Climate model studies of sulphate aerosols and clouds

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SUMMARY

It is generally believed that increases in the atmospheric concentrations of greenhouse gases as a result of man's activities are leading to global warming. It is also believed that the same activities lead to increasing concentrations of sulphate aerosol, which act to cool the climate system and ameliorate the warming. The sulphate cooling may be separated into the direct effect in cloud-free regions and the indirect effect in cloudy regions. This paper summarizes recent work at the Hadley Centre on the indirect effect. Results from two versions of the Hadley Centre Climate Model are shown, using various parameterizations linking the amount of sulphate aerosol and the number concentration of droplets in water clouds. These results illustrate the considerable uncertainties in estimating the indirect effect. It is shown that other naturally occurring components of the aerosol population, in particular sea salt, may be important in reducing the magnitude of the indirect effect.

1. INTRODUCTION

Studies of the impact of human activities on the climate system have identified two opposing forcing factors. The burning of fossil fuels and other industrial activities are increasing the concentrations of greenhouse gases in the atmosphere, leading to global warming. On the other hand, the same activities also lead to increases in the concentration of suspended aerosol particles, which act to cool the system. In the 1970s, concern over the cooling by aerosols led to suggestions that this could hasten the onset of the next ice age. The rapid rise in global temperatures during the 1980s shifted attention to the greenhouse gases, which became the main focus for research. It is now believed that greenhouse gas warming is indeed dominant, but that the aerosol influence leads to a significant moderation of the global warming (IPCC 1996).

The greenhouse gases produced by human activities are long-lived and so are well mixed in the atmosphere. A relatively small number of measurements therefore serves to characterize their distribution and increase with time. In contrast, aerosols have a residence time of only a few days, so their distribution in space and time is extremely heterogeneous. This enormously complicates the calculation of the effect of aerosols on climate, since the global distribution is poorly observed and difficult to model. In considering the impact of atmospheric aerosols on climate, it is conventional to separate the *direct effect* of aerosols on the distribution of radiative heating from the *indirect effect* on the properties of clouds (Charlson *et al.* 1992). The direct effect occurs through the enhanced reflection of sunlight back to space. Provided that the distribution and the chemical and physical composition of the aerosol are known (a formidable requirement), it is a straightforward exercise to calculate the direct radiative forcing. The indirect effect occurs because the water droplets or ice crystals of which clouds are

composed are formed by condensation on a subset of the aerosol population. Changes in the aerosol can alter the size distribution of the droplets or crystals and indirectly modulate the distribution of radiative heating by altering the cloud radiative properties, precipitation efficiency and evolution. The particular difficulty with calculating the indirect effect is that it depends not only on the uncertain distribution and composition of the aerosol, but also on its interaction with clouds and on the subsequent cloud evolution. While all aerosol particles in clear air contribute to the direct effect, only a fraction are activated to become cloud drops, and that fraction depends on many factors which are poorly understood. Estimates of the indirect effect are thus extremely uncertain, as is emphasized in recent reviews (IPCC 1995; Charlson & Heintzenberg 1995; Schwartz & Slingo 1996). This paper provides an update to the work of Jones *et al.* (1994) and Jones & Slingo (1996), who estimated the magnitude and distribution of the indirect effect using the Hadley Centre Climate Model. Much of the emphasis is on the contribution from anthropogenic sulphate aerosol, but because of the competition between different aerosol species in the growth of aerosol to become cloud drops, it is also necessary to consider the importance of other components of the aerosol population, whether anthropogenic or natural in origin. As an example, the importance of naturally occurring sea salt aerosol over the oceans is considered.

2. EXPERIMENT DESCRIPTION

The method used to estimate the indirect effect of anthropogenic sulphate aerosols using a climate model is explained in Jones *et al.* (1994) and Jones & Slingo (1996), and only a brief description will be given here. The model used was the Hadley Centre Climate Model, a version of the UK Meteorological Office's Unified Model (UM; Cullen 1993). The important

Figure 1. Distributions of annual mean indirect radiative forcing by anthropogenic sulphate aerosol from the pre-industrial era to date (Wm^{-2}) using the modelled sulphate distributions of Langner & Rodhe (1991): (a) as estimated by Jones *et al.* (1994); (b) as estimated by Jones & Slingo (1996) using the relation of Jones *et al.* (equation (1)); (c) as (b), but using the relation of Boucher & Lohmann (1995) (equation (2)). Contours are at -0.5 , -1.0 , -3.0 , and -5.0 Wm^{-2} ; regions between -0.5 and -3.0 are lightly shaded, with regions of greater (more negative) forcing heavily shaded.

microphysical parameter to model is the cloud droplet effective radius r_e (Hansen & Travis 1974) which, along with the cloud liquid water path, determines the optical properties of clouds, of which the albedo or reflectivity is the most relevant for estimating the indirect effect. The calculation of the effective radius

for liquid stratiform cloud particles is based on the parameterization of Martin *et al.* (1994), and for convective clouds on Bower & Choulaton (1992) and Bower *et al.* (1994). All ice cloud particles are prescribed a fixed effective radius of $30 \mu\text{m}$, and so make no contribution to the indirect forcing.

Figure 2. As figure 1, but using the HadAM2b version of the Hadley Centre model: (a) using the relation of Jones *et al.* (equation (1)); (b) using the relation of Boucher & Lohmann (1995) (equation (2)).

Estimating the indirect effect of sulphate aerosols since the beginning of the industrial era requires a knowledge of both the current and the pre-industrial distribution of sulphate aerosols. Since such data are not available from observations (even for the present-day), it is necessary to rely on modelled distributions. The data we have used are annual average distributions of sulphate aerosol burdens from a three-dimensional chemical transport model (Langner & Rodhe 1991). This model was run with inputs from both natural and anthropogenic sources (for the 'present-day' scenario), and also using natural sources only (for the 'pre-industrial' scenario).

The key factor in being able to calculate the cloud droplet effective radius is to determine the distribution of the cloud droplet number concentration N_{tot} . In reality, N_{tot} depends on the distribution of cloud condensation nuclei (CCN), which are a subset of the total aerosol population, and consist of many different components. We have had to make the simplifying assumption that N_{tot} is determined solely by the concentration of sulphate aerosol, and it is this connection between sulphate and droplet concentrations which is one of the largest uncertainties in making estimates of the indirect effect.

Indirect forcing is estimated by performing a one-year run of the model (forced with climatological sea

surface temperatures) using the present-day distribution of sulphate. To sample both the annual and diurnal cycles at each timestep (30 min) during this run an additional purely diagnostic call is made to the model's radiation scheme using the pre-industrial aerosol as input. This methodology ensures that no feedbacks can occur, and so the difference in the diagnostic output from the two radiation scheme allows the indirect forcing to be calculated.

3. RESULTS AND DISCUSSION

The first estimate of the indirect forcing of anthropogenic sulphate aerosols obtained using the UM was a global annual mean of -1.26 Wm^{-2} (Jones *et al.* 1994). This estimate was obtained using a low horizontal resolution ($5^\circ \times 7.5^\circ$) configuration of a version of the model known as HadAM2a and used a rather crude method for distributing the sulphate aerosol burden in the vertical. The estimate used measurements made by the UK Meteorological Office's Meteorological Research Flight (MRF) to develop a relation between aerosol number concentration (A) and droplet concentration:

$$N_{\text{tot}} = 375 (1 - \exp(-2.5 \times 10^{-3} A)), \quad (1)$$

where both A and N_{tot} are given in units of cm^{-3} . To obtain a value for A , it was assumed that all the aerosol

Figure 3. Relations used between (a) sulphate aerosol mass concentration m (μg of sulphate ions per m³) and droplet concentration N_{tot} (cm⁻³), and (b) sulphate aerosol number concentration A (number of particles of (NH₄)₂SO₄ per cm³) and N_{tot} . In (b), the data points plotted are those obtained by the Meteorological Research Flight, reanalysed since Martin *et al.* (1994); the diamonds are from maritime air masses, and the crosses from continental air masses.

present was ammonium sulphate having a log-normal size distribution with a median particle radius of 0.05 μm and a standard deviation of 2 (e.g. Kiehl & Briegleb 1993). This single relation (equation (1)) was used for the whole globe, and the resulting distribution of the forcing is shown in figure 1a. The forcing tends to be largest in those regions where there are both significant anthropogenic sulphate emissions and large amounts of low-level (i.e. liquid phase) cloud which is unobscured by higher-level ice cloud.

The more recent estimates of Jones & Slingo (1996) used an updated version of the HadAM2a model and a better method for distributing the aerosol in the vertical. This method tended to reduce the aerosol concentrations in the boundary layer, and the resulting increase in cloud susceptibility (the change in cloud albedo per unit change of droplet concentration; Twomey 1991) led to an increase of about 15% in the estimate of indirect forcing to -1.50 Wm^{-2} , as shown in figure 1b.

One of the other methods for determining N_{tot} evaluated by Jones & Slingo (1996) was to use a relation by Boucher & Lohmann (1995), which related N_{tot} directly to sulphate mass, m :

$$N_{\text{tot}} = 10^{2.21+0.41 \log(m)}, \quad (2)$$

where m is specified in μg (SO₄²⁻) m⁻³ and can be

determined directly from the sulphate distributions without any assumption regarding the aerosol size spectrum. This parameterization gave a considerably lower indirect forcing estimate of -0.61 Wm^{-2} compared with that obtained using equation 1, although with a similar distribution (see figure 1c).

More recent work on the indirect effect has used the next version of the climate model, known as HadAM2b. This incorporates a new radiation scheme (Edwards & Slingo 1996) and the horizontal resolution has been increased to $2.5^\circ \times 3.75^\circ$. These changes reduced the magnitude of the indirect effect by about 10% to -1.37 and -0.55 Wm^{-2} for the Jones *et al.* and Boucher & Lohmann parameterizations respectively, as shown in figures 2a and 2b. Updating the treatment of convective cloud effective radius to be consistent with the results of Bower *et al.* (1994), as opposed to the earlier work of Bower & Choulaton (1992), made only very slight changes to the indirect effect (-1.37 and -1.35 Wm^{-2} , and -0.553 and -0.552 Wm^{-2} ; results not shown).

The radiation scheme used in the earlier work had no dependence on r_e in the longwave (thermal) part of the spectrum and so the indirect forcing was calculated purely in terms of the change in the reflected shortwave radiation. However, the new radiation scheme deals with both shortwave (0.2–5 μm wavelength) and longwave (3–1000 μm) regions of the spectrum identically, and so experiments were conducted to assess the impact of including the changes of r_e in the longwave as well as in the shortwave. Inclusion of indirect aerosol effects in the longwave decrease the global annual mean indirect forcing by between 1 and 2% depending on the N_{tot} parameterization used. Given the large uncertainties in calculating the indirect effect, it is clearly an acceptable approximation to ignore the contribution from the longwave effects.

There is considerable evidence that droplet concentrations are fairly well correlated with sulphate aerosol concentrations in maritime air masses (Charlson *et al.* 1987; Andreae *et al.* 1995), and so using relations like equation (1) to determine N_{tot} from sulphate distributions may be reasonable over the ocean. However, such a link between N_{tot} and sulphate seems far less clear in continental air, which has much higher aerosol concentrations composed of a wide variety of aerosol types (Novakov & Penner 1993). Consequently, using a single relation such as equations (1) or (2) is likely to overemphasize the dependence of N_{tot} on sulphate aerosol concentration. In an attempt to take some account of the difference between maritime and continental air masses in a version of the ECHAM model, Lohmann & Roeckner (1995) use a version of the parameterization of Boucher & Lohmann (1995) which uses different relations for N_{tot} depending on whether the clouds are over land or sea, as opposed to the single relation (equation (2)) presented above.

$$\begin{aligned} N_{\text{tot}}(\text{land}) &= 10^{2.24+0.257 \log(m)} \\ N_{\text{tot}}(\text{sea}) &= 10^{2.06+0.481 \log(m)}. \end{aligned} \quad (3)$$

These relations, along with the single relation of Boucher & Lohmann used previously (equation (2)), are shown in figure 3a. In a similar approach based on

Figure 4. Distribution of annual mean liquid cloud droplet effective radius (μm) (a) from the satellite retrievals of Han *et al.* (1994); (b) from HadAM2b using a single relation (equation (1)) to determine N_{tot} for all areas; (c) as (b), but using equation (4) to determine N_{tot} over land. All data are presented over the same domain as the satellite retrievals. Values less than $8 \mu\text{m}$ are hatched, and those greater than $12 \mu\text{m}$ are stippled.

MRF observations (which have been reanalysed since Martin *et al.* 1994) we have considered the maritime and continental data separately and obtained a new relation for continental clouds.

$$N_{\text{tot}}(\text{cont}) = -7.55 \times 10^{-5} A^2 + 0.252 A + 171.3. \quad (4)$$

This relation is shown in figure 3b, along with that of Jones *et al.* (equation (1)). The use of equation (4) makes droplet concentration much less sensitive to sulphate concentration over land, as observed (Choulaton & Bower, this volume). Equation (1) has been retained for maritime clouds, as it is practically indistinguishable from a quadratic fit to the maritime data within the range of these data (A less than about 300 cm^{-3}), and moreover it does not suffer from the problems of such a fit at higher values of A (values of N_{tot} with a quadratic fit begin decreasing with increasing A for A greater than about 700 cm^{-3}). Although equating clouds over land or sea with clouds in continental or maritime air masses is not ideal, it is

nevertheless a useful distinction. Indeed, we already differentiate between clouds over land and sea in choosing the value of the dispersion parameter k used in the effective radius parameterization (Martin *et al.* 1994), and also for the cloud depth required for convective cloud to be deemed 'deep convection' (and hence having a fixed effective radius; Bower *et al.* 1994).

Using such parameterizations in the UM accentuates the land–sea contrast of effective radius, and tends to improve the distribution of effective radius when compared with the satellite retrievals of Han *et al.* (1994), as shown in figure 4. Indirect forcing over land is significantly reduced in both cases, as shown in figure 5. The global annual mean forcing obtained using equations (1) and (4) is reduced to -0.88 Wm^{-2} ; however, with the relations of Boucher & Lohmann there is a compensation of the effects over land and sea (compared with using a single relation), and the global mean forcing is essentially unchanged at -0.55 Wm^{-2} .

Figure 5. Indirect forcing estimated using different relations for N_{tot} for land and ocean areas; contour intervals are as in figure 1: (a) using equation (1) for the ocean and equation (4) for land; (b) using the relations in equation (3) (Boucher & Lohmann 1995) as used by Lohmann & Roeckner (1995).

Figure 6. Estimate of the change in indirect anthropogenic sulphate aerosol forcing between the present-day and the year 2050 under IPCC scenario IS92a, using equations (1) and (4) to determine N_{tot} . Contours are at 0.0, -0.5, -1.0, -3.0 and -5.0 Wm^{-2} ; regions between -0.5 and -3.0 are lightly shaded, regions of greater (more negative) forcing are heavily shaded. Regions where the forcing is positive (i.e. a decrease in indirect forcing between the present-day and 2050) are hatched.

Figure 7. Change in indirect forcing on the incorporation of an external mixture of sea salt aerosol. Positive numbers indicate a reduction in the magnitude of the indirect effect. Contours are at 0.1, 0.3 and 0.5 Wm⁻²; regions between 0.1 and 0.3 Wm⁻² are lightly shaded, and regions where sulphate forcing is reduced by more than 0.5 Wm⁻² are heavily shaded.

The abundance of other, non-sulphate sources of CCN in continental air, such as nitrates and organic particles, makes it plausible that the link between sulphate and droplet concentrations should be much weaker in these areas, if indeed there is such a link at all. The important question from the standpoint of anthropogenic indirect radiative forcing is how much of these non-sulphate aerosols are *themselves* anthropogenic? If there is only a small anthropogenic contribution to these non-sulphate aerosols, then indirect forcing over the continents since the beginning of the industrial era is likely to be small, whatever the sulphate concentration. However, if these non-sulphate aerosols are largely anthropogenic, then while there may not be substantial *sulphate* indirect forcing, there may still be significant anthropogenic *aerosol* forcings. Whatever the nature of this 'background' aerosol, unless there are dramatic changes in continental aerosol concentrations, there is likely to be little change in indirect forcing over at least northern continental regions in future. Model predictions of sulphate distributions for the year 2050 (H. Rodhe & U. Hansson, personal communication) under the IPCC IS92a scenario (IPCC 1995) show the largest increase in sulphate burdens in South East Asia, and slight decreases over Europe and North America. The change in indirect forcing between the present-day and 2050 is shown in figure 6; the greatest changes are in the more susceptible Southern Hemisphere, with little change in the Northern Hemisphere, and in fact a slight decrease in forcing in the North Atlantic region.

Whilst it is generally accepted that biogenic sulphate is an important source of CCN in maritime air masses (e.g. Charlson *et al.* 1987), there is debate as to the importance of sea salt aerosol in forming cloud droplets, especially under conditions of high windspeed (Blanchard & Cipriano 1987; Latham & Smith 1990; O'Dowd & Smith 1993). Recent work on incorporating

sea salt aerosol into the UM as an external mixture with sulphate (J. Lowe, personal communication) shows a slight decrease in global mean indirect forcing, as shown in figure 7. Due to the logarithmic dependence of sea salt aerosol concentration on windspeed, these changes are concentrated in oceanic areas of high windspeed, typically in the latitude bands 40°–60° N and S. In the Southern Hemisphere these areas tend to be those where, despite the relatively low anthropogenic sulphate concentrations, there was a moderate degree of indirect sulphate forcing due to the presence of clean, susceptible clouds. The inclusion of sea salt aerosol reduces indirect sulphate forcing in these regions by about 50%. These studies indicate the sensitivity of estimates of the indirect effect to the assumptions made about the factors governing the concentration and distribution of cloud droplets. Nevertheless, the indications are that indirect forcing could well be of comparable magnitude to the direct effect, but with a different global distribution. There is a need for much more work in the areas of observations, theory and modelling to improve our understanding of the complex links between aerosol, CCN and cloud droplets to try to narrow the wide limits of uncertainty in estimating the indirect effect.

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Discussion

A. R. RAVISHANKARA (*NOAA Aeronomy Laboratory, Boulder, CO, USA*). Could you please comment on the sensitivity of the calculated indirect effect to the vertical distribution of the sulphate aerosol.

A. SLINGO. The horizontal distribution of the aerosol was taken directly from the chemical transport models. However, for simplicity in performing the simulations, we represented the vertical distribution by assuming that half the mass was uniformly distributed in the boundary layer, with the rest aloft. This approximates the vertical distribution obtained by Langner & Rodhe and also gives values of the cloud effective radius in reasonable agreement with the satellite data. In the future, we will use the full three-dimensional distribution provided by the aerosol scheme now being incorporated into the climate model.

P. S. LISS (*University of East Anglia, UK*). In view of the 'susceptibility' of the Southern Hemisphere atmosphere to increases in CCN, what would be the effect of a several-fold increase in DMS emissions from the southern oceans (as appears to have been the case in the last ice age) on the cloud albedo of that half of the Earth?

A. SLINGO. It seems reasonable to assume that increased DMS emissions would lead to increased CCN, smaller drops, higher cloud albedo and hence a cooling tendency on climate, following the Charlson *et al.* (1987) mechanism. We would need to run a model to estimate just how large the change would be. But the possibility that it might contribute a positive feedback on the ice age is certainly intriguing.

E. WOLFF (*British Antarctic Survey, Cambridge, UK*). You find a strong sensitivity in the mid-to-high latitude Southern Hemisphere. Yesterday we heard suggestions of possible sulphur feedbacks on climate in the last ice age, when, of course, both land and sea ice extent were rather large. Could you confirm my assumption that the indirect effect over ice surfaces will be small, or even possibly in the opposite direction? It would also be interesting (partly because of ice seasonality) to hear about the seasonality of the indirect forcing.

A. SLINGO. We certainly find that the indirect forcing is small over ice surfaces, suggesting that increased sea ice cover might reduce the forcing (but see my response to Liss above). We used annual means of the sulphate aerosol distributions, so we did not examine the seasonality of the forcing, but we will be doing this in the future.

K. P. SHINE (*University of Reading, UK*). There have been several recent attempts at simulating the variation of climate over the past century using coupled general circulation models. These have included the time variation of greenhouse gases and sulphate aerosols and seem to have obtained

reasonable simulations of the evolution of the global mean surface temperature. However, they have, to my knowledge, neglected the negative forcing due to the indirect effect of sulphate aerosols. If it were to be as negative as, or more, negative than -0.5 Wm^{-2} this would seriously degrade the degree of agreement between modelled and observed temperatures. This troubles me somewhat, for it seems to imply that either the indirect forcing estimates are incorrect, or the degree of agreement with observations in recent model experiments has been somewhat coincidental.

A. SLINGO. One could argue that their results provide a clue as to the magnitude of all the non-greenhouse gas forcings, which at present are represented by the forcing they call 'sulphate aerosol'. If our estimates of the indirect effect continue to be larger than this, it could mean that they are missing an unidentified positive forcing (e.g. from increases in absorbing aerosols?). By examining the patterns in temperature change, as opposed to just global means, it may be possible to get some independent information on this problem.