

Accepted Manuscript

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PII: S0169-8095(14)00007-6
DOI: doi: [10.1016/j.atmosres.2014.01.006](https://doi.org/10.1016/j.atmosres.2014.01.006)
Reference: ATMOS 3059

To appear in: *Atmospheric Research*

Received date: 8 March 2013
Revised date: 2 October 2013
Accepted date: 5 January 2014



Please cite this article as: Andrejczuk, M., Gadian, A., Blyth, A., Numerical simulations of stratocumulus cloud response to aerosol perturbation, *Atmospheric Research* (2014), doi: [10.1016/j.atmosres.2014.01.006](https://doi.org/10.1016/j.atmosres.2014.01.006)

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Numerical simulations of stratocumulus cloud response to aerosol perturbation

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Abstract

In this paper results from the 2D numerical model with Lagrangian representation of microphysics are used to investigate the response of the radiative properties of stratocumulus as a result of adding aerosol within the boundary layer. Three different cases characterized by low, moderate and high cloud droplet number and for 3 sizes of additional aerosol 0.01 μm , 0.1 μm and 0.5 μm are discussed. The model setup is an idealization of one of the proposed Solar Radiation Management methods to mitigate global warming by increasing albedo of stratocumulus clouds. Analysis of the model results shows that: the albedo may increase directly in response to additional aerosol in the boundary layer; the magnitude of the increase depends on the microphysical properties of the existing cloud, and is larger for cloud characterized by low cloud droplet number; for some cases for clouds characterized by high cloud droplet number seeding may lead to the decrease in albedo when too large radius of seeding aerosol is used.

Keywords: geoengineering; cloud brightening; LES

1. Introduction

Geoengineering of the stratocumulus clouds is proposed as a one of the methods to offset global warming due to a greenhouse gases emission. Various methods are under consideration, aiming to decrease the flux of the solar radiation reaching the Earth surface (Solar Radiation Management, e.g. Shepherd et al. (2009)). One of the proposed methods is cloud brightening (Latham (1990), Latham (2002), Latham et al. (2008) Latham et al. (2012)). In this method reduction of the solar radiation flux is achieved by increasing

9 the cloud albedo - the first indirect effect Twomey (1977), and longevity - the
10 second indirect effect Albrecht (1989), of the low level stratocumulus clouds,
11 by near surface CCN emission.

12 Climate model simulations (Jones et al. (2009)) indicate that stratocumu-
13 lus cloud seeding may delay global warming by as much as 25 years, giving
14 the time to adopt or to find a better way to deal with the problem. However
15 the cloud-aerosol interactions and aerosol indirect effect is not fully under-
16 stood yet, and representation of these processes in climate models are very
17 simplified (e.g. Ghan et al. (2011)). This uncertainty in representation and
18 understanding of the fundamental processes is a source of uncertainty in the
19 climate prediction. In recent years there have been efforts in the scientific
20 community to asses geo-engineering schemes using climate models (Latham
21 et al. (2012), Korhonen et al. (2010), Jones et al. (2009), Rasch et al. (2009)
22 Latham et al. (2008)), but relatively little research has been devoted to mod-
23 elling details of these processes and in particular, the single cloud response
24 to additional aerosols emitted into the boundary layer. Limited studies with
25 simpler models than used in this paper, have been conducted in the past
26 to address the effect of the aerosol emission on the cloud in the context
27 of the geo-engineering. Bower et al. (2006) and Latham et al. (2012) as-
28 sessed validity of cloud modification as a way to offset global warming with
29 parcel model, without taking into account drizzle. This work confirmed an
30 increase of albedo with an increase of cloud droplet number, with the cloud
31 droplet number being the main factor responsible for cloud albedo change.
32 Wang et al. (2011), and Latham et al. (2012) addressed cloud geo-engineering
33 problem in LES (Large Eddy Simulation) framework, resolving aerosol emis-
34 sion from the surface and transport into the cloud, but with less accurate
35 approach to microphysics, with the similar to Bower et al. (2006) conclusions.

36 In this study a Lagrangian approach to microphysics (Lagrangian Cloud
37 Model) Andrejczuk et al. (2008), Andrejczuk et al. (2010) is used to investi-
38 gate the stratocumulus cloud response to aerosol perturbation. Lagrangian
39 approach to microphysics is a new devepment in cloud modelling, aiming to
40 improve representation of microphysics in numerical models. This study does
41 not intend to model all of the details of the aerosol emission from the spray
42 vessels as proposed by (Latham (1990), Latham (2002)), but rather to look
43 at this proplem in an idealized setup. This paper assumes that emitted
44 aerosol form a well mixed layer below the cloud, with a uniform distribu-
45 tion of aerosol below specified height. Despite this simplification, the process
46 of transport of aerosol from below the cloud into the cloud is represented,

47 and since the boundary layer is typically well mixed there are reasons to
48 believe that aerosol will form such layer with time even when emitted from
49 the surface. We also assume that chemical composition of the aerosol in the
50 boundary layer and that of seeding aerosol is the same ammonium sulphate.

51 The paper is organized as follows: in section 2 numerical model is de-
52 scribed, initial conditions and model setup are described in section 3. Model
53 results are discussed in section 4, and conclusions are in section 5.

54 2. Numerical model

55 Numerical model used to simulate cloud response to aerosol perturbation
56 is the Lagrangian Cloud Model (LCM). Detail of the model formulation can
57 be found in Andrejczuk et al. (2010) (coalescence) and Andrejczuk et al.
58 (2008) (condensation/evaporation). The LCM framework represents the dy-
59 namics and thermodynamics in a traditional, Eulerian framework, with the
60 details of the Eulerian model described in Reisner et al. (2005), Reisner and
61 Jeffery (2009); whilst the microphysics is represented in Lagrangian frame-
62 work, with two way coupling between Eulerian and Lagrangian parts. The
63 microphysical (Lagrangian) part traces millions of parcels, each represent-
64 ing millions of aerosol particles having the same chemical composition and
65 physical properties (location, aerosol size, velocity). Depending on the en-
66 vironmental conditions i.e. the solution of the Eulerian part of the model
67 water can condense/evaporate on the surface of these aerosol. Correspond-
68 ing forces are returned to the Eulerian part to progress model forward in
69 time. Since each Lagrangian parcel represents aerosol having the same phys-
70 ical/chemical properties only one additional parameter to the parcel location,
71 velocity, aerosol and droplet size – number of real aerosol particles Lagrangian
72 parcel represents is required to fully describe properties of the parcel. The
73 model used in this paper is one of three of this type of models recently de-
74 veloped, with other reported by Shima et al. (2009) and Riechelmann et al.
75 (2012).

76 The coalescence algorithm in the Lagrangian microphysics formulation
77 used in simulations reported in this paper maps the collisions between all
78 Lagrangian parcels within the collision grid on a specified two dimensional
79 Eulerian grid (microphysical grid). As a result in coalescence not only droplet
80 sizes are processed but also aerosol sizes, and with time aerosol larger than
81 initially specified can form. New parcels are created only for bins, where
82 number of physical particles is greater than a specified number. Combined

with the parcel merging algorithm, this makes problem numerically solvable, by keeping the number of parcels relatively low. Both mapping and merging processes conserve mass of the aerosol and mass of the water. Based on sensitivity study discussed in appendix of Andrejczuk et al. (2012), in the simulations reported in this paper collision is called every 1s. Additionally each computational grid is split into 4 collision grids.

3. Model set-up and initial conditions

Three 2D idealized cases are considered, with the initial conditions (temperature, q_v , horizontal velocity, aerosol distribution) derived from the VOCALS field campaign, Wood et al. (2011). These cases were based on the cloud droplet concentration and for *HIGH* 250 cm^{-3} , *MED* - 120 cm^{-3} and for *LOW* - 65 cm^{-3} were measured. For all three cases, profiles of potential temperature (θ) and water vapour mixing ratio (q_v) were specified as:

$$\theta(z) = \begin{cases} \theta_B, & z \leq z_B; \\ \theta_C + \alpha z, & z_B < z \leq z_T; \\ \theta_T + (z - z_T)^{2.8}, & z > z_T; \end{cases} \quad (1)$$

$$q_v(z) = \begin{cases} q_{vB} \text{ (or saturation)} & \text{if } z \leq z_T; \\ q_{vT} & \text{if } z > z_T; \end{cases} \quad (2)$$

with the constants for each simulation defined in table 1. Initial profiles for the θ and q_v and profiles derived from a model for the last 3 hours for a model output saved every 6 minutes are shown in figure 1. Additionally, in this figure observed profiles of the LWC and droplet concentration are plotted together with a corresponding profiles diagnosed from a model solution.

A 2D assumption means that the flow evolves only in x-y direction; and the variability of the flow in y direction is neglected. Representation of the atmosphere in two dimension is an approximation, but computational expense of this model prohibits the use of three dimensional domain. A comparison of the solutions between two- and three-dimensional models for a convective planetary boundary layer was discussed by Moeng et al. (2004).

The reference runs use two modal log-normal aerosol distribution fitted to the below the cloud Scanning Mobility Particle Sizer (SMPS) observations (table 2), with the coalescence process active starting from 2nd hour. More details about the setup and comparison of the model results with VOCALS

112 observations can be found in Andrejczuk et al. (2012). Sensitivity runs (table
113 3) were initialized from the reference runs solutions after 4 hours. For the
114 sensitivity runs aerosol of differing concentration and size, were added in the
115 area from 300 meters below the cloud base to the surface. All sensitivity
116 runs were next run for 6 hours with the coalescence process active. The
117 purpose of the sensitivity runs was to determine response of the cloud to
118 the concentration and size of the additional aerosol. Sensitivity runs are
119 identified by the reference run for which additional aerosol is added after 4
120 hours (*HIGH*, *MED*, *LOW*), concentration of the additional aerosol added
121 (100, 200, 400, 800 [cm^{-3}]), and size of the additional aerosol (0.1 - p1, 0.5 -
122 p5 [μm]). See table 3 for an overview of all the runs.

123 For each run, the model output was saved every 10 minutes, and the cloud
124 properties were derived from this output. Three different seeding aerosol sizes
125 were investigated (0.01 μm , 0.1 μm and 0.5 μm), but only simulations using
126 0.1 μm and 0.5 μm are discussed in details. Seeding with the aerosol size 0.01
127 μm has little effect on cloud properties, because a negligible fraction of the
128 aerosol with this size grows to sizes bigger than the activation radius. Note
129 that throughout this paper for the cloud droplet we mean a droplet with the
130 size bigger than 1 μm .

131 4. Results

132 4.1. Cloud properties

133 The aim of the 'cloud brightening' approach is to increase albedo of the
134 stratocumulus clouds. For a plane parallel atmosphere and neglecting ab-
135 sorption, cloud albedo can be approximated Meador and Weaver (1980) (see
136 also Twohy et al. (2005)) as a following function of the optical thickness - τ :

$$A = \frac{0.75(1 - g)\tau}{1 + 0.75(1 - g)\tau}, \quad (3)$$

137 where $g = 0.85$ - asymmetry factor. Optical thickness is defined as (e.g.
138 Stephens (1978)):

$$\tau = \int_0^{\Delta z} dz \int_0^\infty dr Q(x) \pi n(r) r^2, \quad (4)$$

139 where $Q(x)$ is efficiency factor for extinction, $x = 2\pi r/\lambda$, $n(r)$ - cloud droplet
140 size distribution. For the short-wave radiation variability of $Q(x)$ is small and

141 it approaches asymptotic value of 2. This value was used to calculate optical
142 thickness. Additionally only parcels within the cloud having radius bigger
143 than $1 \mu\text{m}$ were used for this calculation. For smaller droplet sizes, the
144 efficiency factor for extinction is small and as a result they do not contribute
145 much to the optical thickness.

146 The left panels of the figure 2 show the evolution in time of the cloud
147 albedo. The solid red colour curve shows the evolution of the albedo for the
148 reference run (without adding aerosol), and solid green/blue/yellow/magenta
149 lines show solutions for perturbation runs with additional aerosol concen-
150 trations $100/200/400/800 \text{ cm}^{-3}$, and a dry radius $0.1 \mu\text{m}$. Dashed lines
151 show corresponding solutions for the cases when seeding aerosol has a radius
152 $0.5 \mu\text{m}$. Each point on the plot represents a space average albedo for the
153 instantaneous solution. The reference runs show that albedo evolution in
154 time is different for each of the cases. It increases with time for the HIGH
155 case, stays approximately constant for the MED and decrease for the LOW.
156 Adding additional aerosol typically increases albedo, with the magnitude of
157 the increase increasing with the increasing concentration of added aerosol.
158 With the few exceptions (MED200p1/MED200p5, HIGH200p1/HIGH200p5,
159 HIGH800p1/HIGH800p5) the effect of seeding aerosol size on albedo change
160 is small - the values of the albedo averaged over the last hour are also shown
161 in table 4.

162 In figure 2 the grey area shows the standard deviation of the albedo for the
163 reference run, indicating a significant variation of the albedo within one time-
164 step. For many cases the mean albedo for the perturbation runs is within the
165 variability of the model solution for the reference run. To determine whether
166 the albedo change for the perturbation runs are statistically different from
167 the reference runs, a two sample tests were performed and the results are
168 presented in table 4, together with the mean increase in the albedo for the
169 last hour. Each sample uses the albedo values for the last 1 hour (what
170 gives length of the sample $6 \times 80 = 480$ points). The null hypothesis is that
171 values in perturbation runs are bigger than in corresponding reference run
172 and significance level is specified as 95%. Out of the 24 perturbed runs only
173 4, for HIGH reference runs, for both seeding aerosol sizes and below the
174 cloud concentrations of 100 and 200 cm^{-3} was rejected. For the remaining
175 20 perturbation runs increases of the albedo are statistically significant. This
176 increase is different for each of the cases; it is smallest - $dA = 1.50 \%$ for the
177 M100p1 and largest - $dA = 16.80$ for the L800p1 run. There is no consistent
178 trend in relation between seeding aerosol size and albedo change and seeding

179 with bigger aerosol sometimes leads to a larger increase in the albedo and
180 sometimes to a smaller increase.

181 One of the possible side effects of the geo-engineering is that it can effect
182 the cloud in the manner to that intended, that is, it may reduce albedo. For
183 example if the cloud is seeded with too big aerosol. Although we do not find
184 it to be a case for seeding aerosol $0.1 \mu\text{m}$, seeding with aerosol $0.5 \mu\text{m}$ for
185 cases HIGH_100 and HIGH_200 leads to a statistically significant decrease
186 in albedo. This finding is consistent with the results reported by Latham
187 et al. (2012), where decrease of the albedo when seeding with large aerosol
188 was also reported.

189 The right hand column of figure 2 shows the cloud droplet number con-
190 centration (N) for the corresponding albedo plots. The transport of the
191 aerosol is relatively rapid and within the 2 hours from the emission start, the
192 maximum of the cloud droplet number within the cloud is reached for most
193 of the cases. The increase of albedo is accompanied by the increase in the
194 cloud droplet number concentration, indicating that indeed, increase in num-
195 ber leads typically to decrease in droplet size and results in a larger albedo,
196 because of the increase in the droplet surface area as discussed by Twomey
197 (1977). There are, however, cases where, the runs with different aerosol
198 seeding size (e.g. M_800p1/M_800p5 8.5-10h), when for larger cloud droplet
199 number albedo is smaller than for small droplet number. This indicate that
200 for these 2 runs adding aerosol may also modify droplet distribution signif-
201 icantly and/or there is a change in LWP leading to smaller in total droplet
202 surface. For these particulate cases, magenta line in figure 3b and 3e there
203 is a significant increase in droplet concentration for sizes less than $2 \mu\text{m}$
204 for the run where the cloud is seeded with aerosol $0.5 \mu\text{m}$ compared tu run
205 with seeding aerosol size $0.1 \mu\text{m}$, and smaller concentration of the 90-100 μm
206 drizzle droplets.

207 Other spectra in figure 3 show that when cloud droplet number increases,
208 droplet concentration increases also, mainly for the sizes smaller than the size
209 for which distribution has maximum (within the range $1 - 10 \mu\text{m}$). Seeding
210 with aerosol of $0.5 \mu\text{m}$ leads to a much larger concentration of the droplet
211 in the range $1-2 \mu\text{m}$ than for corresponding run with seeding aerosol size 0.1
212 μm . For large droplet/drizzle sizes, there is no consistent trend and there
213 are cases where concentration of the drizzle in the perturbed run is bigger
214 than in the reference run, independent on seeding aerosol size.

215 *4.2. Aerosol activation*

216 Since in the microphysical model, full information about aerosol prop-
 217 erties within the cloud droplets is kept, aerosol properties within the cloud
 218 droplets can be examined. Figure 4 shows the scavenged fraction F_i e.g.
 219 G       et al. (2000) for each aerosol bin (the same bin structure as used to
 220 map collisions in coalescence algorithm) defined as:

$$F_i = \frac{N_i}{A_i}, \quad (5)$$

221 where N_i is the number of droplets with radius bigger than $1 \mu\text{m}$ (these do
 222 not have to be bigger than activation radius for given aerosol size) containing
 223 aerosol size from the bin i , and A_i is the total number of aerosol particles
 224 in the bin i . Only model grids with $q_c > 0.001 \text{ g/kg}$ are taken into account
 225 when calculating F_i . The value of 1 indicates that all droplets having aerosol
 226 sizes within a given bin have radius bigger than $1 \mu\text{m}$, and 0 that none of
 227 the aerosol from the bin grew to the size bigger than $1 \mu\text{m}$. Figure 4 shows
 228 that scavenged fraction is smallest for the small aerosol and approach 100 %
 229 for the large aerosol. This is because the small aerosol have small activation
 230 radius but require high saturation to grow to a size bigger than activation
 231 radius. These, high super-saturations are not found often and as a result the
 232 scavenging fraction drops rapidly to 0 for aerosol radius smaller than 0.05
 233 μm . For that reason seeding with the sizes $0.01 \mu\text{m}$ had almost no effect on
 234 the radiative properties of the cloud, almost none of the seeding aerosol had
 235 an opportunity to grow to the droplet size.

236 Figure 4 also shows the different response of the scavenged fraction curves
 237 for small $0.1 \mu\text{m}$ and large $0.5 \mu\text{m}$ seeding aerosol. Seeding with the large
 238 aerosol induces a significant response on the scavenged fraction curves. Curves
 239 move to the right indicating that a smaller fraction of the small aerosol grow
 240 to the sizes bigger than $1 \mu\text{m}$. Since small aerosol activate very quickly it
 241 follows that adding large aerosol leads to a decrease in the supersaturation in
 242 the seeded clouds compared to the reference solution, due to the faster con-
 243 densation rate. For a seeding aerosol size $0.1 \mu\text{m}$, changes in the scavenged
 244 fraction curves are small despite the fact that up to 300 additional droplets
 245 grow to a size bigger than $1 \mu\text{m}$. Note, however, that the difference in the
 246 cloud droplet number between runs with different aerosol seeding size is not
 247 very large because for a seeding size $0.5 \mu\text{m}$, almost all the seeding aerosol
 248 grow to sizes bigger than $1 \mu\text{m}$, and for seeding size $0.1 \mu\text{m}$ around 85 %.

249 4.3. Bulk properties

250 With 24 runs in total there is enough data to examine whether more
 251 general conclusions can be derived from the results. Figure 5 shows the
 252 relationship between the cloud properties and the derived radiative properties
 253 of the clouds for all 24 perturbation runs. Changes are calculated with the
 254 respect to the corresponding reference run (i.e. without adding aerosol):

$$\Delta X = \langle \langle X_p \rangle_x - \langle X \rangle_x \rangle_t, \quad (6)$$

255 where X_p is a vector representing values for the perturbation run and X
 256 values for reference run, subscript x indicate an averaging in space, over the
 257 computation domain; and subscript t indicates an average in time for the
 258 last hour of the simulations. Figure 5 shows the relationship between the
 259 change of the cloud droplet number and the change in albedo (A), Liquid
 260 Water Path (LWP) and effective radius (calculated from relation $r_{eff} =$
 261 $3/2LWP/\tau$ [Stephens (1978)]), and between the change in LWP and the
 262 change in A , together with the last square fit to all data points, and the
 263 correlation coefficient. Expectedly, with the increase of the droplet number
 264 the albedo is increasing (figure 5a) and the effective radius is decreasing
 265 (figure 5b). There is a weak dependence of both dA and dr_{eff} on the seeding
 266 aerosol size, being the result of the variability in the cloud droplet spectrum
 267 and the droplet number. In addition, figures 5a and 5b show also dependence
 268 of the relation between dA/dr_{eff} and dn on a specific case. And for a given
 269 change of droplet number simulations with the initial conditions for HIGH
 270 group respond with the smaller increase of the albedo (larger decrease of
 271 effective radius) than simulations with the initial conditions for the LOW
 272 group. Other two right hand plots show a much larger scattering of the
 273 data, but an increase of the LWP with the increase of the droplet number,
 274 and an increase of albedo with increase of LWP is present. In addition,
 275 examination of figure 5b for runs MED_800p1/MED_800p5 shows that there
 276 was a decrease in LWP for the MED_800p1 run by about 10 [g/m²], which
 277 also may have contributed to the observed smaller albedo for the run with
 278 the larger cloud droplet number.

279 4.4. Comparison with the ship track observations

280 Although, there are no direct measurements of cloud response to aerosol
 281 emission similar to the setup discussed in this paper, there have been mea-
 282 surements of the cloud properties within the ship tracks. Aerosol emitted

283 by ships sometimes effects clouds in a manner similar to proposed by the
 284 geo-engineering approach. At the moment observations in the ship tracks is
 285 the only source of data to validate numerical models, so only a qualitatively
 286 comparison is possible.

287 Analysis of 30 ship tracks by Ackerman et al. (2000) showed that LWC
 288 (Liquid Water Content) on average decreased slightly within the ship tracks
 289 (contrary to results shown in Radke et al. (1989), where a significant increase
 290 in LWP within the ship tracks was observed). The observations analysed by
 291 Ackerman et al. (2000) showed both an increase and a decrease of LWC within
 292 the ship tracks for a single measurement. Results of the simulations discussed
 293 in this papre exhibit similar pattern, and averaged over all perturbation runs
 294 mean decrease of LWP by $\sim 0.6 \text{ g/m}^2$ is present, with both positive and
 295 negative response of LWP to the increase in cloud droplet number (note,
 296 however, that in the analysis we used LWP, whereas in observations LWC
 297 was used).

298 Analysis in Ferek et al. (2000), focused mainly on drizzling cases, demon-
 299 strated an increase in number concentration and a decrease in the size of
 300 cloud droplets within the ship track. Additionally, observations were con-
 301 sistent with the reduction in the drizzle size drops in areas affected by ship
 302 emission. The reduction in the drizzle concentration was also observed by
 303 Radke et al. (1989), where cloud properties for two ship tracks were anal-
 304 ysed. In our simulations, we also observe decrease in the droplet sizes and
 305 an increase in concentration for perturbation runs (disregarding the 2 sim-
 306 ulations where adding aerosol lead to decrease of cloud droplet number).
 307 Especially for the LOW perturbation runs, there is a reduction in maximum
 308 drizzle droplet size (but also drizzle concentration) compared to reference
 309 run. HIGH and MED perturbation runs also show similar tendency, but
 310 there are also cases for these two sets of simulations where either concentra-
 311 tion or drizzle droplet sizes or both are larger for the perturbation run than
 312 for the reference run.

313 As far as impact of pollution on droplet number and size is conserved,
 314 satellite observations are consistent with those from the aircraft and increase
 315 of the cloud droplet number in a ship tracks was reported (Coakley and Walsh
 316 (2002), Segrin et al. (2007)). A liquid water path, drizzle rate and albedo
 317 of the clouds , however, show dependence not only on aerosol emmited from
 318 ships, but also on a cloud regime and vertical structure of the atmosphere
 319 (Segrin et al. (2007), Lebsock et al. (2008), Christensen and Stephens (2011),
 320 Chen et al. (2012)).

5. Conclusions

The results of the 2D numerical simulations shown in this paper indicate that cloud albedo may increase as a result of the seeding if enough aerosol is delivered into the cloud. For seeding to be efficient the aerosol size must be big enough to grow by condensation to the size, where it can effect the radiative properties of the cloud ($\sim 1 \mu\text{m}$) and yet can not be too big to avoid possible problems that may arise when supersaturation field inside the cloud is significantly modified. Based on the scavenged fraction curves, the seeding aerosol size should be bigger than $0.06 \mu\text{m}$ and smaller than $0.5 \mu\text{m}$ to most effectively seed clouds (assuming that the seeding aerosol is ammonium-sulphate). Seeding with too large aerosol may also decrease cloud albedo for specific cases, for clouds characterized by high cloud droplet number concentrations (HIGH setup), when concentration of added aerosol is small. However, for these clouds albedo nevertheless increases in time. The results from the sensitivity studies discussed in this paper and observations of the effect of ship emission on stratocumulus clouds discussed by Chen et al. (2012) indicate that cloud response to aerosol emission is more complex than increase of aerosol \rightarrow increase of cloud droplets \rightarrow increase of cloud albedo. Other factors, such as microphysical properties of existing cloud and the vertical structure of the atmosphere may also effect the outcome of the geo-engineering.

Numerical model results indicate that the transport of aerosol is relatively rapid and the cloud responds with the maximum increase of the albedo within 1–2 hours from the emission for the simulations and the setup discussed in this paper.

Although the model results are consistent with the observations in the ship tracks, dedicated measurements are needed for the quantitative evaluation of the numerical model results.

6. Acknowledgement

This work was supported by NERC grant NE/F018673/1. Observational data was provided by the British Atmospheric Data Centre (BADC). Comments by dr W. Grabowski helped to improve this manuscript.

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Table 1: Constants used to define profiles of the potential temperature, water vapour mixing ratio and cloud water mixing ratio.

<i>RUN</i>	z_B [m]	z_T [m]	θ_B [K]	θ_C [K]	θ_T [K]	q_{vB} [g/kg]	q_{vT} [g/kg]	α [K/m]
HIGH	800	1380	291.1	293.0	302.5	8.3	0.3	3.3×10^{-3}
MED	1000	1400	289.2	290.4	299.0	7.0	0.7	3.0×10^{-3}
LOW	900	1260	290.1	291.1	301.0	7.8	0.7	2.8×10^{-3}

Table 2: Parameters of log-normal distributions used in simulations.

<i>RUN</i>	$N_1[cm^{-3}]$	$r_1[\mu m]$	σ_1	$N_2[cm^{-3}]$	$r_2[\mu m]$	σ_2
HIGH	380	0.071	0.45	160	0.029	0.31
MED	118	0.10	0.43	129	0.022	0.36
LOW	42	0.11	0.25	111	0.023	0.47

Table 3: Overview of the runs. N_a is an aerosol concentration added for each grid from 300 meters bellow cloud base to surface, r_a - radius of the additional aerosol.

RUN	$r_a \mu m$	$N_a [cm^{-3}]$
HIGH	-	-
HIGH100p1	0.1	100
HIGH200p1	0.1	200
HIGH400p1	0.1	400
HIGH800p1	0.1	800
HIGH100p5	0.5	100
HIGH200p5	0.5	200
HIGH400p5	0.5	400
HIGH800p5	0.5	800
MED	-	-
MED100p1	0.1	100
MED200p1	0.1	200
MED400p1	0.1	400
MED800p1	0.1	800
MED100p5	0.5	100
MED200p5	0.5	200
MED400p5	0.5	400
MED800p5	0.5	800
LOW	-	-
LOW100p1	0.1	100
LOW200p1	0.1	200
LOW400p1	0.1	400
LOW800p1	0.1	800
LOW100p5	0.5	100
LOW200p5	0.5	200
LOW400p5	0.5	400
LOW800p5	0.5	800

Table 4: Results of the statistical testing of the null hypothesis that albedo for perturbation run is bigger than for reference run with 5% significance level. Y - hypothesis is true, N - hypothesis is false, * indicates that hypothesis that albedo for reference run is bigger than for perturbation run is true. Additionally averaged over the last hour change in the albedo $\Delta_A = A_p - A_r$ for each perturbation run is also shown.

<i>RUN</i>	$A_p > A_r$	Δ_A (1 hour mean)	Δ_A (1 hour mean)	$A_p > A_r$	<i>RUN</i>
HIGH100p1	N	0.4	-2.5	N*	HIGH100p5
HIGH200p1	N	0.1	-2.0	N*	HIGH200p5
HIGH400p1	Y	2.3	1.1	Y	HIGH400p5
HIGH800p1	Y	3.9	3.6	Y	HIGH800p5
MED100p1	Y	1.5	2.1	Y	MED100p5
MED200p1	Y	4.9	3.0	Y	MED200p5
MED400p1	Y	5.4	4.5	Y	MED400p5
MED800p1	Y	7.8	9.0	Y	MED800p5
LOW100p1	Y	6.0	6.6	Y	LOW100p5
LOW200p1	Y	10.4	9.0	Y	LOW200p5
LOW400p1	Y	11.7	12.4	Y	LOW400p5
LOW800p1	Y	16.8	17.5	Y	LOW800p5

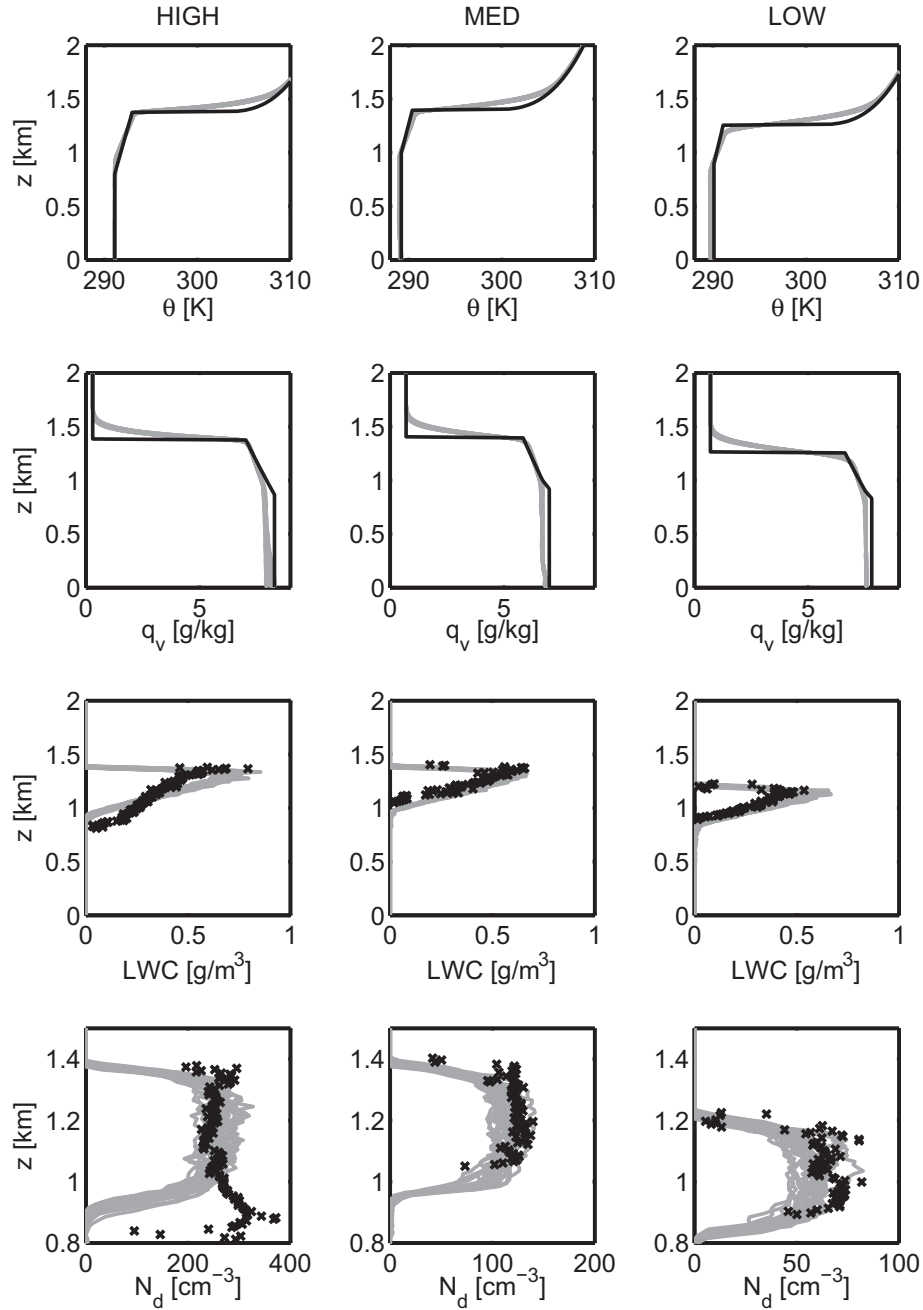


Figure 1: Initial profiles (solid black lines)/observations (black symbols) of potential temperature (θ), water vapour mixing ratio (q_v), Liquid Water Content (LWC), and cloud droplet number (N_d); and model solution for the last 3 hours for a reference setups (gray lines) - HIGH (left column), MED (middle column) and LOW (right column).

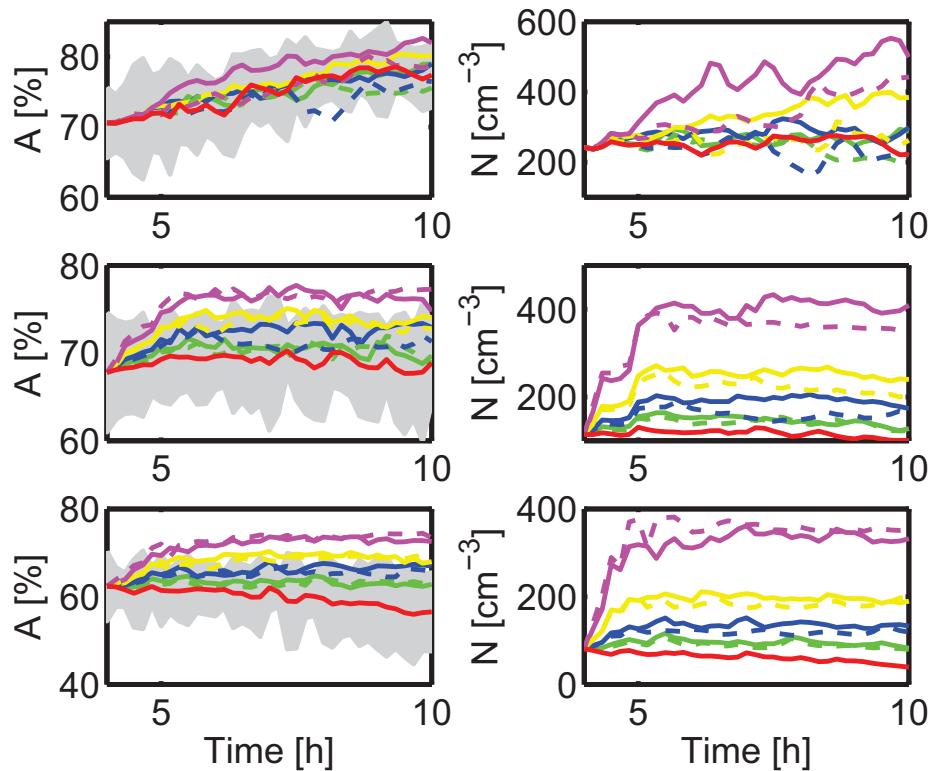


Figure 2: Evolution in time of cloud albedo (left column) and cloud droplet concentration (right panel) for HIGH (a and b), MED (c and d) and LOW (e and f) aerosol distribution. Colors mark different sensitivity simulation: red - reference simulation, green - run with additional aerosol concentration 100, blue - with additional concentration 200, yellow - with additional concentration 400, and magenta - with additional concentration 800 $[\text{cm}^3]$. Solid line - perturbing aerosol size $0.1 \mu\text{m}$, dashed line - perturbing aerosol size $0.5 \mu\text{m}$.

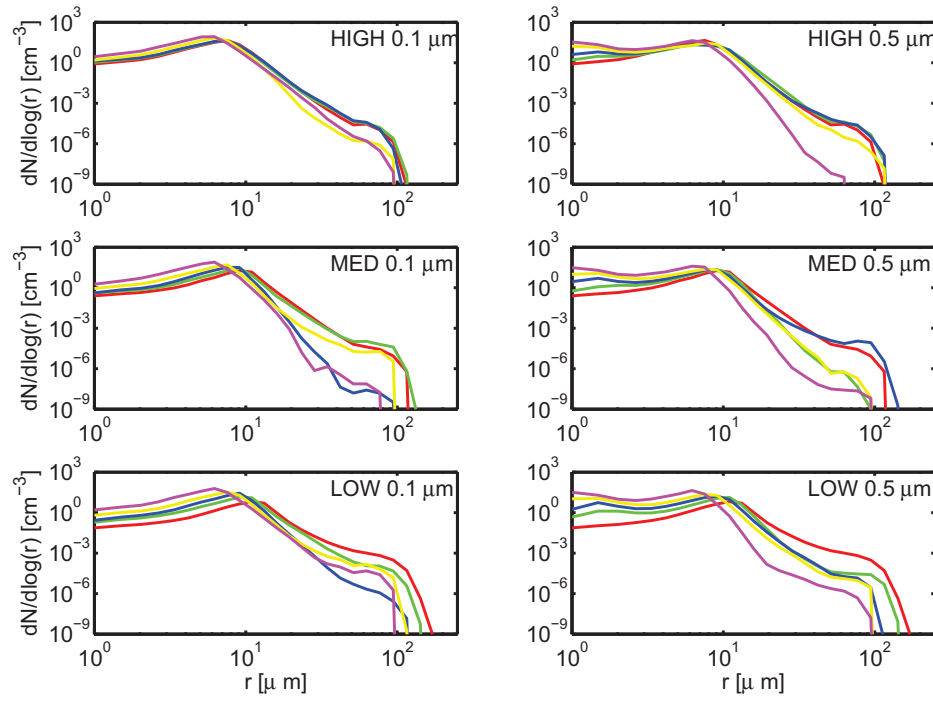


Figure 3: Averaged over the last hour of simulation cloud droplet spectra for perturbing aerosol $0.1 \mu\text{m}$ (left column) and $0.5 \mu\text{m}$ (right column). Colors the same as in Fig. 1.

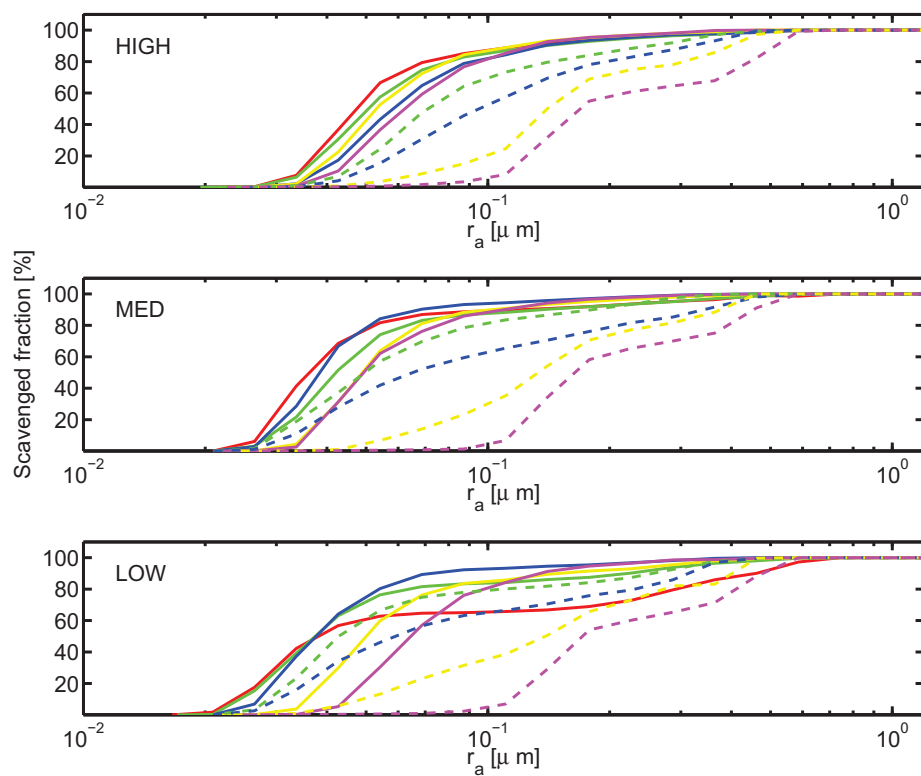


Figure 4: Scavenging fraction averaged over the last hour. Solid line for perturbing aerosol size 0.1 μm , dashed line - perturbing aerosol size 0.5 μm . Colours as in Fig 1.

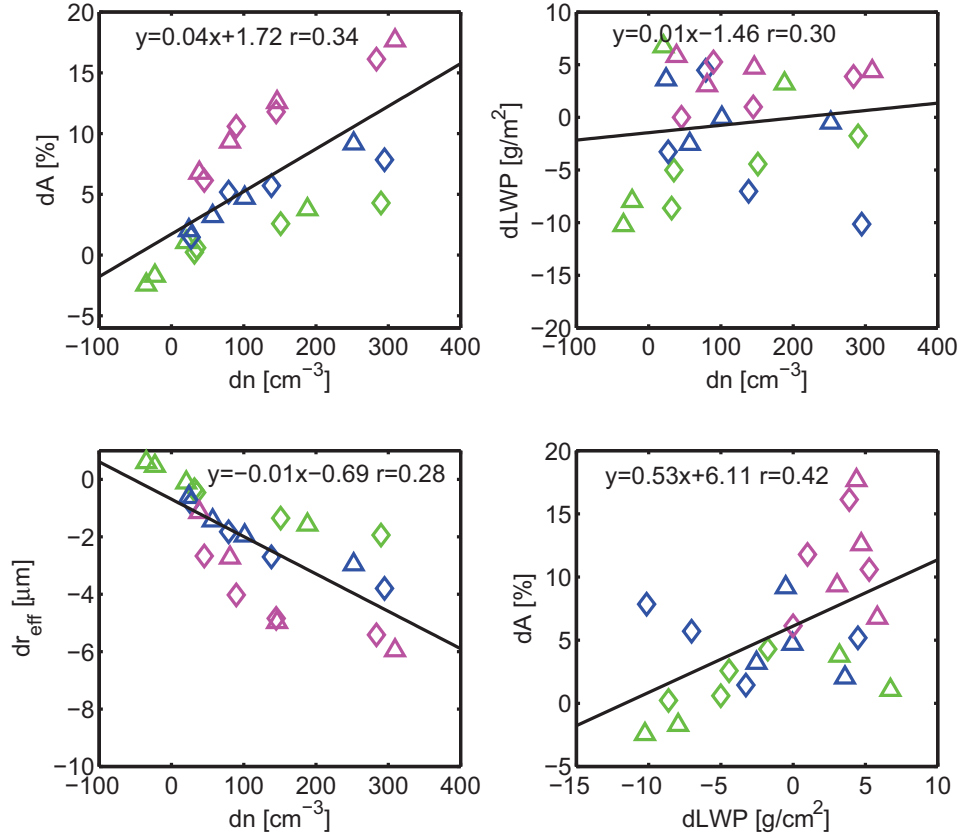


Figure 5: Relation between dn and dA - panel a, dn and $dLWP$ - panel b, dn and dr_{eff} - panel c and $dLWP$ and dA - panel d. Green symbols - perturbation runs with HIGH initial conditions, blue - perturbation runs with MED initial conditions, magenta symbols - perturbation runs with LOW initial conditions. Triangles - perturbing aerosol size $0.5 \mu\text{m}$, diamonds - perturbing aerosol size $0.1 \mu\text{m}$. Black lines - least square fit to all data points. Additionally each figure contains values of the fit and correlation coefficient for each relation.

Highlights:

Numerical simulations aiming to investigate one of the proposed Solar Radiation Management methods in an idealized setup show that:

- 1) the albedo may increase directly in response to additional aerosol in the boundary layer,
- 2) the magnitude of the increase depends on the microphysical properties of the existing cloud, and is larger for cloud characterized by low cloud droplet number,
- 3) for some cases, for clouds characterized by high cloud droplet number, seeding may lead to the decrease in albedo when too large radius of seeding aerosol is used.