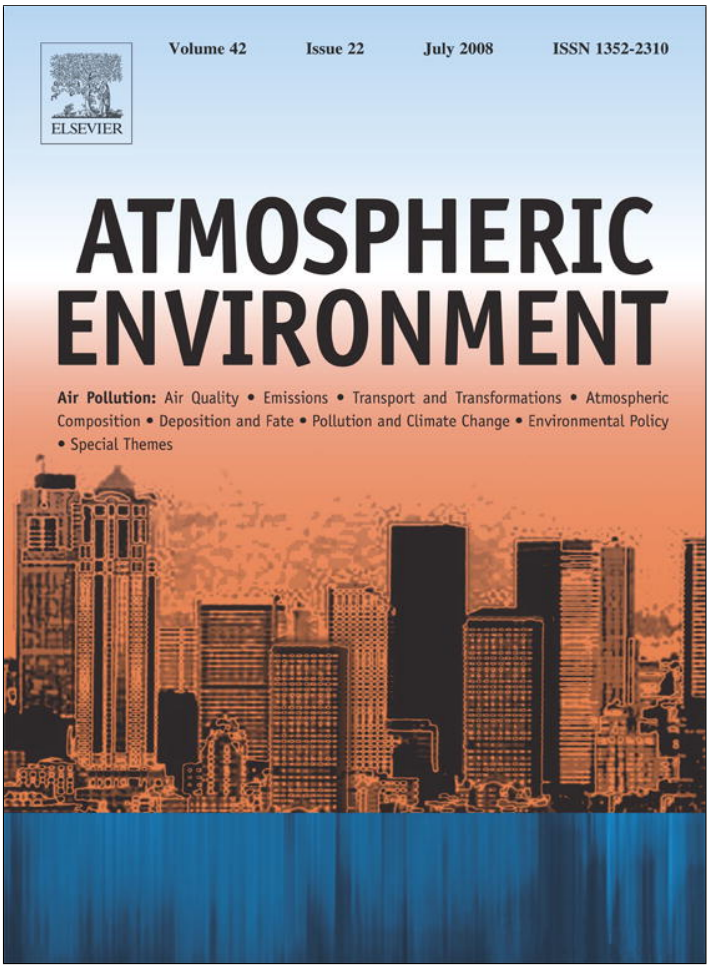

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NEW DIRECTIONS

Atmospheric Environment 42 (2008) 5728–5730

ATMOSPHERIC ENVIRONMENT

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New Directions: The impact of oceanic iron fertilisation on cloud condensation nuclei ☆

There is a growing interest in iron fertilisation of the oceans as a means of mitigating the effects of increased greenhouse gas concentrations. For example, Wingenter et al. (2007) proposed adding iron to an area of the Southern Ocean to encourage phytoplankton growth and stimulate the production of oceanic dimethyl-sulphide (DMS), thereby leading to a surface cooling due to enhanced atmospheric cloud condensation nuclei (CCN) concentrations. Clouds become more reflective in conditions of elevated CCN concentrations through the indirect aerosol effects of increased droplet number, decreased diameter and increased cloud lifetime (given constant cloud liquid water content). The Southern Ocean is a particularly good region in which to undertake such an iron enrichment exercise because much of the water is iron limited and there are relatively few CCN in the atmosphere. The role of DMS in controlling albedo and hence climate has been the subject of much debate and research since the so-called CLAW paper by Charlson et al. (1987). This method of artificial climate control is in contrast to earlier suggestions involving iron fertilisation, which rely on increased oceanic CO_2 uptake. The long-term sequestration of carbon from this has been repeatedly questioned.

Here we present results from a global aerosol model in which DMS emissions have been perturbed in a small region of the Southern Ocean. The GLOMAP-mode model (Manktelow et al., 2007) contains a detailed treatment of aerosol microphysics, including coagulation, condensation, cloud processing and new particle formation with parameterised gas phase sulphur chemistry. The model is driven by ECMWF reanalysed meteorology. Global monthly mean seawater DMS concentrations are prescribed from Kettle and Andreae (2000).

To investigate the impact on CCN concentrations, a perturbation to DMS emissions is applied in the model. Seawater DMS concentrations are increased by factors of 5 (as observed in the Southern Ocean Iron Experiment, SOFeX), 10 and 20, in a patch centred on 56.2°S, 172.0°W (as in SOFeX) covering $2.3 \times 10^6\,km^2$ (5% of the Southern Ocean, assumed to be the oceans south of 50°S). We prescribe this patch in the model at the same concentration for the entire month of December 1999, having been spun up for 2 months without the patch from 1 October.

In the atmosphere, DMS can be oxidised to SO_2, which can dissolve in aerosol/cloud droplets, where it is rapidly oxidised to S(VI) or further oxidised in air to form sulphuric acid (H_2SO_4) vapour. There are then two competing processes for this H_2SO_4 vapour; it can nucleate to form new aerosol particles, which subsequently grow by coagulation and condensation, or can condense on to existing particles, growing them. Both of these pathways can lead to increased numbers of CCN, although new particle formation in the free troposphere has been shown to be the main microphysical pathway by which DMS affects CCN (Korhonen et al., 2008).

The GLOMAP-mode model calculates the impact of DMS on the number of CCN (Figs. 1a, b and Table 1), mechanistically simulating the above processes on a global scale, alongside the atmospheric cycle of other marine species such as sea salt. The most striking feature of the simulation is the position

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1352-2310/$ - see front matter
doi:10.1016/j.atmosenv.2008.05.005

of the extra CCN, which occurs several thousand kilometres downwind of the perturbed DMS patch. The remote impact is due to the several days required for conversion of DMS into CCN.

Even with the most optimistic definition of CCN (aerosol particles with dry diameters greater than 50 nm, corresponding to a cloud supersaturation of 0.36%) the increase in the number of CCN is only 1.4% over the Southern Ocean, substantially lesser than predicted by Wingenter et al. (2007). For the chosen size and geometry of the patch, the maximum increase in CCN over the patch is 4.5% (0.9 CCN cm^{-3}) compared to a maximum increase downwind of 11.0% (1.8 CCN cm^{-3}). When a larger activation diameter is assumed (diameter >70 nm, corresponding to a supersaturation $= 0.21\%$), the impact on CCN is greatly reduced to a fraction of a per cent over the Southern Ocean.

The two processes that lead to aerosol particles reaching CCN sizes have very different spatial

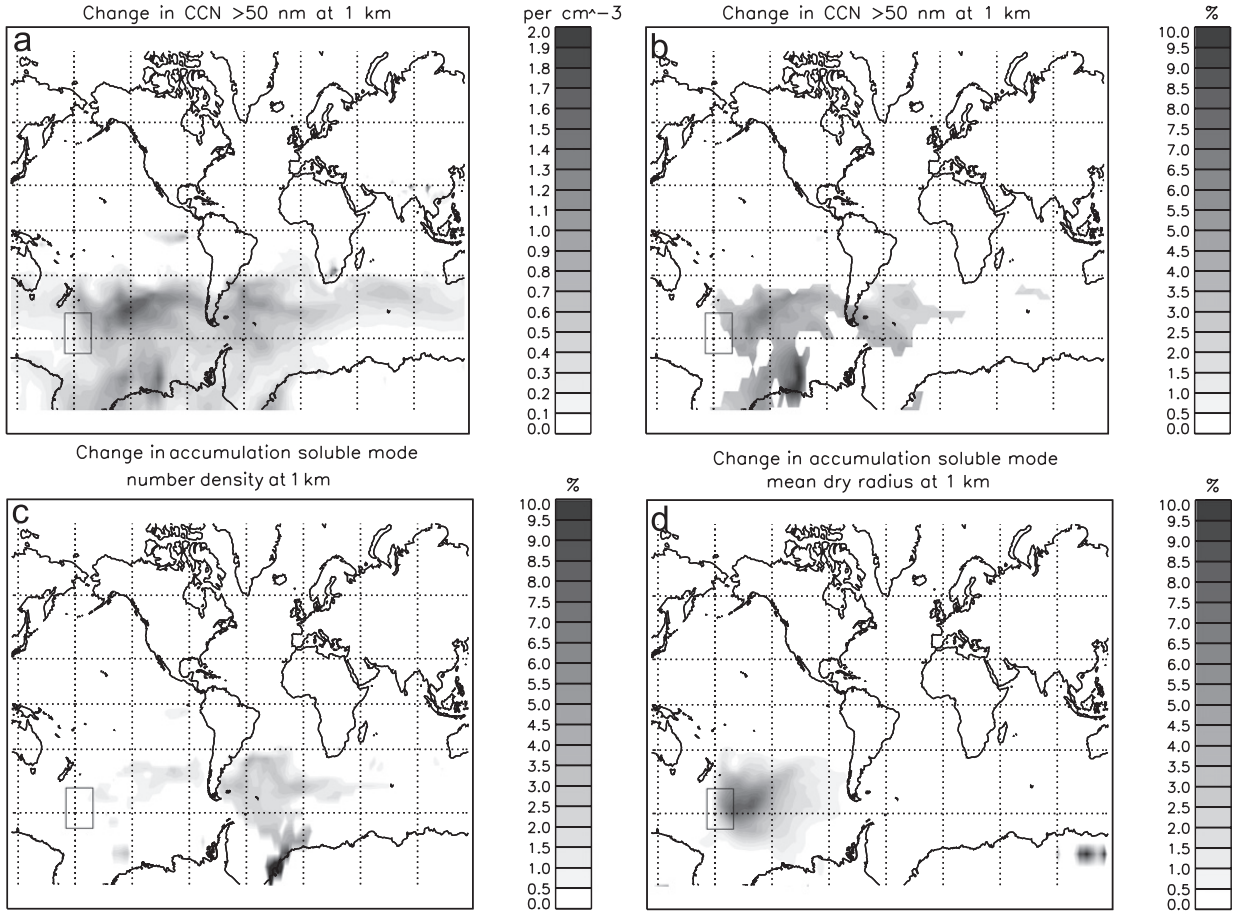

Fig. 1. Impact of the 5× DMS patch on aerosol at 1 km altitude compared to a baseline case. The patch is marked with a black rectangle. (a) Absolute change in the number of CCN >50 nm diameter. (b) Relative change in the number of CCN >50 nm. (c) Relative change in accumulation mode number density. (d) Relative change in accumulation mode mean dry radius.

Table 1
The relative change in DMS flux and CCN over the Southern Ocean (global changes in parentheses), as predicted by GLOMAP-mode for three difference patch concentrations (5×, 10× and 20× normal DMS concentration) at 1 km altitude

	Model 5× patch	Model 10× patch	Model 20× patch
DMS flux change	+8.0% (+1.0%)	+18.0% (+2.3%)	+38.0% (+4.8%)
CCN (>50 nm) change	+1.39% (+0.26%)	+5.35% (+1.05%)	+15.87% (+3.07%)
CCN (>70 nm) change	+0.04% (+0.01%)	+1.73% (+0.36%)	+9.46% (2.14%)

signals in the patch experiment. Nucleation of new aerosol from sulphuric acid occurs mainly in the free troposphere. Production of CCN via this mechanism takes several days, involving vertical transport of DMS, its oxidation, particle nucleation, growth and re-entrainment of particles into the boundary layer. The aerosol is advected large distances during this time and shows up as an increase in the accumulation mode number concentration at 1 km altitude (Fig. 1c). In contrast, condensational growth of existing particles occurs as soon as DMS is oxidised to H_2SO_4 via SO_2 in the marine boundary layer and shows up as an increase in the size of accumulation mode particles quite close to the DMS patch (Fig. 1d). Changes in CCN (Figs. 1a, b) result from these combined changes in particle properties.

The conversion of DMS (and other precursor gases) to aerosol, capable of serving as CCN, involves a spatially and temporally variable set of processes and results in spatially inhomogeneous changes in CCN. An experimental confirmation of the effectiveness of iron fertilisation would need to take account of these complex changes in CCN, which are not related to changes in DMS by a fixed 'production efficiency'.

In summary, by using a microphysical size-resolved global aerosol model, we find that even with an optimistic fivefold enhancement in DMS over an iron-perturbed patch, CCN concentrations over the Southern Ocean would be enhanced by just 1.4%, a factor of 8 less than the 10% estimated by Wingenter et al. (2007). As knowledge develops we should be prepared to re-evaluate these numbers, also taking into account possible changes in primary organic carbon aerosol.

References

Charlson, R.J., Lovelock, J.E., Andreae, M.O., Warren, S.G., 1987. Oceanic phytoplankton, atmospheric sulfur, cloud albedo and climate. Nature 326, 655–661.

Kettle, A.J., Andreae, M.O., 2000. Flux of dimethylsulfide from the oceans: a comparison of updated data sets and flux models. Journal of Geophysical Research—Atmospheres 105, 26793–26808.

Korhonen, H., Carslaw, K.S., Spracklen, D.V., Mann, G.W., Woodhouse, M.T., 2008. Influence of oceanic DMS emissions on CCN concentrations and seasonality over the remote southern hemisphere oceans: a global model study. Journal of Geophysical Research—Atmospheres, in press, doi:10.1029/2007JD009718.

Manktelow, P.T., Mann, G.W., Carslaw, K.S., Spracklen, D.V., Chipperfield, M.P., 2007. Regional and global trends in sulfate aerosol since the 1980s. Geophysical Research Letters 34, L14803.

Wingenter, O.W., Elliot, S.M., Blake, D.R., 2007. New Directions: enhancing the natural sulfur cycle to slow global warming. Atmospheric Environment 41, 7373–7375.

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