

# Enhancement of marine cloud albedo via controlled sea spray injections: a global model study of the influence of emission rates, microphysics and transport

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**Abstract.** Modification of cloud albedo by controlled emission of sea spray particles into the atmosphere has been suggested as a possible geoengineering option to slow global warming. Previous global studies have imposed changes in cloud drop concentration in low level clouds to explore the radiative and climatic effects. Here, we use a global aerosol transport model to quantify how an imposed flux of sea spray particles affects the natural aerosol processes, the particle size distribution, and concentrations of cloud drops. We assume that the proposed fleet of vessels emits sea spray particles with a wind speed-dependent flux into four regions of persistent stratocumulus cloud off the western coasts of continents. The model results show that fractional changes in cloud drop number concentration (CDNC) vary substantially between the four regions because of differences in wind speed (which affects the spray efficiency of the vessels), transport and particle deposition rates, and because of variations in aerosols from natural and anthropogenic sources. Using spray emission rates comparable to those implied by previous studies we find that the predicted CDNC changes are very small (maximum 20%) and in one of the four regions even negative. The weak or negative effect is because the added particles suppress the in-cloud supersaturation and prevent existing aerosol particles from forming cloud drops. A scenario with five times higher emissions (considerably higher than previously assumed) increases CDNC on average by 45–163%, but median concentrations are still below the  $375\text{ cm}^{-3}$  assumed in previous studies. An inadvertent effect of the spray emissions is that sulphur dioxide concen-

trations are suppressed by 1–2% in the seeded regions and sulphuric acid vapour by 64–68% due to chemical reactions on the additional salt particles. The impact of this suppression on existing aerosol is negligible in the model, but should be investigated further in the real environment so that inadvertent impacts can be excluded.

## 1 Introduction

Anthropogenic emissions of greenhouse gases have increased in the last decade faster than ever before (Global Carbon Project, 2008; Raupach et al., 2007). Since no quick reversal for this trend is in sight, several alternative methods to slow down global warming by deliberate manipulation of the climate have been suggested in recent years. These proposed methods, referred to as geoengineering, cover a wide range of technologies aiming to reduce either 1) the atmospheric  $\text{CO}_2$  concentration, e.g. by air capture methods (Bacocchi et al., 2006; Mahmoudkhani and Keith, 2009) or by enhancing the oceanic carbon storage (Khesghi, 1995), or 2) the amount of solar radiation absorbed in the atmosphere and the Earth's surface, e.g. by adding sulphate aerosol to stratosphere (Crutzen, 2006) or by increasing the albedo of the land surface (Akbari et al., 2009; Rigdwell et al., 2009).

In the light of current knowledge, one of the most promising geoengineering methods in terms of effectiveness and affordability is increasing the albedo of low-level marine clouds via controlled injection of sea spray particles into the atmosphere (The Royal Society, 2009; Lenton and Vaughan, 2009; Boyd, 2008). In this method, first proposed by Latham (1990), the aim is to build a fleet of unmanned,



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wind-powered vessels, which could be remotely steered beneath marine clouds and emit sea water droplets into the air at a very high rate (Salter et al., 2008). A significant fraction of the released particles are expected to be transported to cloud altitude where the particles would act as cloud condensation nuclei (CCN) and thus increase the cloud droplet number concentration (CDNC) compared to an unperturbed situation. The increased CDNC would likely lead to an increased cloud albedo since the natural CDNC in remote marine stratocumulus clouds is typically fairly low ( $<150\text{ cm}^{-3}$ ).

Previous climate model studies have suggested that the sea spray geoengineering method could have a significant climate cooling potential. Assuming that all low-level (below 700 hPa) maritime clouds could be seeded with the spray vessels, Latham et al. (2008) predicted a negative cloud forcing large enough to compensate for the positive forcing from doubling the atmospheric  $\text{CO}_2$  concentration. In another study, again using imposed changes in cloud drop concentrations, Jones et al. (2009) limited geoengineering to three persistent stratocumulus regions covering 3.3% of the Earth's surface and found that cloud seeding could offset up to 35% of current greenhouse gas forcing and delay global warming by about 25 years. Rasch et al. (2009) concluded that cloud seeding could restore global averages of temperature, precipitation or sea ice extent to present day values (but not simultaneously) even in the case of doubled  $\text{CO}_2$  concentration, assuming that it is possible to increase the CDNC to a very high value of  $1000\text{ cm}^{-3}$  over the seeded areas that cover 20–70% of the world's oceans.

One limitation in all of these studies is that they do not explicitly simulate the emissions, transport or effects on natural aerosol of the injected sea spray particles, but instead set the CDNC over the seeded regions to a fixed value of  $375\text{ cm}^{-3}$  or  $1000\text{ cm}^{-3}$ . In reality, however, sea spray geoengineering is expected to result in a spatially inhomogeneous CDNC field because of variations in aerosol dilution and deposition rates and because of the wind speed dependence of the spray rate from the planned vessels (Salter et al., 2008). The picture is further complicated by possible “inadvertent” effects on natural aerosol processes. One example of such a possible inadvertent effect is the impact of the injected sea spray particles on natural CDNC by interfering with the formation of sulphate aerosol from dimethylsulphide (DMS), which is an important source of CCN in the remote marine atmosphere (Korhonen et al., 2008a). Such an effect needs to be considered because freshly emitted sea spray particles can reduce the amount of gas phase sulphur compounds transported to the free troposphere (where new particle formation occurs) by two mechanisms: first, by increasing the concentration of alkaline aqueous phase in which  $\text{SO}_2$  oxidation by  $\text{O}_3$  and  $\text{H}_2\text{O}_2$  takes place (Gurciullo et al., 1999) and second, by acting as a condensation sink for  $\text{H}_2\text{SO}_4$  which is currently thought to be the main nucleating and condensing compound in the marine atmosphere. A further inadvertent effect is the impact of the extra sea spray particles on in-cloud supersaturation,

which will also influence the formation of drops from existing aerosol.

In this study, we simulate the emissions and atmospheric processing of deliberately emitted sea spray particles explicitly for the first time. We quantify how wind-speed dependent geoengineering fluxes influence the CDNC at typical marine cloud altitudes and identify how atmospheric processes lead to spatial CDNC variations. Furthermore, we investigate how sea spray geoengineering affects the sources of natural sulphate CCN in the cloud seeding regions.

## 2 Methods

### 2.1 Model description

The model runs were made with the global aerosol model GLOMAP, which is an extension to the TOMCAT 3-D chemical transport model (Chipperfield, 2006; Stockwell and Chipperfield, 1999). A detailed description of GLOMAP is given in Spracklen et al. (2005). The model is run with a T42 spectral resolution ( $2.8^\circ \times 2.8^\circ$ ) and with 31 hybrid  $\sigma$ - $p$  levels extending to 10 hPa. Over the oceans 7 model levels represent the lowest  $\sim 2\text{ km}$ , of which 5 levels are in the lowest  $\sim 1\text{ km}$ . Large-scale atmospheric transport is specified from European Centre of Medium-Range Weather Forecasts (ECMWF) analyses at 6-h intervals. Here, the sectional version GLOMAP-bin is used, which represents the aerosol size distribution with a moving centre scheme using 20 size sections to cover the diameter range of 3 nm to  $25\text{ }\mu\text{m}$ . In the runs presented here, the aerosol composition is described with three internally mixed components: sulphate, sea spray and carbonaceous aerosol (including both organic carbon (OC) and elemental carbon (EC)). The masses of all components along with the number concentration of particles are tracked in each size section. The aerosol processes simulated are: binary homogeneous nucleation of  $\text{H}_2\text{SO}_4$  and  $\text{H}_2\text{O}$  according to Kulmala et al. (1998), condensation of  $\text{H}_2\text{SO}_4$ , hygroscopic growth, coagulation, wet and dry deposition, transport, cloud processing ( $\text{SO}_2$  oxidation by hydrogen peroxide) and sulphate formation in alkaline sea spray particles ( $\text{SO}_2$  oxidation by ozone).

The natural sea spray emission flux is simulated according to Mårtensson et al. (2003) for particles smaller than  $2\text{ }\mu\text{m}$  in dry diameter and according to Monahan et al. (1986) for sizes larger than  $2\text{ }\mu\text{m}$ . Oceanic DMS emissions are calculated using monthly mean seawater concentrations from Kettle and Andreae (2000) and the sea-to-air transfer velocity of Nightingale et al. (2000). Other simulated sources of sulfur species are anthropogenic  $\text{SO}_2$  emissions from Cofala et al. (2005) and volcanic  $\text{SO}_2$  emissions based on Andres and Kasgnoc (1998) and Halmer et al. (2002). We assume that 2.5% of  $\text{SO}_2$  from these continental sources is emitted as primary sulfate particles at particle sizes proposed by Stier et al. (2005). Primary carbonaceous emissions are

taken from van der Werf et al. (2003) for vegetation fires and from Bond et al. (2004) for fossil and biofuels. EC/OC particles are emitted as lognormal modes at sizes proposed in the AEROCOM emissions inventory (<http://nansen.ipsl.jussieu.fr/AEROCOM>).

Monthly mean boundary layer (BL) cloud coverage is specified from the International Satellite Cloud Climatology Project (ISCCP) archive. Aqueous phase  $\text{H}_2\text{SO}_4$  forms in these BL clouds through the reaction of  $\text{SO}_2$  and  $\text{H}_2\text{O}_2$ . Precipitation scavenging in these low-level clouds is currently not described in GLOMAP. Precipitation scavenging (in-cloud and below cloud) of particles and water-soluble gases is considered only for convective and frontal clouds as diagnosed every 6 h in the host model TOMCAT from the ECMWF analyses (and thus separately from ISCCP boundary layer clouds).

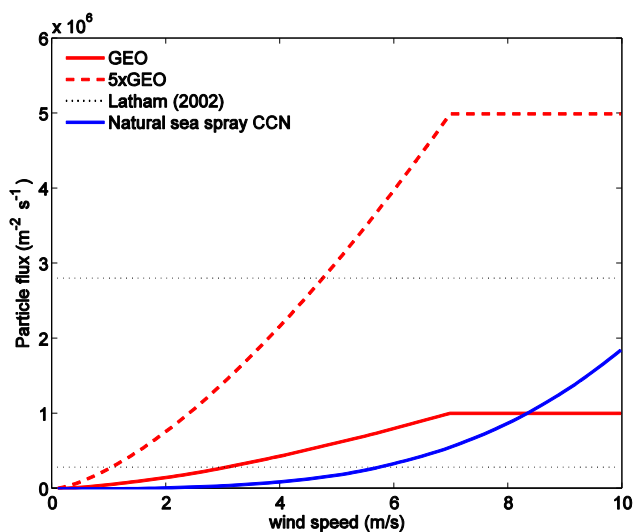
Sulphate formation takes place also in the alkaline aqueous sea spray particles via the reaction of  $\text{SO}_2$  and  $\text{O}_3$ . This reaction is highly pH dependent (important only for  $\text{pH} > \sim 5.5$ ) and is therefore limited by the buffering capacity of the particles as sulphate is formed. It has been found that the buffering capacity of sea spray aerosol is often substantially higher compared to sea water due to calcium enrichment in the aerosol phase (Sievering et al., 1999). As we do not explicitly simulate the aqueous phase chemistry or pH in GLOMAP, we followed the approach of Gurciullo et al. (1999) who (based on ACE-1 campaign measurements) assumed that the maximum amount of sulphate formed via the  $\text{O}_3$  reaction is 6.8 mg of sulphate per 1 g of dry sea spray (three times the amount compared to that without calcium enrichment). This amount is likely to be an upper limit of the actual sulphate production as it represents the maximum enhanced alkalinity observed during ACE-1.

GLOMAP-bin has been evaluated against a wide range of aerosol observations in marine and continental environments (Spracklen et al., 2007, 2008, 2009; Korhonen et al., 2008a, b) and is in good agreement. Relevant to this study, we have also shown that predicted cloud drop number concentrations are in reasonable agreement with a limited set of in situ observations in marine regions (Pringle et al., 2009).

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.

## 2.2 Geoengineering runs

Three model simulations were made: an annual baseline run for the year 2000, which incorporates aerosol processes as described in Sect. 2.1, and two annual runs simulating the proposed geoengineering method by including two intensities of artificial sea spray injection. We assume a wind speed dependence of the sea spray flux based on the spraying ef-



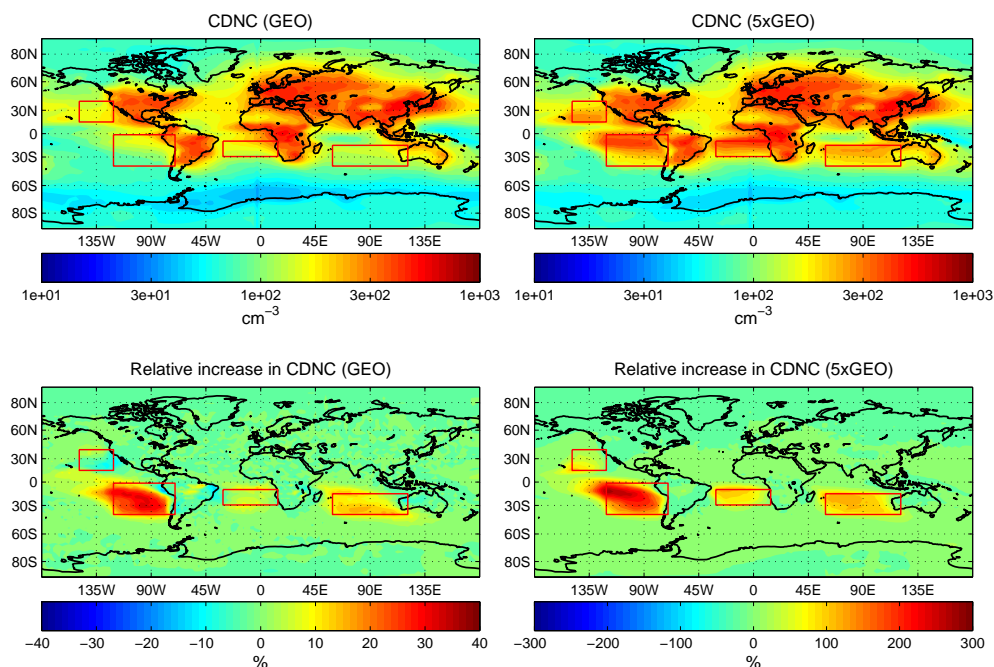
**Fig. 1.** Wind speed dependence of simulated geoengineering fluxes (red lines) and natural flux of sea spray particles larger than 70 nm (blue line). Black dotted lines show the Latham (2002) estimate range for a flux needed to produce 400 additional cloud droplets per  $\text{cm}^{-3}$ .

iciency of the currently planned spray vessels: Salter et al. (2008) anticipate that the vessels reach their maximum output at wind speeds of  $6\text{--}8\text{ m s}^{-1}$  (we assumed a threshold wind speed of  $7\text{ m s}^{-1}$  in this study), and at lower speeds the spray efficiency is assumed to depend on wind speed to the power of 1.5 (S. Salter, personal communication, 2008). 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.

The maximum particle flux at  $>7\text{ m s}^{-1}$  wind speeds was based on the calculations presented in Latham (2002) for the flux required to produce  $400\text{ cm}^{-3}$  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) with the flux of natural CCN-sized ( $>70\text{ nm}$ ) sea spray particles (blue line) (Mårtensson et al., 2003).

The lower of the two geoengineering particle number fluxes (hereafter: GEO) is given by

$$\begin{aligned} F &= 5.4 \times 10^4 \times u_{10}^{1.5} (\text{m}^{-2}\text{s}^{-1}) & \text{when } u_{10} < 7\text{ m/s} \\ F &= 5.4 \times 10^4 \times 7^{1.5} = 10^6 (\text{m}^{-2}\text{s}^{-1}) & \text{when } u_{10} \geq 7\text{ m/s} \end{aligned} \quad (1)$$



**Fig. 2.** Annual mean CDNC (top panels) and relative CDNC change (bottom panels) at 1 km altitude in GEO and  $5\times$ GEO runs.

where  $u_{10}$  is the wind speed 10 m above the ocean surface. The higher flux (hereafter:  $5\times$ GEO) is five times the lower flux. The sea spray particles from geoengineering were emitted at a monodisperse dry diameter of 260 nm following Latham (2002) and corresponding to a particle dry mass of  $1.5\times 10^{-17}$  kg. The particles were emitted into the lowest-altitude grid box, which over the oceans typically covers the lowest  $\sim 60$  m of the atmosphere. Once emitted, these particles underwent the same aerosol microphysical processes (i.e. coagulation, growth due to  $\text{H}_2\text{SO}_4$  condensation and cloud processing, wet and dry deposition) and transport as the natural particles.

In both GEO and  $5\times$ GEO runs the geoengineering flux was limited to four regions: off the coast of California (North Pacific), Chile (South Pacific), Namibia (South Atlantic) and Western Australia (Indian Ocean) (Fig. 2, marine regions inside the red boxes). These regions have been identified as the most favourable all-year sites for sea spray geoengineering in earlier studies (Salter et al., 2008) and together they comprise  $\sim 12.8\%$  of the Earth's surface. Since the model spatial resolution is much too coarse ( $2.8^\circ\times 2.8^\circ$ ) for simulating individual clouds or even cloud decks, we did not attempt to limit geoengineering only to areas with stratocumulus clouds. Instead, cloud seeding was expected to occur continuously in all model grid cells within the chosen regions with the injection rate varying from grid to grid according to the local wind speed.

It is important to appreciate the assumptions in our study concerning the spatial homogeneity of the geoengineering spray emissions. We are assuming that it is technologically

feasible to spray particles homogeneously into the atmosphere on the scale of our model grid boxes of  $\sim 260\times 260$  km (at  $30^\circ$  latitude) and extending over  $6.5\times 10^7$  km<sup>2</sup> of the ocean areas in Fig. 2. A similar assumption was implicitly made by the initial proponents of this geoengineering method (Latham, 2002) when they estimated the particle flux required to alter cloud drop concentrations by a fixed amount. In reality, we believe it would be extremely challenging to spray particles homogeneously without a very large number of vessels. Inhomogeneous spraying would lead to localised high concentrations of sea spray particles and cloud drops, resulting in a different microphysical evolution of these particles compared to the homogeneous case. Clearly there is still much work required to investigate the engineering required to approach homogeneous spraying and distribution. Here we ignore these technological complications.

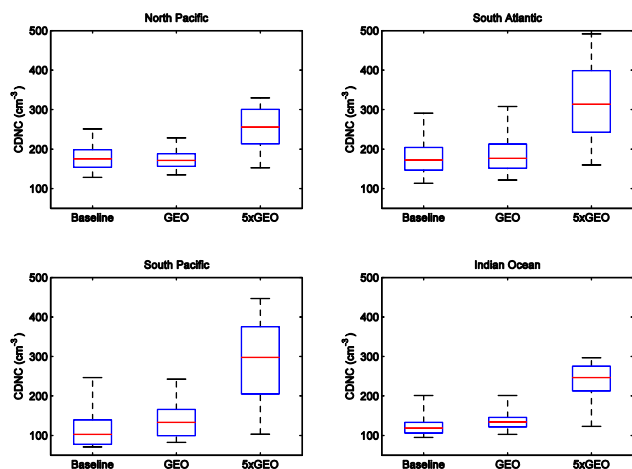
### 3 Results

#### 3.1 Predicted changes in cloud drop number concentration

Figure 2 presents the simulated annual mean values of absolute CDNC in the geoengineering runs as well as the relative CDNC changes compared to the baseline simulation without geoengineering. The CDNC fields were calculated using the modelled monthly mean aerosol at 1 km altitude (approximate cloud base) and a parameterization of drop formation that takes into account the kinetic limitations of droplet growth (Nenes and Seinfeld, 2003). The updraft

**Table 1.** Median CDNC concentrations and mean relative CDNC changes within the four geoengineered regions (indicated by red boxes in Fig. 2). The values are calculated from annual mean CDNC fields.

	GEO		5×GEO	
	CDNC (cm <sup>-3</sup> )	ΔCDNC (%)	CDNC (cm <sup>-3</sup> )	ΔCDNC (%)
North Pacific	171	-2	258	45
South Pacific	133	20	298	163
South Atlantic	177	4	314	77
Indian Ocean	134	11	246	96

**Fig. 3.** The range of predicted annual mean CDNC within the four seeded regions at 1 km altitude. The red line indicates the median, the blue box the 25th and 75th percentiles, and the black whiskers the 5th and 95th percentiles. The number of model grid boxes in the North Pacific is 107, in the South Atlantic 131, in the South Pacific 256, and in the Indian Ocean 217.

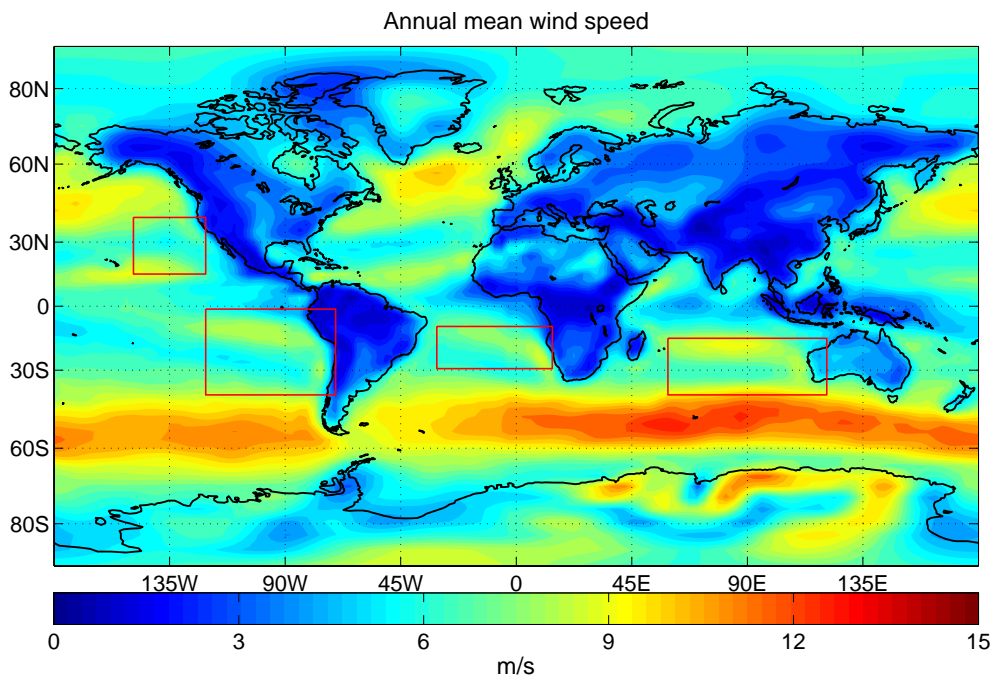
velocities were assumed to follow a Gaussian probability distribution representing typical marine stratocumulus conditions (mean = 0 m s<sup>-1</sup>, standard deviation = 0.25 m s<sup>-1</sup>).

The predicted absolute CDNC varies significantly between and within the four seeded regions (Fig. 2 (top panels) and Fig. 3) which is in contradiction with the assumption of homogeneous CDNC fields over large oceanic regions in previous climate model studies (Latham et al., 2008; Jones et al., 2009; Rasch et al., 2009). The spatial variations are caused by a combination of varying emission rates (which depend on the surface wind speed), transport, and particle loss by deposition and precipitation scavenging. Because all these factors act on the aerosol population continuously, the overall correlation between the surface level wind speed and the predicted CDNC at cloud base in a model column is not very strong (Fig. 2 (top right panel) and Fig. 4), although in some regions a clear correlation is evident (e.g., the high wind speed areas of the North and South Pacific regions).

In the GEO run, the predicted CDNC in the seeded regions increases on average by 20% or less (Table 1), i.e. the sea spray injections fail to increase the CDNC substantially above typical marine background levels (Fig. 2 (top left panel) and Fig. 3). In fact, the model predicts a slight decrease in the CDNC in the North Pacific region as well as off the coast of Chile (Table 1; Fig. 2, bottom left panel). The reasons behind this quite surprising result are discussed in more detail in Sect. 3.2.

In the 5×GEO run the simulated CDNC doubles over large regions, and locally even quadruples in the tropical Pacific (Fig. 2, bottom right panel). A doubled CDNC corresponds to approximately 5% increase in cloud albedo assuming typical marine stratocumulus cloud properties (cloud thickness 500 m, liquid water content (LWC) 0.3 g m<sup>-3</sup>). The simulated absolute CDNC in the seeded regions is in the range of ~100–500 cm<sup>-3</sup> (Fig. 2 (top right panel) and Fig. 3), the high end of which is clearly above marine background values. However, the regional median CDNCs in all four areas remain in the range of 246–314 cm<sup>-3</sup> (Table 1), and thus on average we predict significantly lower CDNC for the spray rates considered here than the 375 cm<sup>-3</sup> assumed in earlier climate model studies of sea spray geoengineering (Latham et al., 2008; Jones et al., 2009).

The relative CDNC change (Fig. 2, bottom panels) depends not only on the spray emission rate and transport but also on the background aerosol concentration. This can be clearly seen by comparing the seeded regions in the Indian Ocean and the North Pacific (Table 1): although the simulated absolute CDNCs in the 5×GEO run are quite similar (medians 246 cm<sup>-3</sup> and 258 cm<sup>-3</sup>) in both regions, the relative CDNC change, which in turn determines the cloud albedo change, is more than double over the Indian Ocean (96% versus 45% over the North Pacific). This difference is because the background CDNC in the North Pacific region off the coast of polluted California is higher than over the Indian Ocean (median values 175 cm<sup>-3</sup> and 119 cm<sup>-3</sup>, respectively). The influence of continental pollution on the relative CDNC change, and thus on the effectiveness of sea spray geoengineering, is also evident close to the shores in all four seeded regions (Fig. 2, bottom right panel). This result suggests that, while regions close to the continents might



**Fig. 4.** Annual mean surface wind speed.

be the most preferable for cloud seeding in terms of practicality (e.g., maintenance of vessels), they are not the most favourable ones in terms of geoengineering efficiency.

### 3.2 Explanation of suppressed cloud drop concentrations

One interesting feature in the GEO simulation results presented above is the relatively small increase of CDNC in the seeded regions and even a decrease in some areas (Fig. 2, bottom left panel). The main reason behind this finding is that the injection of a high number of accumulation mode particles (dry diameter 260 nm) into the aerosol population leads to an increased rate of water vapour condensation in the cloud updraughts, which reduces the maximum supersaturation reached in the cloud. Cloud model studies have previously demonstrated this effect for ship tracks (Russell et al., 1999) as well as sea spray geoengineering (Bower et al., 2006). The suppression of supersaturation means that some of the natural marine particles that form cloud droplets in the baseline run do not activate to droplets in the geoengineering runs. This effect is most significant at the lowest cloud updraughts of the Gaussian distribution we used.

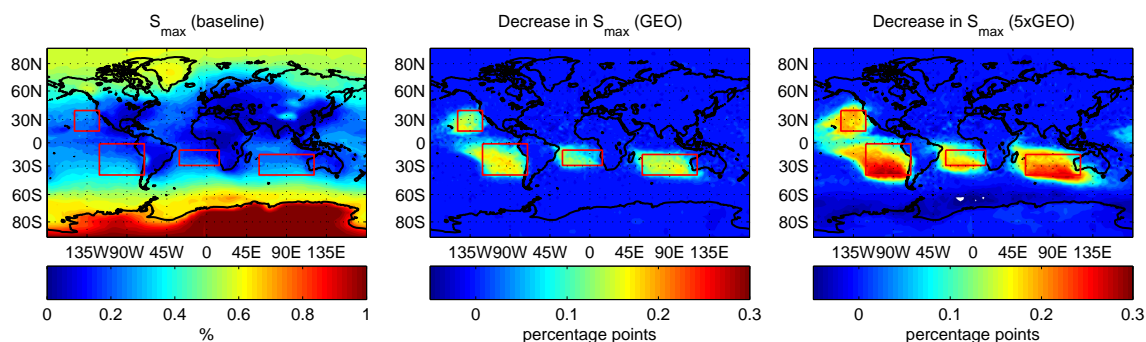
Figure 5 shows the maximum supersaturation reached at 1 km altitude (assumed cloud base) when the updraught velocity is  $0.1 \text{ m s}^{-1}$ . The supersaturation decreases by 0.10–0.13 percentage points from 0.20–0.26% in the baseline run to 0.10–0.14% in the GEO run. Therefore, whereas in the baseline run particles larger than  $\sim 75\text{--}85 \text{ nm}$  in diameter can activate as droplets, in the GEO run only particles larger than

$\sim 110\text{--}140 \text{ nm}$  activate. The effect is even stronger in the  $5\times\text{GEO}$  run (Fig. 5, right panel) in which the supersaturation drops by 0.14–0.20 percentage points and only particles larger than  $\sim 175\text{--}195 \text{ nm}$  in diameter can form cloud droplets. However, the high number concentration of injected sea spray particles in the  $5\times\text{GEO}$  run (Fig. 6, blue line) compensates for the natural particles that do not activate, so the total CDNC increases significantly as seen in Fig. 2.

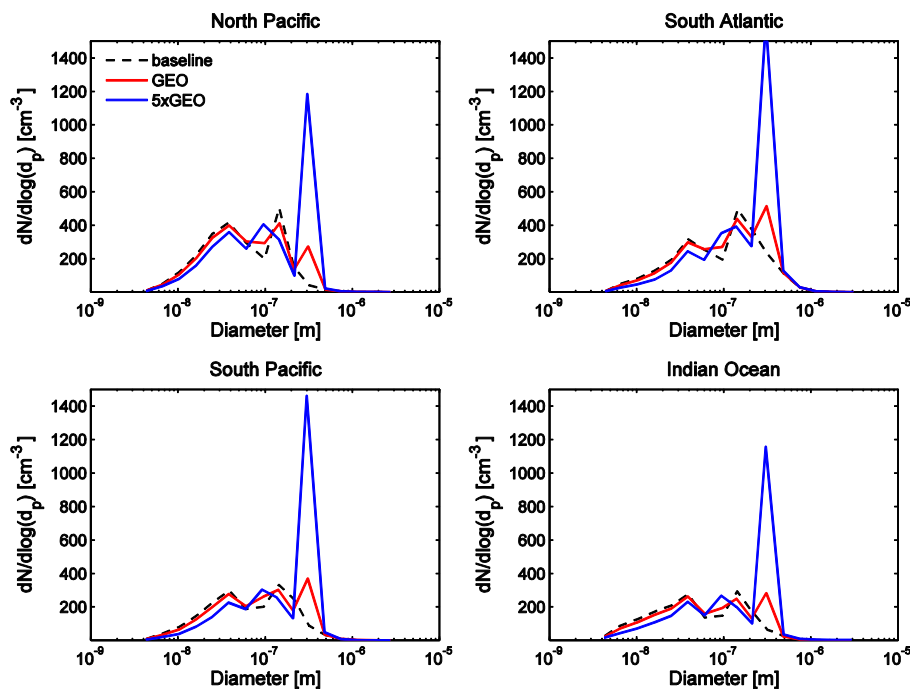
Overall, we predict reduced CDNC in more than 20% of the seeded grid boxes for updraughts less than  $0.4 \text{ m/s}$  in the GEO run. The regional differences are, however, large: while more than half of the seeded area in the North Pacific show CDNC reduction for updraughts below  $0.3 \text{ m/s}$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

It is important to note that the response of CDNC to the emitted particles requires knowledge of not only the cloud updraught velocity but also of the aerosol particle size distribution in the perturbed and unperturbed conditions. The suppression of supersaturation depends on the number, size and growth rate of the droplets. Furthermore, the effect of this suppression on the number of existing particles activated depends on the shape of the number size distribution around the cut-off (activation) diameter.

A second, although less important reason for the predicted suppressed response in CDNC in the GEO run, is that the formation of sulphate in the cloud droplets (via the reaction of  $\text{SO}_2$  and  $\text{H}_2\text{O}_2$ ) takes place predominantly on the injected sea



**Fig. 5.** Maximum supersaturation reached during cloud droplet activation at 1 km altitude in the baseline run (left panel) and the decrease in supersaturation caused by sea spray injections (GEO middle panel; 5×GEO right panel). All values are for updraft velocity 0.1 m/s.



**Fig. 6.** Annual mean size distributions in the four geoengineered regions at 1 km altitude.

spray particles instead of the activated natural particles. As a result, these natural particles grow by cloud processing less rapidly than in the baseline run (Fig. 6, red and black lines). The reduced size of the natural sulphate particles (evident just above 100 nm diameter in Fig. 6) affects their activation to droplets during subsequent cloud cycles. Again, the effect is even stronger in the 5×GEO run (Fig. 6, blue line) but because of the high number of injected sea spray particles, the total predicted CDNC increases.

### 3.3 Effect of sea spray geoengineering on natural sulphate aerosol

Sea spray geoengineering is expected to reduce the atmospheric concentration of sulphur-containing gases in the cloud seeding areas. The concentration of  $\text{SO}_2$  is lowered because the injected sea spray particles increase the amount of alkaline aerosol water, which is important for the pH-dependent oxidation reaction of  $\text{SO}_2$  and  $\text{O}_3$ . The increased oxidation rate of  $\text{SO}_2$  in the aqueous phase reduces its gas phase concentration, which in turn affects the gas phase reaction of  $\text{SO}_2$  and  $\text{OH}$  forming  $\text{H}_2\text{SO}_4$ . The concentration of  $\text{H}_2\text{SO}_4$  is also reduced due to the increased condensation sink provided by the injected particles. The lowered

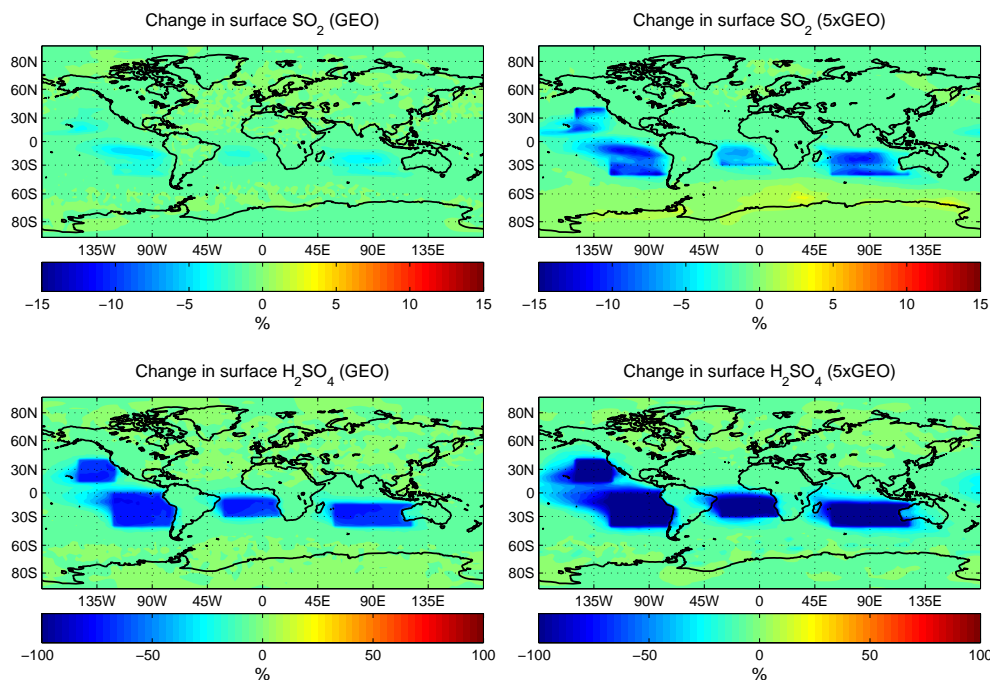


Fig. 7. Effect of cloud seeding on the annual mean surface level concentrations of  $\text{SO}_2$  (top panels) and  $\text{H}_2\text{SO}_4$  (bottom panels).

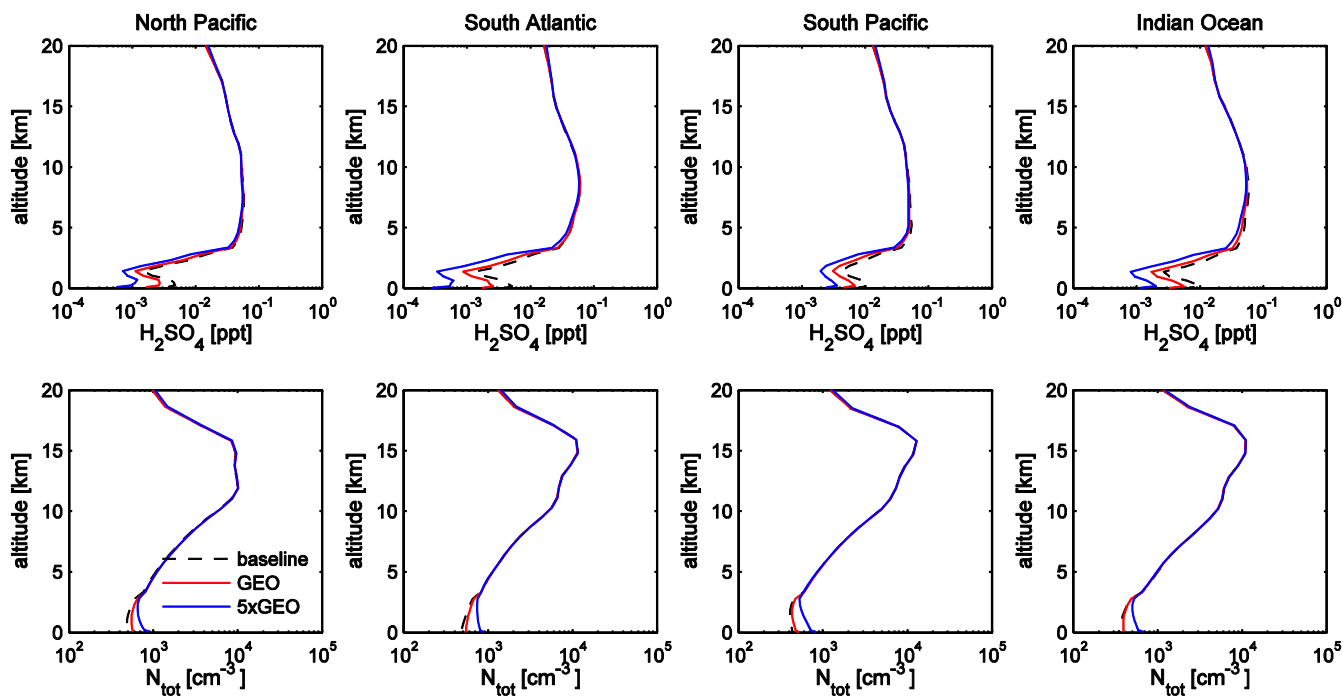


Fig. 8. Effect of cloud seeding on the annual mean vertical profiles of  $\text{H}_2\text{SO}_4$  (top panels) and total particle number concentration ( $N_{\text{tot}}$ ) in the four geoengeered regions.

concentration of  $\text{SO}_2$  and  $\text{H}_2\text{SO}_4$  can influence the formation rate of natural sulphate CCN by 1) decreasing the amount of sulphur transported into the free troposphere where new particles are formed via nucleation of  $\text{H}_2\text{SO}_4$  and  $\text{H}_2\text{O}$  (Ko-

rhone et al., 2008a), and 2) decreasing the concentration of  $\text{H}_2\text{SO}_4$  vapour that contributes to the growth of small particles to CCN sizes.



Figure 7 shows the simulated relative change in surface level  $\text{SO}_2$  and  $\text{H}_2\text{SO}_4$  in the two geoengineering runs. The effect of injected sea spray particles on the  $\text{SO}_2$  concentration is modest: we predict a 1–2% mean decrease in the GEO run and a 5–7% mean decrease (with local maximum decrease up to 14%) in the  $5\times\text{GEO}$  run in the seeded areas (top panels). The reductions are fairly small because the formation of sulphate via the oxidation reaction of  $\text{SO}_2$  and  $\text{O}_3$  (effective only at  $\text{pH} > \sim 5.5$ ) decreases the pH of the aqueous phase and thus shuts off the oxidation reaction relatively rapidly. In our simulations this pH-dependence is approximated by assuming that the oxidation reaction proceeds only when there is less than 6.8 mg of sulphate per 1 g of dry sea salt in the particles. Since the mass of the injected sea spray particles (dry diameter 260 nm) is small in comparison to the bulk natural sea spray mass, most of the sulphate formation takes place in the natural sea spray particles.

On the other hand, cloud seeding decreases the  $\text{H}_2\text{SO}_4$  concentration close to the ocean surface on average by 64–68% in the GEO run and by 96–97% in the  $5\times\text{GEO}$  run (Fig. 7, bottom panels). These large reductions are mainly due to the sea spray injections increasing the condensation sink at the surface by 28–57% and 147–304% in the two runs, respectively. Although this dramatic reduction of  $\text{H}_2\text{SO}_4$  in the lowest atmospheric layers slows down the growth of natural ultrafine particles to larger sizes, it has only a small effect on the natural particle size distribution (Fig. 6) and CCN concentrations. This small impact is consistent with our earlier research which showed that the condensation growth of ultrafine particles is only a minor CCN formation pathway in the marine boundary layer (Korhonen et al., 2008a).

Furthermore, since the reduction in  $\text{H}_2\text{SO}_4$  concentration is limited to the lowest 3–4 km of the atmosphere (Fig. 8, top panels), cloud seeding does not influence CCN formation via free tropospheric nucleation (at  $\sim 5$ –15 km altitude). As a result, the vertical profile of total particle concentration remains unaffected above 4 km (Fig. 8, bottom panels). Below 4 km the change in particle number concentration is determined by the sea spray injections rather than natural sulphate particles.

#### 4 Conclusions

We have presented the first model study of the efficacy of sea spray geoengineering starting from the emissions and going through to the impact on cloud drop concentrations, taking into account the full aerosol microphysical processes. We find that the wind speed dependence of the spray emissions, atmospheric transport and particle loss via deposition and precipitation scavenging lead to a spatially highly inhomogeneous CDNC. Therefore, generating nearly uniform cloud drop fields over large regions of the oceans, as has been assumed in earlier climate model studies, would be extremely challenging.

We also find that the fluxes needed to sustain the desired levels of CDNC (e.g.,  $375 \text{ cm}^{-3}$ ) are likely to be higher than suggested in previous literature. This is because of dilution and removal of particles from the atmosphere but also because the injection of a large number of accumulation mode particles suppresses the cloud supersaturation which in turn prevents some of the natural particles from activating to cloud drops. Our results also suggest that the fractional changes in CDNC, which determine the cloud albedo change, are quite sensitive to the background aerosol concentration. Therefore, persistent stratocumulus regions close to the continents, and thus receiving significant amounts of anthropogenic pollution, may not be the most favourable locations for cloud seeding.

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 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.

We have also been able to assess the inadvertent effects on existing aerosol, which have been neglected in earlier studies. The global model suggests that the impact of sea spray enhancement on the global sulphate aerosol system is negligible. We anticipated that suppression of  $\text{SO}_2$  and  $\text{H}_2\text{SO}_4$  due to reactive chemistry on the particles would affect the formation of aerosol in the free troposphere, which would potentially suppress a very important CCN source to clean marine regions (Merikanto et al., 2009). This effect does not seem to occur, mainly because the mass of salt added to the atmosphere is relatively small and  $\text{SO}_2$  is not greatly depleted.  $\text{H}_2\text{SO}_4$  vapour is strongly depleted in the boundary layer due to increased condensation sink, but this has a negligible effect on aerosol formation, which, in our model, occurs mostly in the free troposphere (Korhonen et al., 2008a; Merikanto et al., 2009). However, further studies of the impact of  $\text{H}_2\text{SO}_4$  suppression on nucleation should be considered, especially taking into account the open cell structure of stratocumulus clouds.

So far the effectiveness of sea spray geoengineering has been studied only with global scale models. However, the spatial resolution in these models is poor, so they cannot resolve the aerosol emissions, transport and activation on a scale of individual stratocumulus cloud cell. Because of this limitation, there is enormous scope for studying this problem using large eddy cloud resolving models where the issues related to the spreading and entrainment of the spray into the clouds, as well as cloud microphysics, can be studied in much more detail.

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