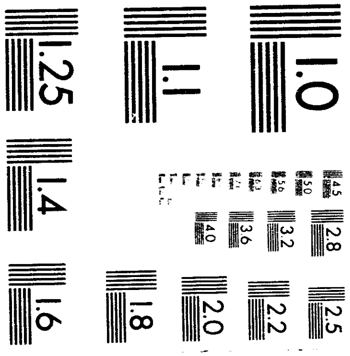
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CLIMATE EFFECTS OF ANTHROPOGENIC SULFATE: SIMULATIONS FROM A COUPLED CHEMISTRY/CLIMATE MODEL

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1. INTRODUCTION

The radiative balance of the earth-atmosphere system is mediated by the greenhouse gases, clouds, and aerosols. The radiative perturbation due to increases in greenhouse gas concentrations between 1765 and 1990 has been estimated to be 2.45 W m^{-2} (IPCC, 1990), with 60% of the forcing attributed to the increase of anthropogenic carbon dioxide. The anthropogenic emissions of CO_2 are primarily from fossil fuel combustion which is, however, also a major source of sulfur dioxide. It has been estimated that anthropogenic sulfur emissions dominate over natural emissions by a factor of 2 on a global average (with 90% of the anthropogenic sources in the Northern Hemisphere) (Spiro et al., 1992), and they have been significantly increased during the past several decades (Charlson et al., 1992; Hameed and Dignon, 1988). Table 1 lists the current estimates of the global gaseous sulfur emissions (Penner, 1993). Model simulations (Penner et al., 1993) illustrate that photochemical reactions of these emitted sulfur compounds lead, through gas-to-particle conversion, to a large increase in the concentration of aerosol sulfate especially over and around industrialized regions. Within these areas the anthropogenic emissions are larger than natural emissions by about a factor of ten or even more (Galloway et al., 1984).

Radiative influences of aerosol sulfate on climate can either be direct or indirect. Particulate sulfate can scatter solar radiation, thereby, directly changing the planetary radiation budget. For the indirect effect, sulfate-containing aerosols modify the microphysics of clouds by acting as cloud condensation nuclei (CCN) and enhancing the cloud reflectivity which is determined in part by the number concentration of CCN. Sulfate-containing aerosols may also alter precipitation development by affecting the mean droplet size and thereby influencing cloud lifetimes and modifying the hydrological cycle.

Several studies have investigated the direct impact of aerosol sulfate on climate using different approaches. For example, Charlson et al. (1991) estimated the global radiative forcing due to scattering by anthropogenic sulfate at about -0.6 W m^{-2}, based on the burden of sulfate simulated by a 3-D chemical-transport model along with estimates of the scattering and back-scattering coefficients per unit mass concentration. A stronger forcing -1 W m^{-2} was predicted by a hemispheric-mean box model calculation (Charlson et al., 1992). These calculated forcings were estimated to be uncertain by a factor of 2. On the other hand, Kiehl and Briegleb (1993) used cloud field from the NCAR CCM2 with monthly mean sulfate distributions from a 3-D chemical-transport model (Langner and Rodhe, 1991), and estimated the direct radiative forcing by sulfate aerosols at only -0.3 W m^{-2}. In addition, their sensitivity study showed a 10% variation in the direct sulfate forcing as a result of changes to the chemical and physical nature of sulfate aerosols (e.g., size distribution and chemical composition). They also indicated that the difference in the magnitude of the direct sulfate forcing between their results and those of Charlson et al. (1991) was due to the difference in modeling of the optical properties of the sulfate aerosols.

The indirect forcing by aerosol sulfate through cloud processes is more difficult to estimate in part because of the large uncertainty in relating aerosol number distribution to sulfate mass concentration. The sulfate-containing aerosol number distribution depends on the processes which form particulate sulfate. H_2SO_4 formed in the gas phase may either condense

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onto existing particles causing them to grow, or form new particles through homogeneous nucleation. Sulfate may also form by aqueous conversion of SO_2 to $SO_4^=$ in cloud drops. The resulting distribution is of importance in evaluating its impact on cloud optical properties. Furthermore, there are also complications as a result of the non-linearity of cloud microphysics where the cloud droplet number is a function of updraft velocity and aerosol characteristics. With a simple radiation balance equation, Charlson et al. (1992) estimated the global shortwave radiative forcing by clouds is about -1 W m^{-2} for an assumed global enhancement in cloud droplet number of 15% in marine stratiform clouds. However, the indirect forcing cannot be quantified until a more precise relationship between sulfur sources, aerosol number distribution, and cloud droplet concentration is available.

In this paper, we use a more comprehensive approach by coupling a climate model with a 3-D global chemistry model to investigate the forcing by anthropogenic aerosol sulfate. The chemistry model treats the global-scale transport, transformation, and removal of SO_2, DMS and H_2SO_4 species in the atmosphere. The mass concentration of anthropogenic sulfate from fossil fuel combustion and biomass burning is calculated in the chemistry model and provided to the climate model where it affects the shortwave radiation. We also investigate the indirect effect, with cloud nucleation parameterized in terms of local aerosol number, sulfate mass concentration, and updraft velocity. Our simulations indicate that anthropogenic sulfate may result in important increases in reflected solar radiation, which would mask locally the radiative forcing from increased greenhouse gases. Uncertainties in these results will be discussed.

Table 1. Estimates of global emission of gaseous sulfur compounds (Tg S/yr)

Source	Annual Flux	Range
Anthropogenic	77	60 - 100
Biomass burning	2.2	1 - 4
Oceans	25	12 - 40
Soil and Plants	1	0.1 - 10
Volcanoes	10	3 - 20
TOTAL	115.2	76.1 - 174

2. COUPLED CHEMISTRY/CLIMATE MODEL

The chemistry model we use is the Lawrence Livermore National Laboratory tropospheric chemistry model (called GRANTOUR). This model treats the global-scale transport, transformation, and removal of trace species in the atmosphere (Walton et al., 1988). GRANTOUR is a Lagrangian parcel model which can be run either off line, using the meteorology fields from a general circulation model, or interactively, in a coupled mode where the meteorology fields are influenced by the calculated sulfate concentration. This chemistry model divides the atmosphere into constant-mass air parcels that are advected by the wind field on a fixed Eulerian grid. There are 50,000 air parcels with dimensions about 100 mb × 330 km × 300 km in present simulations and the time interval is 6 hours. The climate model is the Livermore/NCAR CCM1 with R15 resolution. The solar radiation package in CCM1 has been replaced by a delta-Eddington treatment. The meteorology fields from CCM1 are averaged over a 12-hour time period for use by GRANTOUR.

In the present model simulation, the atmospheric sulfur is supplied by 1) anthropogenic sulfur (SO_2) from fossil fuel combustion, industrial sources, and biomass burning and 2) natural sources such as biogenic ocean sulfur (DMS), terrestrial soils (DMS, H_2S), and vegetation (H_2S). These surface-based sources are adapted from the inventories by Spiro et al. (1992) and Benkovitz (1982) and are input into GRANTOUR with a vertical profile that is assumed to be constant in mixing ratio in the lowest 100 mb. For ocean emissions of DMS, the values from Spiro et al. (1992) were doubled. To treat the gas phase reactions of DMS and SO_2 with OH, a background concentration of OH is specified according to the latitudinally- and seasonally-varying calculated concentrations from the LLNL 2-D model (Penner et al, 1991). The aqueous reaction to convert SO_2 to $SO_4^=$ in clouds was assumed to have an average e-folding lifetime of 30 hours at 40° N at the surface in summer. This e-folding lifetime is scaled to be proportional to the square of the locally specified concentration of OH at other locations. SO_2 and $SO_4^=$ may also be scavenged by precipitation. The scavenging coefficients are set to 0.8 cm hr^{-1} and 1.5 cm hr^{-1} for SO_2 in stratiform and convective precipitation, respectively, and to 5.0 cm hr^{-1} and 1.5 cm hr^{-1} for sulfate in these two precipitation types. In addition, when air parcels are in the lowest 100 mb, deposition velocities of 0.1 and 0.8 cm s^{-1} are applied to $SO_4^=$ and SO_2, respectively. Besides the aforementioned sulfur species, air parcels also carry non-sulfate background aerosols from a uniform source over the continents excluding Antarctica. The background aerosols are assumed to be a mixture of organic matter, NO_3^-, NH_4^+, etc. with the same dry deposition velocity as for $SO_4^=$, and scavenging coefficients of 2.5 cm hr^{-1} in stratiform and 0.75 cm hr^{-1} in convective precipitation. In the coupled mode, GRANTOUR provides sulfate concentrations to CCM1 for use in calculating the radiative forcing and also provides background particle concentrations for use in the cloud droplet parameterization.

The direct effect of aerosol sulfate on climate is calculated by specifying the scattering coefficient for sulfate. Since sulfate particles are hygroscopic, the scattering coefficient depends on the relative humidity. This dependence is adapted from Charlson et al. (1984) where the hygroscopic growth factor is about 1.7 at 80% RH (8.5 m^2 g^{-1} at wavelength 0.55 μm). It is also assumed that the scattering coefficient in the near infrared spectral range is only 40% of the value used for the ultra violet and visible spectral ranges.

To evaluate the indirect effect, we have to know how the aerosol size distribution might change and how many extra cloud droplets might be activated as a result of particulate sulfate, since the optical thickness of cloud is proportional to the cube root of the droplet number (Twomey 1977). However, the relationship between aerosol size distribution and a given mass of aerosol sulfate is not well understood. In order to assess the indirect effect, we assumed the aerosol was an internal mixture, where the aerosol size distribution is determined by condensation of sulphuric acid vapor on a prescribed pre-existing background (non-sulfate) particle distribution and by aqueous-phase oxidation of SO_2 followed by evaporation of the droplets. It is assumed that 25% of aerosol sulfate is distributed to all particles by condensation and 75% to particles which are larger than the minimum size of CCN by cloud processes. This approach will not change the total aerosol number, but the resulting sulfate-containing particle size distribution will grow to a larger size. The background distribution is assumed to be the superposition of three log-normal functions,

$$\frac{d\ N/N_{total}}{d\log d} = \sum_{i=1}^{3} N_i\ \exp\left\{ \frac{\left[\log\left(\frac{d}{D_i}\right)\right]^2}{2\log\sigma_i^2} \right\} \qquad (1)$$

Our assumed size distribution parameters for continental and marine background aerosols are listed in Table 2. The mass fraction of soluble material in the background aerosol is assumed to be 50% for continental and 70% for marine background particles. We parameterize the number of cloud droplets in terms of aerosol size distribution, total number, and vertical velocity using a detailed micro-physical Lagrangian model (Chuang et al., 1992). This Lagrangian model computes the spectral evolution of interstitial aerosols and cloud drops and then predicts the fraction of aerosols nucleated across the cloud base.

Table 2. Parameters of the normalized background size distribution

	N_i	D_i (μm)	$\log\sigma_i$
Continental	1.00	0.035	0.25
	0.50	0.10	0.30
	0.0015	1.20	0.35
Marine	0.50	0.05	0.25
	1.00	0.20	0.15
	0.02	0.70	0.15

3. RESULTS

Climate forcing due to anthropogenic sulfate is calculated as the difference in top of the atmosphere solar radiation with and without anthropogenic sulfate at each time the radiation routine is called. Figure 1 shows the distribution of annual average global sulfate forcing. The range is between 0 and - 5 W m^{-2}. This forcing is relatively large between 10° N and 60° N which is consistent with the large magnitude of anthropogenic sulfur emissions in this latitude belt. Three maxima located at Europe, North American, and East Asia are easily identified.

The climate forcing can be expressed in terms of the changes in the global average solar radiation balance as

$$\Delta F = \Delta F_{clear} + f_c\,(\Delta F_{overcast} - \Delta F_{clear}) \qquad (2)$$

where F is the solar radiative flux at the top of the atmosphere, f_c is the fractional cloudiness, Δ denotes the difference with or without anthropogenic sulfate.

The first term on the right hand side indicates the changes in the average clear sky solar radiative flux and, by itself, measures the direct effect of the sulfate aerosol assuming a cloud-free planet. The second term represents the change in the difference between the net solar radiative flux of the overcast and clear skies (cloud forcing). Magnitudes corresponding to each component in (2) for the simulation presented here are listed in Table 3.

Table 3. Change in solar radiation balance

	N. H.	S. H.	Global
ΔF_{clear}	-1.004	-0.225	-0.615
$f_c(\Delta F_{overcast} - \Delta F_{clear})$	-0.411	-0.204	-0.307
ΔF	-1.415	-0.429	-0.922

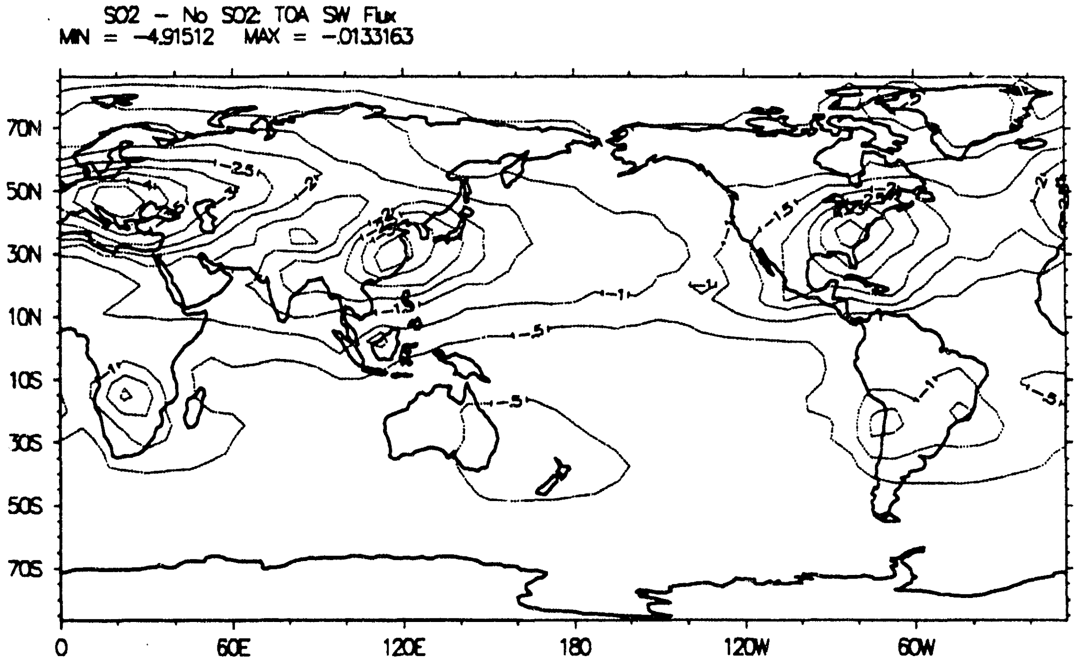

Figure 1. Distribution of annual average global sulfate forcing. Contour is shown every -0.5 W m^{-2}.

4. CONCLUSIONS

The direct sulfate forcing may be isolated from the total sulfate forcing by considering simulations with this model that did not include the indirect effect (Taylor and Penner, 1993). In those model simulations, the total forcing was approximately three-quarters of the clear-sky forcing. The reason for this is that when the sky is overcast, the clouds reduce the sulfate aerosol forcing by roughly 50%. Since the clouds cover roughly half the globe, the global sulfate forcing is reduced by only 25%.

Using the same accounting here, we estimate from Table 1 that the direct forcing of aerosol sulfate is ~ -0.45 W m^{-2}. This implies that the indirect effect due to enhanced cloud albedo is ~ -0.47 W m^{-2}. We conclude that current concentrations of anthropogenic sulfate have direct and indirect effects that may be comparable in magnitude and at least locally will tend to mask the warming effects of increased greenhouse gases.

The magnitude of the indirect effect calculated here is particularly uncertain, because the assumed fraction of particulate sulfate produced by condensation and by in-cloud oxidation for the internal mixing approach may not be correct Uncertainties can also arise from the prescribed 'background non-sulfate particle distribution which must result from a variety of processes (e.g., homogeneous nucleation, coagulation, dry and wet deposition) involving the entire suite of aerosol types such as nitrate and organic carbon. To quantity these uncertainties, sensitivities tests will be performed in a future study.

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5. REFERENCES

Benkovitz, C. M., 1982: Compilation of an inventory of anthropogenic emissions in the United States and Canada. *Atmos. Environ.*, **16**, 1551-1563.

Charlson, R. J., D. S. Covert, and T. V. Larson, 1984: Observation of the effect of humidity on light scattering by aerosols. In *Hygroscopic Aerosols*, L. H. Ruhnke and A. Deepak (Eds.), A. Deepak Publishing, p35-44.

---, J. Langner, H. Rodhe, C. B. Leovy, and S. G. Warren, 1991: Perturbation of the Northern Hemisphere radiative balance by backscattering from anthropogenic sulfate aerosols. *Tellus*, **43AB**, 152-163.

---, S. E. Schwartz, J. M. Hales, R. D. Cess, J. A. Coakely Jr., J. E. Hansen, and D. J. Hofmann, 1992: Climate forcing by anthropogenic aerosols. *Science*, **255**, 423-430.

Chuang, C. C., J. E. Penner, and L. L Edwards, 1992: Nucleation scavenging of smoke particles and simulated drop size distributions over large biomass fires. *J. Atmos. Sci.*, **49**, 1264-1275.

Galloway, J. N., G. E. Likens, and M. E. Hawley, 1984: Acid deposition: Natural versus anthropogenic sources. *Science*, **226**, 829-831.

Hameed, S., and J. Dignon, 1988: Change in the geographical distributions of global emissions of NO_x and SO_x from fossil-fuel combustion between 1966 and 1980. *Atmos. Environ.*, **22**, 441-449.

Intergovernmental Panel on Climate Change (IPCC), 1990: *Climate change, The IPCC Scientific Assessment.* J. T. Houghton, G. J. Jenkins, and J. J. Ephraums (Eds.), 365 pp., Cambridge University Press.

Kiehl, J. T., and B. P. Briegleb, 1993: The relative roles of sulfate aerosols and greenhouse gases in climate forcing. *Science*, **260**, 311-314.

Milford, J. B., and C. I. Davidson, 1987: The sizes of particulate sulfate and nitrate in the atmosphere - A review. *JAPCA*, **37**, 125-134.

Penner, J. E., C. S. Atherton, J. Dignon, S. J. Ghan, J. J. Walton, and S. Hameed, 1991: Tropospheric nitrogen: A three-dimensional study of sources, distributions, and deposition. *J. Geophys Res.*, **96**, 959-990.

---, 1993: Atmospheric chemistry and air quality. In *Changes in Land Use and Land Cover: A Global Perspective*, W. Meyer and B. L. Turner (Eds.), Cambridge University Press, in press.

---, C. S. Atherton, and T. E. Graedel, 1993: Global emissions and models of photochemically active compounds. Global Atmospheric-Biospheric Chemistry: The First IGAC Scientific Conference, OHOLO Conference Series Books, Plenum Publishing, H. Y..

Quinn, P. K., T. S. Bates, J. E. Johnson, D. S. Covert, and R. J. Charlson, 1990: Interactions between the sulfur and reduced nitrogen cycles over the central pacific ocean. *J. Geophys. Res.*, **95**, 16405-16416.

Spiro, P. A., D. J. Jacob, and J. A. Logan, 1992: Global inventory of sulfur emissions with 1° × 1° resolution. *J. Geophys. Res.*, **97**, 6023-6036.

Taylor, K, and J. E. Penner, 1993: Anthropogenic aerosols and climate change. Submitted to *Nature*.

Twomey, S. A., 1977: *Atmospheric aerosols.* Elsevier, 302 pp.

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