

## Model calculations of non-cloud radiative forcing due to anthropogenic sulphate aerosol

M. RAJEEVAN

*Meteorological Office, Pune-411 005, India*

*(Received 25 August 1995, Modified 27 January 1997)*

सार — मानवोद्भवी सल्फेट ऐरोसॉल कण आने वाले सौर विकिरण को फैलाते हैं, जिससे विकिरण प्रक्रिया प्रभावित होने से उसका प्रभाव जलवायु पर पड़ता है। मानवोद्भवी सल्फेट ऐरोसॉल से प्रभावित विकिरण बलन के वार्षिक चक्र का आकलन करने के लिए शोधकर्ता ने दो स्वतंत्र रसायन अभिगमन मॉडलों द्वारा अनुकूल सल्फेट सांद्रता क्षेत्रों के साथ एक त्रिविमीय विकिरण अंतरण मॉडल का प्रयोग किया है। आकलित प्रबलन पैटर्न से पूर्वी संयुक्त राज्य दक्षिणी-पूर्वी यूरोप और पूर्वी चीन के ऊपर इसके अधिकतम विस्तार का पता चलता है। लैंगतर और रोडे (1991) आंकड़ों के अनुसार आकलित वार्षिक भूमंडलीय-औसत विकिरण प्रबलन 0.50 डब्ल्यू. एम.<sup>-2</sup> है और पेन्नर इत्यादि (1994) के आंकड़ों के अनुसार यह मान -0.49 डब्ल्यू. एम.<sup>-2</sup> है। मौसम के अनुसार प्रबलन में भिन्नता पाई गई और शीत ऋतु की तुलना में उत्तरी गोलार्द्ध में ग्रीष्म ऋतु के दौरान अधिक प्रबलन पाया गया। उत्तरी गोलार्द्ध के मध्य-अक्षांशों पर क्षोभमंडलीय निम्न ऊष्मन दर को सल्फेट ऐरोसॉल भी पर्याप्त मात्रा में प्रभावित करता है। भूमंडलीय मेघाच्छन और ऐरोसॉल के ऑप्टिकल गुणों के प्रति भी प्रबलन को संवेदनशील पाया गया है। पिछले आकलनों के परिणाममें भिन्नता के संभावित स्रोतों की इसमें चर्चा की गई है। उत्तरी गोलार्द्ध के मध्य अक्षांशों के ऊपर, ऐरोसॉल के प्रत्यक्ष प्रभाव से उत्पन्न हुए नकारात्मक विकिरण प्रबलन को हरितगृह गैसों में वृद्धि के परिणामस्वरूप सकारात्मक प्रबलन सरलतापूर्वक संतुलित कर देता है।

सल्फेट ऐरोसॉल और हरितगृह गैसों में वृद्धि के कारण तापमान संतुलन प्रोफाइलों के परीक्षण के लिए एक 26 स्तरीय विकिरण संवहनी मॉडल (आर. सी. एम.) का भी प्रयोग किया गया। यह पाया गया कि सल्फेट ऐरोसॉल के प्रभाव से सतह-क्षोभमंडलीय प्रणाली का शीतलन होता है। सल्फेट ऐरोसॉल क्षोभमंडलीय ऊष्मन को कम करता है और हरितगृह गैसों में वृद्धि के बावजूद समतापमंडलीय शीतलन में वृद्धि करता है।

**ABSTRACT.** Anthropogenic sulphate aerosol particles scatter incoming solar radiation thereby perturbing the radiative budget, hence climate. We have used a three dimensional radiative transfer model together with the sulphate concentration fields simulated by two independent chemistry-transport models to calculate the annual cycle of the radiative forcing due to anthropogenic sulphate aerosol. The calculated forcing pattern shows large peaks over the eastern United States, southeast Europe and eastern China. The calculated annual global-mean radiative forcing is  $-0.50 \text{ Wm}^{-2}$  for Langner and Rodhe (1991) data and  $-0.49 \text{ Wm}^{-2}$  for Penner *et al.* [1994 (a&b)] data. The forcing was found to vary with season, with a larger forcing during northern hemispheric summer than winter. Sulphate aerosol also appreciably perturbs the lower tropospheric heating rates over northern hemispheric mid-latitudes. The forcing was also found to be sensitive to the global cloud cover and to the optical properties of the aerosol. The possible sources of the differences in magnitude with previous estimates are discussed. Over northern hemispheric mid-latitudes, the negative radiative forcing due to the direct effect of aerosols appreciably offsets the positive forcing due to increase in greenhouse gases.

A 26-layer radiative-convective model (RCM) was also used to examine the equilibrium temperature profiles due to sulphate aerosols and increase in greenhouse gases. It was found that the effect of sulphate aerosols is the cooling of surface-troposphere system. Sulphate aerosols reduce the tropospheric warming and enhance the stratospheric cooling caused by increase in greenhouse gases.

**Key words**—Radiative forcing, Aerosol, Greenhouse gases, Climate change.

## 1. Introduction

Natural and anthropogenic aerosols are capable of changing the earth's radiation budget through direct back scattering effect and their indirect effect on cloud albedo which arises from their role as cloud condensation nuclei (CCN). The possible role of aerosol particles in changing the earth's radiation budget was addressed beginning in the early 1900's (Angström 1929, 1930, Bergeron 1928). This influence has been assumed to be fairly uniform spatially and constant temporally (Coakley *et al.* 1983).

Human activities have increased global emissions of sulfur gases by about a factor of three during the past century, which has caused the concentrations of sulphate aerosol to increase (Langner *et al.* 1992). This aerosol is distributed quite non-uniformly over the earth and more confined in the Northern Hemisphere (NH). Consequently, it can substantially decrease the incoming solar radiation locally and significantly affect the local radiation budget. Moreover, this aerosol is short-lived in the troposphere, unlike the greenhouse gases, which can lead to temporal non-uniformity of the forcing.

These aerosols perturb the planetary radiative budget in two ways. It reflects the incoming solar radiation back to space and this effect is called direct effect. In addition, this aerosol modifies the microphysical properties of the low level clouds and increases their cloud albedo, thus increasing the planetary albedo. This effect is called indirect effect.

The reflection of solar radiation by the aerosol itself, so-called direct effect of the aerosol, has been estimated to have an annual global mean radiative forcing which ranges from  $-0.25$  to  $-0.9 \text{ Wm}^{-2}$  (Charlson *et al.* 1991, 1992, Kiehl and Briegleb 1993, Taylor and Penner 1994, Haywood and Shine 1995, IPCC 1995).

These estimations of the direct effect differ due to different approximations and simplifications used in the models. Charlson *et al.* (1991) calculated the radiative forcing using the three dimensional sulphate aerosol data simulated by Langner and Rodhe (1991) but with a simple column model to account for the scattering of the aerosol. Kiehl and Briegleb (1993) carried out the calculations with the

same aerosol distribution but with a detailed three dimensional radiative transfer model to account for the scattering of the aerosol in a detailed way. Taylor and Penner (1994) also carried out the calculations with a detailed model but with a different aerosol distribution and different assumption on the treatment of optical properties of the aerosol. Their estimation was about three times higher than the result of Kiehl and Briegleb (1993). Haywood and Shine (1995) recently presented the results of their calculations with the modified version of Charlson *et al.* (1991) model and the sulphate aerosol data simulated by Langner and Rodhe (1991) and Penner *et al.* (1994a). Their calculations give an annually averaged forcing due to sulphate aerosol as  $-0.34 \text{ Wm}^{-2}$ .

Since there is a spread in the estimations of the forcing due to anthropogenic aerosols due to different reasons mentioned above, there is scope for further such calculations so that the uncertainty is reduced and the range is narrowed down as recommended by IPCC (1995). Also it is to be mentioned that all calculations except that of Haywood and Shine (1995) used the sulphate aerosol distribution of Langner and Rodhe (1991). Recently, Penner *et al.* (1994a) simulated the sulphate aerosol distribution with a chemistry-transport model which is different from that used by Langner and Rodhe (1991). Therefore, further scope exists for repeating the detailed radiative transfer calculations with this new data set of aerosol distribution also.

In this paper, we report our calculations of radiative forcing and other climatic implications due to the direct effect of anthropogenic sulphate aerosol using a three dimensional radiative transfer model and a multilevel radiative-convective model. We have used both Langner and Rodhe (1991) and Penner *et al.* (1994a) sulphate aerosol distributions for our calculations. In this study, we present seasonal and meridional variations of this forcing, in addition to the geographical variations.

## 2. Calculation Method

### 2.1. Prescribed geographical distributions of sulphate aerosol

The monthly mean geographical distributions of sulphate aerosol concentrations simulated and

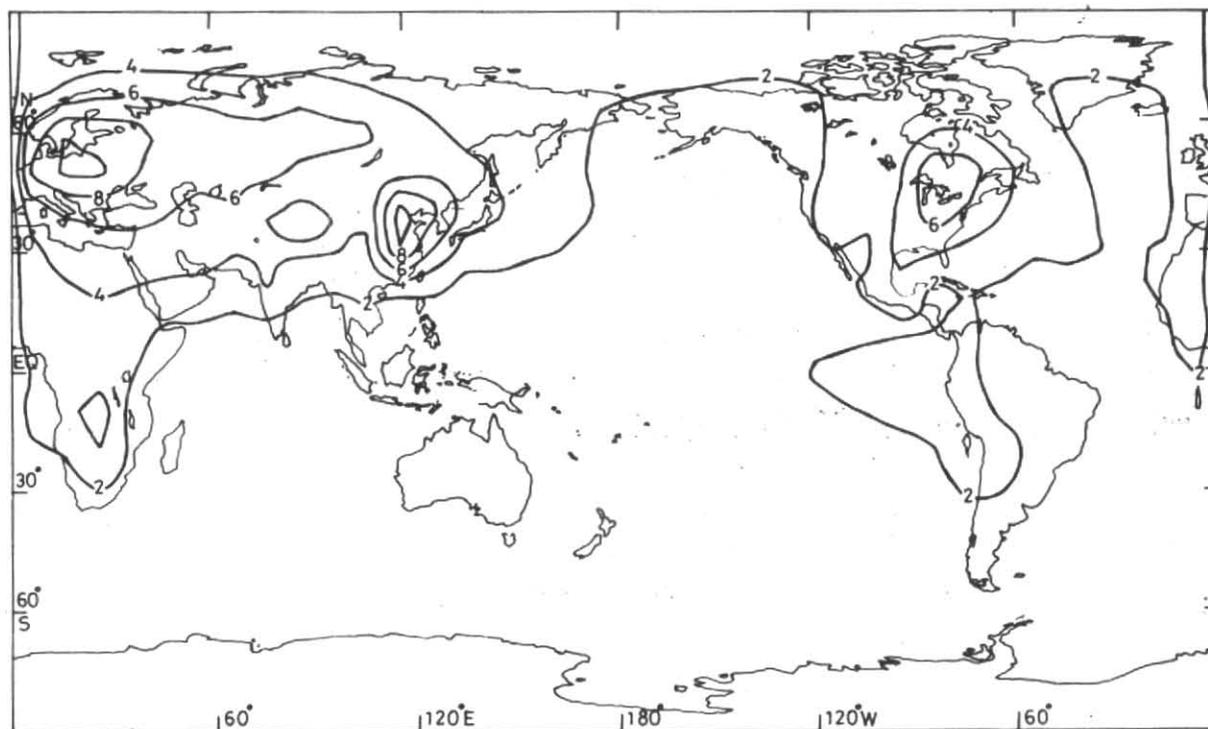


Fig. 1. Geographical distributions of annually averaged anthropogenic sulphate aerosol concentrations ( $\text{gm}^{-2}$ ) from the simulation by Längner and Rodhe (1991)

provided by Langner and Rodhe (1991) and Penner *et al.* (1994a) have been used in this study. Langner and Rodhe (1991) and Penner *et al.* (1994a) simulated these geographical distributions using 3-dimensional chemistry-transport models. The model treats the emission, transport, chemistry and removal processes for three sulphur compounds, dimethyl sulphide (DMS), sulphur dioxide ( $\text{SO}_2$ ) and sulphate. The anthropogenic inputs of sulphur from fossil fuel combustion and various industrial processes are used. The uncertainty in anthropogenic emissions is about 15% while the uncertainty in natural emissions is of the order of a factor of two.

They considered two cases of oxidation of  $\text{SO}_2$  inside clouds. In the standard case, simulation was based on immediate oxidation of  $\text{SO}_2$  as soon as it enters cloud. In the other case, called slow oxidation case, they assumed a delayed rate of oxidation corresponding to some degree of oxidant limitation. We have used the 'slow oxidation case' aerosol. The removal processes considered in the model are in cloud precipitation scavenging, sub cloud scavenging and dry deposition. These simulations are broadly consistent with the observed sulphate

aerosol concentrations in and over polluted regions of Europe and North America. The concentration fields vary with season. Fig. 1 shows the annual geographical distributions of the concentration of anthropogenic sulphate aerosol simulated by Langner and Rodhe (1991) after interpolating to the  $4^\circ \text{ Lat.} \times 5^\circ \text{ Long.}$  Grid of the radiative transfer model. Fig. 2 shows the same distribution as simulated by Penner *et al.* (1994a). Figs. 1 and 2 shows concentration maxima over Europe, southeast Asia, eastern north America and south central Africa. Most of the anthropogenic aerosol is confined in the NH. Annual average zonal distribution of sulphate aerosol is shown in Fig. 3. Most of the anthropogenic aerosols in the NH are confined below 800 hPa.

## 2.2. Radiative properties of sulphate aerosols

To calculate the change in net incoming solar radiation due to aerosols, it is necessary to specify the radiative properties of the aerosol and the amount and spatial distribution of aerosol in the atmosphere. The optical properties are evaluated from standard Mie theory by prescribing the size distribution and index of refraction which depends on the chemical composition. It has been assumed

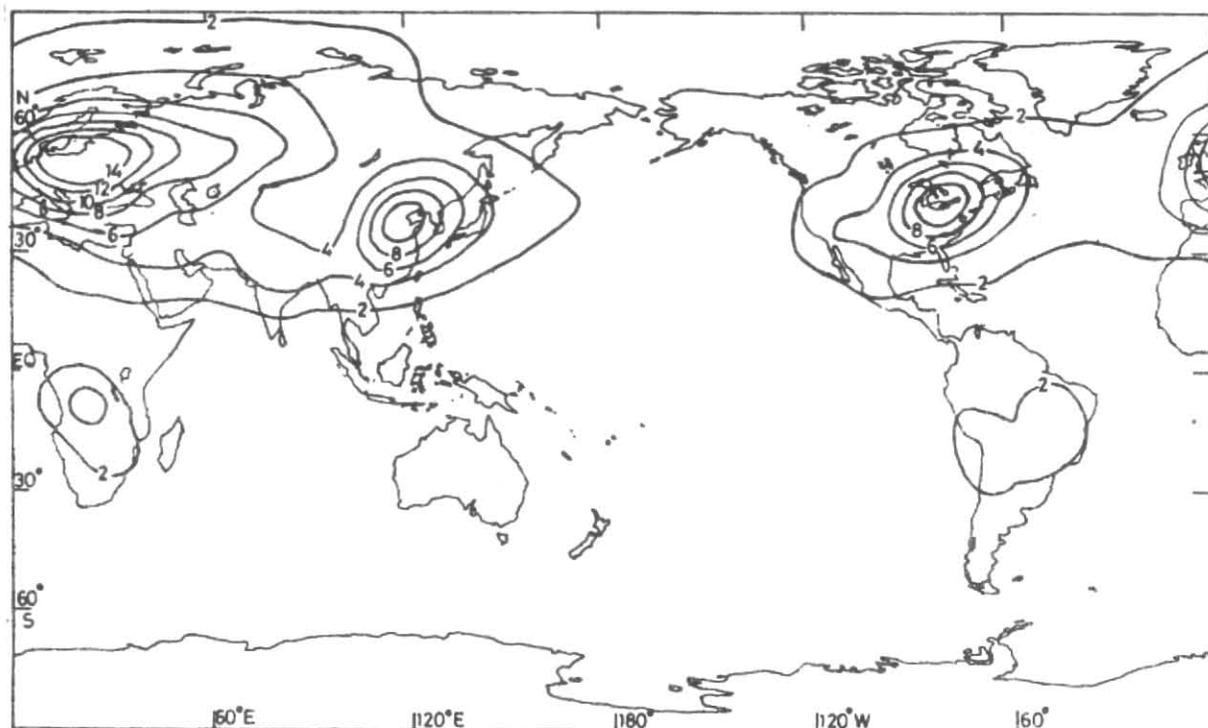


Fig. 2. Same as Fig. 1, but from the simulation by Penner *et al.* (1994)

that the size distribution of the aerosols was log-normal (Whitby 1978). The lognormal distribution is given as,

$$\frac{dN}{d(\ln r)} = C \exp(-P) N_1 \quad (1)$$

where,

$$C = \frac{1}{(2\pi)^{1/2} \ln \sigma}$$

and

$$P = \frac{1}{2} \frac{[\ln(r/r_n)]^2}{[\ln \sigma]^2}$$

$N_1$  is the total number of particles per cubic meter in the accumulation mode,  $r_n$  is the geometric mean radius and  $\sigma$  is the standard deviation of the distribution.

The geometric mean radius by number  $r_n$  was specified as equal to  $0.05 \mu\text{m}$ , the geometric standard deviation  $\sigma$  was 2.0. These values imply

that geometric mean diameter by volume (DGV) is  $0.42 \mu\text{m}$  which falls within the range of observational data. The index of refraction for the sulphate aerosol was based on the assumption that aerosol contained 75%  $\text{H}_2\text{SO}_4$  and 25%  $\text{H}_2\text{O}$ .

The aerosol specific extinction  $\psi_{\text{ext}}(\lambda)$  for a given wavelength is obtained from,

$$\psi_{\text{ext}}(\lambda) = \frac{3}{4 \rho_s \chi_{\text{SO}_4}} \frac{\int Q_e(\lambda, r) r^2 n(r) dr}{\int r^3 n(r) dr} F_n(\text{RH}) \quad (2)$$

where integration is over the particle radius  $r$ ,  $Q_e(\lambda, r)$  is the Mie extinction efficiency,  $n(r)$  is the log-normal size distribution and  $\chi_{\text{SO}_4}$  is the fraction of fine particle mass that is sulphate. This fraction was assumed as 60%. The sulphate dry particle density  $\rho_s$  is assumed to be  $1.7 \text{ g cm}^{-3}$  (Whitby 1978). The size of the aerosol particle depends on relative humidity. Increases in relative humidity results in larger particles which scatter more solar radiation. Charlson *et al.* (1984) reported some observations of the ratio of scattering at a relative humidity to the scattering at 30%. The form of  $F_n(\text{RH})$  was obtained from a fit to these observations.

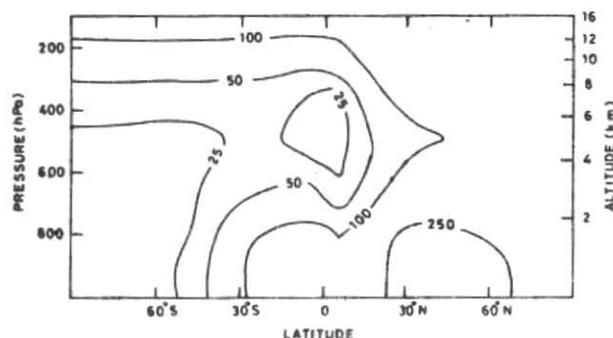


Fig. 3. Zonal-height variation of annually averaged anthropogenic sulphate aerosol concentration of Langner and Rodhe (1991). Units: pptv

The calculated sulphate specific extinction, single scattering albedo  $\omega_0$  and asymmetry parameter  $g$  are shown in Fig. 4.

Specific extinction exhibits a strong wavelength dependence. The variation of  $\omega_0$ , scattering albedo indicates that the sulphate aerosols are non-absorbing in the visible spectral region. The asymmetry parameter  $g$  decreases monotonically in the near infrared region.

### 2.3. Radiative transfer model

The radiative transfer model of the 7-layer tropospheric General Circulation Model (GCM) (Oh and Schlesinger 1991) was used for calculations. This model has 7 layers and the top of the model layer is at 200 hPa. It treats both the solar and infrared radiation in detail. The spectral range of short-wave radiation was divided into three intervals 0-0.44, 0.44-0.69 and 0.69-4.0  $\mu\text{m}$ . The first two intervals are for the treatments of Rayleigh scattering and absorption by ozone and  $\text{CO}_2$ . The last interval is further subdivided into six subintervals. Scattering and absorption by both the gases, cloud droplets and aerosols is calculated using the two stream approach with the delta-Eddington approximation (Geleyn and Hollingsworth 1979). The optical depth and single scattering albedo for cloud droplets are determined following Stephens (1978) for non-ice clouds and Starr and Cox (1985) for cirrus clouds. The parameterization of water vapour absorption is based on Chou and Arking (1981), ozone absorption on Lacis and Hansen (1974) and  $\text{CO}_2$  by Fouqert (1988).

Longwave flux calculations are based on the two stream formula of the flux equations with

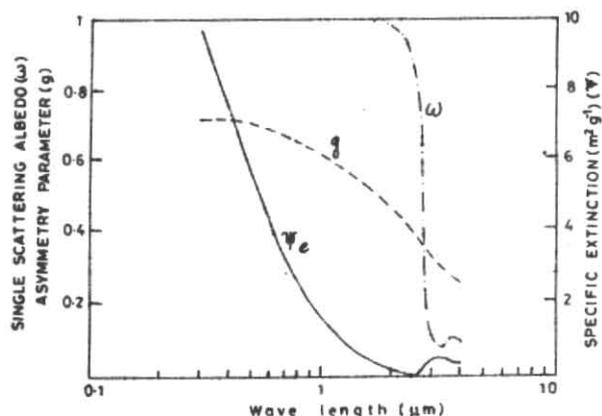


Fig. 4. Radiative properties of sulphate aerosol as a function of wavelength

parameterized optical depths. Gaseous absorptions due to water vapour,  $\text{CO}_2$  and ozone are considered. The parameterization of absorption of water vapour is based on Chou (1984) and Kneizys *et al.* (1983),  $\text{CO}_2$  based on Chou and Peng (1983) and ozone based on Kneizys *et al.* (1983).

In general, several constituents absorb and/or scatter in each homogenous layer. Every constituent is defined in terms of a layer extinction optical depth  $\tau_i$ , single scattering albedo  $\omega_i$  and asymmetry parameter  $g_i$ ,

$$\tau = \sum \tau_i$$

$$\omega = \sum \omega_i \tau_i / \tau$$

$$g = \sum g_i \omega_i \tau_i / \omega \tau$$

where the sums are over all constituents.

To determine the radiative forcing due to the sulphate aerosol, the radiative transfer calculation was performed twice, once with the aerosol present and again with the aerosol absent. The difference between the net radiation at 200 hPa level with and without the aerosol present then gives the radiative forcing by the aerosol.

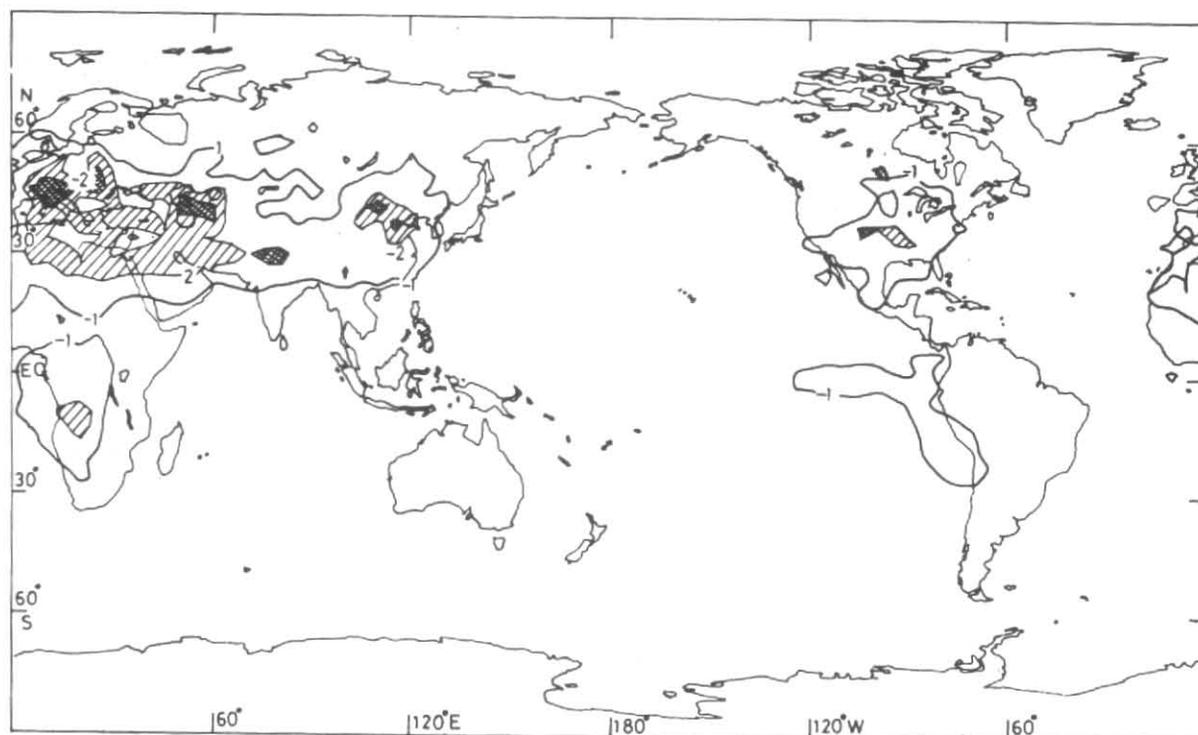
The solar radiation reaching at the top of the atmosphere  $S_\downarrow$  is calculated as

$$S_\downarrow = S_\alpha \cos z \quad (3)$$

$$\cos z = \sin \alpha \sin \delta + \cos \alpha \cos \delta \cos h \quad (4)$$

where,  $S_\alpha$  is solar constant,  $\alpha$  is latitude,  $\delta$  is solar declination and  $h$  is hour angle.

The radiative transfer calculation was performed 24 times per day throughout an entire year and includes both the diurnal and seasonal insolation



**Fig. 5.** Geographical distributions of annual mean radiative forcing ( $\text{Wm}^{-2}$ ) by anthropogenic sulphate aerosols for Langner and Rodhe (1991) data

cycles by suitably changing the solar declination and hour angle.

For each of the 12 calendar months of the radiative transfer calculations, the three dimensional distributions of temperature, water vapour, cloud cover and cloud-water mixing ratio were prescribed from the corresponding monthly mean fields from the last year of a 10-year simulation performed by the University of Illinois 7-layer tropospheric GCM. In this GCM simulation the SST and sea-ice extent were interpolated daily from the corresponding monthly mean observed fields. (This simulation is the control simulation for the 10-year simulation performed as a participant in the Atmospheric Model Intercomparison Project (AMIP)).

Following Kiehl and Briegleb (1993) it is assumed that the sulphate aerosol was located within the lowest 1.5 km of the atmosphere. There are three layers below 1.5 km in the 3-D model. Longwave radiative transfer in the calculations of the radiative forcing of sulphate aerosol has not been included because there is then little temperature difference between the aerosol layer

and the surface and the optical depth in the longwave region is negligible.

### 3. Three dimensional Model Results

#### 3.1. Geographical distributions of the radiative forcing

The annual-mean radiative forcing at the top of the model atmosphere by anthropogenic aerosol is presented in Fig. 5 for Langner and Rodhe (1991) data and Fig. 6 for Penner *et al.* (1994a) data. The spatial pattern of the forcing is similar to those obtained by others. The maximum negative radiative forcing by the anthropogenic aerosols is located over the southeastern Europe, eastern China, southeast United States and south central Africa coinciding with the maximum optical depths. The negative values over these regions often exceeded  $-2 \text{ Wm}^{-2}$ . The radiative forcing by anthropogenic sulphate aerosols is larger in the NH. The contribution from NH to the global forcing is about 75%.

#### 3.2. Zonal means

The annual zonal-mean radiative forcing due to anthropogenic aerosol is presented in Fig. 7. This

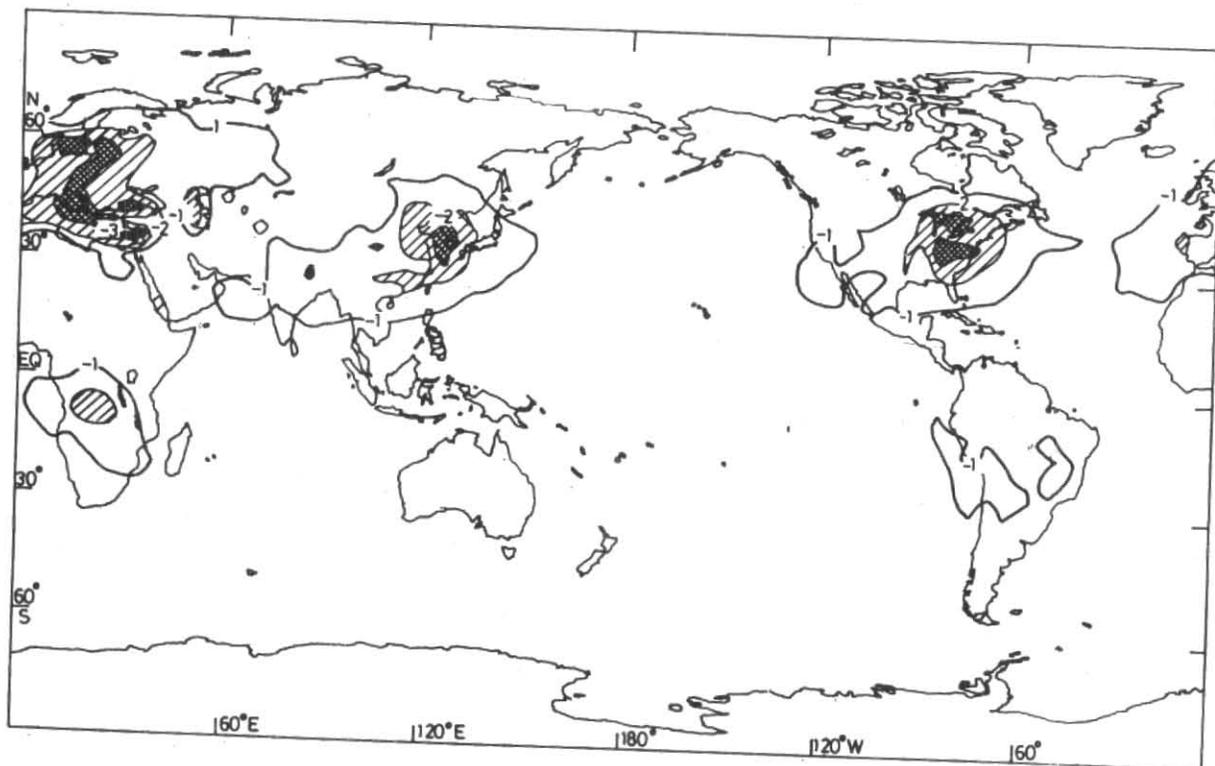


Fig. 6. Same as Fig. 5 but for Penner *et al.* (1994)

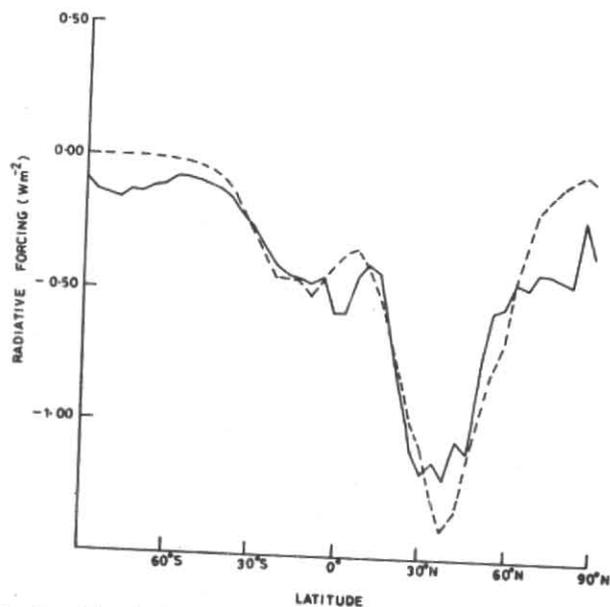


Fig. 7. Zonal mean annual-mean radiative forcing ( $Wm^{-2}$ ) due to anthropogenic sulphate aerosols. Dashed line for Penner *et al.* (1994) data and continuous line for Langner and Rodhe (1991) data

diagram shows a large negative peak in the middle latitudes of the NH. This peak corresponds to maximum abundance of anthropogenic aerosols caused by industrial activities in United States, Europe and China. Comparatively, the forcing by

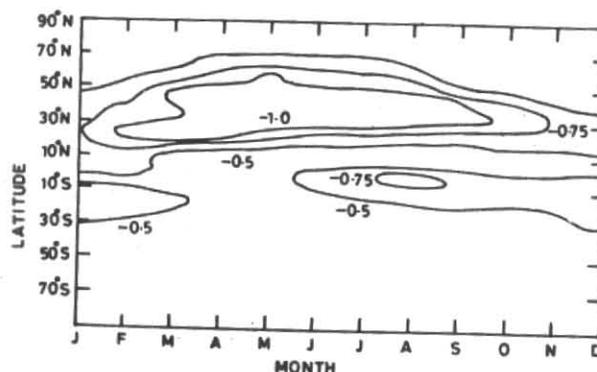


Fig. 8. Month-latitude distribution of the anomaly of radiative forcing from the annual-mean zonal-mean radiative forcing ( $Wm^{-2}$ )

aerosol simulated by Penner *et al.* (1994a) is larger in the NH mid-latitudes.

Fig. 8 shows the month-latitude distribution of the anomaly of monthly mean zonal-mean radiative forcing due to anthropogenic and natural sulphate aerosols from their respective annual-mean zonal means. The largest anomaly was observed over the NH mid-latitudes during the summer months. A large positive anomaly was observed in the Southern Hemisphere (SH). This suggests that direct radiative forcing is dominant during summer months.

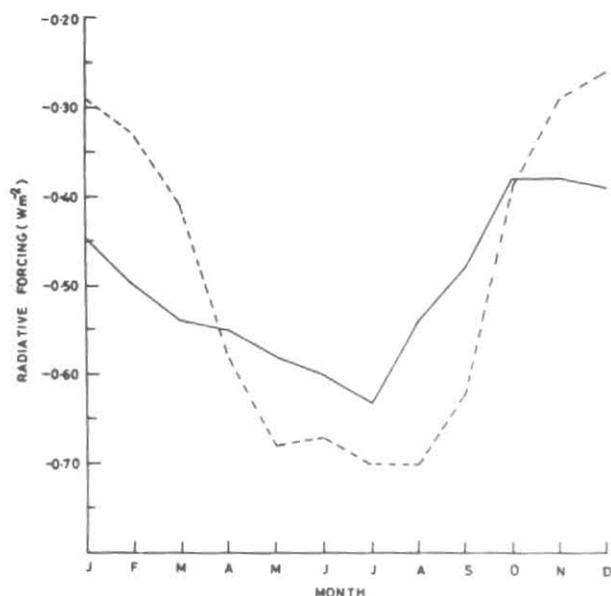


Fig. 9. Monthly variation of the global mean radiative forcing ( $\text{Wm}^{-2}$ ) due to anthropogenic aerosols. Dashed line for Penner *et al.* (1994) data and continuous line for Langner and Rodhe (1991) data

### 3.3. Global and hemispheric means

The calculated annual global-mean radiative forcing due to anthropogenic aerosol is  $-0.5 \text{ Wm}^{-2}$  for Langner and Rodhe (1991) data and  $-0.49 \text{ Wm}^{-2}$  for Penner *et al.* (1994a) data. This forcing is almost one-fourth of the positive radiative forcing of  $2.1 \text{ Wm}^{-2}$  due to increases in greenhouse gases from pre-industrial period to the present. These values are almost 1.7 times more than that obtained by Kiehl and Briegleb (1993). Taylor and Penner (1994) obtained the value as  $-0.95 \text{ Wm}^{-2}$  which is almost twice our value. In section 5, these differences and their possible causes have been discussed.

The monthly variation of global-mean radiative forcing due to anthropogenic aerosol is presented in Fig. 9. The hemispheric mean forcing exhibits a large seasonal variation mainly caused by seasonal variation of incoming solar radiation, with the forcing being largest during summer.

### 3.4. Perturbations in heating rates

In order to examine the possible perturbations in the heating rates due to anthropogenic aerosols, the

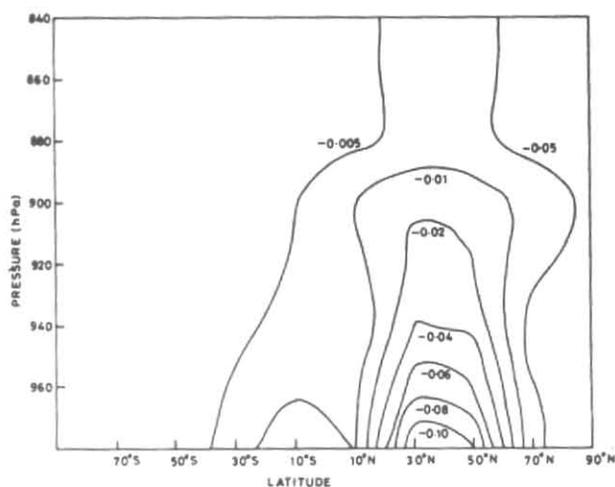


Fig. 10. Annually averaged perturbations of heating rates ( $^{\circ}\text{C}/\text{day}$ ) due to anthropogenic sulphate aerosol Langner and Rodhe (1991) data

heating rate ( $^{\circ}\text{C}/\text{day}$ ) perturbations due to anthropogenic sulphate aerosol have been presented in Fig. 10. It indicates the negative heating rate perturbation of the order of  $0.08^{\circ}\text{C}/\text{day}$  over the NH mid-latitudes in the lower troposphere due to the divergence of the short-wave flux. There are hardly any changes in the heating rates in the SH. In the NH, however, the perturbations are confined below 800 hPa decreasing with height from the surface.

### 3.5. Comparison with the greenhouse forcing

Since the radiative forcing due to sulphate aerosol is showing large regional changes, it is important to compare this forcing with the positive forcing due to increases in greenhouse gases. Thus we have calculated the spatial distribution of the change in the net longwave flux at the top of the model atmosphere for the trace gas increases from pre-industrial period to the present. These values are taken from IPCC (1990). The annual zonal means of the forcing due to greenhouse gases are shown in Fig. 11. The annual zonal means of combined forcing due to aerosols and greenhouse gases are also shown in this diagram. The forcing due to greenhouse gases shows maximum in the subtropical regions. The combined forcing pattern reveals that the negative forcing due to the direct effect of sulphate aerosols offsets appreciably the positive forcing due to greenhouse gases, thus leading to substantial reduction of positive forcing by the greenhouse gases.

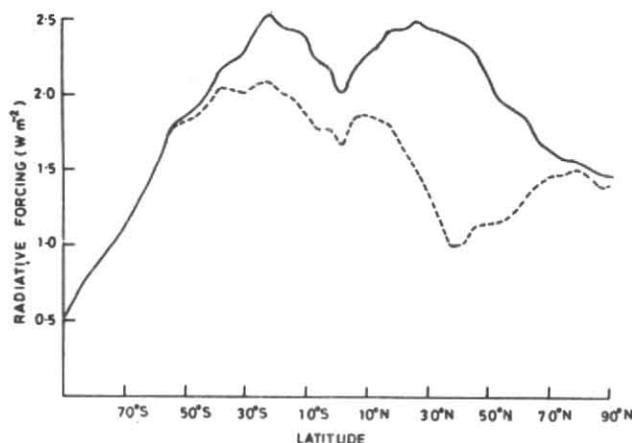


Fig. 11. Meridional variation of annual mean radiative forcing in  $\text{Wm}^{-2}$  due to increases in greenhouse gases (thickline) and combined increases in greenhouse gases and anthropogenic aerosols (dashed line)

### 3.6. Comparison with other studies

The calculated global and hemispheric-mean radiative forcing for the Langner and Rodhe (1991) and Penner *et al.* (1994a) aerosol distributions is compared in Table 1 with the forcing obtained by Kiehl and Briegleb (1993) for the same aerosol distribution. Table 1 also shows the results of Charlson *et al.* (1991) and Haywood and Shine (1995) for the distribution of Langner and Rodhe (1991) and of Taylor and Penner (1994) for a different aerosol distribution. Our global mean radiative forcings for anthropogenic aerosols are 71% larger than the values of Kiehl and Briegleb (1993), 44% larger than Haywood and Shine (1995), 20% smaller than Charlson *et al.* (1991) and 49% smaller than the forcing of Taylor and Penner (1994).

The partition of our global anthropogenic forcing between the Northern and Southern Hemispheres, around 75% and 25%, is identical with the partition obtained by Kiehl and Briegleb (1993), 76% and 24% and more symmetric than the 90% and 10% obtained by Charlson *et al.* (1991) and the 80% and 20% obtained by Haywood and Shine (1995). The percentage ratio of anthropogenic sulphur emission for NH and SH is 90:10. However, the forcing values are not on the same ratio.

The magnitude of the radiative forcing obtained by Taylor and Penner (1994) is about twice our value. There are three possible reasons for their large forcing. First, Taylor and Penner (1994) used a

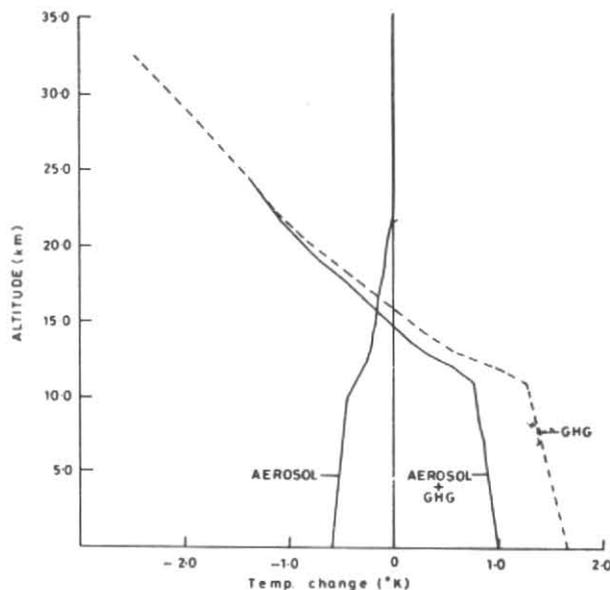


Fig. 12. Equilibrium temperature profiles for the globally averaged sulphate aerosol, increase in trace gases from pre-industrial period and the combined increase in trace gases and sulphate aerosols.

geographical distribution of aerosol concentration field, calculated by a chemistry-transport model different from that of Langner and Rodhe (1991), with the consequence that the sulphate aerosol concentration predicted by Taylor and Penner (1994) is about twice as large as that observed over Europe and varies more strongly with season than the Langner and Rodhe (1991) distributions used here. Secondly, Taylor and Penner (1994) used a constant scattering coefficient of  $8.5 \text{ m}^2/\text{g}$  at  $0.55 \mu\text{m}$ , independent of relative humidity, while our scattering coefficient varies with wavelength with values that decrease rapidly beyond  $0.55 \mu\text{m}$  resulting in smaller forcing.

On the other hand, our results are larger in magnitude than the values obtained by Kiehl and Briegleb (1993), even though we have used the same geographical distributions of sulphate aerosol concentrations. The relative humidity dependent scattering coefficient has also been used which varies with wavelength. Some portion of the difference can be explained by the difference in cloud fields. Whereas Kiehl and Briegleb (1993) have normalized the layer clouds for 60% global cover, we have used the cloud fields whose global mean is 52% which is closer to the climatological cloud cover of 54%.

TABLE 1  
Comparison of radiative forcing ( $\text{Wm}^{-2}$ ) results  
(Percentage values are shown in parentheses)

| Region | Author      |             | Kiehl & Briegleb (1993) | Taylor & Penner (1994) | Charlson <i>et al.</i> (1991) | Haywood and Shine (1995) |
|--------|-------------|-------------|-------------------------|------------------------|-------------------------------|--------------------------|
|        | LR (91)     | PEN (94)    |                         |                        |                               |                          |
| N. H.  | -0.72 (74%) | -0.74 (75%) | -0.43 (76%)             |                        | -1.07 (90%)                   | -0.55 (80%)              |
| S. H.  | -0.25 (26%) | -0.24 (25%) | -0.13 (24%)             |                        | -0.11 (10%)                   | -0.13 (20%)              |
| Global | -0.50       | -0.49       | -0.28                   | -0.95                  | -0.60                         | -0.34                    |

LR (91)—Langner and Rodhe (1991) data. PEN (94)—Penner *et al.* (1994) data.

The direct radiative forcing mechanism mainly acts over cloud-free regions. Similarly, the forcing is sensitive to the relative humidity function. When we change the relative humidity function to unity, we get a radiative forcing of  $-0.31 \text{ Wm}^{-2}$  of Langner and Rodhe (1991) data for anthropogenic aerosol. The relative humidity function was obtained based on a very few observations reported in Charlson *et al.* (1984), hence there may be uncertainties in determining the relative humidity dependence of the scattering coefficient.

The estimations obtained by Haywood and Shine (1995) are smaller than those obtained in this study, even though both studies used the same aerosol distributions. Some portion of the difference can be explained by the fact that Haywood and Shine (1995) did not include the extra scattering due to aerosols in cloudy skies which is considered to be significant (IPCC 1995). In our calculations the extra scattering in cloudy skies contributed about 20% of the total radiative forcing. If we account for this difference, radiative forcing estimated in this study can be reduced to  $0.36 \text{ Wm}^{-2}$  which is close to the result of Haywood and Shine (1995). In addition, they have assumed a growth factor of 1.32 to allow for hygroscopic nature instead of accounting for the changes in scattering efficiency due to changes in relative humidity. Therefore, if we account the extra scattering over cloudy regions and relative humidity dependence then their results would have increased to approximately  $-0.49 \text{ Wm}^{-2}$  which is close to our results.

IPCC (1995) recommended a central value of  $-0.4 \text{ Wm}^{-2}$  with a range from  $-0.25$  to  $-0.9 \text{ Wm}^{-2}$  for the direct radiative forcing due to sulphate aerosols. Present results are about 20% higher than the central value recommended by IPCC (1995).

#### 4. 1-D Model Results

##### 4.1. The 26-layer Radiative-Convective Model

In order to investigate the equilibrium temperature changes due to anthropogenic aerosols, the University of Illinois Multi-layer one dimensional radiative-convective model (Ernwis 1992) was used.

The radiative-convective model is formulated from the thermodynamic energy equation

$$\rho C_p \frac{dT}{dt} = \frac{dS}{dz} - \frac{dR}{dz} + Q_{\text{sfc}} + Q_{\text{conv}} \quad (5)$$

where,  $T$  is temperature,  $t$  is time,  $z$  is altitude,  $\rho$  is density of air,  $C_p$  is the heat capacity at constant pressure.  $S$  is the net downward solar radiation flux,  $R$  is the net upward longwave radiation flux,  $Q_{\text{sfc}}$  is the non-radiative transfer of energy from the surface to the atmosphere and  $Q_{\text{conv}}$  is the convective redistribution of energy within the atmosphere. The radiative treatment in this model is the same as in the 3-D model described in section 2.3. The surface convection parameterization used in the study is that introduced by Manabe and Strickler (1964). The globally averaged profiles of temperature, water vapour and ozone are taken from McClatchy *et al.* (1972). We have assumed clear sky conditions in the model simulations. The surface albedo was assumed as 0.15. There are three model layers below 1.5 km.

The model was integrated forward with the time step of 8 hours with convective adjustment to maintain the critical lapse rate limit ( $6.5^\circ\text{C}/\text{km}$ ). We have also considered the increases in trace gases from pre-industrial period to the present value (IPCC 1990). The model is integrated forward

allowing the model to reach new convective equilibrium with convective adjustment scheme.

Fig. 2 shows vertical profile of the change in temperature due to increases in greenhouse gases, globally averaged sulphate aerosol and a simultaneous increase in both aerosols and trace gases. The increases in trace gases cause a warming to the surface-troposphere system and monotonous cooling in the stratosphere. The surface-troposphere forcing for the increases of GHGs is  $2.1 \text{ Wm}^{-2}$  similar to the value of  $2.04 \text{ Wm}^{-2}$  obtained by Hansen and Lacis (1990). The corresponding surface change is  $1.6^\circ\text{K}$ .

The effect of sulphate aerosols is the cooling of the surface-tropospheric system. The surface-tropospheric radiative forcing due to sulphate aerosols is  $-0.67 \text{ Wm}^{-2}$  and the corresponding surface temperature decrease is  $-0.59^\circ\text{K}$ . Fig. 12 also shows the temperature equilibrium profile for the combined GHG and aerosol. Now the surface temperature is reduced to  $1.1^\circ\text{K}$ , thus offsetting some portion of warming due to GHGs by aerosols. It is also interesting to note that sulphate aerosols also cool the stratosphere due to divergence in the short-wave flux by approximately  $0.15^\circ\text{K}$ , more than the GHG increases alone. Thus sulphate aerosols offset some portion of surface-troposphere warming and enhances the stratospheric cooling due to GHG's.

### 5. Evidence of Influence due to Sulphate Aerosols

There is scanty evidence of the influence of the radiative forcing in inter-hemispheric differences of annual temperature anomaly trend over the industrial era (Wigley 1989). However, comparison of trends of mean daily maximum and minimum temperatures in industrial regions indicates a greater rate of warming at night than during day time, indicative of a short-wave forcing by anthropogenic sulphate aerosol in conjunction with overall global warming over this period. Because of greater insolation in summer than winter the short-wave radiative influence of sulphate aerosol is expected to be greater in summer and might lead to a lesser rate of warming in NH midlatitudes in summer than in winter. To examine the seasonal dependence of this forcing, Hunter *et al.* (1993) examined the seasonal

and latitudinal dependence of temperature anomaly trend evaluated from the monthly combined land and SST anomaly data set of the University of East Anglia Climatic Research Unit (Jones *et al.* 1986). They found pronounced minima in the rate of temperature increase in summer months in the NH midlatitudes which are consistent with the latitudinal distribution of anthropogenic aerosol forcing. However, Hunter *et al.* (1993) have not taken into account the possible influence of volcanic eruptions and ENSO events (Jones *et al.* 1994) while estimating the temperature trends. Jones and Briffa (1992) who analyzed spatial, temporal and seasonal variations of global surface temperatures also suggest that the NH midlatitudes exhibited least warming in summer, whereas SH did not exhibit any significant seasonal cycle. Manabe *et al.* (1992) examined the temperature trend as a function of latitude and season in response to a hemispherically symmetric forcing corresponding to 1% per year increase in  $\text{CO}_2$  in a coupled ocean-atmospheric model. Except for the NH midlatitude summer the model results closely agree with the observed trends at all latitudes and seasons. Little seasonal variation was exhibited in the model temperature trends in the latitude range  $30^\circ$  to  $60^\circ\text{N}$  in contrast to the observations. Recently Mitchell *et al.* [1995 (a&b)] used UK Met Office coupled general circulation model to examine the climate response to increasing greenhouse gases and sulphate aerosols. These studies suggest that the inclusion of sulphate aerosols significantly improves the agreement between the simulated and observed global mean and large scale patterns of temperature in recent decades.

### 6. Conclusions

Using a three dimensional radiative transfer model alongwith two model derived sulphate concentration fields we have calculated the annual cycle of direct radiative forcing due to anthropogenic sulphate aerosols. The spatial pattern shows pronounced maximum values over southeast United States, southeast Europe and eastern China. The global annual radiative forcing due to anthropogenic and natural aerosol was estimated to be  $-0.5 \text{ Wm}^{-2}$  for Langner and Rodhe (1991) and  $-0.49 \text{ Wm}^{-2}$  for Penner *et al.* (1994a) respectively. The zonal profile of the forcing shows pronounced

peak over NH midlatitudes. The forcing also exhibits large seasonal variation with maximum during NH summer. The forcing was found to be sensitive to the optical properties of the aerosol and cloud fields. A 26-layer radiative-convective model was used to examine the equilibrium temperature profiles due to sulphate aerosols, and increase in greenhouse gases. It was found that sulphate aerosols reduces the tropospheric warming and enhances the stratospheric cooling caused by increases in greenhouse gases.

Globally averaged radiative forcing due to increase in greenhouse gases from pre-industrial times was estimated to be  $2.1 \text{ Wm}^{-2}$ . The indirect effect of sulphate aerosol through cloud albedo is not unambiguously determined. Jones *et al.* (1994) estimated global mean radiative forcing due to anthropogenic aerosol as  $-1.3 \text{ Wm}^{-2}$ . Therefore, the total radiative forcing due to sulphate aerosol ranges from  $-1.58$  to  $-2.25 \text{ Wm}^{-2}$ . The higher limit (obtained by Taylor and Penner 1994) is near to the greenhouse forcing, but with opposite sign. However over NH midlatitudes where aerosol negative forcing is more pronounced, the net forcing over that area would be negative, *i.e.*, cooling.

There are still some uncertainties in estimating the radiative forcing due to sulphate aerosol. The sulphate concentration fields predicted by the chemistry-transport model differ and the difference over Europe (where the maximum concentrations are predicted) is a factor of two. The relative humidity dependence of radiative properties are to be determined with more observations. Kiehl and Briegleb (1993) examined the variation of forcing as a function of the width of the particle size distribution and the results indicate that for a 25% increase in width of the size distributions, the direct forcing decreases by roughly 14%. Penner *et al.* (1994b) discussed various options and strategies to quantify and minimize the uncertainty of climate forcing by anthropogenic aerosols. The priorities are for obtaining surface and aircraft based observations of aerosol chemical and physical properties and their influence on the radiative field and satellite observations of aerosol abundance and size distributions.

However, aerosol effects must be taken into account in evaluating anthropogenic influences on

projected future climate, since this forcing is likely to offset the global greenhouse warming to a great extent and this offset would be more pronounced over NH midlatitudes.

#### Acknowledgements

This work in part has been carried out at the Deptt. of Atmospheric Sciences, University of Illinois, USA while the author was on visit under WMO/UNDP fellowship program. The author is grateful to the Director General of Meteorology, India Meteorological Department nominating the author for this fellowship. Thanks are also due to Prof. M.E. Schlesinger, University of Illinois for suggesting this problem and providing necessary facilities and to Dr J.Langner, Swedish Meteorological Services, Stockholm, and Dr Joyce Penner of Lawrence Livermore Laboratory, USA for kindly supplying the sulphate concentration data used in this study and to the anonymous referee whose comments helped to improve the quality of the paper. The preliminary calculations are carried out at CRAY-YMP4/464 at the National Centre for Super Computing Applications, University of Illinois at Urbana-Champaign, USA.

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