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Perturbation of the northern hemisphere radiative balance by backscattering from anthropogenic sulfate aerosols*

By R. J. CHARLSON[1], J. LANGNER[2], H. RODHE[2], C. B. LEOVY[1] and S. G. WARREN[1],
[1]*Department of Atmospheric Sciences and Institute for Environmental Studies, University of Washington, AK-40, Seattle, WA 98195, USA,* [2]*Department of Meteorology*, Stockholm University, S-106 91 Stockholm, Sweden*

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ABSTRACT

Anthropogenic sulfate ($SO_4^=$) aerosol particles play two potential roles in the radiative climate of the earth. In cloud-free air, $SO_4^=$ particles scatter sunlight, some of which is lost to space, thereby reducing solar irradiance at the ground. The same particles can act as cloud condensation nuclei (CCN), the number concentration of which is an important determinant of cloud albedo. This albedo effect, in turn, also influences incoming short-wave solar radiation. Development of a three-dimensional global model for estimating the $SO_4^=$ aerosol mass concentration, along with previously-acquired information on the scattering and back-scattering coefficients per unit mass concentration allow calculation of the effects of anthropogenic $SO_4^=$ aerosol on clear-sky optical depth. Subsequently, this can be used to estimate the change in hemispheric and global average reflected solar radiation. The conclusion is that the change of reflected solar flux due to anthropogenic $SO_4^=$ averaged over the Northern Hemisphere is ca. $-1.1\ Wm^{-2}$, which is comparable but opposite in sign to the present-day radiative forcing by anthropogenic CO_2, $+1.5\ Wm^{-2}$. Because of the spatial variability of the anthropogenic $SO_4^=$ distribution, its meteorological effects must be studied regionally. That is, global models with regional resolution and regional data are required. Unlike the direct effect on solar irradiance, the relationship of CCN number concentration to mass concentration is not known. Thus it is not yet possible to make quantitatively reliable statements about anthropogenic forcing of cloud albedo, although there is qualitative evidence that the CCN effect may also be substantial.

1. Introduction

Considerable attention is being given to changes in the climate of the earth that are anticipated to occur as a consequence of increased concentrations of gases that absorb infrared radiation. While this attention certainly is justified, anthropogenic atmospheric aerosol particles also appear to be capable of changing the radiation budget through their direct backscattering affects as well as through their indirect effect on cloud albedo arising from their role as cloud condensation nuclei (CCN). These effects on the earth radiation budget may today be comparable in magnitude but opposite in sign to those caused by greenhouse forcing (Wigley, 1989; Charlson et al., 1990). Because the global aerosol burden in general and the burden in certain regions of the Northern Hemisphere in particular are calculated to have increased from pre-industrial levels, largely due to the combustion of fossil fuel, it is necessary for prediction of anthropogenic climate change to understand the geographical distribution, time trend and meteorological effects of anthropogenic aerosols.

The possible role of aerosol particles in climate and atmospheric optics is not a newly recognized topic; studies of their influence on incoming solar radiation, usually termed "turbidity", date back to the early 1900's (Ångström, 1929, 1930; Bergeron, 1928). Unfortunately, the numerous national and international monitoring programs that were established in the 1960's and 70's to measure

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atmospheric turbidity have failed to produce a uniformly useful data base, due to both unsatisfactory instruments and a lack of continuity of observation. Satellite-borne sensors may ultimately be capable of providing global maps of both the direct and indirect (CCN-induced) effects; however, analyses to date are limited to the detection of major events such as duststorms and forest fires (Rao et al., 1988) and to measurement of changes in the albedo of marine stratus clouds by effluents from ship stacks (Coakley et al., 1987; Radke et al., 1990). In addition, with the exception of several years of CCN data at Cape Grim, Tasmania (Gras, 1989), there are no organized data on spatial distributions and trends that may be linked to the possible effects of anthropogenic aerosol on clouds. Thus, assessments of hemispheric or global aerosol effects today can only be based on model calculations.

Regional studies of the direct effect of aerosol particles, which scatter solar radiation back into space, have been made; e.g. Ball and Robinson (1982) deduced from measurements of solar irradiance that aerosol in the eastern US caused about a 7% annual average reduction of solar energy reaching the ground over a region about 1000 km in diameter. This is roughly consistent with the regional estimates of 5 to 10% by Bolin and Charlson (1976) in their consideration of the anthropogenic perturbation of the sulfur cycle.

Various model estimates of the global effects of aerosol backscatter, based on very fragmentary data and many assumptions yield differing results but do suggest that the direct effects are of possible importance. Grassl (1988), using a 2-D global model, concluded that anthropogenic aerosol is significant, especially via the indirect effect on clouds. Peak zonal-average cooling rates of 7 Wm^{-2} were estimated for the midlatitudes of the Northern Hemisphere. However, these estimates required assumptions of aerosol size distribution and refractive index and no longitudinal resolution was possible. Shaw (1983) considered the possible role of natural $SO_4^=$ from dimethylsulfide, concluding that its direct effect is currently of marginal climatic importance. Coakley et al. (1983), in a global model calculation, estimated that the current "background aerosol" cools the earth by 2–3 K.

The purpose of this paper is to extend the original appraisal of Bolin and Charlson (1976) by providing an updated and refined estimate of the direct climatic effect of anthropogenic $SO_4^=$ aerosol based on a global three-dimensional numerical calculation of the sulfate aerosol burden and an empirical correlation of optical effect to $SO_4^=$ aerosol mass concentration. This estimate will be compared to a simple burden calculation and calculated forcings due to possible CCN influence on cloud albedo. We will demonstrate the likelihood that aerosol cooling effects are significant in comparison to current climate forcing by anthropogenic CO_2 and other greenhouse gases. Because it is not clear how to relate the *mass* concentration of $SO_4^=$ to CCN *number* concentration, the following calculations will include only the direct effect of aerosol backscatter of solar radiation. However, climate forcing due to CCN changes may also be important. While it seems evident that both chemical and meteorological factors are important in determining the CCN number concentration (Baker and Charlson, 1990), no quantitative relationship is yet agreed upon. It appears likely that the CCN concentration may be a non-linear function of the $SO_4^=$ aerosol particle mass concentration.

2. Optical properties of the aerosol layer

We compute the increment in reflected flux, due to the natural and anthropogenic $SO_4^=$ aerosol layer, ΔF_R, and the corresponding increment to the planetary reflectivity, ΔR_p. The aerosol is assumed to be mainly in the boundary layer. Since it is unable to raise the planetary albedo if it is situated above or below a highly reflective cloud, we consider only the effect on the clear sky energy budget.

The optical thickness δ of the aerosol layer is the sum of absorption and scattering optical thicknesses

$$\delta = \delta_a + \delta_s,$$

but sulfuric acid, its ammonia salts, and water are essentially non-absorptive in the solar spectrum, so $\delta \approx \delta_s$. We show below that the aerosol layer is optically thin ($\delta \ll 1$), so we can neglect multiple scattering within the aerosol layer. The radiative transfer equation then just reduces to Beer's Law. For the aerosol layer alone, the ratio of the scattered flux, F_s, to the incident flux, F_i, for radiation incident at solar zenith angle θ_0 is

$$\frac{F_s}{F_i} = 1 - \exp(-\delta \sec \theta_0) \approx \delta \sec \theta_0, \tag{1}$$

where $\delta \sec \theta_0$ is the slant-path optical thickness. A fraction β of F_s is scattered upward so that the reflectance of the aerosol layer alone is

$$R_a = \beta \frac{F_s}{F_i} = \beta \delta \sec \theta_0. \tag{2}$$

In order to calculate the incremental contribution of the aerosol layer to the planetary albedo ΔR_p, we must correct R_a for three effects: the reflectivity of the underlying surface R_s, the transmissivity of the overlying atmosphere T_i, and the fraction of the area with clear sky conditions $(1 - A_c)$, where A_c is the fractional coverage of clouds.

If the underlying surface has albedo R_s, the reflectance R_{as} of the combined aerosol-surface system can be obtained from a simple multiple reflection model as

$$\begin{aligned} R_{as} &= R_a + T_a^2 R_s [1 + R_a R_s + (R_a R_s)^2 + \cdots] \\ &= R_a + \frac{T_a^2 R_s}{1 - R_a R_s}, \end{aligned} \tag{3}$$

where $T_a = 1 - R_a$ is the transmittance of the (non-absorbing) aerosol layer. The change in system reflectance due to the aerosol layer is

$$\begin{aligned} \Delta R_{as} &= R_{as} - R_s \\ &\approx R_a + (1 - 2R_a) R_s (1 + R_a R_s) - R_s \\ &\approx R_a (1 - R_s)^2, \end{aligned} \tag{4}$$

since $R_a \ll 1$.

This is the change in albedo at the top of the aerosol layer. The change in planetary albedo R_p is less than ΔR_{as} because the overlying atmosphere has non-unit transmittance T_i which diminishes the reflected flux on both the incident and reflected paths. Combining this correction with the correction due to the clear sky fraction, we obtain

$$\begin{aligned} \Delta R_p &\approx T_i^2 \, \Delta R_{as} (1 - A_c) \\ &= T_i^2 (1 - A_c)(1 - R_s)^2 \beta \delta \sec \theta_0. \end{aligned} \tag{5}$$

The corresponding change in reflected solar flux ΔF_R is obtained by multiplying ΔR_p by the incident solar flux $\mu_0 Q_0$, where Q_0 is the solar constant and $\mu_0 = \cos \theta_0$,

$$\begin{aligned} \Delta F_R &= \mu_0 Q_0 \, \Delta R_p \\ &= Q_0 T_i^2 (1 - A_c)(1 - R_s)^2 \beta \delta. \end{aligned} \tag{6}$$

The upward scattered fraction $\beta(\mu_0)$ for a particular solar zenith angle θ_0 is one half of the average over the upward hemisphere of the aerosol phase function $p(\mu, \phi, \mu_0)$ where ϕ is the azimuth angle relative to the solar azimuth angle and μ is the cosine of the zenith angle of the emergent scattered radiance, i.e.,

$$\beta(\mu_0) = \frac{1}{4\pi} \int_0^1 \int_0^{2\pi} p(\mu, \phi, \mu_0) \, d\phi \, d\mu.$$

We can now evaluate approximately the change in reflected solar flux (neglecting correlations between T_i, A_i, R_s, β, and δ),

$$\overline{\Delta F_R} \approx \tfrac{1}{2} Q_0 \overline{T_i^2} (1 - \overline{A_c})(1 - \overline{R_s})^2 \bar{\beta} \delta, \tag{7}$$

where $\bar{\beta}$ is the average over μ_0 of $\beta(\mu_0)$,

$$\bar{\beta} = \int_0^1 \beta(\mu_0) \, d\mu_0,$$

and the factor $\frac{1}{2}$ enters because any point in the globe is illuminated by sunlight only one half of the time over the course of a year. The factor $\bar{\beta}$ has been evaluated by Wiscombe and Grams (1976), and related to the backscattered fraction, b. Because $\bar{\beta}$ includes upward scattered radiation from the forward hemisphere of the aerosol phase function, $\bar{\beta} > b$ in general. For a wide range of aerosol properties satisfying the condition (justified below) that $b \approx 0.15$, Wiscombe and Grams find that $\bar{\beta} \approx 0.29$.

Now using $Q_0 = 1370$ Wm^{-2}, $A_c \approx 0.61$ for the globally averaged cloud cover (Warren et al., 1986; 1988), $\overline{T_i} \approx 0.76$ (based on unpublished calculations of S. G. Warren and W. J. Wiscombe for subarctic summer and a zenith angle of 60°), and $\overline{R_s} \approx 0.15$ (e.g., Robock, 1980), we find

$$\overline{\Delta F_R} \approx 32 \delta. \tag{8}$$

We next estimate the aerosol optical thickness δ as the product of the aerosol burden $B_{SO_4^=}$ (g m^{-2}) and the mass scattering coefficient α(m^2 g^{-1}). We use an empirical value of α based on measurements for sulfate haze in Sweden (Waggoner et al., 1976). The mean value measured for α was 5 $m^2 g^{-1}$, but the air had been heated to reduce the relative humidity (RH) to below 50%. We think 8.5 $m^2 g^{-1}$ is more appropriate for ambient air, because Charlson et al. (1984) showed that α increases by a factor of 1.7 when the RH rises from 50% to the mean RH for the near-surface atmosphere in the Northern Hemisphere, which is 75–80% (London, 1957, Fig. 1). The average RH in cloud-free air may be somewhat lower. This estimate, $\alpha = 8.5\ m^2 g^{-1}$, corresponds to the total scattering by the mixture of $SO_4^=$, water and all other aerosol ingredients that were present in the experiments of Waggoner et al.; however, typically 50–80% of aerosol scattering is by $SO_4^=$ and its chemically associated H_2O. (Acidic sulfate particles are particularly hygroscopic because of the affinity of H_2SO_4 for water.) While there is no global survey of the value of α, this estimate is consistent with extensive measurements in the U.S. (White, 1986) and in remote marine air as well (Covert, personal communication, 1990).

The hemispheric backscattered fraction, b, was determined for sulfate haze in rural air in eastern North America (near St. Louis, Missouri) by Vanderpol (1975) as the ratio of scattering coefficients (σ, units m^{-1}) measured with an angular integrating nephelometer with two angular ranges: $b = \sigma_{bsp}/\sigma_{sp}$, where σ_{bsp} was determined from an integral over scattering angle ϕ of $90° \leqslant \phi \leqslant 170°$; σ_{sp} from an integral over $7° \leqslant \phi \leqslant 170°$. The measured ratio ranged from 0.09 to 0.25 with a mean of 0.15. The relatively small variation in the measured values of α and b indicate that the size distribution of the aerosol also was relatively constant. The limited set of measurements (Vanderpol, 1975) is complemented by Mie calculations for typical lognormal size distribution (geometric mean radius by volume ~ 0.1 to 0.15 μm, $\sigma_g \sim 2$, refractive index $\sim 1.4 - 0i$ to $1.5 - 0i$) in which $0.1 \leqslant b \leqslant 0.15$ (Herbert, 1974). We use this as the basis for assuming that all anthropogenic $SO_4^=$ aerosol has the same value of α and b.

3. Reference box model

To provide a reference against which the global model calculations can be checked and in order to see if the magnitude of δ due to anthropogenic $SO_4^=$ might be of any global or hemispheric significance, the average column burden of anthropogenic $SO_4^=$ can be estimated from the source strength of sulfur in anthropogenic SO_2, F_{SO_2-S}, and the lifetime of $SO_4^=$ aerosol in the troposphere, $\tau_{SO_4^=}$, which is about 6 days ($\sim 5 \times 10^5$ s, Slinn, 1983). The mass flux of $SO_4^=$, $F_{SO_4^=}$ is three times the corresponding mass flux of the element sulfur in SO_2 multiplied by the fraction of SO_2 sulfur converted to $SO_4^=$. As described in Section 4, we estimate that $F_{SO_2-S} \sim 71$ Tg-yr^{-1} (see Table 1, and the sulfur budget tabulations of Möller, 1984; Hameed and Dignon, 1988), and the fraction converted to $SO_4^=$ is estimated to be approximately one half (Rodhe and Isaksen, 1980). Hence, we estimate $F_{SO_4^=} \approx 3F_{SO_2-S}/2 \approx 1.1 \times 10^{14}$ g-yr$^{-1} \approx 3.4 \times 10^6$ gs^{-1}, and the column burden of anthropogenic $SO_4^=$ (averaged over the globe) should be:

$$\overline{B_{SO_4^=}} \cong \frac{F_{SO_4^=}\tau_{SO_4^=}}{A_{earth}}$$

$$\cong \frac{(3.4 \cdot 10^6)(5 \cdot 10^5)}{(5.1 \cdot 10^{14})} \cong 3.3 \cdot 10^{-3}\ gm^{-2}. \qquad (9)$$

Table 1. *Global sulfur emissions included in the model calculations: units: Tg S yr^{-1}*

Emission	Tg S yr^{-1}	Ref.
Anthropogenic (SO_x)*	71	Möller (1984), Hameed and Dignon (1988), Semb (1985)
Biomass burning (SO_2)	2.5	Hao et al. (1991), Andreae et al. (1988)
Volcanoes (SO_x)*	11	Berresheim and Jaeschke (1983)
Oceans (DMS)	16	Bates et al. (1987), Leck and Rodhe (1991),
Soils and plants (DMS, H_2S)	4	Goldan et al. (1987), Lamb et al. (1987), Andreae and Andreae (1988)

* $SO_x = SO_2 + SO_4^=$

This implies that for cloudless skies, $\delta \approx (3.3 \times 10^{-3}) \cdot 8.5 \approx 2.8 \times 10^{-2}$. This is 3 to 6 times the value estimated by Bolin and Charlson (1976) because they underestimated $B_{SO_4^=}$, largely because they underestimated the area under anthropogenic influence (they actually reported $\delta_{bsp} \equiv b\delta$; we have converted it). From (8), $\overline{\Delta F_R} \approx 0.9\ \mathrm{Wm^{-2}}$ (i.e., the planetary albedo is increased by 0.0026) due to the aerosol. This corresponds to a change in global average surface temperature ΔT_s of -0.3 to -0.9 K for climate sensitivity of 0.3–1.0 K/(Wm^{-2}). For the climate sensitivity 0.77 K/(Wm^{-2}) used by Charlson et al. (1987), $\Delta T_s = -0.7$ K, an order of magnitude larger than the upper limit of -0.06 K estimated by Bolin and Charlson (1976) from a simple Stefan-Boltzmann calculation.

Because the anthropogenic $SO_4^=$ must be confined largely to the Northern Hemisphere (NH), the denominator in (9) should be halved, and B and δ doubled for the NH: $\overline{\Delta F_R} \approx 1.8\ \mathrm{Wm^{-2}}$; $\Delta T \approx -1.4$ K. These quantities are close to those in Charlson et al. (1990) which was based on a simpler calculation of loss of solar irradiance at the ground. These are of sufficiently large magnitude to be very important compared to the calculated effects of anthropogenic increases of greenhouse gases or even $2 \times CO_2$ scenarios. However, we know (e.g., from studies of the $SO_4^=$ content of precipitation) that the $SO_4^=$ aerosol of the NH is not at all homogeneous. Thus it is important to carry out a more complete model calculation in order to refine the estimate and reveal the possible distribution of anthropogenic effects.

4. Three-dimensional meteorological/chemical model

The basis for the calculations of the radiative effect of the tropospheric sulfate aerosol described below is a three-dimensional model simulation of the tropospheric part of the atmospheric sulfur cycle. The model is used to calculate the annual mean concentration of $SO_4^=$ aerosol as a function of altitude, latitude and longitude as well as the integral of $SO_4^=$ concentration over altitude which is the burden, $B_{SO_4^=}$, as in (9) above. The three-dimensional transport-chemistry model used in the simulations has a horizontal resolution of 10° longitude by 10° latitude and a vertical resolution of 10 layers between the surface and 100 hPa. Advection takes place by climatological monthly mean winds. Transport processes occurring on smaller space and time scales are parameterized as eddy diffusion. Vertical transport in deep convective clouds is included using the scheme developed by Feichter and Crutzen (1990). Further details regarding the transport part of the model is given by Zimmermann (1984, 1987).

The formulation of the sulfur scheme is based on the work by Rodhe and Isaksen (1980) and has been extended for the application in the three-dimensional model. Three species are carried prognostically in the model, SO_2, $SO_4^=$ (sulfate) and DMS (dimethyl sulfide). The emissions included are given in Table 1. The anthropogenic input of sulfur from fossil fuel combustion and various industrial processes is obtained from the emission inventory by Semb (1985) covering the major source regions in the Northern Hemisphere together with the emission pattern for fossil fuel CO_2 emissions presented by Marland et al. (1985) and Rotty (1987). The emissions of sulfur outside the area covered by Semb is calculated by assuming that the ratio of SO_2 to CO_2 emissions in this area is the same as in the area covered by Semb. The emissions from biomass burning have been calculated by applying a constant emission factor of 0.32×10^{-3} (mol SO_2/mol C) (Andreae et al., 1988) to the estimates of carbon emissions from fires in the tropics presented by Hao et al. (1991). The emission of sulfur from volcanoes (Berresheim and Jaeschke, 1983) is distributed in proportion to the number of active volcanoes per grid square in the model based on the compilation by Simkin et al. (1981). The oceanic (DMS) emission field includes seasonal variations as well as variations in latitude and longitude based on the studies by Bates et al. (1987), and Leck and Rodhe (1991). Estimates of the biogenic sulfur emissions from land have been arrived at using the information on emissions from soils and plants presented by Goldan et al. (1987), Lamb et al. (1987), and Andreae and Andreae (1988). The magnitude of the natural emissions is still uncertain to a least ±50% (Andreae, 1990). However, our main conclusions pertain to the impact of anthropogenic emissions, and recent estimates of the global anthropogenic input of sulfur for the year 1980 (Möller, 1984 (80 Tg yr^{-1}); Hameed and Dignon, 1988 (~62 Tg yr^{-1})) are within ±15% of the value used in this study.

Gas phase oxidation of DMS and SO_2 is assumed to proceed by reactions with OH radicals (Atkinson et al., 1984; Calvert et al., 1978). The aqueous phase oxidation of SO_2 to sulfate in cloud droplets is parameterized as a first order decay process. The efficiency of this process is based on an estimate of the probability for a molecule of the trace species to be exposed to cloud water in the atmosphere, together with an assumed lifetime of 10 hours for SO_2 before oxidation in cloud droplets. The mean rate of liquid phase transformation in the layer from 1–5 km is about 0.23 day^{-1}. Removal by precipitation is calculated from information on climatological rainfall rates and scavenging efficiencies following the approach by Junge (1963). The maximum rate of removal by precipitation is 1.3 day^{-1} for $SO_4^=$ and 0.4 day^{-1} for SO_2 and occurs in the lowest 2 km in the mid-latitudes and in the lowest 5 km in the tropics. Dry deposition is included using surface deposition velocities of 0.2 cm s^{-1} for $SO_4^=$ and 0.8 and 0.6 cm s^{-1} for SO_2 over ocean and land respectively. Further details regarding the sulfur simulations have been reported by Langner and Rodhe (1991).

5. Calculations

5.1. Column burden of $SO_4^=$

The annual mean column burden of $SO_4^=$, $B_{SO_4^=}$, in the troposphere (1000–100 hPa) is calculated for two different model runs; including and excluding natural emission of sulfur. Fig. 1a shows the horizontal distribution of the vertically integrated, annual mean amount of $SO_4^=$ in mg $SO_4^=$ m^{-2} for the case including both natural and anthropogenic emissions, and Fig. 1b shows the difference between this case and natural emissions only.

5.2. Total optical depth (δ) of the tropospheric sulfate

The total optical depth is given by

$$\delta = \int_0^{z_{\mathrm{top}}} \sigma_{\mathrm{sp}}\, \mathrm{d}z \qquad (10)$$

where

$$\sigma_{\mathrm{sp}} = \alpha m_{SO_4^=}, \qquad (11)$$

and $m_{SO_4^=}$ is the mass concentration of $SO_4^=$, gm^{-3}. For the calculations in Figs. 1 and 2, α is taken to have a value of 8.5 m^{2}g^{-1} for sulfate at ambient humidity and is assumed to be constant. Figs. 1a and b can also be read as the approximate horizontal distribution of the annual mean value of 100δ for the two situations considered because $100\delta = B_{SO_4^=} = (\mathrm{mg\ m^{-2}})$ if $\alpha \approx 10$ m^{2}g^{-1}, which is close to 8.5 m^{2}g^{-1}.

5.3. Radiation loss

To obtain a first order estimate of the change in annual average reflected solar radiance $\Delta F_R(\phi, \lambda)$ as a function of latitude ϕ and longitude λ, we use the observed space-dependent annual mean value of $A_c(\phi, \lambda)$ and calculated $\delta(\phi, \lambda)$, but retain the global mean estimates for $\overline{T_i^2}$, $\overline{R_s}$, and β, to give

$$\Delta F_R(\phi, \lambda) \approx \tfrac{1}{2} Q_0 [1 - A_c(\phi, \lambda)] \times \overline{T_i^2} [1 - \overline{R_s}]^2 \beta \delta(\phi, \lambda), \qquad (12)$$

where the factor $\frac{1}{2}$ again arises from the fact that any point on the globe is illuminated by the sun only one half the time.

5.4. Hemispheric and global averages

For comparisons with other radiative forcings of the earth-atmosphere system hemispheric and global averages have been calculated. Global and hemispheric values of ΔF_R and δ from the model calculations are given in Table 2.

6. Discussion

Fig. 1 shows the calculated global distribution of sulfate aerosol with all emissions in Table 1 included (1a) and the calculated difference in sulfate loading between the case with all emissions included and the case with anthropogenic emissions excluded (1b). The expected bulges of $SO_4^=$ over North America, Europe and coastal Asia are clearly evident. Figs. 1a and b also show the corresponding calculated aerosol optical depth. This can be compared directly to data, e.g. as reviewed by Bolin and Charlson (1976) and Husar

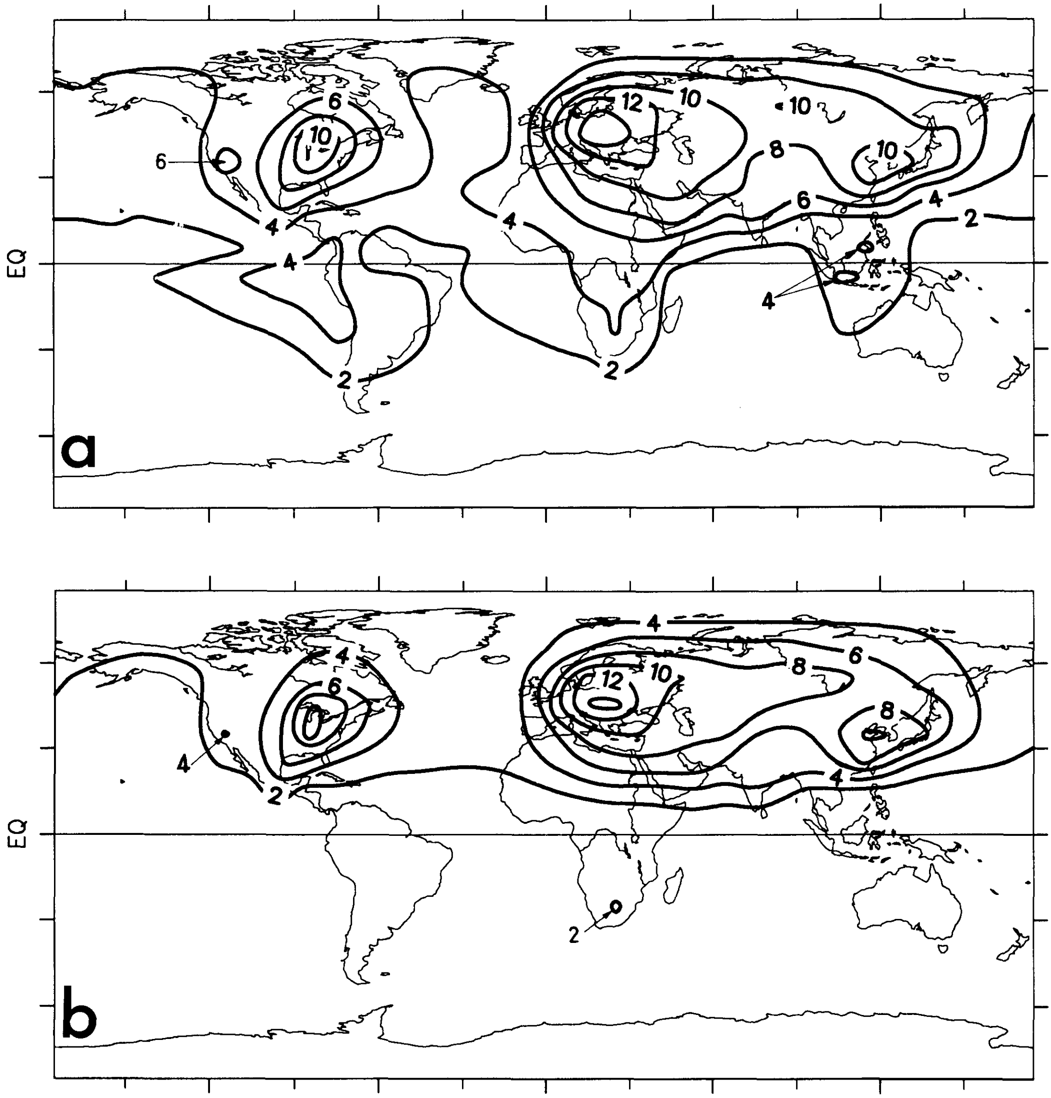

Fig. 1. (a) Calculated distribution of the column burden of SO$_4^=$ aerosol, $B_{SO_4^=}$, including natural and anthropogenic emission. Also, $100\delta \cong B_{SO_4^=}$ (mg m^{-2}). (b) Calculated $B_{SO_4^=}$ including only anthropogenic emissions, mg m$^{-2} \approx 100\delta$. All emission data are in Table 1.

et al. (1981). The peak optical depth reported in North America is ca. 0.3 which agrees remarkably well with the model which gives 0.12. We note that the coarse resolution of our model implies a flattening of all peaks. The geographical distribution also agrees with maps of turbidity (Husar et al., 1981).

Fig. 2 shows the increase in reflected irradiance at the top of the atmosphere from Eq. (12) with actual cloud fraction into account. Fig. 2a shows ΔF_R including all emissions and Fig. 2b including only anthropogenic emissions. The maxima of loss of solar radiation in Fig. 2 indicate anthropogenic influence ranging from close to zero to over 4 Wm^{-2} (Fig. 2b). Again this compares well with the observed loss of solar irradiance at the surface

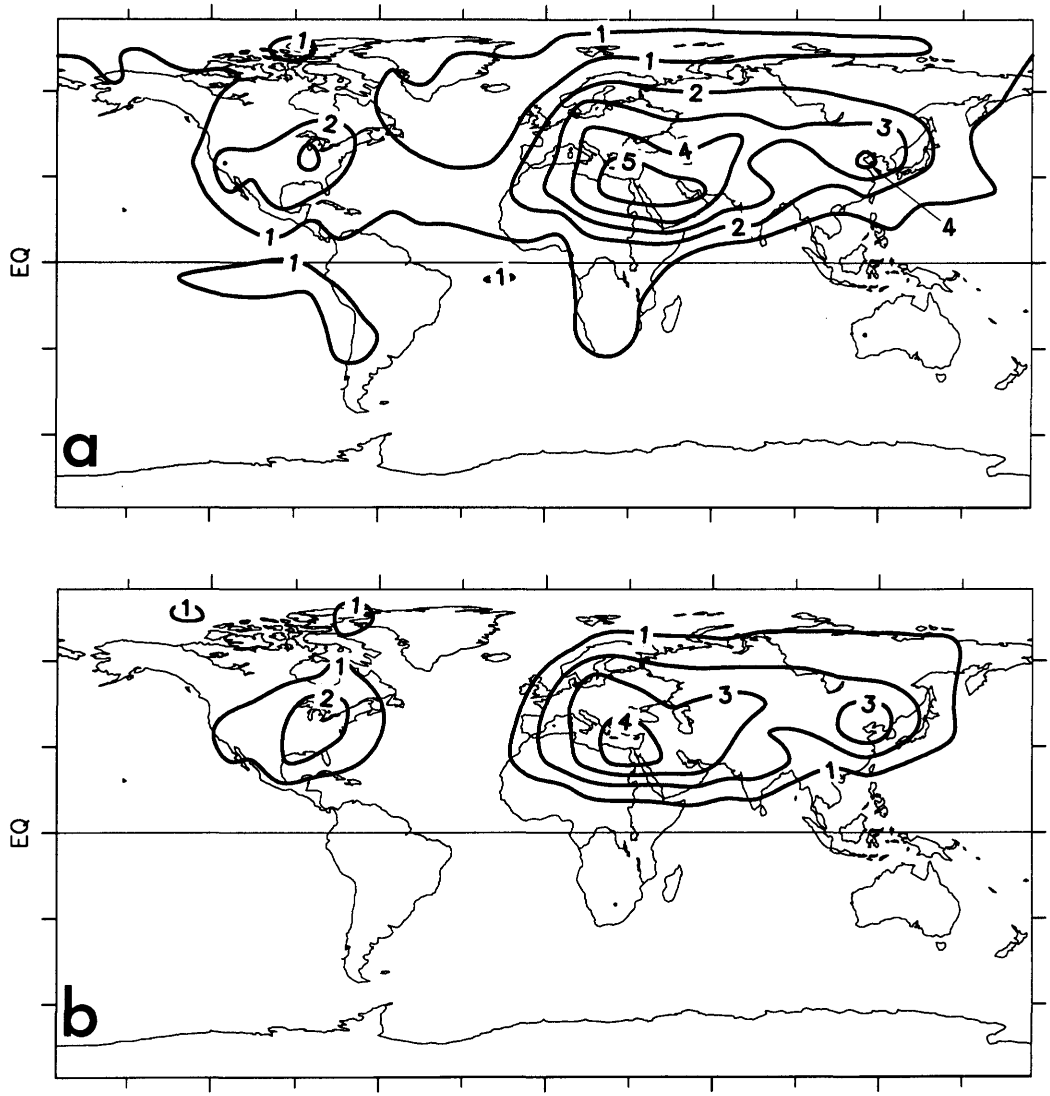

Fig. 2. (a) Calculated increase of reflected flux to space, ΔF_R, due to tropospheric $SO_4^=$ aerosol, including anthropogenic and natural sources. Units are Wm^{-2}. (b) Calculated ΔF_R including only anthropogenic sources (Wm^{-2}). Both 2a and b are based on estimated cloud fraction, A_c, per Warren et al. (1986, 1988).

due to light extinction around 7.5% (Eastern U.S., Ball and Robinson, 1982), which was composed of backscatter and absorption of approximately equal magnitude. The geographical distribution of loss of irradiance is shifted from that for δ (compare Fig. 2a with Fig. 1a), due to the geographical dependence of A_c.

We can also compare these results for the anthropogenic $SO_4^=$ part of the total non-cloud aerosol radiative forcing with other global estimates. First, we are unaware of any previous models of the radiative effect of aerosol based on a global chemical model. Coakley et al. (1983) summarized the then existing estimates of the effect of tropospheric aerosol on the earth's radiation budget, all of which were based on hypothetical

Table 2. *Area averaged optical depth,* δ, *and reflected solar radiation due to tropospheric* $SO_4^=$, $\Delta F_r (W\,m^{-2})$

	Anthropogenic emissions included		Only natural emissions		Difference between case with and without anthropogenic emissions	
Area	δ	ΔF_r	δ	ΔF_r	δ	ΔF_r
N. Hemisphere	$4.43 \cdot 10^{-2}$	1.57	$1.39 \cdot 10^{-2}$	0.50	$3.04 \cdot 10^{-2}$	1.07
S. Hemisphere	$1.48 \cdot 10^{-2}$	0.46	$1.13 \cdot 10^{-2}$	0.35	$0.35 \cdot 10^{-2}$	0.11
Globe	$2.96 \cdot 10^{-2}$	1.02	$1.26 \cdot 10^{-2}$	0.42	$1.70 \cdot 10^{-2}$	0.60

aerosol models. Coakley et al. employ aerosol optical parameterization only for the Northern Hemisphere; i.e., their globe had two Northern Hemispheres. They concluded that the "background tropospheric aerosol" cools the earth surface by 2–3 K. From our analysis above, we think that a significant fraction—half or more—of their calculated effect is probably anthropogenic rather than natural as they claim (p. 132). In fact, their optical depths (ranging from $0.07 \leqslant \delta \leqslant 0.16$; average ≈ 0.14 were substantially larger than those currently observed in clean, Southern Hemisphere marine air (Forgan, 1987) and in air over the eastern North Pacific Ocean ($\delta \sim 0.05$). Hence, we conclude that anthropogenic aerosol backscatter in cloud-free air may contribute ca. 1–2 K of cooling to the Northern Hemisphere, roughly comparable to the ca. 1 Wm^{-2} from Table 2 and a climate sensitivity of 1 K/Wm^{-2}.

We cannot be as quantitative in comparing the direct effect with the possible indirect CCN effect of anthropogenic $SO_4^=$. A change of 30% of CCN over the oceans, influencing only marine low clouds (stratus) is calculated to change the global surface temperature by -1 K (Charlson et al., 1987). A mere 20% increase of CCN in the NH is calculated to yield a cooling of ca. 1 Wm^{-2} (Wigley, 1989). While there certainly is potential for the anthropogenic $SO_4^=$ to have increased the CCN number concentration, there are as yet no data and there is no agreed-upon theory relating number concentration of CCN to mass concentration of $SO_4^=$.

It is clearly important to refine calculations for both clear sky and calculations of direct aerosol effect in order to reduce the factor of two uncertainty that is implicit in our above calculations of ΔF_R, with an ultimate goal of arriving at uncertainties in the anthropogenic perturbation of radiative forcing at the surface comparable to those for greenhouse gases—perhaps 20%. This leads to a list of questions and research topics that are suggested by these results.

7. Large remaining questions and problems

The first main category concerns the refinement of the model. Better knowledge of α and b, their variability and dependence on controlling factors, would increase confidence in the calculated effects. Subsequently, solar zenith angle should be included for each geographical grid point with time of day and seasonal factors included. Seasonal variation of $B_{SO_4^=}$ should be added, although it is likely to vary by only a factor of two or less (Husar et al., 1981). Addition of the underlying surface albedo and clear sky transmission for each grid point would allow a calculation of local heat balance. Finally, the calculated radiative effect should be compared to measurements, both ground and satellite based. Improvements in the uncertainty of natural and anthropogenic emission fluxes of gaseous sulfur compounds are also needed as are improvements in modelling the transformation and removal processes.

The numbers that we have estimated for the climatic forcing due to sulfate aerosol scattering refer to the change that has occurred due to anthropogenic sulfur emissions up to the present time. Any future changes in this forcing will depend on how these emissions will vary. Because of the short atmospheric lifetime of sulfate and its precursors, the atmospheric burden will adjust within a few weeks to changes in emissions. This is a very different situation than for most greenhouse gases which have effective lifetimes of decades to centuries. For example, the concentration of CO_2 will continue to rise for more than a century even if emissions are kept constant at today's level. This difference is illustrated schematically in Fig. 3 which shows how the climatic forcings due to CO_2 and aerosol sulfate would change if the global fossil fuel consumption leveled off and eventually was reduced. Because of the rapid growth in emissions during the past decades both the greenhouse forcing due to CO_2 and the opposite forcing due to aerosol sulfate have grown accordingly. During a leveling off phase the greenhouse forcing will continue to grow whereas the aerosol forcing will remain constant. During a decay phase, the greenhouse forcing due to CO_2 will start to level off and the aerosol cooling decline. This simple example illustrates that the relative importance of these two major counteracting anthropogenic forcing agents in the future will deend critically on changes in the fossil fuel use (large scale desulfurization measures would of course also influence the sulfur emissions). Thus, the current tendency towards a balance in the Northern Hemisphere is likely to change.

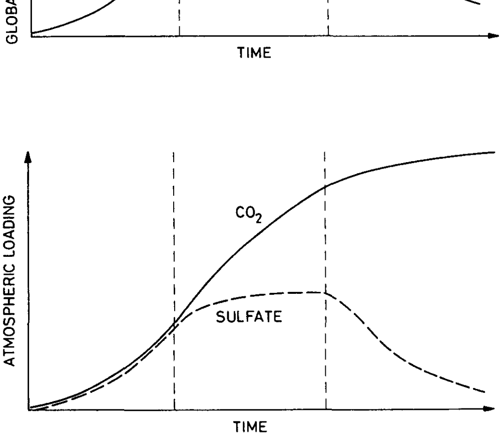

Fig. 3. Schematic illustration of the difference between response times of climate forcing due to CO_2 (heating) and sulfate (cooling) during different patterns of global fossil fuel consumption.

Another important factor to consider is the geographical distribution of future sulfur emissions. The largest aerosol effects on the radiation balance are likely to occur for emissions in tropical and subtropical areas. This is because of:

(i) larger insolation;

(ii) lower cloudiness (especially in the subtropics);

(iii) larger convective mixing of boundary layer air into the free troposphere leads to a longer lifetime and more widespread distribution of aerosol sulfate.

The regional scale of the climate forcing due to aerosol sulfate makes it difficult to compare this forcing with that caused by the globally distributed greenhouse gases. It is far from clear that global, or even hemispheric, averages of the aerosol forcing provide a proper basis for comparison. We recommend that general circulation models be used to simulate the climate response to current and possible future regional scale changes in forcing caused by sulfate aerosols.

8. Conclusion

Backscatter of solar shortwave radiation by anthropogenic, non-cloud $SO_4^=$ aerosol particles is calculated to cause a loss of solar irradiance of ca. $-1\ \mathrm{Wm}^{-2}$ averaged over the Northern Hemisphere. This is comparable to but opposite in sign to the current greenhouse forcing by increased CO_2 to date (the total greenhouse forcing to date, including all greenhouse gases, is about 1.6 times the CO_2 forcing). Uncertainty exists in this calculation; however we estimate that these results are within a factor of about two of reality because the bases of the meteorological model, the chemical model and the optical calculation are all empirical. That is, almost all the quantities used in these calculations are data from measurements, most of which have been or can be verified as being realistic. These results agree with more complex radiative transfer models (e.g., Coakley et al., 1983) and indicate the likelihood that what has been considered to be the "natural background tropospheric aerosol" is in fact significantly influenced by anthropogenic $SO_4^=$ over large regions of the NH. There are few outstanding questions regarding the fundamental basis for understanding the direct, non-cloud effect of $SO_4^=$ aerosol on solar radiation. However, the indirect effect of anthropogenic $SO_4^=$ aerosol particles acting as CCN remains largely an open question. Other open questions remain regarding the nature and magnitude of meteorological effects due to the NH/SH asymmetry of the greenhouse and aerosol radiative forcings, as well as the latitudinal and longitudinal asymmetry within the NH.

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