Model Simulations of the Competing Climatic Effects of SO2 and CO2

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ABSTRACT

Sulfur dioxide-derived cloud condensation nuclei are expected to enhance the planetary albedo, thereby cooling the planet. This effect might counteract the global warming expected from enhanced greenhouse gases. A detailed treatment of the relationship between fossil fuel burning and the SO2 effect on cloud albedo is implemented in a two-dimensional model for assessing the climate impact. Although there are large gaps in our knowledge of the atmospheric sources and sinks of sulfate aerosol, it is possible to reach some general conclusions. Using a conservative approach, results show that the cooling induced by the SO2 emission can presently counteract 50% of the CO2 greenhouse warming. Since 1980, a strong warming trend has been predicted by the model, 0.15°C during the 1980–1990 period alone. The model predicts that by the year 2060 the SO2 cooling reduces climate warming by 0.5°C or 25% for the Intergovernmental Panel on Climate Change (IPCC) business as usual (BAU) scenario and 0.2°C or 20% for scenario D (for a slow pace of fossil fuel burning). The hypothesis is examined that the different responses between the Northern Hemisphere (NH) and the Southern Hemisphere (SH) can be used to validate the presence of the SO2-induced cooling. Despite the fact that most of the SO2-induced cooling takes place in the Northern Hemisphere continents, the model-predicted difference in the temperature response between the NH and the SH of ~0.2°C in 1980 is expected to remain about the same at least until 2060. This result is a combined effect of the much faster response of the continents than the oceans and of the larger forcing due to CO2 than due to the SO2. The climatic response to a complete filtering of SO2 from the emission products in order to reduce acid rain is also examined. The result is a warming surge of 0.4°C in the first few years after the elimination of the SO2 emission.

1. Introduction

Fossil fuel combustion has two competing effects on the climate system: a warming due to the emission of CO2 and other trace gases (Manabe and Wetherald 1980; Hansen et al. 1988) and a cooling due to sulfate particles formed from the SO2 emission (Mitchell 1975; Charlton and Sellers 1980; Twomey et al. 1984; Wigley 1989; Kaufman et al. 1991). At the present emission level, the SO2-induced cooling has the potential of at least partially offsetting the CO2-induced warming (Mitchell 1975; Twomey et al. 1984; Wigley 1989; Hansen and Lacis 1990; Charlson et al. 1991; Kaufman et al. 1991). The radiative effect of CO2 is relatively easy to assess based on its infrared absorption and emission properties. The influence of this radiative effect on climate is, however, complicated by the oceanic response and the feedbacks among various components of the climate system (Cess et al. 1990). The induced cooling by SO2 is a consequence of the oxidation of SO2 into sulphuric acid and salts (Rodhe 1978; Georgii 1979; Husar et al. 1981; Seinfeld 1986). These sulfuric compounds form hygroscopic submicron particles that scatter solar radiation back to space, thus causing a direct cooling (Mitchell 1975; Coakley et al. 1983; Charlson et al. 1991), referred to here as the direct effect. These particles also increase the concentration of cloud condensation nuclei (CCN) from as low as 30–50 cm−3 in remote oceanic regions where anthropogenic SO2 is minimal (Twomey 1959; Jiusto 1967; Twomey and Wojciechowski 1969; Radke 1989) to 1000–4000 cm−3 in regions contaminated by industrial pollution (Braham 1974) or by organic smoke particles from fires (Warner and Twomey 1967; Radke 1989). An increase in the CCN concentration can cause an increase in the cloud droplet concentration (Twomey 1959; Twomey and Warner 1967; Twomey 1977; Leitch et al. 1986; Radke 1989). This was demonstrated in a systematic analysis of hundreds of cloud water samples taken over eastern North America (Leitch et al. 1986; Leitch et al. 1992). It was shown that an increase in the cloud water sulfate concentration is associated with an increase in both the aerosol particle concentration and the cloud droplet concentration, but not with liquid water content. An increase in the cloud droplet concentration will cause an increase in the total reflective surface area within the cloud, thus leading to an increase in the cloud albedo (Twomey 1977; Twomey et al. 1984; Coakley et al. 1987; Radke et al. 1989; Nakajima and King 1990; Nakajima et al. 1991)—referred to here as the indirect
effect. This effect, however, can be complicated by various factors, such as a change in the cloud liquid water content (Leaitch et al. 1992). There are also indications that the increased CCN concentration, at least in the case of stratiform clouds, prolongs the cloud lifetime by suppressing drizzle (Radke et al. 1989; Nakajima and King 1990; Nakajima et al. 1991; Albrecht 1989). This process enhances the planetary albedo and further contributes to global cooling (Liu and Ou 1989).

One of the major difficulties in the study of the SO$_2$ climatic effect is the estimation of the CCN concentration (Twomey et al. 1984; Hansen and Lacis 1990; Charlson et al. 1990; Baker and Charlson 1990). This uncertainty results from the fact that atmospheric sulphur compounds have a short lifetime, a nonhomogeneous distribution of sources, and a variable vertical distribution of concentration.

It has been postulated that the SO$_2$ effect can be tested by observing the difference in the warming of the Northern Hemisphere (NH) and Southern Hemisphere (SH) (Schwartz 1988; Wigley 1989). Since most of the SO$_2$ emission is in the NH, one might expect that the effect of SO$_2$ on climate is larger in the NH than in the SH. Due to a lack of evidence for a difference in the temperature changes between the hemispheres, Schwartz (1988) postulated that the effect of SO$_2$ on climate is insignificant. Wigley (1989) found that the temperature change is larger in the SH than in the NH by $0.1^\circ$ and that this difference has an uncertainty of $\pm 0.2^\circ$. Due to this large uncertainty in the difference between the hemispheric responses, one cannot conclude that the effect of SO$_2$ on climate is insignificant (Wigley 1989). It is interesting to test if in the future the SO$_2$-induced cooling generates a larger difference in the hemispheric responses. Although they did not examine the indirect SO$_2$-induced forcing, Charlson et al. (1990) suggested that the direct SO$_2$-induced forcing is comparable but opposite to the anthropogenic CO$_2$-induced forcing. In the next section we shall show that the indirect SO$_2$ forcing is expected to be much larger than the direct forcing.

In this paper we present a detailed analysis of the relationship between fossil fuel burning, CO$_2$ and SO$_2$ emission, the concentration of anthropogenic CCN, and cloud albedo, along with a simulation of the combined CO$_2$ and SO$_2$ climatic effect using the two dimensional climate model of Peng et al. (1987). No attempt is made to account for the SO$_2$-induced increase in cloud lifetime (Radke et al. 1989; Albrecht 1989). It is assumed that the effect of graphitic carbon on the reduction of cloud albedo can be neglected (Twomey et al. 1984).

2. Direct and indirect SO$_2$-induced forcing

In a recent review article, Charlson et al. (1992) emphasized the significance of the direct SO$_2$-induced effect on climate and the uncertainties in the indirect effect. On the other hand, Twomey et al. (1984) pointed to the strong leverage of the SO$_2$-induced indirect effect on climate as compared to the direct effect. This leverage arises from the formation of a cloud droplet several micrometers in radius from a nucleus that in the dry state may have been 0.01–0.02 $\mu$m. Cloud formation increases the scattering cross section by 7–8 orders of magnitude. As a result, it is important to find a direct way to compare the two effects. In this section, we compare the direct and indirect effects of an increase in the sulfate concentration. The advantage of such a comparison is that the ratio between the two effects is independent of the rate of emission, the rate of conversion of SO$_2$ to sulfates, and the geographic distribution of sulfates.

For an increase in the concentration of the SO$_2$-derived aerosol particles $\Delta N_a$, the first-order estimation of the increase in planetary albedo, $\Delta A_d$, induced directly by the aerosol particles can be computed from

$$\Delta A_d = 2(1 - C_f)(1 - A_f^2)H\beta\Delta N_a \int_0^\infty \pi r^2 Q_s \frac{dn}{dr} \frac{dr}{dr}$$

In this equation, single scattering is assumed. The total cloud fraction is $C_f$, $A_f$ is the surface albedo, $H$ is the thickness of the aerosol layer, $\beta$ is the backscattering ratio, $dn/dr$ is the normalized particle size distribution, $r$ is the particle radius, $Q_s$ is the scattering efficiency, and therefore, $\pi r^2 Q_s$ is the scattering cross section of the particle. Equation (1) is similar to (2) of Charlson et al. (1992), except that the aerosol scattering cross section per unit volume and the vertical distribution are introduced instead of the aerosol optical thickness. We shall show that this substitution helps in the comparison between the direct and the indirect effects for a given size distribution. The factor of 2 accounts for the average solar zenith angle. Note that $C_f$ should exclude thin cirrus clouds that do not mask the aerosol effect. Due to the difficulties in estimating the fraction of the earth covered by the thin cirrus clouds, the total cloud cover is used.

Estimation of the indirect forcing is based on the relationship between the increase in the aerosol particle concentration, $\Delta N_a$, and a corresponding change in the cloud albedo, $\Delta A_c$. Following Kaufman et al. (1991), for a small $\Delta N_a$,

$$\Delta A_c = 0.17(\Delta \tau / \tau) = 0.057(\Delta N_d / N_d)$$
$$= 0.04(\Delta N_a / N_a) = 0.04 f_c(\Delta N_a / N_a)$$

where $\tau$ is the cloud optical thickness, $N_d$ is the concentration of the cloud droplets, $N_a$ is the initial CCN concentration. Respectively, $\Delta N_d$ and $\Delta N_a$ are the increases in $N_d$ and $N_a$, resulting from the increase in the aerosol particle concentration $\Delta N_a$. The fraction of aerosol particles, $f_c$, that can form CCNs depends on the maximum supersaturation reached in the cloud
TABLE 1. Results of Mie computations for the integrals in Eq. (5) for three log normal distributions and two refractive indexes. Types A, B, C, and D refer to various aerosol size distributions and refractive indices. The number distribution is computed for supersaturation of 0.1% and 0.5%.

<table>
<thead>
<tr>
<th>Aerosol type</th>
<th>A</th>
<th>B</th>
<th>C</th>
<th>D</th>
</tr>
</thead>
<tbody>
<tr>
<td>Refractive index—(n_r)</td>
<td>1.43</td>
<td>1.43</td>
<td>1.43</td>
<td>1.55</td>
</tr>
<tr>
<td>Particle mode radius—(R_g (\mu m))</td>
<td>0.03</td>
<td>0.03</td>
<td>0.05</td>
<td>0.03</td>
</tr>
<tr>
<td>Dispersion of the log-normal particle size distribution—(\sigma)</td>
<td>0.35</td>
<td>0.60</td>
<td>0.60</td>
<td>0.35</td>
</tr>
<tr>
<td>Scattering cross section, (\sigma_s)</td>
<td>5.9 (10^{-5})</td>
<td>1.2 (10^{-3})</td>
<td>1.1 (10^{-2})</td>
<td>9.4 (10^{-3})</td>
</tr>
<tr>
<td>Fraction of activated particles:</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>For supersaturation of 0.5%, (f_c (r_{min} = 0.02))</td>
<td>0.87</td>
<td>0.75</td>
<td>0.93</td>
<td>0.87</td>
</tr>
<tr>
<td>For supersaturation of 0.1%, (f_c (r_{min} = 0.07))</td>
<td>0.008</td>
<td>0.08</td>
<td>0.29</td>
<td>0.008</td>
</tr>
<tr>
<td>(R_{eff} (mass) (\mu m))</td>
<td>0.04</td>
<td>0.05</td>
<td>0.09</td>
<td>0.04</td>
</tr>
<tr>
<td>(R_{eff} (mass, super sat. 0.5%) (\mu m))</td>
<td>0.04</td>
<td>0.06</td>
<td>0.09</td>
<td>0.04</td>
</tr>
<tr>
<td>(R_{eff} (mass, super sat. 0.1%) (\mu m))</td>
<td>0.18</td>
<td>0.12</td>
<td>0.13</td>
<td>0.18</td>
</tr>
<tr>
<td>(\eta (super sat. 0.5))</td>
<td>10(^{-4}) - 8 (10^{-4})</td>
<td>2 (10^{-3}) - 0.02</td>
<td>0.014 - 0.13</td>
<td>1.4 (10^{-4}) - 1.3 (10^{-3})</td>
</tr>
<tr>
<td>(\eta (super sat. 0.1))</td>
<td>0.01 - 0.04</td>
<td>0.02 - 0.2</td>
<td>0.05 - 0.44</td>
<td>0.016 - 0.14</td>
</tr>
</tbody>
</table>

\[ f_c (r_{min} = 0.02) = \int_{r_{min}}^{\infty} \frac{dn}{dr} (\text{super sat. 0.5%}), \quad f_c (r_{min} = 0.07) = \int_{r_{min}}^{\infty} \frac{dn}{dr} (\text{super sat. 0.1%}), \quad \sigma_s = \int_0^{\infty} \pi r^2 Q_r \frac{dn}{dr} \]

and on the aerosol size distribution; \(f_c\) can be computed as the fraction of the number of particles that are larger than the minimum radius, \(r_{min}\), required for activation.

\[ f_c = \int_{r_{min}}^{\infty} \frac{dn}{dr} dr. \] (3)

According to Twomey (1977), \(r_{min}\) ranges from 0.02 \(\mu m\) for a supersaturation of 0.5% to 0.07 \(\mu m\) for a supersaturation of 0.1%. Examples for the values of \(f_c\) for three aerosol size distributions are given in Table 1.

The increase in the planetary albedo, \(\Delta A_i\), caused by the indirect forcing can be estimated by

\[ \Delta A_i = \Delta A_r C_{\gamma r} = 0.04 f_c (\Delta N_r / N_r) C_{\gamma r}, \] (4)

where \(C_{\gamma r}\) is the fraction of the earth’s surface covered by clouds that are affected by pollution (low to middle clouds). The ratio of the direct forcing to the indirect forcing, \(\eta\), is then

\[ \eta = \frac{\Delta A_d}{\Delta A_i} = 50 \left( (1 - C_{\gamma}) / C_{\gamma r} \right) (1 - A_i)^2 \times H\beta N_r \int_0^{\infty} \pi r^2 Q_r \frac{dn}{dr} \left( \int_{r_{min}}^{\infty} \frac{dn}{dr} dr \right)^{-1}. \] (5)

The aerosol particles are assumed to have a lognormal size distribution (Whitby 1978), \(\beta\) and \(C_{\gamma}\) are taken to be 0.29 and 0.6 (Charlson et al. 1991), \(H\) is 1 to 2 km, \(A_i\) is taken to be 0.15, \(C_{\gamma}\) is 0.2 to 0.3, and \(N_r\) is assumed to be in the range 100–300 particles cm\(^{-3}\) (Kaufman et al. 1991). The ratio of the two integrals in (5) is the most variable part that determines the ratio between the direct and indirect sulfate effects. This ratio is tabulated in Table 1 for three aerosol lognormal distributions and two refractive indexes, \(n_r\), which determines the scattering efficiency, \(Q_r\). The choice of \(n_r = 1.43\) is for a mixture between sulfate and water, while the choice of \(n_r = 1.55\) is for dry sulfate (Jursa 1985). The fine particle mode of Whitby (1978) and the AFGL model (Jursa 1985) are used as well as an intermediate model. For comparison, the mass-weighted effective radius defined below is also tabulated;

\[ R_{eff}(mass) = \left( \int_0^{\infty} r^3 \frac{dn}{dr} dr \right)^{1/3}. \]

The mass-weighted effective radius for the CCNs, also given in Table 1, is computed as the integral from the minimum activation radius for supersaturations of 0.5% and 0.1% (0.02 and 0.07 \(\mu m\), respectively):

\[ R_{eff}(mass, supersatur. 0.5%) = \left( \int_{0.02}^{\infty} r^3 \frac{dn}{dr} dr \right)^{1/3}, \]

\[ R_{eff}(mass, supersatur. 0.1%) = \left( \int_{0.07}^{\infty} r^3 \frac{dn}{dr} dr \right)^{1/3}. \]

The range of the ratio \(\eta\) for each combination of \(n_r\), \(\sigma\), and \(R_g\) and for the ranges of \(H\), \(C_{\gamma r}\), and \(N_r\) is given in the table. For the range of parameters given here and for the lower supersaturation of 0.1%, the ratio \(\eta\) is in the range from 0.01 to 0.5. For the supersaturation of 0.5%, it decreases to as low as 0.0001. Therefore, the direct effect is expected to be significantly smaller than the indirect effect, though the uncertainties in the computation of the indirect effect are large. In this paper we concentrate on the climatic cooling induced indirectly by the SO\(_2\)-derived particles. The aerosol model \(C\) with a corresponding ratio of the indirect to direct sulfate effects of \(\eta = 0.5\) is used in this paper. This model represents the lower limit of the predicted sulfate effect.

The direct effect of sulfates is estimated to be 0.6 W m\(^{-2}\) by Charlson et al. (1991). This result, together with the present estimate of \(10^{-4}-0.5\) in the ratio between the direct and indirect effects of sulfates, would
translate to a very large indirect forcing and, hence, an unreasonably strong climate cooling. This consideration shows the importance and uncertainties in the indirect sulfate effect. Therefore, further studies are required for the parameters given in Table 1 and for the assumptions on which computations of these parameters are based.

3. CO₂ and CCN concentrations

Anthropogenic CO₂ is emitted into the atmosphere mainly by fossil fuel and biomass burning. The past and anticipated CO₂ concentrations used in this study, are based on the consumption rates given in the Intergovernmental Panel on Climate Change (IPCC) (Houghton et al. 1990; Woodwell et al. 1983) and are reproduced in Fig. 1. The business as usual (BAU) scenario represents a continued increase in the consumption rate of fossil fuel at the present pace. The B and C scenarios represent a lower rate of increase. Scenario D represents a leveling off of the consumption rate close to the 1990 consumption rate and a decrease after the year 2010.

The rate of SO₂ emission, \( P_s(t) \), for the period 1900–1985 used in this study is taken from Möller (1984). The emission of SO₂ after 1985 is estimated using the IPCC scenarios for fossil fuel consumption. To convert the fossil fuel consumption rate into the SO₂ emission rate, it is assumed that the present ratio between SO₂ emission (Möller 1984) and fossil fuel consumption remain unchanged in the future. Therefore, the future emission rate of SO₂, \( P_s(t) \), is given by:

\[
P_s(t) = P_s(1985)P_f(t)[P_f(1985)]^{-1} \quad \text{for} \quad t > 1985,
\]

where \( P_f(t) \) is the rate of fossil fuel consumption according to the IPCC scenarios and \( P_f(1985) \) is the rate of fossil fuel consumption in 1985; \( P_s(1985) \) is the SO₂ (Woodwell et al. 1983; Houghton et al. 1990) will emission rate in 1985 taken from Möller (1984). Plots for \( P_s(t) \) are shown in Fig. 2 for the four scenarios.

In computing the increase in CCN, the following factors need to be considered: 1) The fraction of the sulfur that forms aerosol particles, \( f_a \); 2) The fraction of aerosol particles that have a potential to form cloud droplets, \( f_c \); 3) The lifetime of sulfate particles, \( T_s \), ranging from five (Garland 1978) to six days (Charlson et al. 1990); 4) The spatial distribution of sulfate particles. The sulfate particles, because of the short lifetime, are assumed to be confined to a fraction of the earth’s surface area, \( \alpha \), up to a height \( H \) above the surface.

These factors are a simplification of the atmospheric processes and transport that determine the sulfate lifetime and spatial distribution. A more accurate treatment of the evolution and spatial distribution of sulfates in the atmosphere can be obtained through the use of global models of the atmospheric sulfur cycle (Charlson et al. 1990; Charlson et al. 1991; Charlson et al. 1992), coupled with a three-dimensional global circulation model. By considering these factors, the density of sulfur that forms potential CCN is

\[
\rho_s(t) = f_c f_a P_s(t) T_s (\alpha S_e H),
\]

where \( S_e \) is the area of the earth’s surface. Based on the studies of Seinfeld (1986) and Wixson et al. (1980), Kaufman et al. (1991) estimated the values of \( f_a \) and \( f_c \) as 0.4 and 0.5 respectively. Charlson et al. (1990) used \( f_c = 0.5 \).

Anthropogenic CCN in ambient conditions of 70% relative humidity contains roughly half water and half nonaqueous material (Hänel 1981; Kaufman and Fraser 1983). The nonaqueous part contains sulfuric acid (H₂SO₄) or ammonium sulfate, (NH₄)₂SO₄ (Whitby 1978). Therefore, roughly a quarter of the nonaqueous mass of the CCN or an eighth of the total mass is sulfur (Fraser et al. 1984; Kaufman et al. 1991).
The density of sulfate particles is 1.5 g cm\(^{-3}\) (Fraser et al. 1984). Therefore, the relationship between the concentration of the anthropogenic CCN and the density of sulfur is given by

$$\Delta N_c(t) = 1.33 \rho_s(t) / r_m,$$

(8)

where \(r_m\) is the mass-weighted mean radius of the sulfate aerosol particles. (The units are g cm\(^{-3}\) for \(\rho_s\), cm for \(r_m\), and particles cm\(^{-3}\) for \(N_c\).) The mean value of \(r_m\) was estimated by Whitby (1978) to be \(\approx 1.5 \times 10^{-7}\) cm. Similar values (0.15 ± 0.02 \(\mu\)m) for the radius that corresponds to the peak in the volume distribution, were compiled by Hidy (1984) for urban and smoke aerosol. Integrals of the size distributions, reported in Table 1, suggest that the value of \(r_m\) for sulfate particles with radius above the minimum radius required for activation is between 0.04 and 0.18 \(\mu\)m. Thus, the large value of 0.15 \(\mu\)m that is used in the present climate modeling is a conservative value that may underestimate the cooling effect by the sulfate particles.

The present model assumes that there is a representative value for the lifetime and the size of the particles, independent of the rate of SO\(_2\) emission. In this case the density of sulfur that forms potential CCNs can be described by (7). The linear relationship between the emission of SO\(_2\) and the increase in the CCN concentration [Eqs. (7) and (8)] is a first-order approximation to the problem. It ignores, for example, the effect of particle concentration on cloud processes that affect the removal rate and consequently the life time of the particles (Baker and Charlson 1990). A dependence of the removal rate on SO\(_2\) concentrations can introduce a nonlinearity in the relation between SO\(_2\) concentration and the CCN concentration. An example for this nonlinearity is found in the characteristics of straitiform clouds measured off the coast of California, which shows an increase in the liquid water concentration in ship tracks. This is associated with a suppression of drizzle and increases in the lifetime of the cloud, the CCN, and the interstitial particles (Radke et al. 1989; Albrecht 1989; Nakajima et al. 1991). Some nonlinearities have been incorporated into a simpler model of the CCN concentration in the maritime boundary layer (Baker and Charlson 1990). Improvements to the model discussed here would require further measurements and analysis of the processes that determine the lifetime and size of the particles.

According to the studies of Twomey (1959, 1977) and Kaufman et al. (1991), the number of cloud droplets, \(N_d\), is related to the number of CCN, \(N_c\), by the empirical relationship

$$N_d(t) = N_c(t)^{0.71}.$$

(9)

The relationship between the cloud optical thickness, \(\tau\), and the density of cloud droplets for a constant liquid water content is given by Twomey (1977),

$$\frac{\tau(t)}{\tau_0} = \left( \frac{N_d(t)}{N_{d0}} \right)^{1/3} = \left( 1 + \frac{\Delta N_c(t)}{N_{c0}} \right)^{0.24},$$

(10)

where the subscript 0 denotes unperturbed situations.

There is a strong nonlinearity in the relationship between the change in the cloud optical thickness and the CCN concentration given by the exponent 0.24 in (10). Due to this nonlinearity, it is expected that, while changes in the fossil fuel consumption may at the present time cause a substantial forcing on the climate system (Kaufman et al. 1991), a constant increase in the emission rate in the future will cause a smaller forcing. Note that the dependence of the infrared forcing on the CO\(_2\) concentration is less nonlinear (power of 0.71—Kaufman et al. 1991) than the indirect sulfate effect.

The change in CCN due to fossil fuel burning as given by (8) is an averaged quantity over latitude bands. Because the source of anthropogenic sulfate aerosol is over land, it is expected that \(\Delta N_c\) is much larger over land than over oceans. Estimates of sulfate aerosol concentrations show that while the concentration over the ocean increased by a factor of 2–4 from preindustrial time, over the continents the concentration increased by a factor of 4–15 (Langner and Rodhe 1990; also in Houghton et al. 1990). Therefore, in this study we assume that the anthropogenic CCN concentration, \(\Delta N\) is three times larger over land than over oceans. However, the concentration of unperturbed CCN, \(N_{c0}\), is also expected to be larger over land than over oceans (Kaufman et al. 1991). As a result, the difference in the ratio \(\tau/\tau_0\) between land and ocean is not expected to be large. The concentration of CCN for unperturbed situations, \(N_{c0}\), is taken to be 100 cm\(^{-3}\) over land and 50 cm\(^{-3}\) over oceans (Kaufman et al. 1991). A zonal mean value of \(\tau/\tau_0\) is derived by averaging the values over land and oceans. Based on computations of anthropogenic sulfate distribution of Charlson et al. (1991), it is assumed that the anthropogenic sulfate aerosol is confined in the 20°–60°N band, which covers 26% of the earth’s surface, and is uniformly mixed in the lowest 2 km near the surface. Since only a small fraction of the sulfate aerosol is injected into higher altitudes, this fraction is ignored in the computations of the aerosol density in the lowest 2 km. Figure 3a shows the zonally averaged trends of \(\tau/\tau_0\) for the IPCC scenarios computed from (7), (8), and (10), with \(P(t)\) given by (6). A comparison between the evolution of CO\(_2\) (Fig. 1) and the cloud optical thickness (Fig. 3a) shows the difference between the characteristics of the forcing by the SO\(_2\)-induced cooling and the forcing due to the greenhouse effect of CO\(_2\). The much longer CO\(_2\) lifetime causes a monotonous increase in the CO\(_2\) concentration for all IPCC scenarios, while the cloud optical thickness, due to the short lifetime of atmospheric sulfate aerosol, closely follows the emissions scenarios. Following Charlson et al. (1987) and Wigley (1989), it is assumed that low clouds (cumulus, stratocumulus,
and stratus), as well as middle clouds (altocumulus), are affected by the SO$_2$-derived particles. For the middle clouds, the change in the CCN concentration is much smaller than that for low clouds, but the unperturbed CCN concentration, $N_{00}$, should also be much smaller. Therefore, the relative change in the CCN concentration, $\Delta N_c / N_{00}$, and, hence, the ratio of the optical thicknesses, $\tau / \tau_0$, are assumed to be the same for low and middle clouds.

By combining Eqs. (7), (8), and (10), the fractional change in cloud optical thickness is given by

$$\frac{\Delta \tau}{\tau_0} = \left(1 + \frac{1.33 f_f f_a P_0 T_s}{N_{00} r_m \alpha S_c H} \right)^{0.24} - 1. \quad (11)$$

The sensitivities of $\Delta \tau / \tau_0$ to the parameters $f_f$, $f_a$, $T_s$, $N_{00}$, $r_m$, $H$, and $\alpha$ are computed from the above equation and are given in Table 2. Since the change in the cloud albedo is proportional to $\Delta \tau / \tau_0$ [Eq. (2)], the radiative forcing due to emission of anthropogenic SO$_2$ is also proportional to $\Delta \tau / \tau_0$. The radiative forcing shown in Table 2 was computed using the climate model described in section 4.

The perturbed values are specified according to the uncertainty in these parameters (Kaufman et al. 1991). Uncertainties in the particle size cause the largest uncertainty in the cloud optical thickness and in the radiative forcing. It is possible that a substantial increase in pollution, accompanied with an increase in the CCN concentration, may reduce the supersaturation reached in the cloud, causing an increase in the effective mass-weighted particle radius (as indicated in Table 1) and a reduction of the efficiency of the effect of aerosol on clouds.

In Table 2, two cases of uncertainties associated with the fraction of the globe covered by pollution, $\alpha$, are given. The first case is a direct result from the uncertainty in the sulfate dispersion model (i.e., Charlson et al. 1991) between the present value of 0.26 and a

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**Table 2.** The sensitivity of the cloud optical thickness and the radiative forcing to uncertainties in the parameters that define the radiative forcing of sulfate particles. Results are given for the BAU IPCC scenario. For simplicity, all perturbations from the base model were performed in the direction of increasing the forcing. An uncertainty of a factor of 17 in the product of the parameters (see text) causes an uncertainty of a factor of 4 in the optical thickness in 1980 and a factor of 3 in 2060. The last line in the table represents the results from a possible expansion of the fraction of the earth area covered by pollution from 0.26 in 1980 to 0.6 in 2060 due to industrial development in countries south of 20° lat.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Perturbation</th>
<th>1980</th>
<th>2060</th>
<th>1980</th>
<th>2060</th>
</tr>
</thead>
<tbody>
<tr>
<td>Base value</td>
<td>none</td>
<td>0.40</td>
<td>0.80</td>
<td>0.38</td>
<td>0.76</td>
</tr>
<tr>
<td>$f_f$</td>
<td>0.5 → 0.7</td>
<td>0.49</td>
<td>0.94</td>
<td>0.47</td>
<td>0.89</td>
</tr>
<tr>
<td>$f_a$</td>
<td>0.4 → 0.6</td>
<td>0.51</td>
<td>0.97</td>
<td>0.48</td>
<td>0.92</td>
</tr>
<tr>
<td>$T_s$</td>
<td>6 → 10 days</td>
<td>0.54</td>
<td>1.02</td>
<td>0.52</td>
<td>0.97</td>
</tr>
<tr>
<td>$N_{00}$</td>
<td>100 → 50 cm$^{-3}$</td>
<td>0.60</td>
<td>1.10</td>
<td>0.57</td>
<td>1.05</td>
</tr>
<tr>
<td>$r_m$</td>
<td>0.15 → 0.10 μm</td>
<td>0.79</td>
<td>1.37</td>
<td>0.75</td>
<td>1.31</td>
</tr>
<tr>
<td>$H$</td>
<td>2 → 1.0 km</td>
<td>0.60</td>
<td>1.10</td>
<td>0.57</td>
<td>1.05</td>
</tr>
<tr>
<td>$\alpha$</td>
<td>0.26 → 0.4</td>
<td>0.3</td>
<td>0.64</td>
<td>0.46</td>
<td>0.94</td>
</tr>
<tr>
<td>Combined uncertainty</td>
<td>1.0 → 17</td>
<td>1.7</td>
<td>2.6</td>
<td>1.6</td>
<td>2.4</td>
</tr>
<tr>
<td>$\alpha$</td>
<td>0.26 → 0.6 (for 2060)</td>
<td>0.51</td>
<td>1.12</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

$f_f$ is the fraction of sulfate particles that have the potential to form cloud drops, $f_a$ is the fraction of the emitted sulfur that form sulfate particles, $T_s$ is the sulfate particles lifetime, $N_{00}$ is the preindustrial concentration of CCN in the NH over the land, $r_m$ is the mass-weighted average particle size, $H$ is the aerosol scale height, and $\alpha$ is the fraction of the globe that is covered by pollution.
high value of 0.4. The second case represents the possibility that future increase in fossil fuel consumption may not only in the NH but also in countries located south of the present 20°-60° latitudinal zone. In this case the present value of $\alpha = 0.26$ is increased to $\alpha = 0.60$ by the year 2060. In both cases, an increase in $\alpha$ causes a reduction in the change of the cloud optical thickness, but an increase in the global radiative forcing. The increase in the forcing results from the nonlinearity of the model as indicated by (11).

The large uncertainties in Table 2 do not allow a linear summation of the individual errors in order to compute the total error. As a result, we used an empirical approach to compute the total error. A simulation was performed in which each parameter in Table 2 had the same probability to be equal to the base value or the perturbed value. The combined product of the parameters in the parentheses of Eq. (11) was computed for each case and compared to the unperturbed values. For the 40 cases studied, the product differed from the base value on average by a factor of 14. For 65% of the cases the ratio of the perturbed to unperturbed values were under a factor of 17. Therefore, the overall uncertainty in the product is assumed to be a factor of 17. The corresponding perturbed values of $\Delta r/\tau$ and of the forcing are given in the table. Due to the nonlinear dependence of $\Delta r/\tau$ on the parameters [exponent of 0.24 in Eqs. (10) and (11)] the overall uncertainty in the effect of the sulfate aerosol on the cloud optical thickness [using Eq. (11)] is a factor of 4 for 1980 and 3 for 2060.

Examples of the time dependence of the sensitivity are shown in Fig. 3b. The change in the cloud optical thickness due to a 50% increase in aerosol lifetime and a 33% decrease in particle size are plotted. An increase of 50% in the particle lifetime causes the optical thickness to increase by only $\sim 10\%$. An increase of 33% in the particle size represents more than a doubling of the particle volume. The corresponding change in the cloud optical thickness is $\pm 35\%$.

4. The climate model

The climatic effects of anthropogenic CO$_2$ and SO$_2$ are studied using the zonally averaged multilayer energy balance model of Peng et al. (1987). It has five layers in the atmosphere, one mixed layer in the upper ocean, and one simple two-dimensional advective-diffusive deep ocean in order to simulate long-term climate response to external forcing. Seasonal effects are included. Snow and sea ice are computed. Relative humidity and clouds are specified parameters.

The model includes a rather detailed treatment of radiative transfer. In computing the radiative heating/cooling for the five atmospheric layers, each layer is divided into three sublayers for proper flux integrations in the vertical. The thermal IR fluxes are computed using the algorithm of Chou et al. (1991), which includes the absorption due to water vapor, CO$_2$, O$_3$, CH$_4$, and N$_2$O. Except for cirrus, clouds are assumed to be black in the thermal IR region. The cirrus clouds are treated as greybodies with an emissivity of 0.8. The solar fluxes are computed using the algorithms of Chou (1986, 1990) for the absorption due to water vapor, CO$_2$, and O$_3$, and using the algorithm of Lacis and Hansen (1974) for the absorption due to O$_3$. The cloud albedo is computed using the discrete-ordinate algorithm of Stamnes et al. (1988).

Heat transports in the atmosphere and at the earth's surface are parameterized as functions of temperature. The effect of large-scale eddies is included in the regions poleward of 20° latitude and is parameterized on the basis of quasigeostrophic theory. The effect of the mean meridional circulation is included in the tropics but is neglected in the extratropical regions. The heat convergence due to small-scale eddies is parameterized as a diffusion process. The release of latent heat is computed from the difference between the total moisture convergence due to all scales of motion and the increase in atmospheric moisture storage (with the relative humidity fixed in the model, the atmospheric moisture storage is a function of temperature).

The deep ocean has a large heat capacity and delays the climate response to external forcing. It is represented in the model by two polar regions of downwelling and a vast region of upwelling between $\pm 60°$ latitudes. Each of the three regions is divided into 10 layers of 400-m thickness. The heat exchange between the deep ocean and the ocean-mixed layer in the upwelling region depends significantly on the diffusion process. A constant diffusion coefficient of 0.65 cm$^2$ s$^{-1}$ is used in the upwelling region.

Clouds are grouped into five types: Ci—cirus, above 10 km; As—altostratus, 4–5 km; Cu—cumulus, 1–5 km; St—stratus, 0–1 km; Ns—nimbostratus, 2–5 km;Cb—cumulonimbus, 3–12 km. Seasonal cloud cover and heights for the Northern Hemisphere are taken from London (1957). The cloud cover for the Southern Hemisphere is assumed to be 8% larger than the Northern Hemisphere. By assuming random overlapping between clouds at different heights, the cloud cover as viewed from space is used for radiative transfer calculations. For the low (Cu, St) and middle (As) clouds, which are assumed to be affected by sulfate aerosols, the range of aerial coverage as viewed from space is 0.13–0.18 for the former and 0.05–0.1 for the latter, depending upon the season and latitude. Thus, the average aerial coverage of clouds affected by sulfate aerosols is $\approx 0.23$. Cloud optical thicknesses are extracted from the aircraft measurements of Feigelson (1978) and are specified in the model; these values are given in Peng et al. (1982).

In response to a doubling of atmospheric CO$_2$ concentration, the model predicts a warming of 2.6°C in the global average surface temperature. If we assume that the response to a doubled CO$_2$ is half of the re-
sponse to a quadrupled CO₂, as is suggested by Manabe and Stouffer (1980) and Wetherald and Manabe (1981), then the latitudinal and seasonal distribution of the equilibrium response of surface temperature as simulated in our model is in good agreement with that of Manabe and Stouffer (1980), which also has specified cloud parameters. By analyzing some GCM results, Arking (1991) and Schlesinger (1989) determined that the cloud feedback factor is ≈0.1 for the GFDL model (Wetherald and Manabe 1988) and ≈0.2 for the GISS model (Hansen et al. 1984). These values translate to an amplification of global warming by a factor of ≈1.5 (Arking 1991). If this effect of cloud feedback is included in our model, then the global warming due to a doubled CO₂ would be ≈4.0°C, which is consistent with most recent GCM simulations.

5. Climatic effects of fossil fuel burning

The climatic effect of sulfate aerosol is simulated by scaling the cloud optical thickness according to the curves given in Fig. 3a for the two extreme IPCC scenarios: BAU and D. The values of the cloud optical thickness specified in the climate model (Peng et al. 1987) are assumed to be representative of the year 1980 and the cloud optical thickness at other years is scaled by the optical thickness ratio plotted in Fig. 3a.

The radiative forcing of CO₂ and SO₂ are shown in Fig. 4 for both the BAU and D scenarios. The radiative forcing is computed as follows. 1) The climate model is run to equilibrium with the atmospheric CO₂ concentration and the value of ΔT/ΔT representative of the year 1990. 2) The computation is repeated, one year at a time, for one full year by specifying the CO₂ concentration and ΔT/ΔT shown in Fig. 3a; the model temperatures throughout the one-year course are fixed at the same values as the equilibrium run for 1990. 3) Annual and global radiative budgets at the top of the atmosphere are computed for each year. 4) The differences in the radiation budgets at the top of the atmosphere between the perturbed runs and the equilibrium run are defined as the forcing to the climate system.

While in 1980 the SO₂-induced forcing reduced the forcing due to CO₂ by 50%, the reduction in 2060 is 20% for the BAU scenario and 15% for scenario D. The smaller effect in 2060 than in 1980 results from the nonlinear dependence of the cloud optical thickness on the pollution concentration. The effect is still smaller for scenario D because the atmospheric SO₂ responds to the decrease in fossil fuel consumption much faster than the CO₂ concentration. The SO₂-induced forcing in 1990 is 0.45 W m⁻², which can be compared with the direct SO₂-induced forcing of 0.6 W m⁻² as estimated by Charlson et al. (1991). The two forcings are comparable, even though we would expect a larger forcing for the indirect effect as discussed in section 2. The difference may arise largely due to the conservative values of the aerosol parameters used in this paper, which tend to reduce the indirect SO₂-induced effect.

The simulated transient responses of the global surface temperature to the anthropogenic CO₂ and SO₂ are shown in Fig. 5. Table 3 shows the changes in the surface temperature simulated for the years 1980 and 2060. The surface temperature response is directly related to the radiative forcing shown in Fig. 4. In the year 1980, the cloud optical thickness increased by 40% from the prehistorical era (see Fig. 3) due to a threefold increase in the CCN concentration. A 40% increase in the optical thickness is equivalent to a 0.06 increase in the fractional cloud albedo. As a result, the global warming is reduced from 0.3°C to 0.1°C. By the year 2060, the effect of the SO₂-derived CCN on clouds will

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**Fig. 4.** Radiative forcing relative to the preindustrial climate for the IPCC BAU scenario (curves a and b) and for the IPCC scenario D (curves c and d). Curves a and b are for the CO₂ forcing and curves b and d for the effect of SO₂-induced cooling.

**Fig. 5.** Transient response of the global surface temperature to the anthropogenic CO₂ and SO₂ emissions. Radiative forcing due to CO₂ only is given by curve a (BAU scenario) and curve b (scenario D). Surface temperature response due to both CO₂ and SO₂ are given by curve c (BAU scenario) and curve d (scenario D).
Table 3. The CO₂ and SO₂-induced changes in the surface temperature (°C) for the years 1980 and 2060 for the Southern and Northern hemispheres (BAU stands for business as usual).

<table>
<thead>
<tr>
<th>Emission scenario</th>
<th>Change from 1900 to 1980</th>
<th>Change from 1900 to 2060</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>NH</td>
<td>SH</td>
</tr>
<tr>
<td>CO₂ only</td>
<td>0.31</td>
<td>0.29</td>
</tr>
<tr>
<td>CO₂ + SO₂</td>
<td>0.31</td>
<td>0.29</td>
</tr>
<tr>
<td>SO₂-induced cooling</td>
<td>0.30</td>
<td>0.08</td>
</tr>
<tr>
<td>Scenario D</td>
<td></td>
<td></td>
</tr>
<tr>
<td>CO₂ only</td>
<td>0.31</td>
<td>0.29</td>
</tr>
<tr>
<td>CO₂ + SO₂</td>
<td>0.31</td>
<td>0.29</td>
</tr>
<tr>
<td>SO₂-induced cooling</td>
<td>0.30</td>
<td>0.08</td>
</tr>
</tbody>
</table>

mostly be saturated for the BAU scenario, due to the nonlinearity of the SO₂-induced forcing. It will also be very small for scenario D, due to the decrease of the emission rates. On the other hand, due to the long CO₂ lifetime, the CO₂ concentration continues to increase. As a result, by the year 2060 the CO₂ warming continues to increase, but the SO₂ cooling is only moderate. The model predicts a warming of 1.4°C and 0.9°C, respectively, for the BAU and D scenarios. The reduction of the global warming in 2060 due to SO₂ is 25% for the BAU scenario. Note that there is a trend of strong warming after 1980. The surface temperature increases by 0.15° from 1980 to 1990. This strong warming is similar to the increase in the global temperature record of 0.2° during the same period (IPCC report).

There are large uncertainties in the estimation of the SO₂-derived CCNs (Wigley 1989; Kaufman et al. 1991; Hansen and Lacis 1990; Charlson et al. 1990; Baker and Charlson 1990). In section 3 we discussed the effect of uncertainties in the aerosol properties on the cloud optical thickness and radiative forcing (see Table 2 and Fig. 3b). The uncertainty in the SO₂-induced radiative forcing and climate cooling is a factor of 3–4. We have also studied the sensitivity of the SO₂-induced cooling to two additional parameters: (a) the type of clouds affected by the pollution, and (b) the latitudinal distribution of the pollution. It is found that elimination of the exposure of middle clouds to the pollution decreases the SO₂-induced cooling by almost a factor of 2. On the other hand, an increase in the latitudinal extent of the pollution from 20°–60°N to 20°–90°N increases the cooling by only <0.1°C.

Due to increasing concern for the environmental impact of fossil fuel combustion, an international concerted effort to reduce the CO₂ and SO₂ emission to the atmosphere is expected to be forthcoming. The emission of SO₂ may be filtered or the consumption of fossil fuel may even be reduced by using other forms of clean energy (e.g., nuclear power or solar energy), as modeled by the IPCC scenario D. In Fig. 6, we simulate the impact of filtering SO₂ from the emission products in order to reduce acid deposition (Wigley 1989). The fossil fuel consumption is the same as in the BAU scenario, but the SO₂ emission to the atmosphere after the year 2020 is drastically reduced to zero. The dashed curve in Fig. 6 shows that the surface temperature increases steeply by 0.4°C in the first ten years of the cessation of SO₂ emission. This strong effect should be taken into consideration when planning a reduction in acid deposition.

6. The difference in the response between the hemispheres

As was pointed by Schwartz (1988) and Wigley (1989), the difference in the surface temperature responses to the radiative forcing between the NH and

![Fig. 6. Transient responses of the global surface temperature to the anthropogenic CO₂ and SO₂ emissions for the BAU scenario. After the year 2020, the SO₂ is filtered from emitting to the atmosphere in an effort to reduce acid rain (dashed curve). The responses for the BAU scenario are given by curve a for the change in CO₂ only and curve c for changes in both CO₂ and SO₂.](image)
of burning fossil fuel on climate. Although the prediction of future climate is very uncertain, it is concluded that the emission of SO$_2$ for the BAU scenario has a potential of offsetting the CO$_2$-induced warming by 60% at the present time (1990) and by 25% by the year 2060. An effort to reduce smog and acid rain by reducing the emission of SO$_2$ to the atmosphere could enhance the warming due to increasing concentration of greenhouse gases.

Every attempt has been made to simulate as accurately as possible the climatic effect of anthropogenic SO$_2$. Due to the short lifetime of sulfate aerosol and the uneven spatial distribution of sources, however, a number of difficulties are encountered. The major uncertainties concern the relationship between the SO$_2$ emission and the CCN production, the relationship between the CCN concentration and the cloud optical thickness, the value of the background CCN concentration, and the vertical distribution of the SO$_2$-derived CCN. In order to fully assess the impact of the anthropogenic SO$_2$ on climate, we need to improve our understanding of these processes.

This study addresses only two aspects of the climatic effect of burning fossil fuels, namely, the SO$_2$ effect on cloud albedo and the CO$_2$ greenhouse effect. Trace gases other than CO$_2$ have a combined greenhouse effect nearly comparable to that of CO$_2$ (Ramanathan et al. 1985). On the other hand, the reduction of cloud droplet size caused by the SO$_2$-derived CCN has an effect of prolonging the cloud lifetime (Radke et al. 1989; Albrecht 1989), thereby increasing the planetary albedo. The aerosol backscattering of radiation to space can also increase the planetary albedo (0.6 W m$^{-2}$, Charlson et al. 1991). These effects are not addressed in this study.

Despite the uncertainty of a factor of 3 to 4 in the radiative forcing and the temperature response, it is concluded that changes in the cloud albedo due to the emission of SO$_2$ and other particles (e.g., smoke and dust) should be accounted for in modeling the climatic effect of fossil fuel burning, since they have the potential of partially offsetting the greenhouse warming.

Even though the SO$_2$-induced cooling takes place mainly in the Northern Hemisphere, and is stronger over the continents than over the oceans, the model predicts that the difference in the temperature response of $-0.2^\circ$C can be compared with the difference of $-0.1 \pm 0.2^\circ$C between the two hemispheres, as indicated from historical records (Wigley 1989). The ocean plays a complex role in generating this difference. According to our model, the ocean response to the forcing of CO$_2$ only is much smaller than that of the land (see Fig. 7), mainly due to the large heat capacity of the oceans.

7. Summary and conclusions

A two-dimensional model with a detailed treatment of the SO$_2$ effect on cloud condensation nuclei, optical thickness, and albedo is used to simulate the impact

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REFERENCES


