



# Computational assessment of a proposed technique for global warming mitigation via albedo-enhancement of marine stratocumulus clouds

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Accepted 15 November 2005

## Abstract

A simplified version of the model of marine stratocumulus clouds developed by Bower, Jones and Choularton [Bower, K.N., Jones, A., and Choularton, T.W., 1999. A modeling study of aerosol processing by stratocumulus clouds and its impact on GCM parameterisations of cloud and aerosol. *Atmospheric Research*, Vol. 50, Nos. 3–4, The Great Dun Fell Experiment, 1995-special issue, 317–344.] was used to examine the sensitivity of the albedo-enhancement global warming mitigation scheme proposed by Latham [Latham, J., 1990. Control of global warming? *Nature* 347, 339–340; Latham, J., 2002. Amelioration of global warming by controlled enhancement of the albedo and longevity of low-level maritime clouds. *Atmos. Sci. Letters* (doi:10.1006/Asle.2002.0048).] to the cloud and environmental aerosol characteristics, as well as those of the seawater aerosol of salt-mass  $m_s$  and number concentration  $\Delta N$ , which—under the scheme—are adverbly introduced into the clouds. Values of albedo-change  $\Delta A$  and droplet number concentration  $N_d$  were calculated for a wide range of values of  $m_s$ ,  $\Delta N$ , updraught speed  $W$ , cloud thickness  $\Delta Z$  and cloud-base temperature  $T_B$ : for three measured aerosol spectra, corresponding to ambient air of negligible, moderate and high levels of pollution. Our choices of parameter value ranges were determined by the extent of their applicability to the mitigation scheme, whose current formulation is still somewhat preliminary, thus rendering unwarranted in this study the utilisation of refinements incorporated into other stratocumulus models.

In agreement with earlier studies: (1)  $\Delta A$  was found to be very sensitive to  $\Delta N$  and (within certain constraints) insensitive to changes in  $m_s$ ,  $W$ ,  $\Delta Z$  and  $T_B$ ; (2)  $\Delta A$  was greatest for clouds formed in pure air and least for highly polluted air. In many situations considered to be within the ambit of the mitigation scheme, the calculated  $\Delta A$  values exceeded those estimated by earlier workers as being necessary to produce a cooling sufficient to compensate, globally, for the warming resulting from a doubling of the atmospheric carbon dioxide concentration.

Our calculations provide quantitative support for the physical viability of the mitigation scheme and offer new insights into its technological requirements.

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**Keywords:** Climate; Modification; Albedo; CCN

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## 1. Introduction

Low-level marine stratocumulus clouds cover about a third of the oceanic surface and characteristically possess albedos,  $A$ , in the range 0.3 to 0.7 (Schwartz and Slingo, 1996). They therefore make a significant (cooling) contribution to the radiative balance of the Earth. Latham (1990, 2002) proposed a possible technique for ameliorating global warming by means of controlled enhancement of the droplet concentrations  $N_d$  in such clouds, with a corresponding increase  $\Delta A$  in their albedo (and also possibly in their longevity), thereby producing a cooling effect. The technique involves dissemination—at or close to the ocean surface—of seawater (NaCl) droplets around 1  $\mu\text{m}$  in size, which are sufficiently large to act as cloud condensation nuclei (CCN) when they rise into the bases of these clouds. The central physics behind this scheme, which have been authoritatively treated—both theoretically and experimentally—in a considerable number of studies (for example, Twomey, 1977, 1991; Charlson et al., 1987; Albrecht, 1989; Wigley, 1989; Slingo, 1990; Ackerman et al., 1993; Pincus and Baker, 1994; Rosenfeld, 2000; Brenguier et al., 2000; Peng et al., 2002; Stevens et al., 2005), is that an increase in droplet concentration  $N_d$  causes the cloud albedo to increase because the overall droplet surface area is enhanced and can increase cloud longevity (tantamount to increasing cloudiness) because the growth of cloud droplets by coalescence to form drizzle or raindrops—which often initiates cloud dissipation—is slowed down, since the droplets are smaller and the clouds correspondingly more stable. In some circumstances, increasing  $N_d$  may completely suppress the production of precipitation. Ship-tracks are, of course, examples of inadvertent and uncontrolled albedo increase in such clouds. Calculations by above-mentioned workers indicate that a 50–100% increase in  $N_d$ , in marine stratocumulus clouds, on a global basis, would be sufficient to produce a cooling, which would more than compensate for the warming (roughly  $4 \text{ W m}^{-2}$ ) associated with a doubling of the atmospheric carbon dioxide concentration. The studies of above-mentioned and other workers indicate that the associated changes  $\Delta A$  in planetary and top-of-cloud (marine stratocumulus) albedo are about 0.005 (1.5%) and 0.02, respectively. These calculations of required values of  $\Delta A$  take no account of possible further cooling resulting from increased cloud longevity/cover and may thus be regarded as upper limits.

Advertent generation of sea-salt CCN at the ocean surface, thereby enhancing  $N_d$ , is, of course, basically a

version of a process that happens naturally. Latham showed that the seawater dissemination rates required to produce the above-mentioned value of top-of-cloud  $\Delta A$  were modest (around  $10^8$  to  $10^9 \text{ kg h}^{-1}$  (globally) or  $0.1$  to  $1.0 \text{ kg h}^{-1} \text{ km}^{-2}$ ). This is because the required values of droplet size and associated salt-mass  $m_s$  are so small:  $m_s$  values as low as  $10^{-17} \text{ kg}$  (corresponding to a droplet diameter  $d$  of about  $0.8 \mu\text{m}$ ) being adequate for activation as CCN. Two advantages of this proposed global warming mitigation technique, if it were to be deployed operationally, are that: (1) albedo-control could be exercised by measuring  $A$  from satellites and switching seawater droplet disseminators on or off as required; (2) the only raw material needed is seawater, which would largely fall back into the oceans within a few days.

Further support for the quantitative validity of this scheme for global warming mitigation is provided by the computations of Jones et al. (submitted for publication) using the Hadley Centre's (UK Meteorological Office) HadGAM1 general circulation model (GCM). These indicate that warming due to a doubling of the carbon dioxide content of the atmosphere could be roughly compensated for—when taking account of the negative forcing due to the production of anthropogenic aerosol to date—by a doubling of the droplet number concentration  $N_d$  in three extensive regions of maritime stratocumulus clouds (off the West coasts of Africa and North and South America), which together cover about 3% of the global surface. If the anthropogenic aerosol factor is discounted,  $N_d$  would need to be roughly quadrupled. If only clouds covering this specially selected 3% of the Earth's surface were modified, instead of all marine stratocumulus clouds, the critical value of top-of-cloud albedo-change required to compensate for a doubling of carbon dioxide concentration would rise from 0.02 (mentioned earlier) to about 0.16. The associated values of enhanced  $A$  are within natural bounds.

The principal goal of the study described in this paper was to employ a simple model of marine stratocumulus to establish—over wide ranges of parameter values—the conditions under which this mitigation technique might be able to produce a cooling comparable with global warming. More specifically, we explore the sensitivity of albedo-change  $\Delta A$  to the properties of the natural aerosol, the clouds and the advertently created seawater aerosol introduced into them. (The calculations of Latham (1990, 2002) were much more limited and did not include estimates of  $\Delta A$ , while the GCM calculations of Jones et al. (submitted for publication) did not lend themselves to sensitivity calculations of this kind.)

It is important to stress that our primary objective was to assess the quantitative validity of the proposed global cooling technique, not to advance our understanding of the development and properties of marine stratocumulus clouds. A worthwhile study of the latter topic would have required a fully developed dynamical and chemical model with treatment of entrainment, drizzle formation, the possible interrelationship between the advertently added aerosol and cloud thickness, and other refinements. However, in our case, with our focus on the former topic, it was deemed adequate to utilise our much simpler model (with prescribed dynamics and no entrainment, droplet coalescence—and thus drizzle formation—or chemistry), on the grounds that the mitigation scheme is currently still at an early stage of development, and further sophistication in the modelling would not add significantly to our understanding. On the question of possible drizzle formation, we note that, since the adulterated cloud droplet size distributions are significantly narrower than the natural ones, the calculated values of  $\Delta A$  are likely to err on the side of being too low.

In Section 2, we present a brief description of our model, together with the ranges of parameter values employed, and details of the three (experimentally observed) aerosol spectra utilised. The results of our computations are presented and discussed in Section 3. In Section 4, we discuss the implications of these calculations with respect to the proposed albedo-enhancement global cooling scheme, and outline the current status of important technological and meteorological questions regarding it.

## 2. The model, input parameter values and aerosol spectra

In our modelling study, marine stratocumulus clouds (assumed to be adiabatic, of infinite width and in a steady-state condition) are formed in air whose natural aerosol characteristics are one of three specified alternatives, based on observation and corresponding, in turn, to negligible, modest or significant levels of pollution. In some further calculations, these clouds are inoculated with additional NaCl aerosol of prescribed concentration  $\Delta N$  and constant salt-mass  $m_s$ , which are added to the original aerosol spectrum. For all of these scenarios, calculations are made of the in-cloud vertical distributions of cloud droplet concentration  $N_d$ , liquid-water-content  $L$  and supersaturation  $S$ . We also compute the changes,  $\Delta A$ , in the top-of-cloud albedo, resulting from adding additional aerosol as described above. These calculations are made for a range of specified

values of cloud-base temperature,  $T_B$ , cloud thickness,  $\Delta Z$ , updraught speed,  $W$  (assumed constant with altitude),  $\Delta N$  and  $m_s$ .

Our model (Bower et al., 1999) calculates the growth of an aerosol spectrum of specified size distribution and chemical composition as it enters and passes through cloud. For the purposes of this work, it was assumed that species in the gas phase have no effect on the cloud formation and that the aerosol composition does not change throughout the experiment (i.e. the chemistry module was turned off). Observed aerosol size distributions (see below) were input to the model at their equilibrium wet size at 99% relative humidity, calculated assuming—for computational simplification—that the particles were comprised entirely of sodium chloride. The activation of the aerosol population and the subsequent cloud droplet growth was then calculated assuming a constant, specified updraught until cloud-top was reached. The cloud was assumed to be adiabatic and drizzle was not allowed to form.

Calculations of  $N$ ,  $S$ ,  $L$  and  $\Delta A$  were made for: four values of  $W$  (0.05, 0.2, 0.5 and  $1.0 \text{ m s}^{-1}$ ), three values of  $T_B$  ranging from 5 to 25 °C, and  $\Delta Z$  values of 100 and 210 m. Values of cloud-base height were set to 300 m in all model simulations. Values of albedo-change,  $\Delta A$ , were calculated from the droplet number concentrations using the method described by Schwartz and Slingo (1996).

The three aerosol size spectra (A, B, C) utilised in our computations are illustrated in Fig. 1. Spectrum B corresponds to a relatively typical clean Northern Hemisphere maritime aerosol sample measured during the ACE-2 experiment in summer 1997 (Bower et al., 2000). It was measured at Punta del Hidalgo on the North coast of Tenerife at 03:30 on 2 July 1997. Back-trajectory analysis indicated that the air had previously spent the last 6 days prior to arrival at the site over the open Atlantic Ocean to the northwest of the Canaries, and was therefore free of any recent anthropogenic influence from Europe. This spectrum, a combination of a size distribution measured by an electrical mobility sizing technique (using a Vienna Style Differential Mobility Particle Sizer, DMPS) in the size range 3–650 nm and a laser light scattering technique (using an Active Scattering Aerosol Spectrometer Probe, ASASP-X) in the size range 0.1–3.0  $\mu\text{m}$ , was used for most of the sensitivity tests carried out in the work described herein.

Spectrum A corresponds to an aerosol size distribution measured in extremely clean Southern Hemisphere maritime air from a German ship, Polarstern, as it cruised south in the southern Atlantic off the west coast

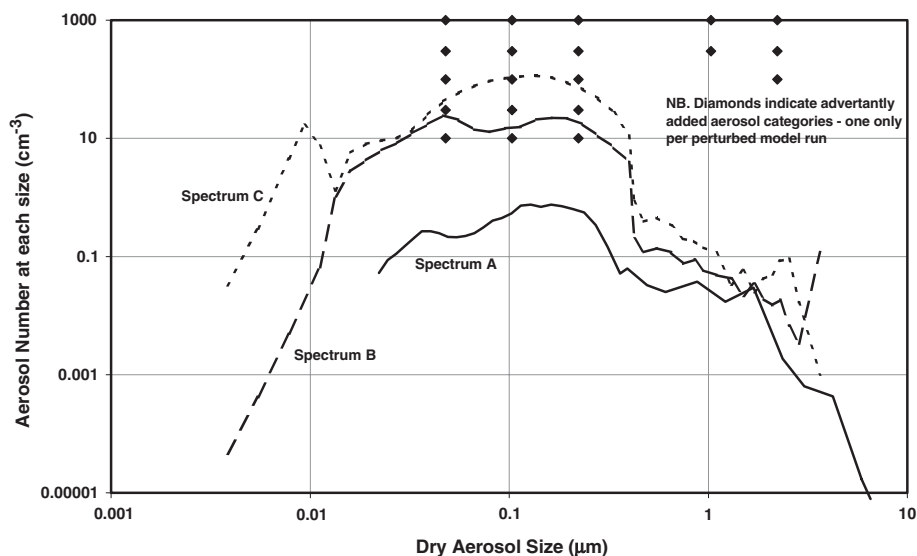


Fig. 1. The observed aerosol spectra used as inputs for the model runs. Spectrum A is for clean maritime conditions over the South Atlantic, spectrum B for unpolluted conditions in the subtropical North Atlantic and spectrum C for polluted conditions in the North Atlantic, the air having recently travelled over western Europe. Further details are given in the text.

of southern Africa en-route from Bremerhaven to Cape Town. This spectrum was again measured by a combination of electrical mobility and laser light scattering techniques as in ACE-2 (this time using a DMPS and a Grimm dust monitor Model 1.108). Spectrum A is an average of the distribution measured between midnight and 09:00 on 8 November 2003, at an average position of 8S, 5W. Back-trajectory analysis indicated that this air mass had previously traveled northwards, parallel to the coast of Africa from an area originating in the clean southern oceans hundreds of miles to the southeast of southern Africa. Thus, this air had been free of the influence of pollution over many days prior to our measurements.

Spectrum C was measured at 06:00 on 17 July 1997, again at the Punta del Hidalgo site on Tenerife during ACE-2. On this occasion, the air mass was classified as being moderately polluted and back trajectories indicated that the air had previously passed over parts of the Iberian Peninsula some 1.5–2 days earlier.

The air masses in which both spectra B and C were measured subsequently flowed up into and interacted with a hill cap cloud system in which cloud droplet number concentrations of 100–150 and 600–700  $\text{cm}^{-3}$ , respectively, were measured. By contrast, total droplet concentrations (over the described size ranges) observed during the period spectrum A was measured aboard the Polarstern were typically 7–10  $\text{cm}^{-3}$ .

In this modelling sensitivity study, the values of total cloud-top droplet number concentration  $N_d$  for aerosol

spectra A, B and C (for  $T_B=15\text{ }^\circ\text{C}$  and  $W=0.2\text{ m s}^{-1}$ ) are 8, 133 and 453  $\text{cm}^{-3}$ , respectively. For spectrum B, with  $U=1.0\text{ m s}^{-1}$  and  $T_B=15\text{ }^\circ\text{C}$ ,  $N_d=301\text{ cm}^{-3}$ . Although in the ACE-2 cases (curves B and C) the submicron particle aerosol composition was measured and was found to consist predominantly of ammonium sulphate mixed with some organic and nitrate as well as sodium chloride, tests showed that the effects of using the simplified 100% NaCl composition assumption on the results of the current work were small.

For calculations involving spectrum A, we employed five values of  $\Delta N$ , the advertantly added aerosol number concentration (in addition to zero). These are 10, 30, 100, 300 and 1000  $\text{cm}^{-3}$ . For spectra B and C (which contain many more cloud droplets than for spectrum A), we performed calculations only for the three highest of these values of  $\Delta N$ . For all three spectra,  $m_s$  values employed were  $10^{-18}$ ,  $10^{-17}$  and  $10^{-16}$  kg. However, for spectrum B,  $m_s$  values of  $10^{-15}$ ,  $10^{-14}$  and  $10^{-13}$  kg were also used. The solid diamonds displayed in Fig. 1 illustrate the above-mentioned totality of the number concentrations and dry sizes of the advertantly added aerosol.

### 3. Results of the computations: discussion

Table 1 presents, for all three aerosol spectra, calculated values of cloud droplet concentration  $N_d$ , maximum supersaturation  $S$  and top-of-cloud albedo-change  $\Delta A$  for the full range of values of additional

Table 1  
 Calculated values (for the three aerosol spectra, A, B and C) of total droplet number concentration  $N_d$  ( $\text{cm}^{-3}$ ), maximum supersaturation  $S$  (%) and albedo-increase  $\Delta A$ , for specified values of updraught speed  $W$  (m/s), salt-mass  $m_s$  (kg) and additional droplet concentration  $\Delta N$  ( $\text{cm}^{-3}$ )

Spectrum	$W$	$m_s = 10^{-18}$ kg				$m_s = 10^{-17}$ kg				$m_s = 10^{-16}$ kg							
		Base run	10	30	100	300	1000	10	30	100	300	1000	10	30	100	300	1000
A	$N_d$	8.23	17.8	37.3	106	301	986	17.6	36.9	105	300	984	17.6	36.7	105	299	981
	$S$ (%)	0.751	0.710	0.695	0.685	0.68	0.676	0.582	0.479	0.393	0.342	0.303	0.554	0.425	0.302	0.218	0.148
	$100\Delta A$	5.8	11.4	19.2	27.1	36	5.7	11.3	19.1	27	35.9	5.7	11.2	19.1	27	35.9	1040
B	$N_d$	133	133	133	133	133	0.307	0.307	0.298	0.289	0.274	0.251	0.251	0.202	0.143	0.143	15.47
	$S$ (%)	0.307	0.307	0.307	0.307	0.307	0	0	4.16	8.77	15.9	3.67	8.22	15.47	15.47	15.47	15.47
	$100\Delta A$	301	301	301	301	497	1183	277	473	1139	277	473	1139	277	453	1125	1125
C	$N_d$	0.733	0.733	0.733	0.733	0.729	0.723	0.664	0.591	0.498	0.623	0.505	0.623	0.505	0.354	0.354	0.354
	$S$ (%)	0.203	0.203	0.203	0.203	0.203	0.203	0	3.76	10.3	9.98	0.623	3.06	9.89	9.89	9.89	9.89
	$100\Delta A$	453	453	453	453	453	453	453	453	453	453	453	453	453	453	453	453

For all model runs,  $T_B=15$  °C,  $\Delta Z=210$  m and cloud-base is at 300 m.

aerosol concentration  $\Delta N$  and salt-mass  $m_s$  employed. These calculations are for the specimen values of  $T_B$ ,  $\Delta Z$  and  $W$  displayed in the table or its legend. Table 2 presents further results for spectrum B, with larger salt-mass values.

As mentioned earlier, the focus of this study is on our proposed albedo-change global warming mitigation technique, so we devote only cursory attention below—in our presentation and analysis of results—to features not concerned specifically with this technique, as these have already been treated in considerable detail—using more comprehensive models—by workers cited in Section 1. It is reassuring to note, however, that our general results are not inconsistent with those reported by these earlier workers.

Tables 1 and 2 show that, for the intermediate aerosol spectrum B, calculated values of albedo-change  $\Delta A$  and cloud droplet concentration  $N_d$  are insensitive to salt-mass  $m_s$  over the range  $10^{-17}$  to  $10^{-13}$  kg. Above  $m_s=10^{-17}$  kg, values of  $N_d$  decrease slightly with further increases in  $m_s$ , as these lead to an increased uptake of water vapour and a reduction in peak supersaturation, as shown in Tables 1 and 2. This in turn prevents the activation of the smallest previously activated aerosol in the base runs and in runs with smaller  $m_s$ . We see that, for the smallest salt-masses employed in our calculations, the highest supersaturations are achieved, since there is less competition for the available water vapour, and thus more CCN are activated. The concomitant increase in  $N_d$  is accompanied by an increase in  $\Delta A$  because of the associated increase in total cloud droplet surface area. Increasing the updraught speed  $W$  from 0.2 to 1.0 m/s results in an increase in cloud droplet concentration  $N_d$ , for both the unadulterated and adulterated clouds, with a corresponding slight diminution in the values of albedo-change  $\Delta A$ .

For  $m_s=10^{-18}$  kg, the lowest value of salt-mass employed, the advertently added CCN were activated only above a threshold updraught speed, which increased as the number of droplets in the unadulterated spectrum increased. In the case of spectrum B, this threshold was 0.5 m/s, for spectrum A it was 0.05 m/s and for spectrum C nucleation of the advertently added sea-salt aerosol did not occur for the range of  $W$  values utilised. We see from the table that, for spectrum B, with  $W=1.0$  m/s, the albedo-changes  $\Delta A$  for  $m_s=10^{-18}$  kg are slightly greater than those calculated for larger salt-masses (Ghan et al., 1998).

For spectrum A, corresponding to clouds formed in very pure air, Table 1 reveals that (for all values of  $\Delta N$ ) the values of  $\Delta A$  are insensitive to salt-mass  $m_s$  over the

Table 2

As for Table 1, except for spectrum B only, over the  $m_s$  range  $10^{-15}$  to  $10^{-13}$  kg

Spectrum	$W$	Base run	$m_s=10^{-15}$ kg					$m_s=10^{-14}$ kg					$m_s=10^{-13}$ kg							
			$\Delta N$ ( $\text{cm}^{-3}$ ) $\rightarrow$	0	10	30	100	300	1000	10	30	100	300	1000	10	30	100	300	1000	
B	0.2	$N_d$	133				201	354	1040				355	1040				159	333	1000
		100A					3.12	7.38	15.47				7.39	15.5				1.37	6.92	15.2

range  $10^{-18}$  to  $10^{-15}$  kg, and are typically several times greater (for the same values of  $\Delta N$ ) than those for spectrum B.

For spectrum C, corresponding to clouds formed in highly polluted air, the additional CCN were not nucleated for salt-masses  $m_s$  less than  $10^{-16}$  kg, and the  $\Delta A$  values, for conditions where nucleation occurred, were much lower—for the same values of other salient parameters—than those for aerosol spectra A and B.

Table 1 shows that, for all three aerosol spectra, the calculated values of albedo-change  $\Delta A$  and total droplet concentration  $N_d$  are highly sensitive to the imposed additional aerosol concentrations  $\Delta N$ , over the range of values employed. This is because, for almost all conditions specified in the table, all the adventerly introduced CCN are activated, with a concomitant significant increase in  $N_d$  and therefore  $\Delta A$ , for reasons given earlier.

A similar pattern of results to that portrayed in the preceding paragraph was found over the entire range of  $T_B$ ,  $\Delta Z$  and  $W$  employed.

The influence of cloud-base temperature  $T_B$  on albedo-change  $\Delta A$  is small,  $\Delta A$  increasing steadily with  $T_B$  presumably because the additional droplets grow to somewhat larger sizes, as a consequence of the greater amount of water vapour available.

The values of  $\Delta A$  were found to be larger (by a factor ranging from 1.3 to 2.0) for the clouds of depth  $\Delta Z=110$  m, compared with those for  $\Delta Z=210$  m, shown in the tables. The sensitivity of  $\Delta A$  to  $\Delta Z$  increases as  $\Delta Z$  decreases (e.g. Twomey, 1971).

Apart from defining, as described earlier, whether or not the adventerly added aerosol are nucleated, the influence of updraught speed  $W$  on albedo-change is significantly smaller than that of  $\Delta N$ , over the range of parameter values considered. However,  $\Delta A$  is found to decrease appreciably as  $W$  increases, presumably because fewer additional CCN are available to be activated. This interpretation is consistent with the observed reduction, as  $W$  increases, in the rate of production of new droplets. A typical result, for spectrum B, is that with  $\Delta N=1000 \text{ cm}^{-3}$ ,  $T_B=15 \text{ C}$ ,  $\Delta A$  decreases from 0.19 to 0.14 as  $W$  increases from 0.05 to 1.0 m/s.

We conclude from Tables 1 and 2 and the above discussion that the values of albedo-change  $\Delta A$  are much more sensitive to the number of additional CCN,  $\Delta N$ , than to any other parameter. The relation between  $\Delta N$  and  $\Delta A$  is seen to be strongly nonlinear (e.g. Twomey, 1991; Pincus and Baker, 1994). For example, from Table 1, for spectrum B, with  $U=1$  m/s and  $m_s=10^{-17}$  kg, we see that, for  $\Delta N=100 \text{ cm}^{-3}$ ,  $\Delta A=3.0$ , while for  $\Delta N=1000 \text{ cm}^{-3}$ ,  $\Delta A=13$ . The greater the number of droplets added, the slower the increase in  $\Delta A$ . This non-linearity is seen to exist for all spectra and cases displayed in the tables.

The tables also show that the albedo increases  $\Delta A$  resulting from advertent addition of seawater aerosol are by far the greatest for spectrum A, where the naturally occurring values of  $N$  are lowest: and are smallest for the most polluted air mass (spectrum C).

The general conformity between the pattern of results emanating from our model, and those which have been obtained from more rigorous and comprehensive models bolsters our confidence in the (approximate) quantitative validity of our values of albedo-change calculated for parameter values pertaining to the global warming mitigation technique, outlined earlier, which we now discuss in the light of these results.

#### 4. Application of calculations to proposed mitigation scheme: discussion

The calculations presented in Section 3 yield some significant implications with respect to technological aspects of the proposed global warming mitigation technique:

- Since—as long as activation of the added CCN occurs—the calculated albedo-changes are highly insensitive to the values of salt-mass  $m_s$ —and therefore seawater droplet size—for the range considered, the choice of  $m_s$  can, to a considerable extent, be dictated by technological convenience. It would probably be optimum for effective albedo-change to confine our salt-mass range to about  $10^{-17}$  to  $10^{-15}$  kg (corresponding to seawater droplets in the approximate size range 0.8 to 4  $\mu\text{m}$ ) because smaller

particles may not be nucleated and larger ones could act as ultra-giant nuclei (Johnson, 1982) and thus perhaps promote drizzle onset and concomitant cloud dissipation.

- b. In terms of influencing values of albedo-change,  $\Delta A$ , there is no advantage in striving for monodispersity of the seawater droplet spectrum: though we note that monodispersity is likely to enhance cloud stability and therefore longevity. As mentioned earlier, since the adulterated droplet size distributions are much narrower than the natural ones, the calculated values of  $\Delta A$  are likely to be underestimates.
- c. Table 1 reveals that for all cases considered in which advertently added CCN are activated, the top-of-cloud albedo increases  $\Delta A$  exceeded the above-mentioned critical value of 0.02 (which, as mentioned earlier, pertains to advertent adulteration of all marine stratocumulus clouds, covering about a quarter of the Earth's area (Charlson et al., 1987)), and that values of  $\Delta A$  can be achieved which—especially for spectrum A—are many times larger than this critical value. It follows that the areal fraction of maritime stratocumulus clouds which needs to be adulterated in order to compensate for warming resulting from the burning of fossil fuels can be correspondingly lower than unity; therefore, rendering less daunting the practical problem of achieving adequate lateral dispersal of advertently disseminated seawater CCN. The GCM calculations of Jones et al. (submitted for publication), also mentioned earlier, yielded a similar conclusion, with adequate cooling being predicted for an areal fraction of about 0.03.
- d. A possible consequence of the fact that the largest albedo-changes occur for clouds formed in pure air (spectrum A) is that it may prove helpful—in any practical attempt to enhance cloud albedo—to focus attention on pure oceanic regions/clouds: however, we envisage that the choice of cloud regions to be adulterated would be largely dictated by their extensiveness (Jones et al., submitted for publication). We note also that the non-linearity between  $\Delta A$  and  $\Delta N$  (e.g. Twomey, 1991) illustrated in Table 1 and discussed earlier suggests that, in order to achieve the degree of cooling required, it may prove adequate, in practice, to enhance the cloud droplet concentration  $N_d$  by a significantly smaller factor than previously envisaged.

We conclude, therefore, that the calculations presented herein provide support for the quantitative validity of our proposed mitigation scheme, and reveal some degree of easement of requirements and additional

flexibility which may prove useful practically. However, many problems would need to be satisfactorily resolved before justification would exist for the deployment of an operational system on the massive scale required. Most of these could be classified as either technological or meteorological and we outline important ones in each category below.

Two crucial technological questions so far unanswered are: (a) how do we produce the seawater aerosol of the required sizes and number concentrations? (b) How do we disseminate these particles to ensure that sufficient numbers of them enter the clouds to be adulterated?

There are, under examination, several possible techniques for atomising seawater to produce droplets of the size and quantity required.

Ultrasonic excitation of a liquid using a piezo-electric transducer (Barreras et al., 2002) will induce Faraday waves at a free surface. If the wave steepness is sufficient, drops will be thrown from the wave crests. The spread of drop diameters is acceptably narrow and the mean diameter can be predicted from the frequency of excitation. Frequencies of several megahertz would be needed. Small commercial units are available (APC, 2005), but unfortunately the energy used is about 300 kJ/kg of water sprayed. This is very much higher than would be predicted for the creation of the new surface area against surface tension, which would be only 470 J/kg for 1  $\mu\text{m}$  sized drops. The difference may be because the viscous losses in high frequency capillary waves are very high. There may be many crests, which are dissipating energy but only a few crests with steepness above the ejection threshold. Lozano et al. (2003) suggest lower energy requirements of only 20 kJ/kg for pulsed excitation. It may be possible to use transducers with a cylindrical surface to concentrate energy to a smaller region and also to combine ultrasonics with electrostatic methods using a charged knife-edge at the focus of the transducer cylinder.

A novel technique involving electrostatic atomisation of seawater drops appears highly promising but requires more work.

The spray platforms will have to be dispersed over wide oceanic areas, operate for long periods and have sufficient mobility to allow them to follow changing weather patterns, return to land for maintenance and avoid the cost of anchoring. One possibility is to use unmanned sailing vessels which would move back and forth across a track perpendicular to the prevailing wind direction, changing position from time to time as redirected by satellite communications. The problems of trimming cloth sails and handling ropes would be

avoided by the use of Flettner rotors for propulsion. These are vertical spinning cylinders which develop very large forces perpendicular to the apparent wind direction with aerodynamic lift coefficients up to 9, far above that of any sail. The original Flettner ship Buchau, renamed Baden-Baden, was launched in 1924 and made a successful Atlantic crossing in 1926 (Seufert and Seufert, 1983). Even though the rotors were made from steel, they were only one quarter the weight of the previous rig. Modern materials would give an even greater advantage. The system allowed Baden-Baden to sail 20° closer to the wind than a conventional sailing ship, apply ‘brakes’, make turns about her own centre and go directly into reverse. This manoeuvrability would be useful for directing the spray dissemination vessels as required. A spinning cylinder allows the effects of rolling motions of the vessel to be swamped by centrifugal acceleration if the spray source is built into the inner, vertical wall of a Flettner drum. Energy for the spray mechanism and drum drive could come from a larger-than-normal ‘propeller’. It would normally be used in reverse as a turbine, taking energy from the forward motion of the vessel, but could also drive it using battery power for delicate manoeuvres in windless conditions. At moderate speeds, batteries can give 24 h endurance. The power level at the cut-out wind speed of 12 m/s would be of the order of 1 MW.

Latham (2002) identified and/or examined a number of unresolved meteorological/scientific issues which require further attention. His analysis, not repeated here, concerned: aerosol losses between creation and cloud entry; drizzle formation and its impact on cloud longevity; the role of ultra-giant nuclei; the impact of advertent aerosol dissemination on the exchange of heat and water vapour; influence of the disseminated aerosol on higher-level clouds, particularly their production of precipitation; effects (which might be multifarious and profound) of inevitable modification to the temperature structure of the atmosphere. We could add other issues to this list, including: radiative effects associated with aerosol which enter the atmosphere but not clouds and meteorological changes associated with a cooling which occurs predominantly in the Southern Hemisphere. All these, and others, need to be addressed before we can establish whether the proposed mitigation technique is operationally feasible and safe (and if so, for what optimal conditions).

Our view regarding priorities for future work is that in the immediate future we should focus attention on the technological and meteorological issues outlined earlier in this section, while simultaneously planning and executing a limited-area field experiment in which

selected clouds in a field of marine stratocumuli are inoculated with seawater aerosol, as proposed earlier, and airborne and satellite measurements are made to establish, quantitatively, the concomitant microphysical and radiative differences between adulterated and unaffected adjacent clouds.

### Acknowledgements

Part of this work was conducted while JL was in receipt of a Tyndall Fellowship.

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