

Climate response to tropospheric absorbing aerosols in an Intermediate General-Circulation Model

By J. COOK and E. J. HIGHWOOD*
University of Reading, UK

(Currently under review by Q.J.R. Meteorol. Soc., April 2003. Please do not copy or cite without permission)

SUMMARY

This study uses idealised aerosol distributions with the Reading Intermediate General-Circulation Model to assess and explain the climate response in that model to absorbing tropospheric aerosol. We find that the sign of the direct aerosol radiative forcing is not a good indication of the sign of the resulting global and annual mean surface temperature change. The climate sensitivity parameter for aerosols which absorb some solar radiation is much larger than that for CO₂ or solar experiments with the same model.

Reasons for the enhanced surface temperature response in the presence of aerosol are examined. Significant changes in cloud amount occur, some of which appear most influenced by the change in surface temperature and may be generic to any mechanism that warms the surface. A reduction in low cloud amount occurs when the aerosol single-scattering albedo is less than 0.95; the so-called “semi-direct” effect of aerosols is clearly evident in this model. We suggest that this aerosol-cloud feedback is present in all GCMs which include absorbing tropospheric aerosol but remains largely undiagnosed. Comparisons with a previous study and further sensitivity tests suggest that the magnitude of this effect and the mechanisms behind it are strongly dependent on the cloud scheme employed.

KEYWORDS: Climate sensitivity Cloud feedback Critical single-scattering albedo Semi-direct effect

1. INTRODUCTION

Tropospheric aerosols are an important component of the present day climate system. These submicron particles from both natural and anthropogenic sources scatter and absorb solar radiation and, in the case of dust, absorb and emit long wave terrestrial radiation. The inclusion of tropospheric aerosols in simulations of radiative transfer through the atmosphere has been shown to be necessary to bring atmospheric climate models and models of radiative transfer into agreement with satellite data (Haywood *et al.*, 2000). In addition, there have undoubtedly been large changes in both the magnitude and spatial distribution of the atmospheric loading of aerosols due to changes in fossil fuel burning and other practices during the industrialisation of the globe. In particular, in recent decades, Asia has become a particularly strong source of black carbon, which forms the major absorbing component of aerosols, while the sources from Western Europe and the USA have decreased due to regulation of emissions (Tegen *et al.*, 2000).

Despite the importance of tropospheric aerosols, our knowledge and understanding of their properties and their interactions with the climate are at a relatively unsophisticated level compared to our understanding of the well-mixed greenhouse gases such as carbon dioxide (IPCC, 2001). This is partly due to the complexity of tropospheric aerosols; they never occur as individual species, but rather are present in some mixture of different components. The way and proportion in which they are mixed has profound implications for the aerosol optical properties and their effect on climate (Haywood and Boucher, 2000). In addition, aerosols possess a lifetime on a scale of days to weeks in the troposphere

* Corresponding author: Department of Meteorology, University of Reading, PO Box 243, Reading, Berkshire RG6 2BB, UK. Email: e.j.highwood@reading.ac.uk

© Royal Meteorological Society, 2003.

and, therefore, their spatial and temporal distributions are very inhomogeneous. Measurement campaigns can only sample aerosol in limited locations and for short periods of time, for example the Tropospheric Aerosol Radiative Forcing Observational Experiment (TARFOX) (Hegg *et al.*, 1997), the Indian Ocean Experiment (INDOEX) (Clarke *et al.*, 2002), ACE-Asia (Wang *et al.*, 2002), and the Southern African Regional Science Initiative (SAFARI) (Haywood *et al.*, 2003). As a consequence, it is difficult to build up a global picture of tropospheric loading and composition, or to model its impact on climate. For example, Wild (1999) reports that GCM calculations of aerosol absorption are not in good agreement with observations. Observations of baseline pre-industrial and natural aerosol loading are practically non-existent.

Different climate mechanisms are often compared using radiative forcing, a measure of the perturbation to the Earth's energy balance on altering the composition of the atmosphere or the properties of the surface. Radiative forcing is defined as the change in net irradiance at the tropopause after the stratosphere has been allowed to return to radiative equilibrium. In global mean terms, tropospheric aerosols of different types produce radiative forcings of different magnitudes and even sign (IPCC, 2001). Underlying the concept of radiative forcing as a comparative measure of potential climate change is an assumed linear relationship between global mean radiative forcing, ΔF , and the global mean equilibrium surface temperature response, ΔT_s , described by:

$$\Delta T_s = \lambda \Delta F \quad (1)$$

where λ is the parameter that describes climate sensitivity. The usefulness of this concept, however, relies on a constant λ that is independent of the forcing mechanism. The climate sensitivity to globally distributed forcings such as increases in carbon dioxide or changes in solar forcing does indeed appear to be broadly consistent, at least to within 20% (e.g. Hansen *et al.*, 1997; Forster *et al.*, 2000; Joshi *et al.*, 2003). There have been indications, however, that for spatially varying radiative forcings, such as changes in ozone and aerosols, a fixed forcing-response relationship begins to break down (e.g. Ponater *et al.*, 1999).

To date, there has been only one systematic study covering a wide range of forcing mechanisms using only a simplified general-circulation model (Hansen *et al.*, 1997) and the results suggested that this problem is particularly apparent for absorbing aerosol. Hansen *et al.* (1997) found dramatic differences in λ for absorbing aerosol, especially if clouds were allowed to vary. In that study, the absorption of solar radiation by the aerosol with low single-scattering albedo heats the atmosphere and produces a decrease in the local large-scale cloud cover. This could either be regarded as a positive feedback or, as Hansen *et al.* (1997) termed it, the ‘‘semi-direct effect’’ of tropospheric aerosols.

One observational and modelling study appears to confirm that this type of response occurs in the real atmosphere. During the Indian Ocean Experiment (INDOEX) campaign, Ackerman *et al.* (2000) observed only sparse cloud in regions affected by continental haze. This seemed contradictory to the standard assumption that the presence of aerosol augments cloud depth or lifetime (the conventional aerosol indirect effect). They hypothesised that it was the absorbing nature of the aerosol which produced the reduction in cloud cover. Solar heating could reduce the relative humidity and speed up cloud evaporation or, as in the cloud feedback to greenhouse warming described by Senior and Mitchell (2000), a change in the static stability of the atmosphere due to the solar heating

could affect cloud cover. Using a large-eddy simulation model together with representations of absorbing aerosol, Ackerman *et al.* (2000) found a decrease in local cumulus cloud fraction of 5 - 10% in the polluted regions, apparently due to the “burning off” of cloud as solar radiation was absorbed by the soot aerosols. The effect of this process on climate response to absorbing tropospheric aerosol depends crucially on the level at which cloud is altered, since low cloud tends to cool as their albedo effect dominates over the greenhouse effect, while higher cloud tends to warm. This would also seem to implicate some further dependence on the altitude of aerosol particles and their position relative to cloud cover. Krüger and Graßl (2002) examined satellite imagery and found evidence of increased cloud absorption of solar radiation in regions close to sources of black carbon. Although this latter effect is somewhat different to the semi-direct effect that is the focus of this study, it is important observational evidence of the impact of absorbing aerosol on cloud.

Other studies suggesting a link between absorbing aerosol and the hydrological cycle include that by Ramanathan *et al.* (2001), who suggest that the large decrease in solar radiation reaching the surface due to both scattering and absorbing aerosol, must be balanced by a similar decrease in evaporation, thereby spinning-down the hydrological cycle. This suggestion is somewhat contrary to the statement by Allen and Ingram (2002) who suggest that since absorbing aerosol mainly warms the troposphere (similar to increases in carbon dioxide), the hydrological cycle should respond in the same way, increasing in strength.

This study aims to investigate whether the results of Hansen *et al.* (1997) are corroborated using a somewhat more sophisticated model and to determine the involvement of cloud interaction in this model’s climate response to aerosol forcing. The climate sensitivity to various aerosol types, as well as doubled carbon dioxide concentration and increased solar irradiance, is calculated for our model for varying and fixed cloud situations. We extend the analysis to cover diagnosis of climate response in terms of variables other than surface temperature.

2. MODEL DESCRIPTION AND EXPERIMENTAL DESIGN

(a) *Model description*

This investigation is based on experiments using a version of the Reading Intermediate General Circulation Model (IGCM) with imposed aerosol. The IGCM was developed to retain the physics necessary for a realistic representation of climate processes while remaining computationally inexpensive and suitable for running many multi-decadal integrations. This enables exploration of the large parameter space and aids investigation of the mechanisms involved in climate forcing and response. Further details on the development of the IGCM are provided in the study by Forster *et al.* (2000).

The IGCM has previously been used to investigate climate response to variations in carbon dioxide and solar irradiance (Forster *et al.*, 2000) and, more recently, also tropospheric and stratospheric ozone (Joshi *et al.*, 2003). It was shown that the model produces a response to a wide range of perturbations that is in good agreement with more sophisticated GCMs although it tends towards the low end of the climate sensitivity parameter range (Joshi *et al.*, 2003). Modifications have been made to the model set-up used in these earlier studies and provide a stable control climate which is suitable for the purposes of this investigation. Principal changes to the model include: substitution of the existing

radiation scheme for that of Morcrette (1990); tuning of super-saturation levels for low, mid and high level clouds to bring the top-of-the-atmosphere energy budget of a fixed sea surface temperature control run into equilibrium; and derivation of new ocean heat fluxes from the fixed sea surface temperature run for use in mixed-layer ocean experiments. The model's slab ocean is set at a depth of 2m to allow a steady state surface temperature to be reached quickly in mixed-layer ocean experiments. This is necessary for calculating equilibrium surface temperature changes in order to derive the climate sensitivity parameter. The IGCM reaches equilibrium after approximately five years with a 2m mixed-layer ocean, but 30-year integrations have been performed to give sufficient degrees of freedom.

Clouds in the IGCM are calculated using a simplified version of the Slingo (1987) cloud scheme. Five cloud types are described mainly by vertical placement in the model's atmosphere, cloud fraction and liquid water path. Cloud fractions for low, mid and high level cloud are parameterised using relative humidity while deep convective cloud is parameterised using precipitation. Shallow convective cloud fraction is fixed at a value of 0.35. The global distributions of low, mid and high level cloud have been compared with data from the International Satellite Cloud Climatology Project (ISCCP). Mid and high level cloud cover in the IGCM was found to give a reasonable approximation of the satellite observations. Low cloud cover is broadly similar except for regions off the west coasts of Africa and South America where the absence of stratocumulus in the IGCM is evident (Forster *et al.*, 2000) in common with many GCMs.

The IGCM offers significant advances over the simplified Wonderland model employed by Hansen *et al.* (1997). The resolution of the IGCM in these experiments is considerably higher with 22 levels in the vertical and a $5^\circ \times 5^\circ$ horizontal global grid. In comparison, the model used by Hansen *et al.* (1997) only covered 120° of longitude with a horizontal resolution of $7.83^\circ \times 10^\circ$, and included only nine vertical layers, of which two or three represented the stratosphere. This enabled long integrations of up to 1300 years but could not be described as a global model.

(b) *Aerosol specification and experimental design*

Initially we use the IGCM to repeat the experiments performed by Hansen *et al.* (1997) and then extend the analysis of model output to gain a clearer understanding of the basic mechanisms that are initiated by aerosol radiative forcing. Aerosol effects are modelled in the IGCM by describing three key parameters: optical depth, τ , defined as the product of aerosol burden and the extinction coefficient; single-scattering albedo, ω ; and the asymmetry factor, g . These are sufficient for defining the optical properties that factor heavily in the calculation of radiative forcing (Haywood and Boucher, 2000). We assume constancy over a band of 0.3 to $4\mu\text{m}$ and no impact on long-wave radiation. The dependence of aerosol properties on geographical distribution and source of aerosol, and, consequently, the influence of external conditions such as relative humidity on radiative effects, is neglected.

We adopt a similar approach to the study by Hansen *et al.* (1997) whereby we fix total aerosol optical depth and the asymmetry factor, and characterise aerosol types solely by the single-scattering albedo. All values are applicable at $0.55\mu\text{m}$. Hansen *et al.* (1997) specified an optical depth of 0.1 in their study. In the IGCM we choose $\tau = 0.2$ to amplify climate response in order that statistically significant results may be achieved within a practical time frame. This figure is

not unrealistic. An average total optical depth of 0.28 was measured in TARFOX (Hegg *et al.*, 1997), and ground-based measurements from the Aerosol Robotic Network (AERONET) program suggest values of a similar magnitude for a range of global sites, although there exists high seasonal variability (Holben *et al.*, 2001). The asymmetry factor is set at 0.7 in both the IGCM and the study by Hansen *et al.* (1997). Haywood and Shine (1995) use Mie calculations to provide an estimate of $g = 0.7$ for sulphate aerosol with $\omega = 1$ at $0.55\mu\text{m}$, and the IPCC (2001) lists asymmetry factor values in the region of 0.65 for various aerosol types compiled from a range of literature sources.

Five single-scattering albedo, $\omega = 0.8, 0.85, 0.9, 0.95, 1$, have been chosen to cover the majority of observed values, ranging from purely scattering, sulphate-based aerosol ($\omega = 1$) to aerosol containing substantial amounts of black carbon ($\omega = 0.8$). Bulk visible wavelength single-scattering albedo in the real atmosphere have been measured between 0.85 and 0.96 (e.g. Hegg *et al.*, 1997; Dubovik *et al.*, 2002).

Our investigation begins with experiments in which the IGCM's climate is perturbed by a globally uniform layer of aerosol in the lower troposphere (between approximately 930mb and 695mb of the model's atmosphere where surface pressure is 976mb). This is equivalent to grid points at which low cloud may be present. Each level has equal aerosol optical depth. All feedback mechanisms are enabled and climate response is identified via comparison with a 30-year mixed-layer ocean control run with no aerosol. Hansen *et al.* (1997) also present results for experiments in which the aerosol layer is shifted to a higher altitude to study height dependence of the aerosol forcing-response relationship. We include in section 3(e) results for which aerosol is placed in the mid troposphere, above low cloud, for comparison.

An indication of the semi-direct effect contribution to climate response, as it exists in the IGCM, is obtained by removing inter-annual variability of cloud properties and their capacity for reacting to changes in the climate. This eliminates the semi-direct effect, as defined by Hansen *et al.* (1997), and any additional cloud feedback, and enables an assessment of the impact of cloud response on the model's climate sensitivity to aerosol forcing. A one-year cloud climatology was created using monthly mean cloud variables outputted from the 30-year varying cloud control run. This is imposed on a second set of mixed-layer ocean aerosol experiments to fix the clouds. Cloud fractions are specified at each time step of the cloud climatology by linearly interpolating between monthly mean values to derive intermediate time steps. The discrete cloud level values were held fixed at the monthly mean for the duration of each month. A fixed-cloud control integration was completed to give a fair indication of changes in climate in response to aerosol perturbation in this particular set of experiments.

Radiative forcing, ΔF_a , in this study, is calculated as the net change in irradiance at the tropopause using the fixed dynamical heating approximation, after the stratosphere has returned to radiative equilibrium. Monthly mean climatologies of atmospheric variables are produced from an IGCM control run and employed as input in the ΔF_a calculations. The choice of control run has important implications for ΔF_a . Although the mean cloud fields in the fixed and varying cloud control runs are identical, removing inter-annual cloud variability impacts on other variables such as surface albedo and planetary albedo, and, consequently, the energy budget. Hence, a more accurate calculation of the radiative forcing that is closely related to the surface temperature response in

the IGCM is achieved by using either the varying cloud or fixed cloud control climatologies where appropriate.

It is essential that we identify changes in climate that are unique to aerosol and those that might be common to other forcing agents, for example, carbon dioxide or solar irradiation perturbation. By fixing both land and ocean surface temperatures, we are able to separate the atmospheric response to a change in surface temperature, and therefore that which may also result from a different forcing leading to a similar surface temperature response, from the response solely attributable to aerosol.

3. RESULTS

(a) *Climate sensitivity*

We begin by examining the global and annual-mean adjusted radiative forcing and surface temperature response to each aerosol species, given in Table 1. The equilibrium response is calculated using values from the last 20 years of each 30-year run. The climate sensitivity parameter, λ , is derived using Eq. 1. We also present here IGCM results from standard carbon dioxide and solar irradiance perturbation experiments. For the carbon dioxide experiment the concentration level of the control run is increased from 336ppmv by a factor of 2 ($2\times\text{CO}_2$). For the solar experiment the solar constant is raised from 1376Wm^{-2} by 2% ($+2\%\text{S}_0$). The purpose of these runs is, in part, to provide a measure of the IGCM's performance in an area that is well documented in other models, but mainly to highlight the idiosyncratic behaviour of climate response to aerosol forcing in comparison with other forcing agents.

TABLE 1. GLOBAL AND ANNUAL MEAN RADIATIVE FORCINGS AND SURFACE TEMPERATURE RESPONSE DUE TO AEROSOL FOR VARYING CLOUD, FIXED CLOUD AND CLOUDLESS EXPERIMENTS.

Experiment	ΔT_s , K		ΔF_a , Wm^{-2}		λ , $\text{K}/(\text{Wm}^{-2})$	
	Varying clouds	Fixed clouds	Varying clouds	Fixed clouds	Varying clouds	Fixed clouds
<i>Lower troposphere</i>						
$\omega = 1$	-1.70	-2.20	-4.72	-4.58	0.36	0.48
$\omega = 0.95$	-0.60	-1.00	-3.02	-3.04	0.20	0.33
$\omega = 0.9$	0.60	0.00	-1.40	-1.57	-0.43	0.00
$\omega = 0.85$	1.80	1.10	0.14	-0.18	12.86	-6.11
$\omega = 0.8$	2.90	2.10	1.61	1.15	1.80	1.83
<i>Mid troposphere</i>						
$\omega = 1$	-1.80	-	-4.90	-	0.37	-
$\omega = 0.95$	-0.90	-	-2.28	-	0.39	-
$\omega = 0.9$	-0.10	-	0.23	-	-0.43	-
$\omega = 0.85$	0.60	-	2.64	-	0.23	-
$\omega = 0.8$	1.20	-	4.95	-	0.24	-
$2\times\text{CO}_2$	1.90	-	3.81	-	0.50	-
$+2\%\text{S}_0$	1.90	-	4.85	-	0.39	-

Climate sensitivity parameter, λ in $\text{K}/(\text{Wm}^{-2})$, calculated using $\lambda = \Delta T_s / \Delta F_a$.
Uniform global distribution of forcing agent. Mixed-layer ocean experiments.

Studies including Forster *et al.* (2000) and Joshi *et al.* (2003) have already provided detailed comparisons of climate response to CO_2 and S_0 perturbations in the IGCM with other models. Climate sensitivity in the IGCM was found to

be at the low end of the scale comprising of a number of other models but the increased sensitivity to CO₂ relative to solar irradiance changes which is apparent in other models (Joshi *et al.*, 2003; Hansen *et al.*, 1997) is maintained. Despite the differences between the version of the IGCM used in this study and the version employed by Forster *et al.* (2000), climate sensitivity is found to be consistent ($\lambda_{CO_2} = 0.47$ and $\lambda_{S_0} = 0.4$ in Forster *et al.* (2000)) and the global distribution patterns of radiative forcing and surface temperature response retain key features noted in Forster *et al.* (2000) and Joshi *et al.* (2003).

Forster *et al.* (2000) discuss in detail the difference between the climate sensitivities for +2% S₀ and 2×CO₂ in the IGCM and it is maintained that for these forcings, at least, radiative forcing provides a useful indication of surface temperature response. We now consider the results in Table 1 for aerosol perturbation experiments. Climate sensitivity varies spectacularly across the range of single-scattering albedo represented in this investigation, not only in magnitude but also in sign. Evidently, a linear forcing-climate response relationship does not exist in the IGCM, especially when clouds are able to respond freely to aerosol perturbation. In particular, note that there are cases where radiative forcing does not even predict the sign of the surface temperature response (cases where λ is negative in Table 1). These results are consistent with those obtained by Hansen *et al.* (1997) and add weight to the hypothesis that the lack of fixed proportionality, at least, is model independent.

The critical single-scattering albedo, ω^* , has been used as a guide for inferring global mean climate response from a given aerosol single-scattering albedo (e.g. Haywood and Shine, 1995; Hansen *et al.*, 1997; Liao and Seinfeld, 1998). It has frequently been defined as the single-scattering albedo at which ΔF switches from positive to negative, ω_F^* , and several studies have provided a way of calculating this value when using simple radiation models (e.g. Haywood and Shine, 1995; Liao and Seinfeld, 1998). If Eq. 1 held true for aerosols with a constant λ regardless of the single-scattering albedo, the implication is that ω_F^* provides a means of determining climate response to aerosol such that for $\omega < \omega_F^*$, an aerosol causes global mean warming and for $\omega > \omega_F^*$, an aerosol leads to global mean cooling. The results given in Table 1, however, clearly show that λ is not constant.

We define ω_T^* as the single-scattering albedo at which ΔT_s changes from warming to cooling. This implicitly includes any additional feedback effects that are present in the IGCM. Hansen *et al.* (1997) found $\omega_F^* = 0.85$ whereas $\omega_T^* = 0.91$ when all feedbacks were enabled. From Fig. 1 we estimate the values of ω_F^* and ω_T^* for varying cloud experiments in the IGCM to be 0.85 and 0.93, respectively in good agreement with Hansen *et al.* (1997). The value of ω_F^* here falls below the majority of observed single-scattering albedo and could be used wrongly to imply that these observed aerosol mostly cool the atmosphere. ω_T^* , however, lies within the range of observed values and indicates that the degree to which these aerosols cool may be smaller than expected as less absorption is required to induce a global mean warming. We also notice that inhibiting cloud response causes a very small change in ω_F^* but, in agreement with Hansen *et al.* (1997), ω_T^* is quite substantially lowered to about 0.9.

The difference between the values of ω_F^* and ω_T^* is large and serves to highlight the inadequacy of defining ω^* as a function of ΔF . ω_F^* , in these cases, fails to reflect the impact of feedback mechanisms on climate response to aerosol, and the use of this to indicate a surface temperature response in models is unreliable.

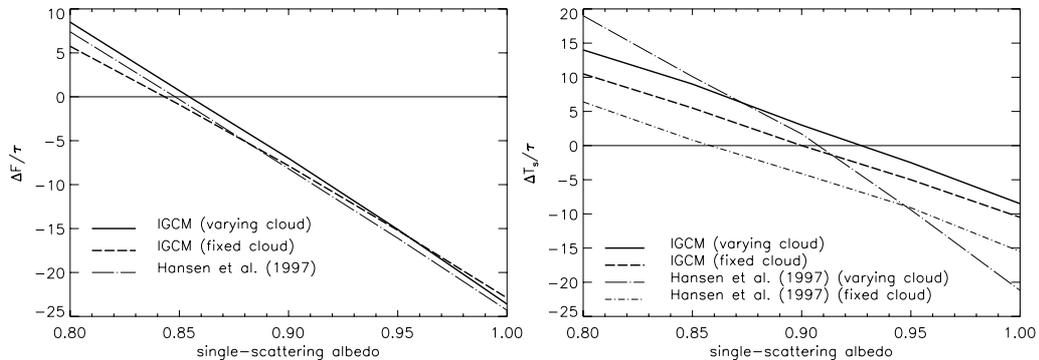


Figure 1. Graphs showing (left) global and annual mean radiative forcing per unit optical depth against ω ; (right) global annual mean surface temperature response per unit optical depth against ω . $\omega_T^* \approx 0.85$ for both fixed and varying cloud IGCM experiments and the study by Hansen *et al.* (1997). Hansen *et al.* (1997) calculated a single value of ΔF_a for each aerosol type using a climatology from a mixed-layer ocean control run with all feedback mechanisms disabled. In this study we find values of 0.93 and 0.9 for ω_T^* for the varying cloud and fixed cloud experiments, respectively. Hansen *et al.* (1997) have equivalent values of 0.91 and 0.86 for varying and fixed cloud experiments

(b) Cloud response to aerosol

In an extension to the work by Hansen *et al.* (1997), we must now examine in more detail the mechanisms by which λ is enhanced for absorbing aerosol. The results of Hansen *et al.* (1997) suggest a large role for cloud feedback. It is appropriate, therefore, to begin with an analysis of cloud changes within the IGCM.

Cloud response to atmospheric perturbations plays a significant role in climate response but still represents one of the largest uncertainties in climate modelling (IPCC, 2001). A substantial proportion of the variation in climate change due to radiative forcing between studies stems from model-specific cloud parameterisation schemes. A number of investigations have examined cloud response to atmospheric forcing across a range of models (e.g. Cess *et al.*, 1996) and within the same model using different cloud schemes (e.g. Senior and Mitchell, 1993; Lee *et al.*, 1997). The general consensus among these studies is that there exists significant variation in cloud response in models to the degree that it cannot be ascertained whether clouds act as positive or negative feedback mechanisms.

In this study we simplify investigation of cloud response to analysis of cloud fraction at different levels in the IGCM's atmosphere and use this to estimate the contribution of clouds to changes in the radiation budget of the model's climate. The first column of Fig. 2 shows the zonal mean cloud fractions for varying cloud control runs with a mixed-layer ocean (varying surface temperatures) and with fixed sea and land surface temperatures. The cloud cover responses to perturbation of both the fixed surface temperature and varying surface temperature climate systems by scattering and absorbing aerosol ($\omega = 1$ and $\omega = 0.8$, respectively), $+2\%$ S_0 and $2\times\text{CO}_2$ are also given. By comparing the varying and fixed surface temperature experiment responses, it is possible to identify the cloud response specifically attributable to absorbing aerosol and, hence, determine the semi-direct effect, as it exists in the IGCM.

Global and annual mean values show that cloud amount decreases linearly with aerosol single-scattering co-albedo ($1 - \omega$) for $\omega < 1$ in the varying surface

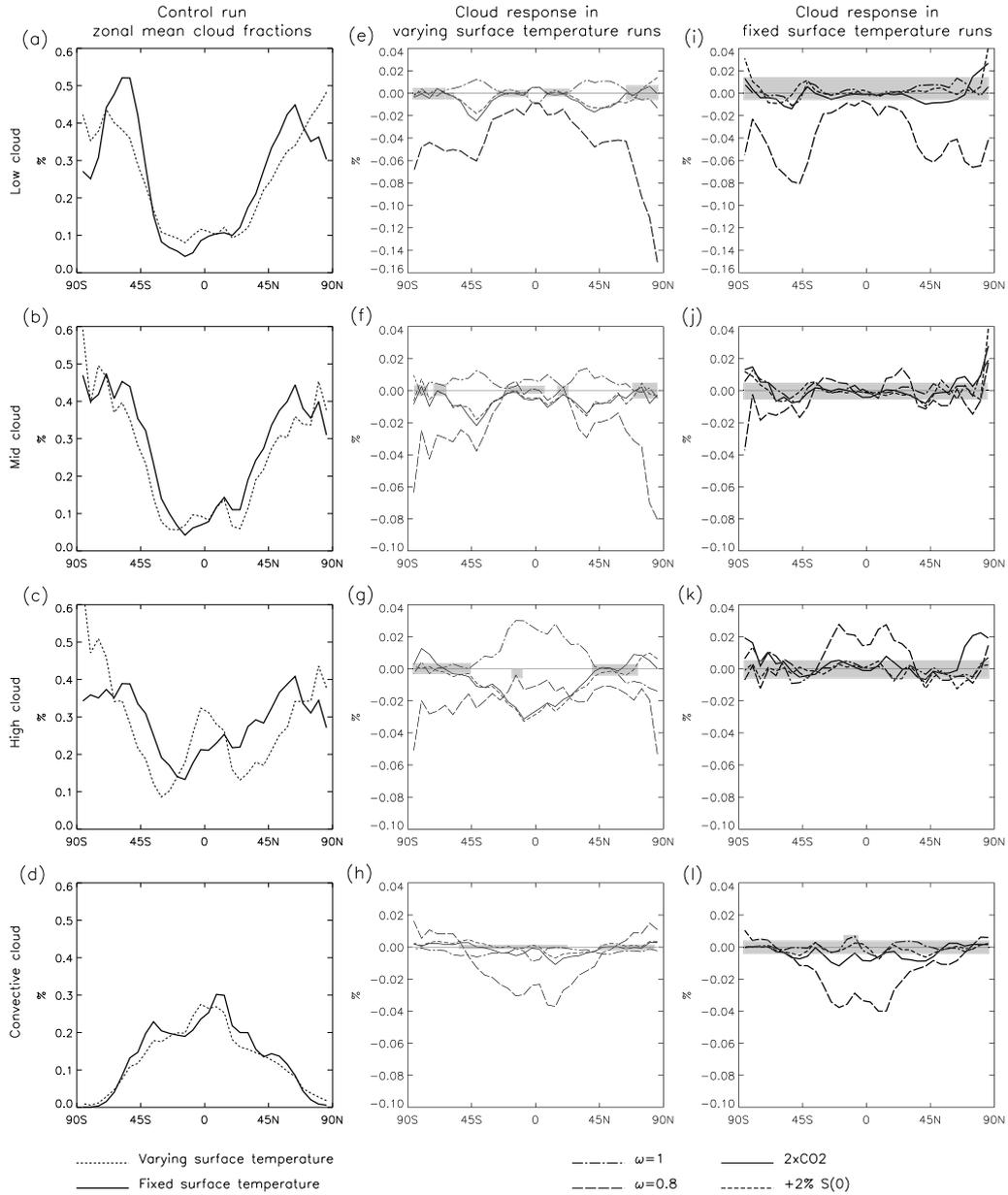


Figure 2. Zonal mean cloud fractions in the control runs of IGCM, (a)–(d), and change in zonal mean cloud fraction (aerosol-control) in the model for varying surface temperature runs, (e)–(h), and fixed surface temperature runs, (i)–(l). The first row is low cloud response, the second is mid cloud response, the third is high cloud response, and the fourth row is convective cloud response. Shaded areas represent values which are not significant at the 95% confidence level.

temperature experiments for all cloud types, with $\omega = 0.8$ leading to the greatest cloud loss. In Fig. 2 (second column) absorbing aerosol in the varying surface temperature experiment is shown to cause reduction in clouds across all latitudes with large decreases occurring where cloud fraction is greatest in the control climate (at the poles for low, mid and high cloud; in the tropics for convective cloud). In contrast, the cloud response to purely scattering aerosols in the varying surface temperature run is small but there are statistically significant increases in low and mid level clouds at mid latitudes and a large increase in tropical high cloud. Convective cloud sees a small overall reduction. By fixing both land and sea surface temperatures, however, the atmospheric responses to surface temperature and albedo effects are eliminated and an assessment of the cloud changes attributed directly to the presence of the two aerosol types in the column leads to a different picture (Figs. 2, third column). The low cloud reduction for $\omega = 0.8$ forcing is clearly a response to the absorbing aerosol within this level itself and increased static stability remains to decrease convective cloud. Mid level cloud changes, however, become generally statistically insignificant and we find that, in fact, the response of high cloud (Fig. 2k) to direct atmospheric changes is an increase in contrast to the varying surface temperature runs (Fig. 2g). It is also apparent that the cloud changes seen in the $\omega = 1$ varying surface temperature case are not directly due to scattering aerosol and there are virtually no significant changes in cloud cover when surface temperature and albedo responses are removed.

These responses may be described in terms of feedback response to identify the semi-direct effect. Cloud feedback is defined as the change in cloud forcing in response to a perturbation of the climate (Cess *et al.*, 1996). In the IGCM low and mid cloud cover causes a negative forcing (cools the climate) and high cloud causes a positive forcing (warms the climate). Firstly, we consider the impact of direct perturbation of a column in the model by aerosol alone, that is, when surface temperature is fixed. As absorbing aerosol reduces low cloud, the magnitude of the negative cloud forcing is decreased and implies a positive feedback, which becomes a source of additional warming in the system. High cloud is also increased, therefore, raising the magnitude of positive forcing, another positive feedback. Overall, the impact of $\omega = 0.8$ aerosol in the column is to instigate a positive cloud feedback.

Allowing surface temperature to react to aerosol forcing and the changes in the column, low cloud is still reduced with mostly similar magnitudes of change seen across the meridional plane and, therefore, this results in a positive feedback. We also see a decrease in mid cloud cover, another positive cloud feedback. This would seem to be a response to changes in surface temperature rather than aerosol. However, including the change in surface temperature also appears to effect a reduction in high cloud cover that is sufficient to counteract the increase seen due to aerosol heating in the column itself and cause an overall decrease in high cloud cover everywhere outside of the poles. This implies a negative feedback. It seems that this effect is strong enough to negate the positive low and mid cloud feedback and moderate some of the direct warming from $\omega = 0.8$ aerosol. This is consistent with the climate being less sensitive to strongly absorbing aerosol when clouds are allowed to respond as shown by λ in Table 1.

(c) *Cloud response to $2\times CO_2$ and $+2\% S_0$*

The extent to which these results are common to other forcing agents that produce an increase in surface temperature is evident in the cloud response to $2\times CO_2$ and $+2\% S_0$. In the perturbed fixed surface temperature climate (Fig. 2, third column) there are virtually no significant effects on cloud. Cloud does not react as a feedback and any resultant change in surface temperature that could have been detected would have been directly attributable to the positive forcing. This provides a strong indication that the low cloud response to $\omega = 0.8$ aerosol forcing is unique to absorbing aerosol. There are significant changes in cloud in response to $2\times CO_2$ and $+2\% S_0$ in the varying surface temperature experiments and it is particularly striking that the response to the two forcings are almost identical despite their differences in the impacts on the radiation budget; CO_2 forcing is predominantly in the long wave whereas solar irradiance forcing impacts only on the short wave flux with different latitudinal variation (Hansen *et al.*, 1997). The cloud response to $2\times CO_2$ and $+2\% S_0$ is more modest than for absorbing aerosol and statistically significant changes in low and mid cloud cover occur only at mid latitudes at which there are decreases resulting in relatively small positive feedbacks. Again we see a decrease in high cloud which confirms that there is a negative feedback mechanism in place in the IGCM which reacts to suppress a positive change in surface temperature. The zonal pattern of this change, however is different from that for absorbing aerosol and may have some bearing on the strength of the feedback.

The uniqueness of the positive cloud feedback to absorbing aerosol is further highlighted by the impact of purely scattering aerosol in comparison with $2\times CO_2$ and $+2\% S_0$. The changes in cloud fractions for $\omega = 1$ are almost precisely equal in magnitude and opposite in sign to that for $2\times CO_2$ and $+2\% S_0$ and is consistent with a climate sensitivity, $\lambda_{\omega=1}$ that is similar to λ_{CO_2} and λ_{S_0} .

(d) *Surface energy balance response to absorbing aerosol*

Having discussed the changes in cloud cover due to both surface temperature changes and due to the presence of the aerosol itself, it is important to remember that changes in components of the surface energy balance, such as latent heat, can also affect cloudiness. The global and annual mean surface energy balance in the control run (mixed layer ocean and varying clouds) is small and negative. ($-2.57Wm^{-2}$). The addition of scattering aerosol produces a slightly more negative surface energy balance, but as ω decreases the surface energy balance becomes smaller in magnitude. For all the experiments included here, the change in surface energy balance compared to the control simulation is less than 10%. Examination of the individual components reveals that the surface upward long wave flux increases as ω increases, since the surface is warming and therefore emitting more radiation. However, this increase is almost totally balanced by an increase in surface downward LW radiation and so has little effect on the overall surface energy balance. The net SW downward radiation decreases compared to the control and as ω decreases, due to the increased absorption of solar radiation. For predominantly scattering aerosol this decrease is almost entirely balanced by decreases in both the sensible heat and latent heat, such that the surface energy balance change is small compared to the control run. For more absorbing aerosol the warming of the surface reduces the decrease in latent heat that was observed for scattering aerosol compared to the control run, although it is still less than in

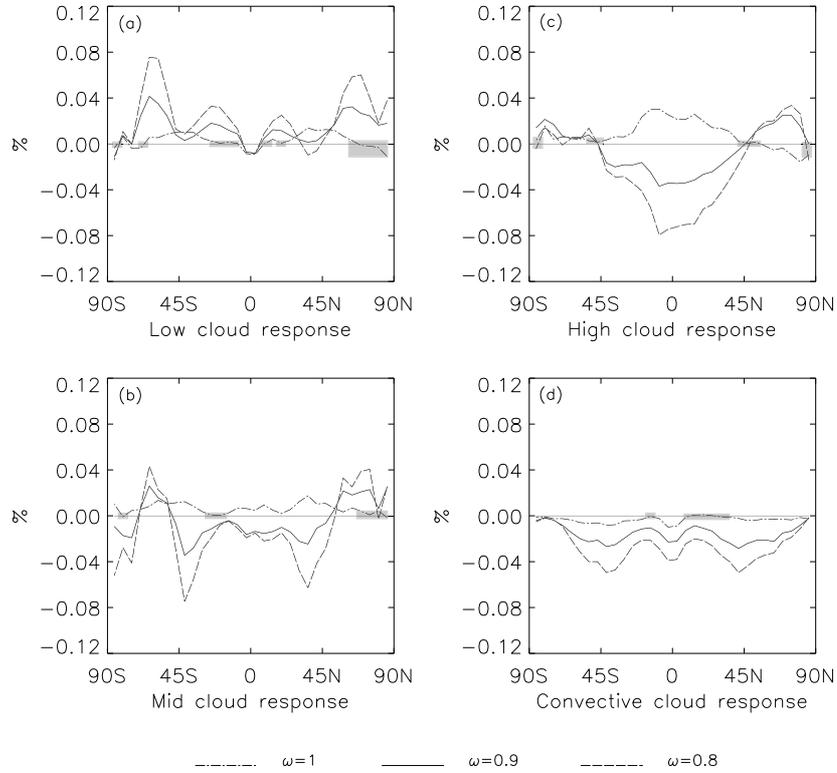


Figure 3. Zonal mean change in (a) low, (b) mid, (c) high and (d) convective cloud fraction, (aerosol-control, $\omega = 0.8, 0.9, 1$) placed in the IGCM's mid troposphere. The zonal mean cloud fractions for the relevant varying surface temperature control run are given in Fig. 2 (first column). Shaded areas represent values which are not significant at the 95% confidence level.

the control run. The sensible heat decreases further from the control run value as ω decreases. In the increased carbon dioxide simulation, the latent heat increases compared to the control run. In so far as surface latent heat can be considered as a proxy for the strength of the hydrological cycle, it appears that the addition of aerosol reduces its strength, as discussed by Ramanathan *et al.* (2001) whereas the increase in CO_2 increases it. However, when the aerosol is absorbing, the weakening is less severe despite the continuing decrease in solar radiation at the surface, presumably due to a mitigating effect of a warmer lower atmosphere and surface. Further investigation of the impact of aerosols on surface energy balance and the hydrological cycle should be the subject of future study.

(e) Sensitivity to vertical placement of aerosol

The magnitude of direct aerosol radiative forcing is heavily dependent on the albedo of the underlying region. Absorption by dark aerosol has been shown to be greatly enhanced by high surface albedo (Haywood and Shine, 1997) as both the reflected short wave radiation and incoming solar radiation are incident on the aerosol. This effect is illustrated by the ΔF_a values in Table 1 for lower and mid tropospheric aerosols; the absorbing aerosol placed at a higher altitude, and therefore situated above bright low cloud, imposes a much stronger direct radiative forcing than the same aerosol placed within the lower troposphere.

By placing the aerosol layer at a higher altitude we gain further insight to the mechanisms by which cloud response operates and uncover a strong height dependence. This appears to have important implications for climate sensitivity to absorbing aerosol. In this set of experiments, the varying surface temperature control run with varying clouds is perturbed by an aerosol layer situated between 310mb and 400 mb of the IGCM's atmosphere. This places aerosol mainly within mid level cloud and partially within the high cloud level but entirely above low level cloud. Total column aerosol optical depth is maintained to be 0.2.

The zonal mean cloud response to aerosol with $\omega = 0.8$, 0.9, and 1 aerosol are given in Fig. 3 and represent the changes in cloud fractions of the varying surface temperature control run, shown in Fig. 2. The mechanism which induced changes in cloud fraction in response to scattering aerosol ($\omega = 1$) in the lower troposphere was shown to arise from changes in the surface temperature and was also found to be the cause of the responses to $2\times\text{CO}_2$ and $+2\%$ S_0 forcing. In Fig. 3, it is evident that the response of clouds at all levels to $\omega = 1$ aerosol placed at a higher altitude is almost identical to the low aerosol response in Fig. 2 and it may be deduced that the IGCM's response mechanism to scattering aerosol remains the same irrespective of the altitude of the aerosol layer, as we may expect of a response due to a change in surface temperature. This appears to be confirmed by the climate sensitivity given in Table 1 which is also very similar to λ for $\omega = 1$ aerosol in the lower troposphere. Hansen *et al.* (1997) also find that λ is not reliant on the altitude of scattering aerosol.

As the absorbing fraction of an aerosol is increased, the vertical position of the aerosol layer presents further complications in the analysis of the forcing-climate response relationship. The semi-direct effect of absorbing aerosols is clearly seen to be in operation at the level of the aerosol in Fig. 3b, which, in this case is within the mid-cloud layer. Both $\omega = 0.8$ and $\omega = 0.9$ have caused a reduction in the cloud cover and, consequently, a positive cloud feedback. These aerosols also generally lead to a decrease in zonal mean high cloud but, while there is a more even zonal pattern for low aerosol, the changes here resemble those observed under CO_2 and S_0 perturbation, and may be driven by the surface temperature change.

Below the aerosol layer, however, we see distinct differences between low cloud response to mid-tropospheric absorbing aerosol (Fig. 3a) and that for absorbing aerosol in the lower troposphere (Fig. 2c). While a warm aerosol layer within the low cloud level decreases cloud cover, heating above it results in cloud increases that vary greatly in magnitude with latitude. The absorbing aerosol semi-direct effect not only has to compete against the high cloud negative feedback, but also against a negative feedback that arises from an increase in low cloud cover. This low cloud negative feedback does not appear to be specific to the IGCM. A comparable result has been simulated by Johnson *et al.* (2003) using a large-eddy model. In that study, the effects of absorbing aerosol on marine stratocumulus were examined. Their results showed that liquid water path increased when the absorbing aerosol layer ($\omega = 0.88$, $\tau = 0.2$) was situated above the boundary layer but decreased for an aerosol layer within the boundary layer. Although the mechanism behind this response in the large-eddy model may not be the same mechanism that leads to an increase in cloud cover in the IGCM, we still find that the thickened cloud implies a stronger negative cloud forcing and, therefore, a negative cloud feedback.

The overall balance of these feedback effects, again, varies with the single-scattering albedo and this impact of raising the height of the aerosol layer is conveyed in the climate sensitivity given in Table 1. For the more absorbing aerosols ($w = 0.8, 0.85$) the negative feedbacks strongly suppress the direct and semi-direct effects and λ is lowered to a level below that for $2\times\text{CO}_2$ and $+2\%$ S_0 forcing. It may appear at first glance that for $\omega = 0.9$ aerosol, as with $\omega = 1$, the climate sensitivity is not altered by raising the height of the aerosol layer but on closer inspection we actually find that, in contrast to the lower tropospheric aerosol results, the negative λ is due to a positive ΔF_a and negative ΔT_s . When the aerosol layer is placed at a higher altitude, $\omega_T^* < \omega_F^*$.

The vertical profile of temperature response is examined for aerosol signals in Fig. 4. The distinctive response to $\omega = 0.8$ absorbing aerosol and its reliance on the altitude of the aerosol layer is easily detected and especially emphasised by the contrasting response to $\omega = 1$ scattering aerosol. The greatest temperature response to $\omega = 0.8$ aerosol occurs within the aerosol layer irrespective of the height, as expected. The maximum temperature increase is seen to be higher for absorbing aerosol at a higher altitude. This result may be due to the effect of ascribing the same aerosol optical depth to both the high and low aerosol layers. As a consequence, heating is more concentrated in the thinner high aerosol layer. The cooling response to scattering aerosol displays no aerosol height dependence; the responses to high and low $\omega = 1$ aerosol are very similar. Again, we find that this response is largely equal in magnitude and opposite in sign to the response to $2\times\text{CO}_2$ and $+2\%$ S_0 forcing (not shown here).

The changes in cloud cover in response to aerosol in the IGCM (Figs. 2 and 3) show a high degree of consistency with the vertical temperature response. The level of the maximum heating correlates with the level of cloud which suffered the greatest decrease. The temperature increases also lead to increased stability, particularly in the case of absorbing aerosol in the lower troposphere, and, therefore, a reduction in convective cloud (Fig. 2h). The cooling due to $\omega = 1$ aerosol agrees well with the decreases seen in low, mid and high level cloud (Fig. 2e – g). The contribution of surface temperature changes to the overall climate response is also clear in the cases where the surface temperature is fixed. It confirms that climate response to purely scattering aerosol is due to surface temperature changes while absorbing aerosol heats the layer in which it is placed, causing cloud decreases in that layer (Fig. 2i – j). Above the $\omega = 0.8$ aerosol layer at high altitude, there is a small temperature decrease which may be responsible for the increased high cloud seen in Fig. 2k.

(f) Summary of results

To summarise, the large decreases in low cloud cover seen in the varying surface temperature climate are unique to absorbing aerosol within the same level, as is the reduction in low cloud in the fixed surface temperature run. When absorbing aerosol is placed in the mid troposphere, cloud cover at that level is reduced. These results seem to be in agreement with the aerosol semi-direct effect hypothesis put forward by Hansen *et al.* (1997) in which a reduction in large-scale cloud at the same level as the absorbing aerosol leads to a positive feedback. The high cloud in the IGCM, however, acts as a strong negative feedback regardless of the forcing agent investigated here but the pattern of the response, and possibly the relative strength of the feedback, is dependent on the climate change mechanism. We propose that there also exists a second part to

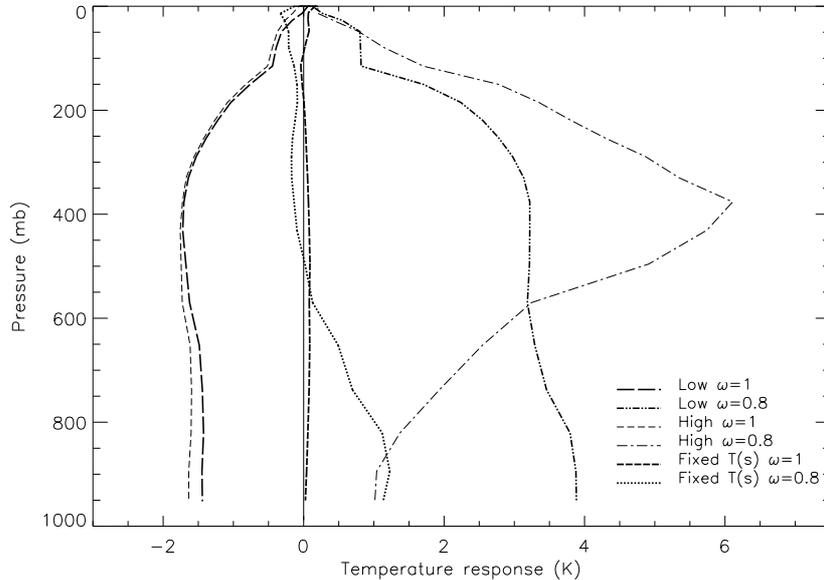


Figure 4. Global and annual mean vertical temperature responses to purely scattering ($\omega = 1$) and strongly absorbing ($\omega = 0.8$) aerosol placed below 695mb (low aerosol) and between 310 and 400mb (high aerosol) in the IGCM's atmosphere. Vertical temperature responses to $\omega = 1$ and $\omega = 0.8$ aerosol when the surface temperature is fixed are also included. Surface temperature responses are omitted (surface pressure = 976mb in the IGCM).

the cloud feedback mechanism and that its contribution is model dependent and determined, to some degree, by the single-scattering albedo. In the IGCM this is a strong negative cloud feedback from high cloud changes, for strongly absorbing and purely scattering aerosols, and accounts for the decreased sensitivity in the varying cloud runs in relation to the fixed cloud runs. The results by Hansen *et al.* (1997) suggest that in their Wonderland model it is a positive cloud feedback and leads to a more sensitive climate in their varying cloud experiments although they do not report a detailed analysis of cloud response.

4. CONCLUSION AND FUTURE WORK

We have shown that the IGCM's climate displays varying levels of sensitivity to different forcing agents and that, in particular, the value of λ for an idealised globally uniform distribution of aerosols differs from that for carbon dioxide and solar irradiance forcing when the single-scattering albedo is less than unity. It is also clear that, in agreement with the findings of Hansen *et al.* (1997), λ is not constant for changing ω . The contribution of feedback mechanisms is implicit in the definition of λ as employed in this study and the dependence of these mechanisms on the forcing agent leads to a variable climate sensitivity. Hence, ΔF_a is rendered a poor predictor of the sign and magnitude of climate response to absorbing aerosol forcing.

As a consequence of the feedback processes initiated by absorbing aerosol forcing, we also find that ω_F^* is unreliable as an indicator of the sign of ΔT_s and should not be used to ascertain the surface temperature response to observed aerosol single-scattering albedo. The values of ω_T^* found in this study suggest

that the overall global mean negative ΔT_s implied by calculation of ΔF_a in simple models is an overestimation.

Cloud plays a vital role in the climate response to aerosol in the IGCM. The semi-direct effect does exist as a feature of this model but it may be separated into two distinct processes. Firstly, there is the large-scale low cloud reduction in response to low altitude absorbing aerosol forcing which leads to a positive feedback and is expected to be similar but not identical in most GCMs. A small part of this is common to other positive forcing agents but much is characteristic of the change in the atmospheric heating profile due to absorbing aerosol. This constitutes the semi-direct effect as defined by Hansen *et al.* (1997). A second model specific cloud feedback response to surface temperature change has also been identified which is a negative feedback in the IGCM but positive in the Wonderland model and would also seem to depend, to an extent, on the forcing agent. The overall climate sensitivity to absorbing aerosol results from a balance between these two effects, and will be highly model dependent.

Whilst the experiments performed here use highly idealised and globally uniform aerosol distributions, it is evident that the response of models to even simple absorbing aerosol fields is not well understood. It has been shown that clouds play an extremely important role in determining this balance. The continuing large difficulties in modelling cloud in global models suggest we may be some way from being able to tease out the mechanism of the aerosol semi-direct effect and other cloud feedbacks. Nonetheless, the semi-direct effect and other aerosol-cloud feedbacks described here exist, mainly undiagnosed, in any model that includes absorbing aerosol. Further examination and diagnosis of the effects in existing models, with both idealised and more realistic cloud distributions would undoubtedly lead us to a better understanding of climate model response to absorbing aerosol, and to the frailties of model cloud representation.

ACKNOWLEDGEMENTS

J. Cook was supported by NERC grant NER/M/S/2001/00065. We would like to thank Manoj Joshi for developing the radiative forcing code. Keith Shine and Ben Johnson are also thanked for their helpful comments.

REFERENCES

- | | | |
|--|------|---|
| Ackerman, A. S., Toon, O. B.,
Stevens, D. E.,
Heymsfield, A. J.,
Ramanathan, V.,
Welton, E. J. | 2000 | Reduction of tropical cloudiness by soot. <i>Science</i> , 288 , 1042–1047 |
| Allen, M.R., and Ingram, W. J. | 2001 | Constraints on future changes in climate and the hydrologic cycle. <i>Nature</i> , 419, 224 |

- Cess, R. D., Zhang, M. H., Ingram, W. J., Potter, G. L., Alekseev, V., Barker, H. W., Cohen-Solal, E., Colman, R. A., Dazlich, D. A., Del Genio, A. D., Dix, M. R., Dymnikov, V., Esch, M., Fowler, L. D., Fraser, J. R., Galin, V., Gates, W. L., Hack, J. J., Kiehl, J. T., Le Treut, H., Lo, K. K.-W., McAvaney, B. J., Meleshko, V. P., Morcrette, J.-J., Randall, D. A., Roeckner, E., Royer, J.-F., Schlesinger, M. E., Sporyshev, P. V., Timbal, B., Volodin, E. M., Taylor, K. E., Wang, W. and Wetherald, R. T. 1996 Cloud feedback in atmospheric general circulation models: An update. *J. Geophys. Res.*, **101**, 12791–12794
- Chylek, P. and Wong, J. 1995 Effect of absorbing aerosols on global radiation budget. *Geophys. Res. Lett.*, **22**, 929–931
- Clarke, A.D., Howell, S., Quinn, P. K., Bates, T.S., Ogren, J. A., Andrews, E., Jefferson, A., Massling, A., Mayol-Bracero, O., Maring, H., Savoie, D., and Cass, G. 2002 INDOEX Aerosol: A comparison and summary of chemical, microphysical and optical properties observed from land, ship and aircraft. *J. Geophys. Res.*, **107**, DOI10.1029/2001JD000572–
- Dubovik, O., Holben, B., Eck, T. F., Smirnov, A., Kaufman, Y. J., King, M. D., Tanré, D., Slutsker, I. 2002 Variability of absorption and optical properties of key aerosol types observed in worldwide locations, *J. Atmos. Sci.*, **59**, 590–608
- Forster, P. M. de F., Blackburn, M., Glover, R. and Shine, K. P. 2000 An examination of climate sensitivity for idealised climate change experiments in an intermediate general circulation model. *Climate Dynamics*, **16**, 833–849
- Hansen, J., Sato, M. and Ruedy, R. 1997 Radiative forcing and climate response. *J. Geophys. Res.*, **102**, 6831–6864
- Haywood, J. and Boucher, O. 2000 Estimates of the direct and indirect radiative forcing due to tropospheric aerosols: a review. *Rev. of Geophysics*, **38**, 513–543
- Haywood, J., Osborne, S. R., Francis, P. N., Keil, A., Formenti, P., Andreae, M. O., Kaye, P. H. 2003 The mean physical and optical properties of regional haze dominated by biomass burning aerosol measured from the C-130 aircraft during SAFARI 2000. *J. Geophys. Res.*, **108**, 8473–
- Haywood, J., Ramaswamy, V. and Soden, B. J. 2000 Tropospheric aerosol forcing in clear sky satellite observations over oceans. *Science*, **28**, 1299–1303
- Haywood, J. and Shine, K. 1995 The effect of anthropogenic sulphate and soot aerosol on the clear sky planetary radiation budget. *Geophys. Res. Lett.*, **22**, 603–606
- Haywood, J. and Shine, K. 1997 Multi-spectral calculations of the radiative forcing of tropospheric sulphate and soot aerosols using a column model. *Q. J. R. Meteorol. Soc.*, **123**, 1907–1930
- Hegg, D., Livingston, J., Hobbs, P. V., Novakov, T. and Russell, P. 1997 Chemical apportionment of aerosol column optical depth off the mid-Atlantic coast of the United States. *J. Geophys. Res.*, **102**, 25293–25303

- Holben, B. N., Tanré, D., Smirnov, A., Eck, T. F., Slutsker, I., Abuhassan, N., Newcomb, W. W., Schafer, J. S., Chatenet, B., Lavenu, F., Kaufman, Y. J., Vande Castle, J., Setzer, A., Markham, B., Clark, D., Frouin, R., Halthore, R., Karneli, A., O'Neill, N. T., Pietras, C., Pinker, R. T., Voss, K., Zibordi, G. 2001 An emerging ground-based aerosol climatology: Aerosol optical depth from AERONET. *J. Geophys. Res.*, **106**, 12067–12097
- IPCC 2001 Climate change 2001: the scientific basis, contribution of working group I to the third assessment report of the Intergovernmental Panel on Climate Change. Cambridge
- Johnson, B., Shine, K., Forster, P. M. de F. 2003 Impact of absorbing aerosol on marine stratocumulus, *Q. J. R. Meteorol. Soc.*, submitted.
- Joshi, M., Shine, K., Ponater, M., Stuber, N., Sausen, R. and Li, L. 2003 A comparison of climate response to different radiative forcings in three general circulation models: towards an improved metric of climate change. *Climate Dynamics*, **20**, 843–854
- Krüger, O., and Graßl, H. 2002 The indirect aerosol effect over Europe. *Geophys. Res. Lett.*, **29**, 31-1–31-4
- Lee, W.-H., Iacobellis, S. F., Somerville, C. J. 1997 Cloud radiation forcing and feedbacks: General Circulation Model tests and observational validation. *J. Climate*, **10**, 2479–2496
- Liao, H. and Seinfeld, J. H. 1998 Radiative forcing by mineral dust aerosol: sensitivity to key variables. *J. Geophys. Res.*, **103**, 31637–31645
- Morcrette, J.-J. 1990 Impact of changes to the radiative transfer parameterisations plus cloud optical properties in the ECMWF model. *Mon. Weather Rev.*, **118**, 847–873
- Ponater, M., and Sausen, R., and Feneberg, B., and Roeckner, E. 1999 Climate effect of ozone changes caused by present and future air traffic. *Climate Dynamics*, 15631–642
- Ramanathan, V., Crutzen, P. J., Kiehl, J. T., and Rosenfeld, D. 2001 Aerosols, climate and the hydrological cycle. *Science* 29421192124
- Senior, C. A. and Mitchell, J. F. B. 1993 Carbon dioxide and climate: The impact of cloud parameterization. *J. Climate*, **6**, 393–418
- Senior, C. A. and Mitchell, J. F. B. 2000 The time dependence of climate sensitivity. *Geophys. Res. Lett.*, **15**, 124–130
- Slingo, J. M. 1987 The development and verification of a cloud prediction scheme for the ECMWF model. *Q. J. R. Meteorol. Soc.*, **113**, 899–927
- Tegen, I., Koch, D., Lacis, A. A. and Sato, M. 2000 Trends in tropospheric aerosol loads and corresponding impact on direct radiative forcing between 1950 and 1990: A model study. *J. Geophys. Res.*, **105**, 26971–26989
- Wang, J., Flagan, R. C., and Seinfeld, J. H. 2002 Airborne measurements of atmospheric carbonaceous aerosols during ACE-Asia. *J. Geophys. Res.*, **107**, D23–art. no. 4704
- Wild, M. 1999 Discrepancies between model-calculated and observed short-wave atmospheric absorption in areas with high aerosol loading. *J. Geophys. Res.*, **104**, 27361–27371