

Figure 1: Particles from commercial shipping create ship tracks of increased reflected sunlight in marine clouds off the west coast of the United States

Observation of the effect of aerosol particles on marine low clouds (Figure 1) led British scientist John Latham to propose using an analogue from nature - particles of salt from sea water - to brighten clouds over parts of the ocean, making them more reflective, and thus cooling climate (Latham 1990). For this marine cloud brightening (MCB), optimized particles of sea salt would be dispersed from ships and ingested into marine low clouds (Latham et al., 2012). The particles reside in the atmosphere for a few days so brightening is limited to localized areas. Dispersal is continuous and targeted to susceptible areas.

Marine clouds occurring over cool oceans (known as "stratocumulus") appear particularly susceptible to additions of aerosol, with large stratocumulus cloud decks present off the west coasts of California, Chile and south-central Africa. Early climate modeling studies suggest that using ships to deliver optimized sea salt particles into 10-25% of the Earth's marine clouds could provide enough cooling to offset a doubling of CO₂, extending the time available to reduce greenhouse gases (Rasch et al., 2009; Jones et al., 2009).

Although global climate modeling has demonstrated potential for a significant cooling effect/negative radiative forcing from MCB (Fig. 2), many questions remain:

1. What fraction of the low cloud regions must be seeded to produce a given radiative forcing?
2. How many spray sites are required?
3. What is the maximum possible forcing one could produce using MCB?
4. What is the optimal size range for the injected particles?

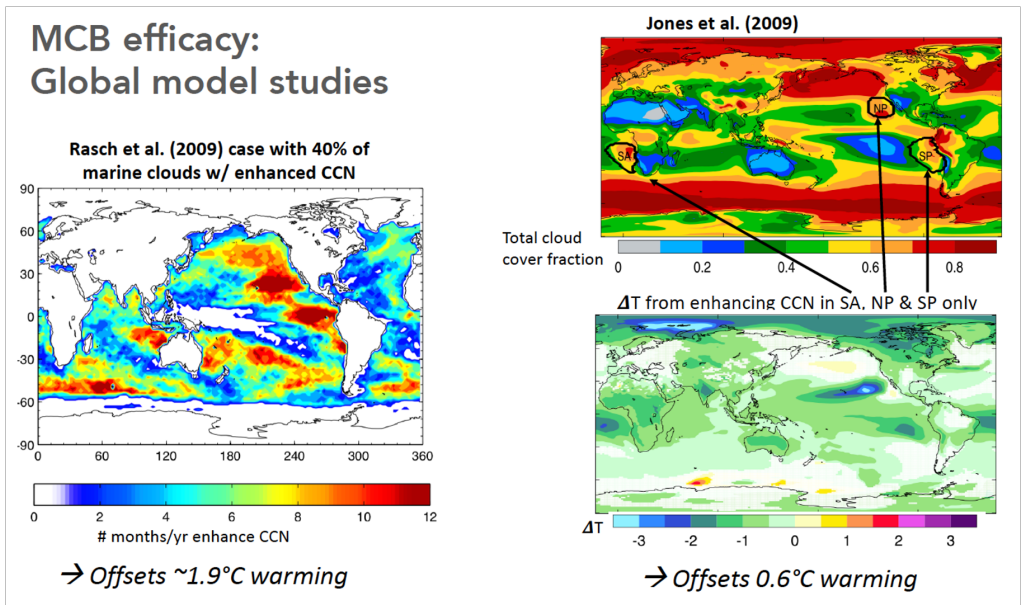


Figure 2: Existing climate modeling of MCB has made a range of different assumptions about the locations of seeding and the rates and sizes of injected particles. Significant cooling can be obtained in these simulations, but thus far there has been little attempt to optimize the characteristics of the seeding (number of sprayers, particle injection rates and sizes, etc.).

- Twomey (1977) formulation for the susceptibility of cloud albedo α_c to increases in cloud droplet concentration N_d :

$$\frac{d\alpha_c}{dN_d} = \frac{\alpha_c(1-\alpha_c)}{3N_d} \quad [1]$$

- Integrate [1] to give an expression for the increase in cloud albedo $\Delta\alpha_c$ caused by an increase in N_d , assuming no change in cloud liquid water:

$$\Delta\alpha_c = \phi_{\text{atm}} \frac{\alpha_c(1-\alpha_c)(r_N^{1/3}-1)}{1+\alpha_c(r_N^{1/3}-1)} \quad [2]$$

where $r_N = N'_d/N_d$ is the ratio of the droplet concentration for the seeded clouds to that in the unseeded cloud and ϕ_{atm} (~ 0.7) converts to TOA albedo accounting for the absorption and scattering of solar radiation by the atmosphere above cloud (Diamond et al., 2020).

- MCB is deployed over the fraction of Earth f_{ocean} ($=0.7$) covered by ocean; only a fraction f_{low} ($=0.29$) of the ocean is covered with low clouds suitable for seeding. Outside of these areas, MCB is assumed to exert no radiative forcing.
- A fraction f_{seed} of the regions covered with marine low cloud is subject to seeding.
- The global mean shortwave radiative forcing ΔF from MCB can be written as

$$\Delta F = -F_{\odot} f_{\text{ocean}} f_{\text{low}} f_{\text{seed}} \phi_{\text{atm}} \Delta\alpha_c \quad [3]$$

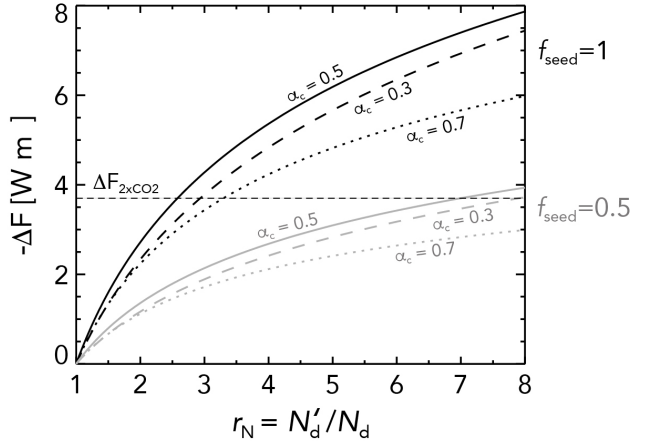
where F_{\odot} is the diurnal mean incoming solar irradiance, here assumed to be equal to the global mean solar irradiance $F_{\odot} = 342 \text{ W m}^{-2}$.

- Assuming all stratiform low clouds are seeded ($f_{\text{seed}}=1$), then from [3], if cloud albedo is increased by $\Delta\alpha_c = 0.01$, then $\Delta F = -0.57 \text{ W m}^{-2}$. A cloud albedo increase of $\Delta\alpha_c = 0.065$ produces $\Delta F = -3.7 \text{ W m}^{-2}$, which would balance the longwave radiative forcing $\Delta F_{2\times\text{CO}_2}$ from doubling CO_2 .
- Assuming $\alpha_c = 0.56$ (Bender et al. 2011), then a **relative increase in droplet concentration of $r_N = 2.25$ would produce a forcing with a magnitude equal to $\Delta F_{2\times\text{CO}_2}$** . This is within the range of N_d increases over the ocean (2.10-2.85) needed to counter CO_2 doubling in an analysis of three variants on a climate model (Slingo 1990).

Global SW radiative forcing

Figure 3: Global SW radiative forcing estimated using Eqn. 3, as a function of the ratio of perturbed to unperturbed cloud droplet concentration $r_N = N'_d/N_d$

Curves are shown for the case where all stratiform low cloud regions are seeded ($f_{\text{seed}}=1$, black lines) and where only 50% are seeded ($f_{\text{seed}}=0.5$, gray lines), for cloud albedo α_c ranging from 0.3-0.7.



TREATMENT OF AEROSOL and CLOUD DROPLET ACTIVATION

- The Abdul-Razzak and Ghan (2000) activation scheme is used to determine N_d for seeded and unseeded regions. Background aerosol (sulfate) is lognormally distributed with geometric mean diameter D_0 , geometric standard deviation S_0 , and number concentration N_0 . Injected aerosols are sodium chloride, distributed lognormally with GMD D_s and GMS S . Updraft speed is fixed. See Table 1 for details.
- Any practical deployment of MCB would be unable to produce uniform increases of N_d because seeding is not distributed evenly. We assume an array of N_{ships} sprayers distributed randomly throughout the seeded fraction of the regions with stratiform low clouds.

PARTICLE INJECTION AND AREA PERTURBED

- Each sprayer injects NaCl particles continuously at a salt mass rate \dot{M}_s . The total number of particles sprayed per second from each sprayer \dot{N}_s for a lognormal relationship of particles with density ρ_s is:

$$\dot{N}_s = \frac{6\dot{M}_s}{\pi\rho_s D_s^3 e^{9\ln^2 S/2}} \quad [4]$$

- The area affected by a single sprayer assumes that sprayers are stationary; air masses pass over them with wind speed U_0 , assumed constant (Table 1). Injected particles have a residence time τ_{res} . The length scale $L_t = U_0 \tau_{\text{res}}$ is the distance downstream over which the particle concentration is affected by spraying. The area perturbed by each sprayer A_t is then determined by multiplying this length scale by a mean track width W_t , i.e. $A_t = L_t W_t = U_0 \tau_{\text{res}} W_t$. We assume that the injected particles are distributed evenly within the perturbed area A_t .
- Particles are assumed to mix rapidly in the vertical through the depth h of the MBL. In a real ship track, further dilution then occurs by lateral diffusion, which is approximated using a lateral plume spreading rate κ (Table 1). For the heuristic model, each track has uniform width $W_t = \kappa \tau_{\text{res}}/2$ and uniform concentration:

$$N_s = \frac{\dot{N}_s \tau_{\text{res}}}{h A_t} = \frac{2\dot{N}_s}{h \kappa U_0 \tau_{\text{res}}} \quad [5]$$

- This idealized rectangular track produces a radiative forcing that is slightly weaker (<25%) than that for a more realistic track in which particle concentrations dilute along the track and are removed gradually with an e-folding time τ_{res} (see Fig. 4)
- The number of (non-overlapping) rectangular tracks required to cover the 29% of the ocean covered by stratiform low clouds ($\sim 1.1 \times 10^{14} \text{ m}^2$), assuming $\tau_{\text{res}}=2$ days, $W_t=44 \text{ km}$, and $U_0=7 \text{ m s}^{-1}$ (i.e., $L_t=1200 \text{ km}$; $A_t = 5.3 \times 10^{10} \text{ m}^2$) is ~ 2000 .
- Overlapping tracks are unavoidable because air mass trajectories are not constant in time. We conducted Monte Carlo simulations, placing N_{ships} randomly-oriented or aligned rectangular tracks at random over a large domain of area A . The probability of k tracks overlapping is well-predicted by a Poisson distribution:

$$p(k) = \frac{\zeta^k e^{-\zeta}}{k!} \quad [6]$$

where $\zeta = N_{\text{ships}} A_t / A$ is the mean track density, i.e., the mean number of superimposed tracks. $p(k)$ is insensitive to both the track aspect ratio (L_t/W_t) and whether tracks are aligned with their long sides in one direction or are randomly oriented. We use $p(k)$ to model the superposition of tracks by assuming that the injected aerosol concentration in regions with k tracks is kN_s .

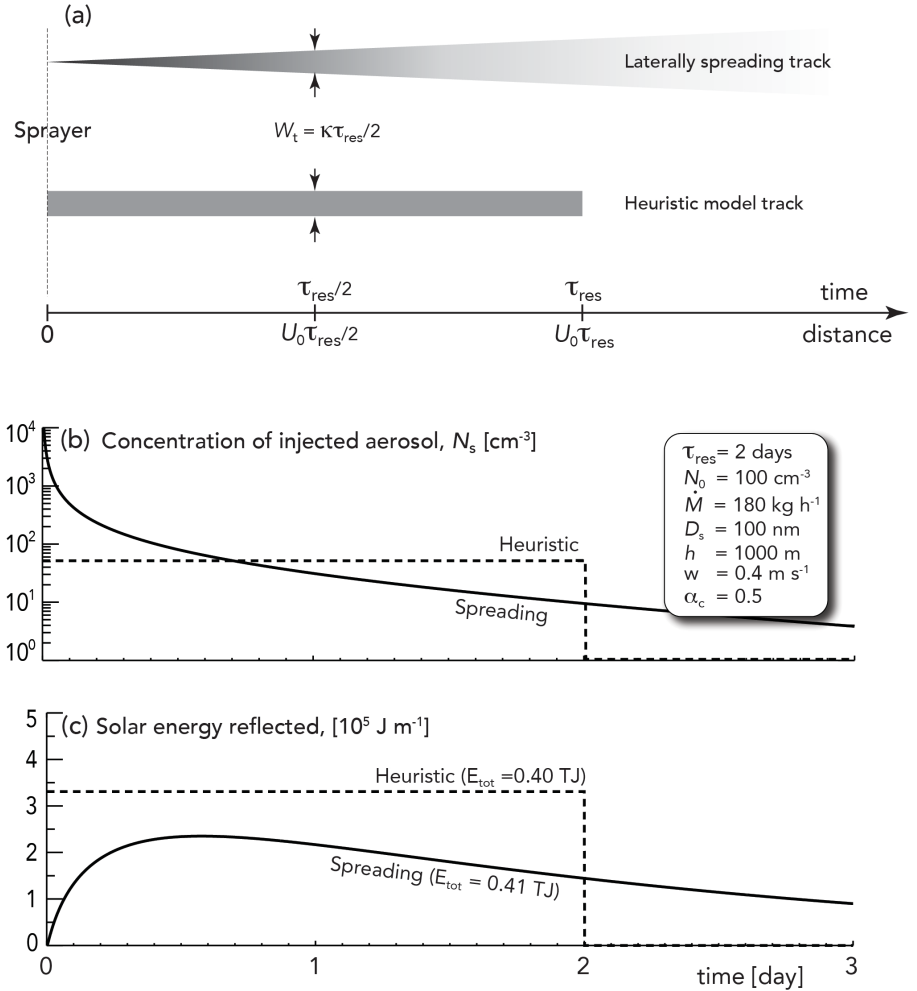


Figure 4: (a) Schematic showing laterally-spreading track and a rectangular shaped track assumed in the heuristic model. Lower panels show an example given parameters shown in the box, of (a) the number concentration of injected aerosol; (b) the solar energy reflected per meter in the heuristic and spreading tracks. The total mass and number of injected particles is exactly the same in each case. Note that in this case, as under most circumstances, the overall energy reflected E_{tot} for the two tracks is almost equal.

Table 1: Parameters used in the heuristic model and their assumed values

Symbol	Parameter	Assumed value(s)	Justification
α_c	Unperturbed cloud albedo	0.5	Bender et al. (2011). See text.
ϕ_{atm}	Atmospheric correction factor	0.67	Based on Diamond et al. (2020).
f_{ocean}	Fraction of Earth's surface covered by ocean	0.7	
f_{low}	Fraction of ocean covered by stratiform low clouds unobscured by high clouds	0.29	36% of the ocean covered by stratiform low clouds (Hahn and Warren 2007). Assume 80% of these are unobscured by high clouds.
f_{seed}	Fraction of stratiform low cloud regions in which sprayers operate	0.5-1.0	
F_{\odot}	Solar irradiance	342 W m ⁻²	Assume global mean solar irradiance.
\dot{M}_s	Rate of NaCl injection by each sprayer	1-1000 kg hr⁻¹	Variable
N_{ships}	Number of sprayer vessels deployed	10³-10⁵	Variable
D_s	Geometric mean diameter of injected NaCl particles	10-1000 nm	Variable
S	Geometric standard dev. of injected NaCl particles	1.6	
τ_{res}	Residence time of injected particles	2 days	Based on Wood et al. (2012)
D_0	Geometric mean diameter of background aerosol	175 nm	Assumed to be sulfate
S	Geometric standard dev. of background aerosol	1.5	Values based on marine accumulation mode aerosol climatology of Heintzenberg (2000)
N_0	Number concentration of background aerosol	50-150 cm ⁻³	
w	Updraft speed for aerosol activation	0.4 m s ⁻¹	Approximate value based on numerous stratocumulus field experiments
U_0	Mean surface wind speed	7 m s ⁻¹	Mean near-surface wind over global ocean (Archer and Jacobson 2005)
h	Marine PBL depth	1 km	Typical mean value for marine low clouds over oceans
κ	Plume lateral spread rate	1.85 km h ⁻¹	Based on observed ship track spreading rate Durkee et al. (2000)

GLOBAL RADIATIVE FORCING FOR MCB

- Heuristic model used to estimate global radiative forcing.→→

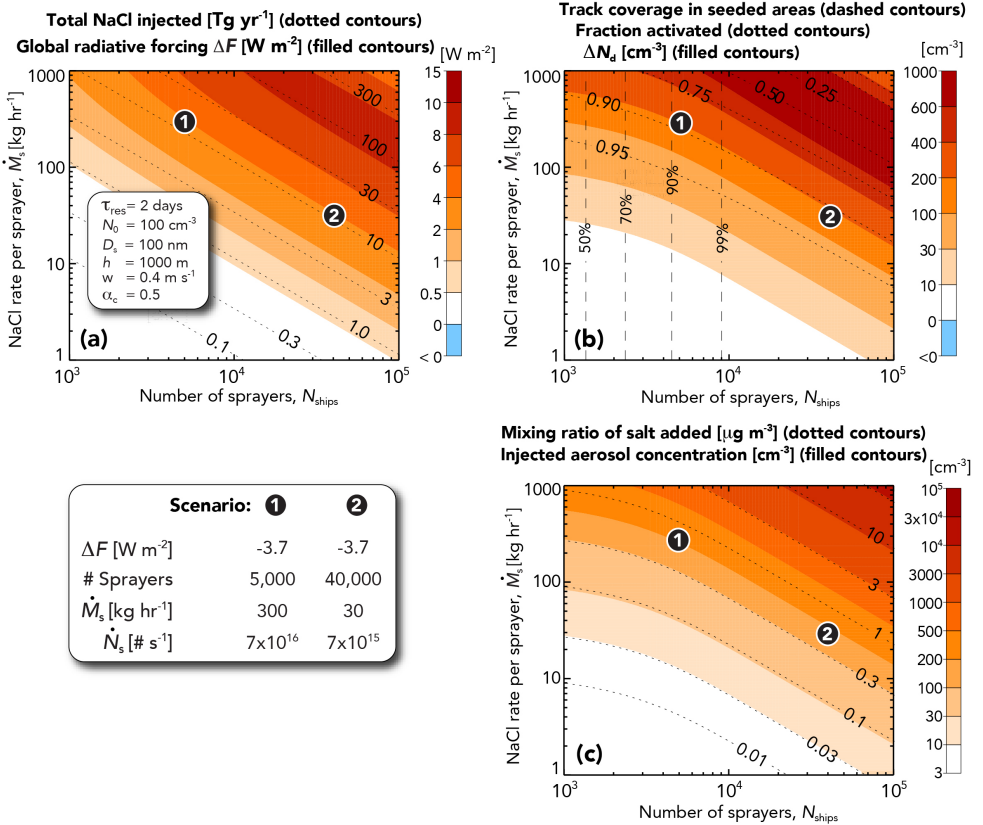


Figure 5: (a) Global mean radiative forcing ΔF (colors) and total flux of sodium chloride (dotted contours) from MCB applied to all marine low cloud regions (29% of the ocean area) as a function of the number of sprayers and the salt mass injection rate \dot{M}_s for each sprayer; (b) increase in cloud droplet concentration ΔN_d (colors), mean fraction of aerosol activated in tracks (dotted contours), and track coverage (dashed contours); (c) injected aerosol concentration in tracks (colors) and mixing ratio in the MBL of injected salt (dotted contours).

- Two scenarios (1) and (2) highlight that a certain level of radiative forcing can be achieved with different spray strategies (fewer ships with higher injection rate vs more ships with lower rate).
- Radiative forcing to offset doubled CO_2 can be achieved with global mean salt spray rates of $\sim 10 \text{ Tg yr}^{-1}$. This is much lower than the natural sea salt flux, which range from 3,000 to $>70,000 \text{ Tg yr}^{-1}$ (Grythe et al., 2014).
- Injected salt burden for scenarios is 0.06-0.07 Tg, much lower than the natural salt burden in the atmosphere of 3-18 Tg (Textor et al. 2006).

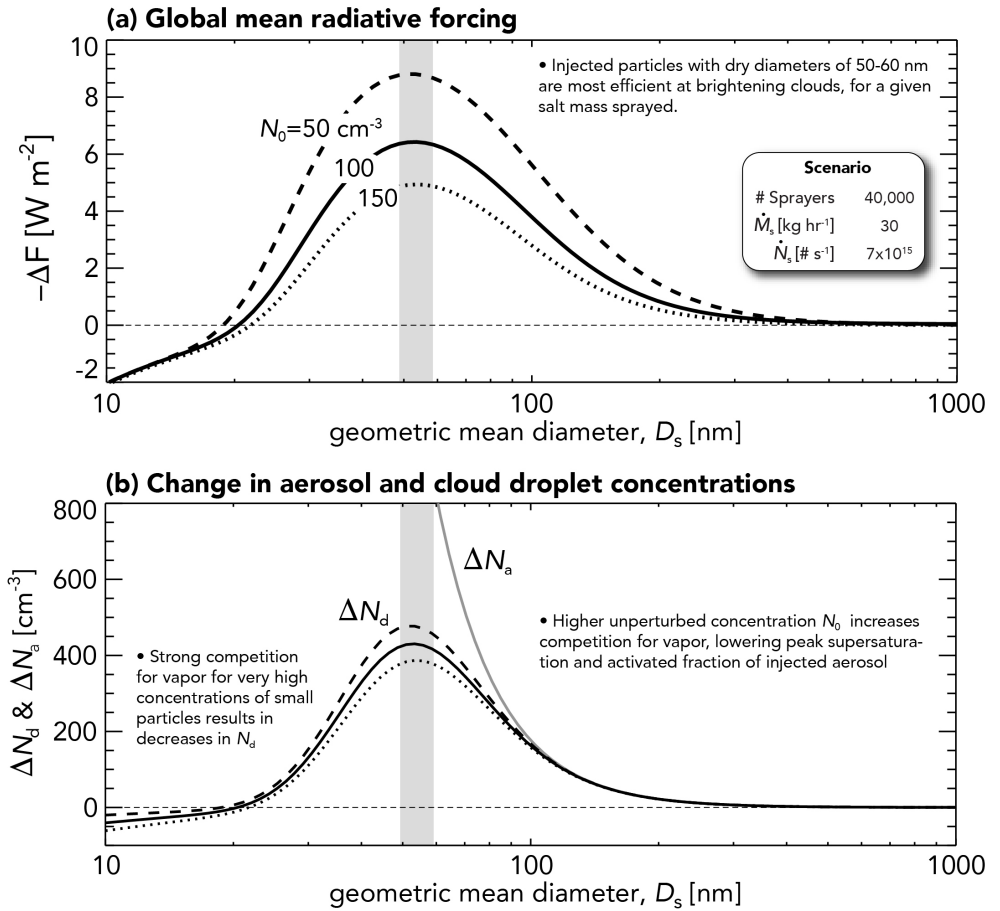


Figure 6: **(a)** Global mean radiative forcing ΔF for a fixed salt mass spray rate as a function of injected particle diameter for three unperturbed aerosol concentrations; **(b)** Change in mean cloud droplet concentration and aerosol concentration in sprayed regions.

- Injected particles with dry geometric mean diameters of 50-60 nm are most efficient at brightening clouds for a fixed mass of salt injected.
- In addition to reducing albedo susceptibility, higher unperturbed aerosol concentrations also lead to lower perturbed N_d , which further weakens the Twomey effect.
- Marine cloud *darkening* can occur for very high concentrations of injected particles smaller than 20 nm because of increased competition for vapor.

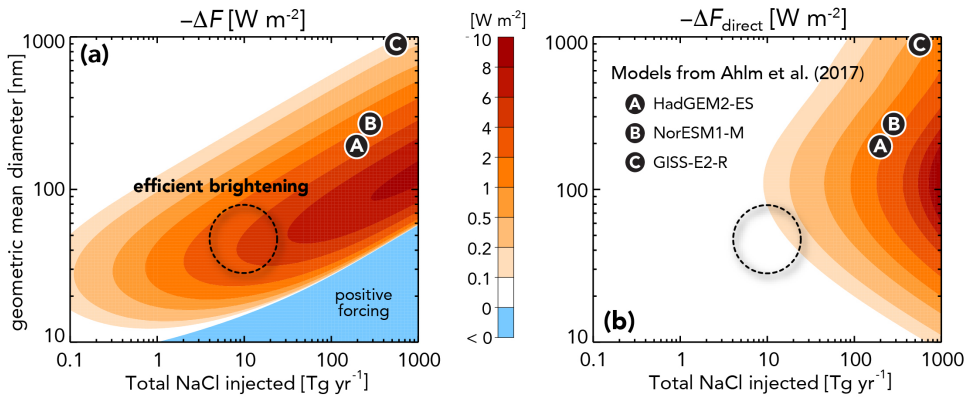


Figure 7: Global mean radiative forcing from (a) MCB; (b) direct effects, plotted as a function of the total NaCl spray rate and the geometric mean dry diameter of injected particles; (c) ratio of MCB to total radiative forcing.

Assumes spraying occurs over entire oceans. Direct radiative forcing ΔF_{direct} for injected particles estimated using Mie scattering and assumed MBL mean relative humidity of 90%. Spraying assumed to take place over entirety of global oceans. Spray configurations for three models used in the recent GeoMIP assessment are shown, highlighting significant direct contribution to radiative forcing.

- ΔF_{direct} significant only for total salt injection rates exceeding ~ 100 Tg yr⁻¹
- There is little direct forcing occurring with efficient application of MCB
- Models used in recent GeoMIP assessment (Ahlm et al., 2017) use injected particles that are much larger than needed for efficient brightening, and hence require much greater salt injection to provide required aerosol forcing

HOW WELL CAN THE HEURISTIC MODEL REPRODUCE BRIGHTENING IN LARGE EDDY SIMULATIONS (LES)?

- LES studies of ship tracks/MCB are compared against the heuristic model.
- LES domain sizes are much smaller than the area of the Earth over which seeding is required to produce a significant global cooling effect, but we scale the results to produce LES domain-mean radiative forcing estimates (Fig. 8a).
- Heuristic model radiative forcing estimates are within a factor of 3 of those from the LES studies in almost all cases (dotted lines, Fig. 8a).
- **Brightening efficiency = solar energy reflected per mass of salt sprayed** (Fig 8b) varies considerably more than the brightening itself, because the injected dry particle diameters vary dramatically: 50 nm (C20a, C20b); 200 nm (W11, J13); 600 nm (P18).
- Small injected particles ($D_s \sim 50$ nm) are most efficient at brightening clouds in the LES models (Fig. 8b)
- Brightening in the heuristic model and the LES both scale approximately inversely with the unperturbed droplet concentration. This scaling is much stronger than that expected from Twomey susceptibility alone.

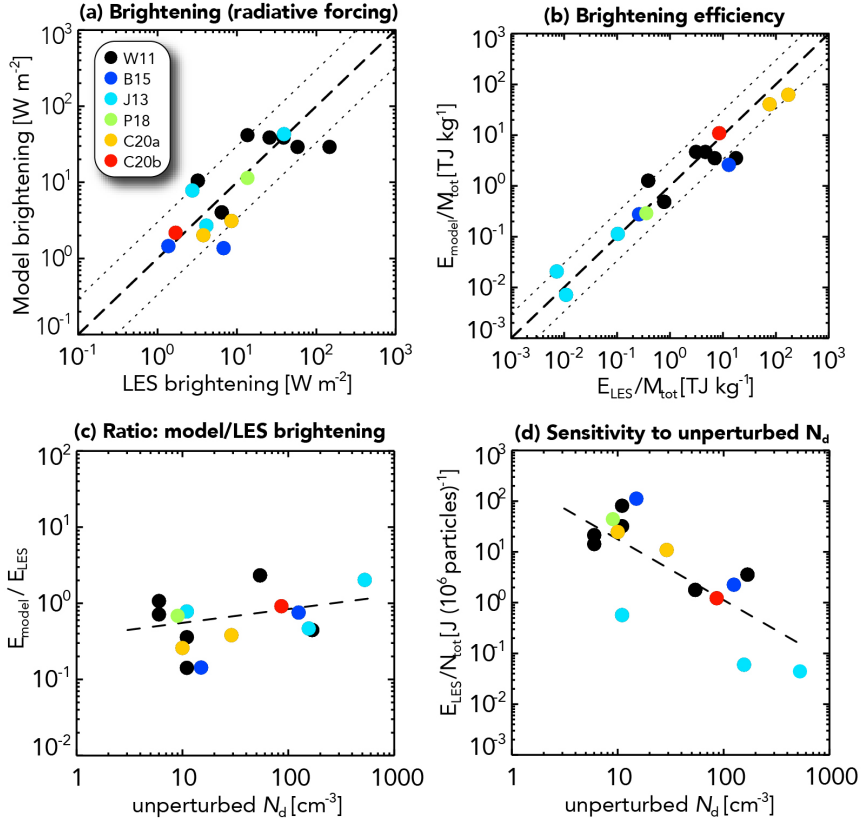


Figure 8: Comparison of heuristic model (“model”) with results from large eddy simulations reported in the literature. (a) domain mean brightening from the heuristic model and the LES; (b) efficiency of brightening as expressed in terms of total energy reflected over the duration of the LES experiment per kg of NaCl injected; (c) ratio of the heuristic model to LES brightening as a function of the unperturbed domain mean cloud droplet concentration N_d ; (d) efficiency of brightening as expressed in terms of total energy reflected per number of particles injected as a function of unperturbed N_d .

LES Studies: W11 (Wang et al., 2011); B15 (Berner et al., 2015); J13 (Jenkins et al., 2013); P18 (Possner et al., 2018); C20a, C20b (Chun et al., 2020). Some of the LES studies include multiple cases, each plotted with a separate symbol.

CONCLUSIONS

- Radiative forcing to offset doubled CO₂ can be achieved with global mean salt spray rates of ~10 Tg yr⁻¹. This is much lower than the natural sea salt flux, and much lower than spray rates used in global models, which have injected larger particles than are needed to efficiently brighten clouds.
- Injected particles with dry geometric mean diameters of 50-60 nm are most efficient at brightening clouds for a fixed mass of salt injected.
- Marine cloud darkening can occur for very high concentrations of small (<20-30 nm) injected particles because of increased competition for vapor.
- Competition for vapor effectively limits maximum magnitude of radiative forcing from MCB to no more than 10 W m⁻² assuming all low clouds are seeded, and that negative cloud adjustments are small.
- Heuristic model radiative forcing estimates are within a factor of 3 of those from large eddy simulations, across a range of different spray and unperturbed conditions.

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