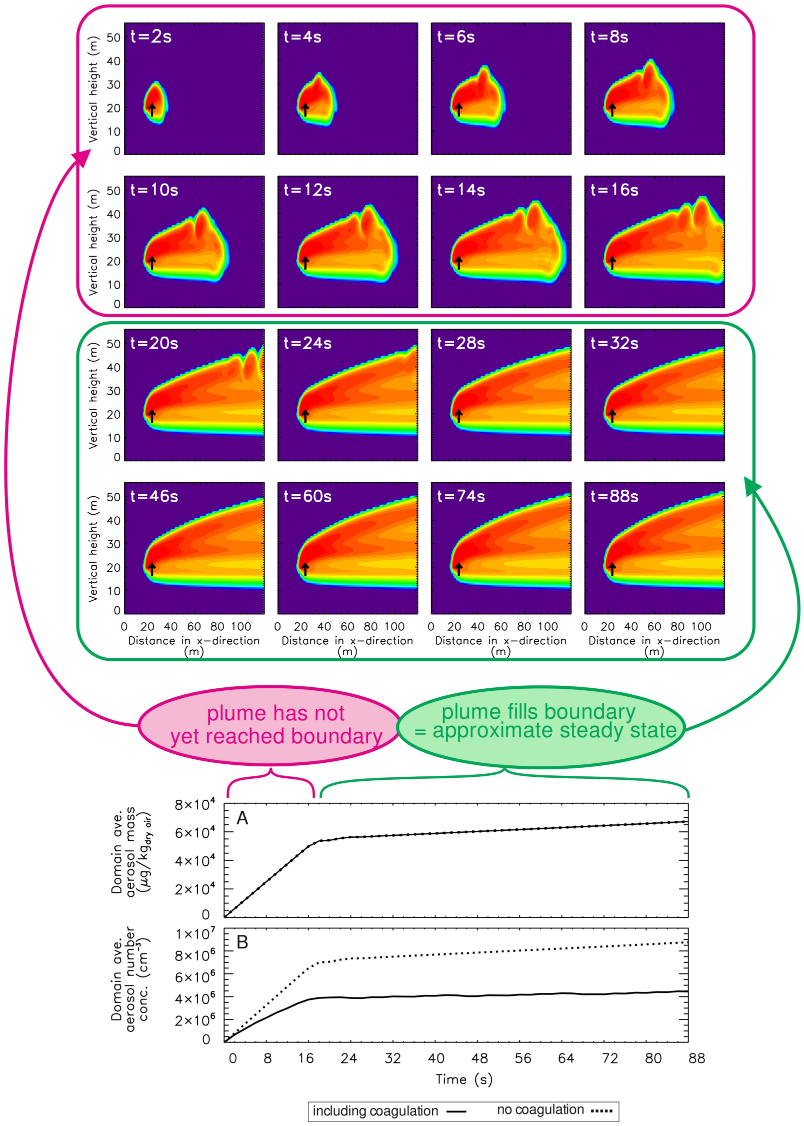

FIGURE C:

Upper panels – Aerosol concentrations (averaged over the 'into page'-direction) through time. Crosswind flows from left to right. Black solid arrows indicate the location of the aerosol emission.

Lower panels – Time series for: (a) domain average aerosol mass ($\mu g/kg_{dry\ air}$); (b) domain average aerosol number concentration (cm^{-3}).

Referee #2:

3) One technical concern is about running the WRF/Chem model as such a high spatial and temporal resolution. Is this justifiable? How was the turbulence developed in seconds? It would more convincible to show the key dynamical features produced in the simulation that described in the text (on page 18686, the first paragraph).

Response: The results of the LES modelling were intended to supplement the Gaussian plume modelling that is taken forward in developing the in-plume aerosol coagulation parameterization. Thus, in order to maintain the flow of the manuscript, a comprehensive presentation of these auxiliary LES modelling results was not included. However, in view of your comments, it is clear that further justification of the model use in this context is necessary.

With regards to the development of turbulence within the model, the high resolutions that are needed to capture both the high aerosol concentrations and dynamics within the plume necessitate the limited domain size. Because of this limited domain size, the LES simulation captures the aerosol plume alone, independent of boundary layer turbulence and dynamics. Thus, the development of boundary layer turbulence is not necessary within this simulation. It is assumed that the omission of marine boundary interactions here will not be significant as the vertical velocity of the aerosols upon emission (12 ms-1) is higher than typical up and downdraft velocities within the marine stratocumulus deck (< 1m/s (Pringle et al., 2012)). Therefore, for this limited domain region (close to the aerosol emission point) the dynamics resulting from this upward flow velocity would dominate over marine boundary layer turbulence. This point is now clarified in the text.

In terms of the plume features created within the WRF/Chem model at these resolutions, several characteristic properties have been compared with understanding of the analogous jet-in-cross-flow process. As seen in the upper panels of Figure C the interaction of the upward flow (at 12 ms^{-1}) and the crosswind (at 6 ms^{-1}) causes the plume to develop rapidly. The shape of a typical simulated plume is illustrated in Figure D. Analysis of the trajectory of the plumes agrees reasonably with empirical data for observed jets-in-cross flows (Margason, 1993, Muppidi and Mahesh, 2005). Additionally, the interaction of the upward and crosswind flows produce a counter rotating vortex pair (Figure D) which is characteristic of a jet-in-cross flow. The distributions of the aerosols through these counter rotating vortices (including the effects of coagulation) are shown in Figure E. As expected for aerosols of this size, they are preferentially located at the centres of these vortices. The high concentrations in these locations results in higher rates of coagulation. This produces the higher concentrations of larger aerosols located within the counter rotating vortices, particularly further from the aerosol emission source. These model outputs indicate that the interaction of upward and crosswind flows, and the distribution of the aerosols within these flows simulated by the LES model match with expectations based on current knowledge.

We agree that the manuscript benefits from further justification of the use of the LES modelling, and illustration of its output. As such, we include the illustrative output shown in Figure D in the revised manuscript as Figure 2 in order to orientate the reader to the LES modelling output. Additionally, further descriptions of the outputs are included in the text.

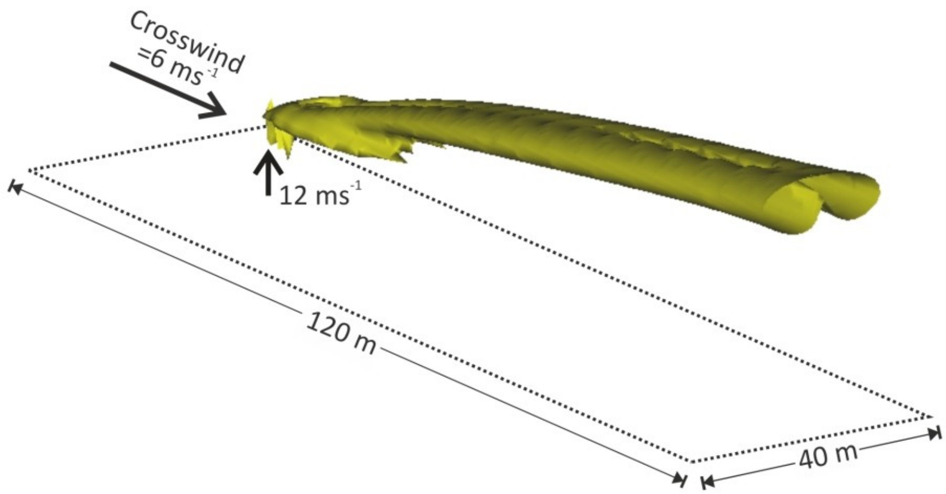

FIGURE D:
Instantaneous isosurface for a given aerosol concentration, illustrating the typical structure of a simulated JICF in the absence of a rotor during the approximately steady-state phase. (The case shown is a simulation used during trials of the methodology)

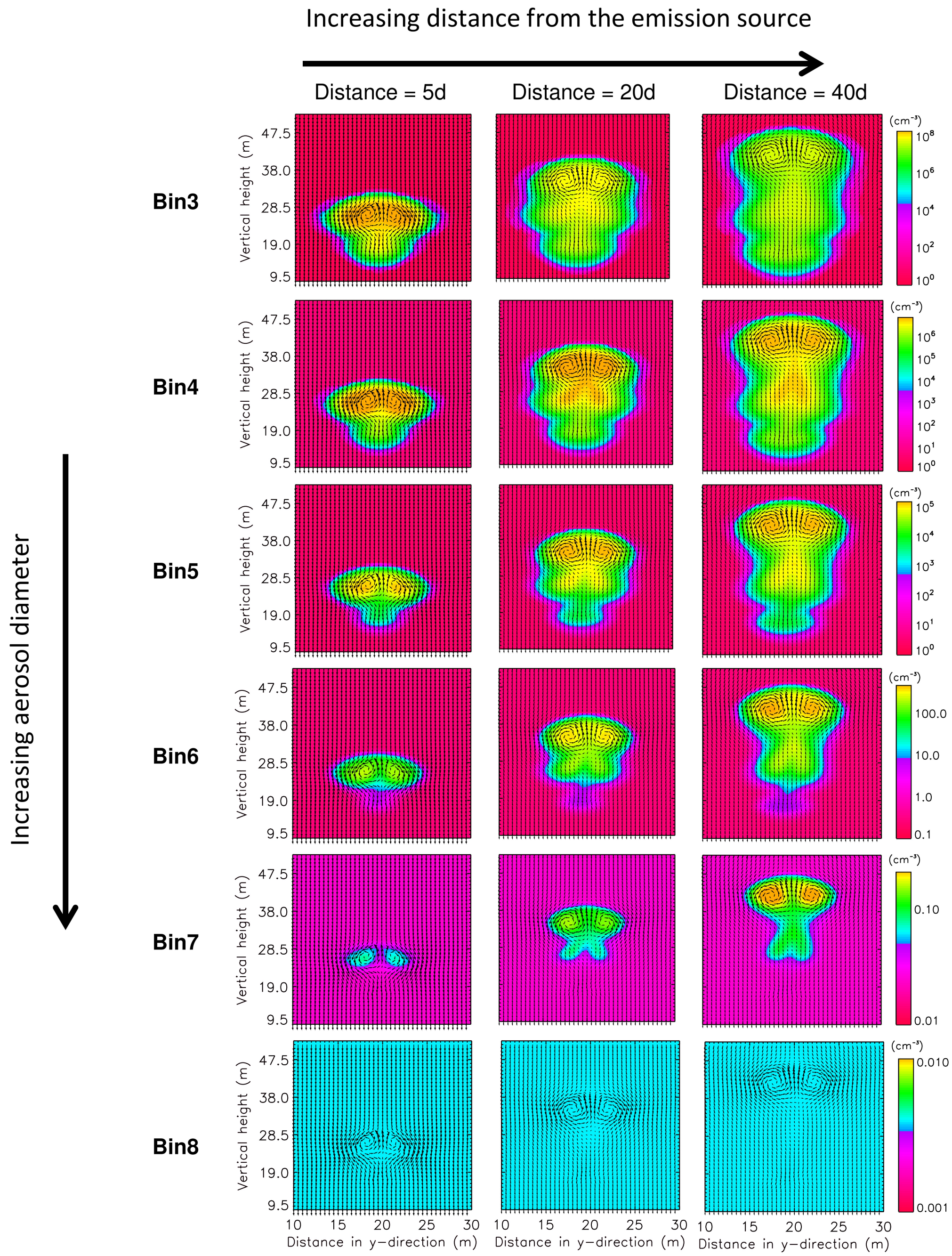

FIGURE E:
Sections of planes perpendicular to the trajectory at increasing distances from the aerosol emission source (at 5d, 20d and 40d, where d=diameter of the rotor) showing the distribution of Bin 3 to Bin 8 aerosol concentrations (cm^{-3}) for the simulation which includes coagulation. Aerosol concentrations are overlaid by arrows showing velocities of the flow. All show 44 seconds from simulation start. Note the different contour colour ranges. Aerosols are emitted into Bin 3.

Referee #2:
4) The calculations were based on dry particle sizes, but sea spray particles are water droplets (large sizes and different coagulation efficient) upon injection. The sizes will decrease as evaporation occurs, which will also induce cooling and change to the kinematics of flow. Without considering these effects in the calculations, the conclusions of the paper become less relevant to the sea-spray injections. It should be made very clear in the paper.

Response: While the parameterization takes aerosol dry diameter as input, the Gaussian-plume model calculations were performed using the wet diameters assuming 80% relative humidity. We have attempted to make this more clear in the text. We state in Sect. 2.1 of the manuscript that the assumption that wet aerosol and air instantly reach equilibrium at 80% relative humidity will result in uncertainties due to possible errors in the size of the aerosol and possible effects on the dynamics of the plume. We further discuss these uncertainties in Sect. 5, where we state that because our results were not strongly dependent on the inital particle size (the difference in the fraction of particles remaining between an initial dry diameter of 200 nm and 400 nm was on the order of 0.05 or less), we do not expect that errors in the wet diameter would strongly affect our results. We also note that "Jenkins and Forster (2013) found that including water with the emitted aerosols [...] led to evaporation and reduced buoyancy within the plume. This caused a reduced vertical plume height but increased horizontal dispersion. As such, the particle concentrations within the plume were not significantly affected, suggesting that this effect would not significantly alter [the fraction of particles remaining after coagulation]." We have added the following to our conclusions:

"We do not include the decrease in the wet diameter of the particles during transport due to evaporation or effects on the dynamics of the plume due to evaporative cooling. However, the results of the model are not strongly dependent on the wet diameter of the particles, and the results of Jenkins and Forster (2013) suggest that the effects of evaporative cooling on the dynamics of the plume would not strongly affect our results."

Referee #2:
Technical edits:
1) P18685, line16: it doesn't make much sense to call it "longitudinal boundary" for such a small model domain.
Response: The term 'longitudinal' is removed, as suggested.

Referee #2: *2) p18686: the Mahesh (2013) reference is missing.*
Response: The missing reference has been included.

Referee #2: *3) P18690, line5: no need to spell out LES again here*
Response: The words "Large Eddy Simulation" have been removed.

Referee #2: *4) P18694, equation (6): needs improvement. This is for number flux Fn, not F? The "1 m/s" inside the expression is rather confusing. The equation can be much simplified, for example, by using "min(7, u)" for wind speed.*
Response: The equation is indeed for number flux F_n, not fraction of particles remaining F. We have edited the equation as suggested.

Referee #2: *5) P18695, line15: misspelled "CDNC"*
Response: The misspelling has been corrected.

Referee #2: *6) P18695, line16: using "non-zero" is better than "positive" here*
Response: "positive" has been changed to "non-zero", as suggested.

REFERENCES

Jenkins, A. K. L. and Forster, P. M.: The inclusion of water with the injected aerosol reduces the simulated effectiveness of marine cloud brightening, accepted by Atmospheric Science Letters, 2013.

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Salter, S., Sortino, G. & Latham, J. 2008. Sea-going hardware for the cloud albedo method of reversing global warming. *Philosophical Transactions of the Royal Society A: Mathematical, Physical and Engineering Sciences,* 366**,** 3989-4006.