MAUSAM, 49, 2 (April 1998), 195-202

551.510.42: 551.576.11:551.58:551.521.14

Aerosol - cloud - climate effect: Study with a radiative transfer model

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(Received 25 January 1996, Modified 27 January 1997)

सार — त्रिविमीय विकिरणात्मक ट्रांसफर मॉडल का उपयोग करते हुए, मानवजनित सल्फेट वायुविलय से उत्पन्न अप्रत्यक्ष विकिरणशील द्रुतगति (मेघ एल्बीडो में वृद्धि द्वारा) का परिकलन किया गया है। गणना दैनिक एवं मौसमी काल चक्रों तथा सल्फेट वायुविलय सांद्रण आँकड़ों के त्रिविमीय वितरणों से की गई है। भूमंडलीय स्तर पर ज्ञात किए गए अप्रत्यक्ष द्रुतगति औसत वार्षिक माध्यमान का अनुमान —1.13 प्रति वर्ग वॉट मीटर लगाया गया है, जिसकी तुलना हाल ही के जी. सी. एम. की गणनाओं से की जा सकती है। औद्योगिकीकरण युग के पहले से लेकर अब तक ग्रीन हाऊस गैसों में वृद्धि के कारण —1.13 प्रति वर्ग वॉट मीटर की इस द्रुतगति की तुलना 2.12 प्रति वर्ग वॉट मीटर के साथ की गई है। मानवजनित वायुविलय से उत्पन्न यह अप्रत्यक्ष द्रुतगति, मूलतः उत्तरी गोलार्द्ध के औद्योगिक क्षेत्र में ग्रीन हाऊस गैसों के उष्ण प्रभाव की तुलना में कहीं उच्च है। इस तरह से इन क्षेत्रों में सकल विकिरणात्मक द्रुतगति विरोधी है अर्थात् यह शीतलन है। अप्रत्यक्ष द्रुतगति के आकलन से संबंधित कुछ ऐसी अनिश्चितताएं जिनके बारे में आगे और अनुसन्धान की आवश्यकता है, उनका यहाँ उल्लेख किया गया है।

ABSTRACT. The indirect radiative forcing (through the enhancement of cloud albedo) due to anthropogenic sulphate aerosols was calculated using a three-dimensional radiative transfer model. The calculations were made with diurnal and seasonal cycles and three-dimensional distributions of sulphate aerosol concentration data. The globally averaged annual mean indirect forcing is estimated to be -1.13 Wm^{-2} which is comparable with the recent General Circulation Model calculations. This forcing of -1.13 Wm^{-2} is compared with 2.12 Wm^{-2} due to increase in greenhouse gases from the pre-industrial period to the present. This indirect forcing due to anthropogenic aerosol is substantially higher than the warming effect due to greenhouse gases over the industrial regions in the northern hemisphere, so that the net radiative forcing over these regions is negative, *i.e.* cooling. Some uncertainties in the estimation of the indirect forcing that needs further research are pointed out.

Key words — Radiative forcing, Aerosol, Climate change, Cloud albedo.

1. Introduction

Anthropogenic aerosol compounds in the atmosphere have increased dramatically primarily during the course of industrialization and most rapidly since about 1950. Of all the particulate pollutants that humans create, sulphate aerosols are the best understood because of the availability of a large body of data. Unlike the greenhouse gases this aerosol is distributed quite non-uniformly over the earth and more confined in the northern hemisphere (NH).

Radiative influences of aerosols on climate may be divided as direct, referring to scattering and absorption of radiation by the aerosol particle themselves and indirect referring to the influence of aerosols on cloud radiative properties. Apart from backscattering and absorption of solar radiation, aerosol particles also absorb infra red (IR) radiation. But this effect is usually small because the opacity of aerosols decrease at longer wavelengths. Previous estimates (Charlson *et al.* 1992, Kiehl and Briegleb 1993, Taylor and Penner 1994) of the direct forcing by anthropogenic sulphate aerosols indicate that this forcing is of comparable magnitude to the greenhouse gases forcing but opposite in sign.

The indirect way in which the sulphate aerosols cool the earth is by influencing the albedo of clouds. In clouds some of the sulphate particles act as nuclei for condensation. The density of cloud condensation nuclei determines the number density and the size of cloud droplets. For a given amount of condensed water, the number density in turn affects the albedo of the cloud. Thus by changing the droplet size distribution and concentration the optical properties of the cloud may be altered sufficiently to change the global energy budget and thus the climate. This effect is called Indirect effect or ' Twomey effect " (Twomey 1974, Twomey *et al.* 1984). Charlson *et al.* (1992) and Schlesinger

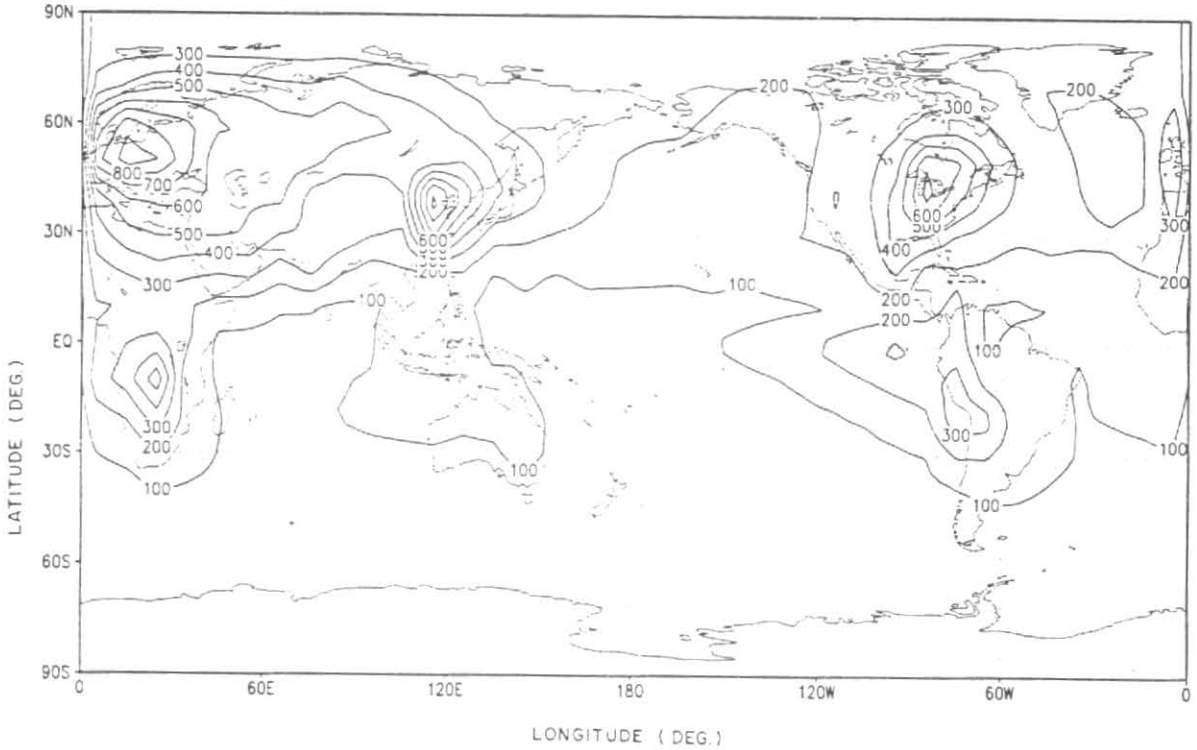

Fig.1 (a). Annual mean total (natural +anthropogenic) sulphate aerosol concentration, Global mean : 180 cm^{-3}

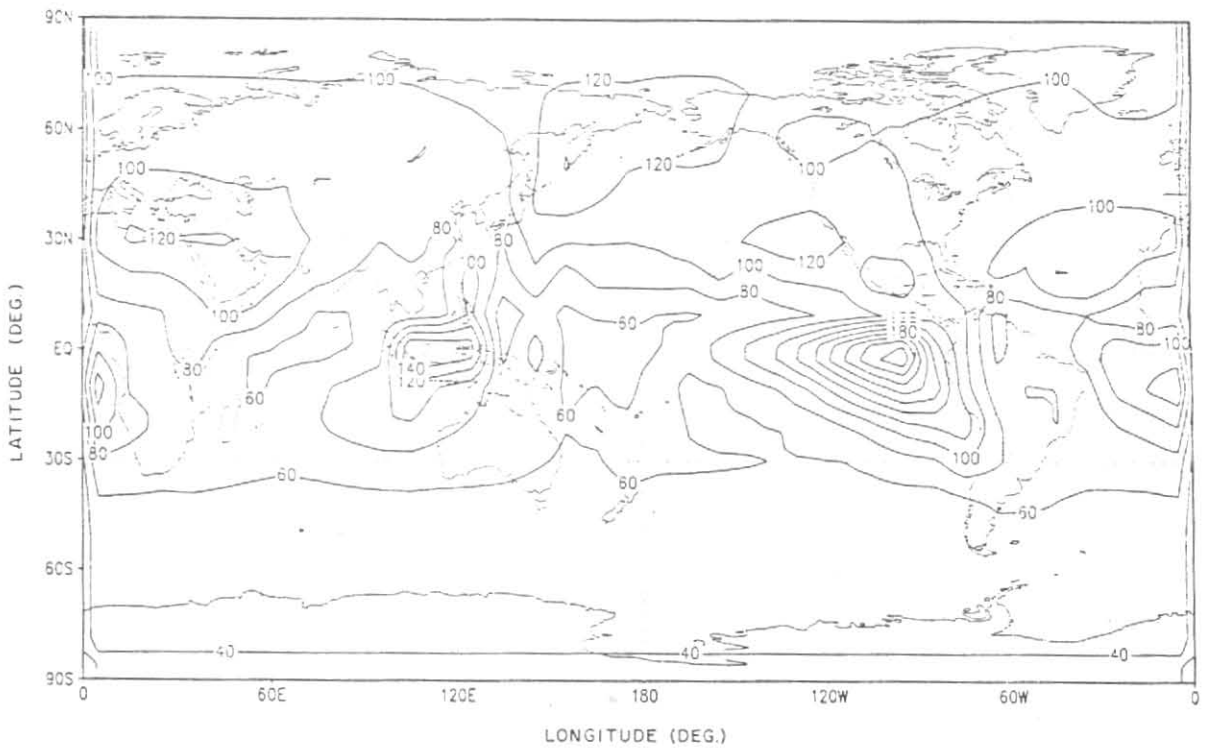

Fig.1(b). Annual mean natural sulphate aerosol concentration, Global mean : 83 cm^{-3}

et al. (1992) made a preliminary estimate of this forcing roughly as - 1 Wm^{-2} by simple calculations.

There are also some observational evidences in which the anthropogenic aerosols influence the albedo of the low

level clouds. Shiptrack observations reported by Coakley *et al.* (1987) reveal a simultaneous decrease in cloud droplet radii and increase in cloud albedo. Kim and Cess (1993) reported enhanced cloud albedo off the east coasts of industrialized regions using ERBE data. However, it was not known on how difference in the number of cloud condensation nuclei and cloud droplets relate to the changes in the amount or mass of anthropogenic aerosols. Thus without this specific knowledge it was not possible to estimate this forcing more accurately.

Recently Martin *et al.* (1994) and Leaitch *et al.* (1992) analysed the aircraft observations of the microphysical characteristics of warm strato-cumulus clouds from a wider range of locations and proposed parameterization relating droplet concentration and aerosol concentration. They also proposed a useful relationship for the effective radius (r_e) of the droplet size spectrum in terms of droplet concentrations. Jones *et al.* (1994) made use of these parameterizations to calculate the indirect radiative forcing with the Hadley Centre atmospheric general circulation model. Recently Boucher and Lohmann (1995) studied this effect using the data of Leaitch *et al.* (1992) and others. They simulated this effect using two general circulation models.

In this study, similar calculations of indirect radiative forcing due to anthropogenic aerosols using the observations of Martin *et al.* (1994) and Leaitch *et al.* (1992) and a three-dimensional radiative transfer model are reported and compared this forcing with the positive forcing due to the greenhouse gases. Some sensitivity studies have also been carried out in this context are also reported.

2. Sulphate aerosol concentration data

The monthly mean geographical distributions of sulphate aerosol concentrations simulated by Langner and Rodhe (1991) are used in the present study. They simulated these geographical distributions using a three dimensional chemical-transport model which treats the emission, transport, chemistry and removal processes for three sulphur compounds, dimethyl sulphide, sulphur dioxide and sulphate. These processes were resolved using an Eulerian transport model with a horizontal resolution of 10° long.× 10° lat. and ten vertical layers between the earths surface and 100 hPa. Sources were divided into anthropogenic and natural emissions (from oceans, plants, soils and volcanoes). Emissions of sulphur components used in the Chemistry - transport model are given in Table 1. These simulations are broadly consistent with the observational sulphate aerosol concentrations in and over polluted regions of Europe and North America.

For the present calculations it was assumed that the sulphate is in the form of ammonium sulphate. To calculate

TABLE 1

Sources of sulphur components used in the Chemistry-transport Model (Units : Tg S yr^{-1})

Sources	Industrial case	Pre-industrial case
Anthropogenic SO_2	66.5	0.0
Anthropogenic SO_4	3.5	0.0
Biomass burning (SO_2)	2.5	0.25
Volcanoes	8.5	8.5
Oceans (DMS)	16.0	16.0
Soils and Plants	1.0	1.0

the distributions of aerosol particle number concentration a log-normal size distribution (Appendix A) was assumed using a median particle radius of 0.05 mm and a geometric standard deviation of 2.0. The vertical distribution of the aerosols was approximated by assuming that half of the column integrated mass was located in the lowest 1.5 km of the atmosphere. This yields annual average total (natural + anthropogenic) sulphate aerosol number concentration ranging from $<100 cm^{-3}$ over the oceans to $>800\ cm^{-3}$ over central Europe. (Fig.1a.) When only preindustrial case aerosols are considered to have the maximum concentrations of the order of 200 cm^{-3} and are located over eastern parts of Pacific (Fig.1b). Thus the maximum anthropogenic aerosol concentrations are observed over central Europe, Eastern China and eastern United States.

3. Radiative transfer model and calculation method

A three dimensional radiative transfer model of (Oh and Schlesinger 1991) was used in the calculations. This model has seven vertical layers from surface to 200 hPa and a horizontal resolution of 5° long. × 4° lat. The model treats both solar and IR fluxes in detail. The scattering and absorption by both gases, cloud droplets and aerosols are calculated using the two stream approach with the delta-Eddington approximation (Geleyn and Hollingsworth 1979). The spectral range of solar radiation is divided into eight intervals. Fractional cloud cover is treated by assuming maximum overlap for vertically contiguous cloud layers and random overlap for vertically noncontiguous cloud layers.

We have used the parameterizations proposed by Slingo (1989) for the optical depth and single scattering albedo for cloud droplets as the functions of the effective radius.

The radiative properties of the clouds are calculated as follows:

$$\tau_i = \mathrm{LWP}\ (a_i + b_i / r_e)$$

$$1 - \omega_i = C_i + d_i * r_e$$

$$g_i = e_i + f_i \cdot r_e \qquad (1)$$

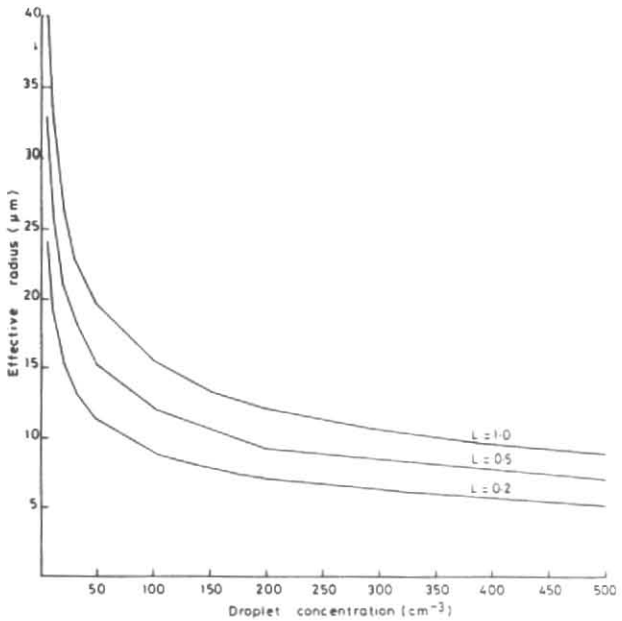

Fig.2. The variation of cloud droplet effective radius with cloud droplet concentration for continental clouds with different cloud liquid water content (gm^{-3})

where τ_i is the cloud optical depth in the g_i th spectral interval, ω_i is the single scattering albedo and g_i the asymmetry parameter. LWP is the liquid water path and r_e is the equivalent radius of the drop size distribution. r_e is defined as the ratio of the third to the second moment of the size spectrum,

$$r_e = \frac{\int_0^\infty n(r) r^3 dr}{\int_0^\infty n(r) r^2 dr} \qquad (2)$$

where $n(r)$ is dropsize distribution function.

$a_i, b_i, c_i, d_i, e_i, f_i$, are constants.

The cloud droplet effective radius (r_e) are stratiform and shallow convective clouds can be related to cloud droplet number concentration N_{tot} (cm^{-3}) as

$$r_e = (3L/4 \pi\rho K N_{tot})^{1/3} \qquad (3)$$

where L is the cloud liquid water content and r, the density of water. The value of the constant K depends on whether the cloud is maritime ($K = 0.80$) or continental ($K = 0.67$). This can be further related to the sulphate aerosol concentrations, A (cm^{-3}) based on aircraft observations from a wide range of locations (Martin *et al.* 1994).

$$N_{tot} = 375\ [1\text{-}\exp(-2.5 \times 10^{-3} A)] \qquad (4)$$

This equation was derived by Jones *et al.* (1994) as a single continuous function of both maritime and continental aerosol concentrations based on 110 pairs of aircraft observations.

The relationship between effective radius (r_e) and droplet concentrations (cm^{-3}) is shown in Fig.2. The effective radius (r_e) changes more appreciably with droplet concentration when the mean droplet concentration is small.

From Eqns. (1-4), it is evident that an increase in aerosol concentration would lead to an increase in droplet concentration (Eqn.4) resulting in a smaller cloud droplet effective radius r_e (Eqn.3). Smaller cloud droplet effective radius r_e would ultimately lead to higher cloud optical depth at a constant liquid water path (Eqn.1).

Another relation between cloud water sulphate and N_{tot} deduced from data taken over North America derived by Leaitch *et al.* (1992) was also used for calculations. This equation is

$$\log_{10}(N_{tot}) = 0.257\ (\pm 0.052)\ \log_{10}(0.122\ A) + 1.95\ (\pm 0.21) \qquad (5)$$

The numbers in parentheses are the standard errors.

In this equation N_{tot} was limited to ensure that $N_{tot} \leq A$, at low values of A.

In this study, an assumption was made that clouds over land are continental and those elsewhere are maritime. For deep convective clouds the value of r_e used was 9.5m for continental clouds and 13.5m for maritime clouds as done by Jones *et al.* (1994).

For radiative transfer calculations both diurnal and seasonal cycles of solar radiation were taken into account. 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 Atmospheric General Circulation Model (AGCM). This simulation is the controlled simulation performed as a participant in the Atmospheric Model Inter-comparison Project (AMIP).

The indirect forcing [change in top of atmosphere (TOA) net radiation] was estimated by performing radiative transfer calculations with only natural aerosol concentrations and with the total (industrial + pre-industrial) aerosol concentrations. The difference between these two calculations gives the indirect radiative forcing due to anthropogenic aerosols.

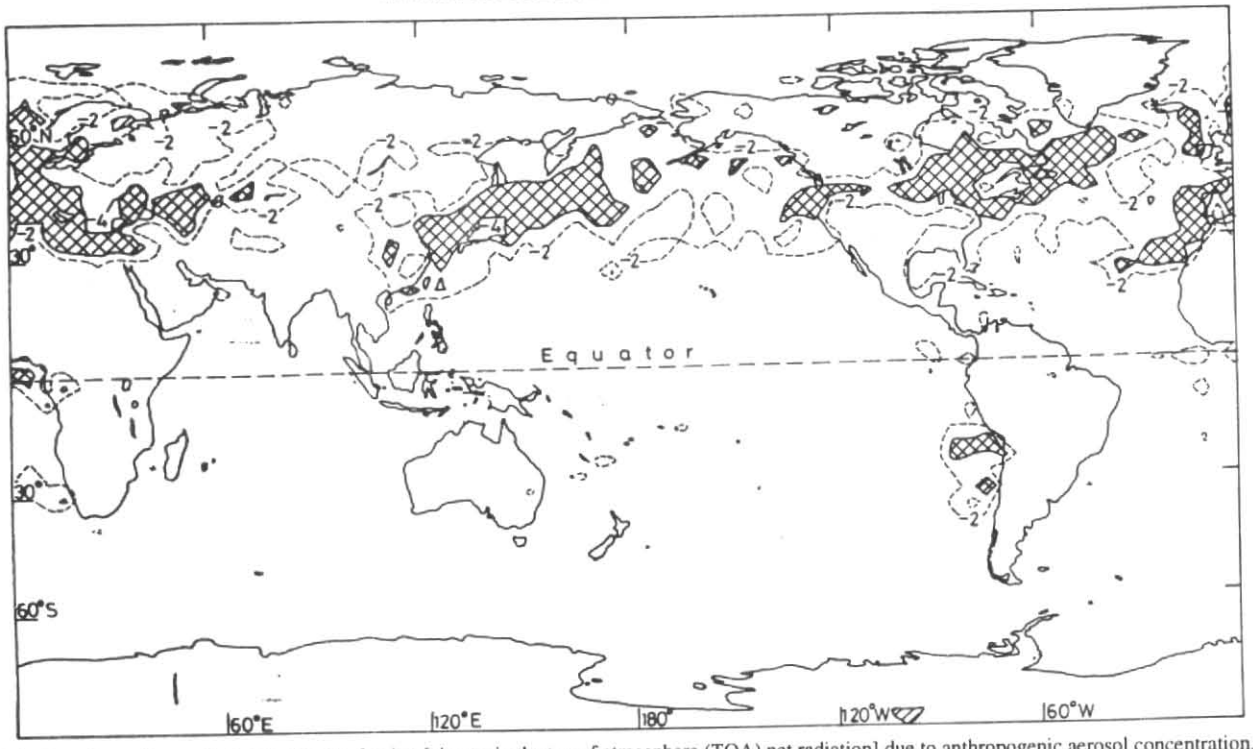

Fig.3. Annual mean indirect radiative forcing [change in the top of atmosphere (TOA) net radiation] due to anthropogenic aerosol concentration. Units : Wm^{-2}. Contour interval : 2 Wm^{-2}. Areas of Negative forcing larger than -3.5 Wm^{-2} are shaded.

4. Results and discussion

4.1. *Indirect climate forcing*

The calculated values of the low cloud r_e are comparable with the observed values published by Han *et al.* (1994). The annual mean spatial distributions of low cloud r_e in case of natural aerosol and total aerosol concentration (not shown here) revealed that generally there is a decrease of low cloud r_e throughout most of the NH due to anthropogenic aerosols in accordance with Eqns. (3 and 4). This decrease is most pronounced near the major industrial regions which produce the largest amounts of aerosols.

The annual mean change in the top of the atmosphere (TOA) net radiation due to the indirect effect of anthropogenic sulphate aerosols is shown in Fig.3. The cooling due to enhanced cloud albedo is the greatest near polluted regions of Europe, North America and China. These maximum changes in net radiation occur due to large changes in low cloud r_e and due to sufficient low clouds which are unobscured by higher level clouds.

The annual mean anthropogenic indirect forcing is estimated as - 1.13 Wm^{-2}. In NH there are some regions where the negative forcing even exceeds -4 Wm^{-2}. The mean NH forcing (-1.59 Wm^{-2}) is larger compared with - 0.69 Wm^{-2} in the southern hemisphere (SH). This is due to the reason that more anthropogenic aerosols are located in the NH. The global mean value of -1.13 Wm^{-2} is comparable with the simple estimates of Charlson *et al.* (1992) and Schelesinger

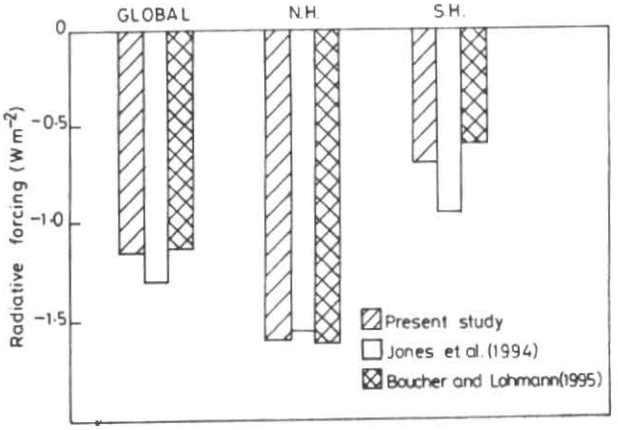

Fig.4. Comparison of the calculated annual mean indirect radiative forcing (Wm^{-2}) with the results of Jones *et al.* (1994) and Boucher and Lohmann (1995)

et al. (1992) and the recent General Circulation Model (GCM) estimates of Jones *et al.* (1994) and Boucher and Lohmann (1995). The comparison of the present estimates with those of Jones *et al.* (1994) and Boucher and Lohmann (1995) is shown in Fig.4. The estimate of Jones *et al.* (1994) for SH is larger than the current estimate. This difference in the estimates may be due to different sets of cloud cover and cloud liquid water content distributions used for the calculations. In the present calculations the forcing in the Southern Hemisphere is 43% of NH value.

It is interesting to note that there is appreciable contribution to the globally averaged forcing from SH even with

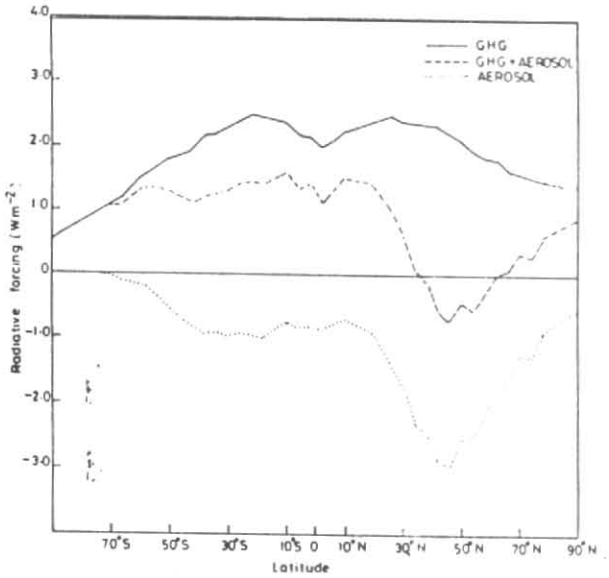

Fig.5. Zonal variation of the positive radiative forcing due to the increase in the greenhouse gases, negative radiative forcing due to the indirect effect of anthropogenic aerosol and the net forcing. Units : Wm^{-2}

the modest aerosol loading. This may be because the indirect forcing depends also on the susceptibility of the clouds to the changes in aerosol concentrations (Platnick and Twomey 1994). Cloud susceptibility is defined as the derivative of cloud albedo with respect to cloud droplet number concentration for a given liquid water content. Clouds of SH are more susceptible than those of NH because the cloud droplet concentration is smaller in SH. It can be noted from Fig.3 that there is appreciable contribution from the oceanic regions affected by the anthropogenic aerosols. The seasonal variation of the radiative forcing is shown in Table 2. The forcing is maximum during NH summer (June-July August, JJA) due to the seasonal cycle of the insolation in NH.

With the parameterizations of Eqn. (5), based on observations of Leaitch *et al.* (1992) the forcing was estimated to be -1.26 Wm^{-2}, and 70 % of the forcing is contributed from NH.

This indirect forcing due to anthropogenic aerosol is substantially larger than the direct radiative effect of -0.3 Wm^{-2} β estimated by Kiehl and Briegleb (1993) using the same aerosol distribution. However, as in the direct effect the indirect forcing is mainly concentrated in NH where maximum anthropogenic aerosols reside. This cooling, moreover, is relatively localized which has implications for the regional radiative budget.

To compare this indirect forcing with the warming effect due to greenhouse gases, the radiative forcing due to increase in greenhouse gases was calculated using the same three dimensional radiative transfer model which considered the increase in greenhouse gases from pre-industrial period to the present. These values are those defined by the Intergovernmental Panel on Climate Change (IPCC). These values are given in Table 3.

TABLE 2
Seasonal variations of the Indirect Forcing (Units : Wm^{-2})

S.No.	Region	DJF	JJA
1.	Global	-1.146	-1.211
2.	Northern hemisphere	-1.423	-1.848
3.	Southern hemisphere	-0.869	-0.574

DJF : December - January - February
JJA : June - July - August

TABLE 3
Concentrations of greenhouse gases during pre-industrial period and the present (IPCC 1990) in ppmv

Greenhouse gas	Pre-industrial period	Present
CO_2	280.000	353.000
CH_4	0.800	1.720
N_2O	0.288	0.310
CFC 11	0.0	$0.280 * 10^{-3}$
CFC 12	0.0	$0.484 * 10^{-3}$

The global annual mean positive forcing due to increase in greenhouse gases from the pre-industrial period to the present is estimated as 2.12 Wm^{-2} which is similar to the previous estimate (Kiehl and Briegleb 1993, Houghton *et al.* 1990). The combined effect of increase in greenhouse gases and the indirect effect of anthropogenic sulphate aerosols revealed that in the northern hemisphere there are regions where the aerosol indirect effect is actually larger than the greenhouse effect. Net negative forcing occurs over eastern parts of the United States, south central Europe and eastern China where there are large negative forcing due to this indirect effect. The zonal variation of aerosol indirect forcing and the net forcing are shown in Fig.5, which clearly indicates pronounced reduction of warming effect due to greenhouse gases over NH mid-latitudes by aerosols. The net forcing over these regions is negative, *i.e.*, a cooling effect.

4.2. *Uncertainties in the radiative forcing*

There are some uncertainties in the estimation of indirect forcing which could lead to a much wider spread of estimates. The possible uncertainties are: (*i*) The sulphate aerosol distribution in the pre-industrial case (*ii*) the dependence of the physical characteristics of the aerosol on indirect forcing (*iii*) the generality of Eqns. (3-4) derived from the aircraft observations (*iv*) the details of sulphur chemistry and (*v*) the dependence of the results on the quality of the GCM cloud simulations.

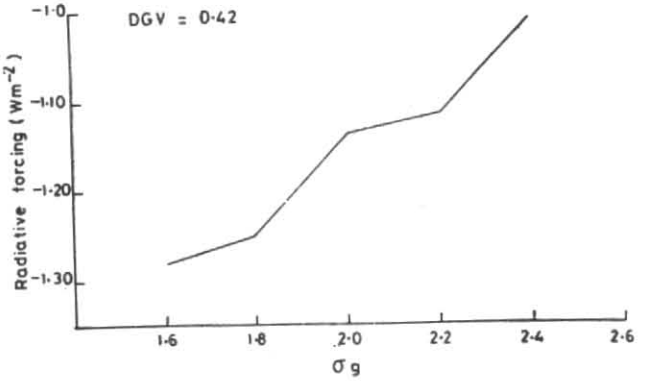

Fig.6. Variation of the radiative forcing with the width of particle size distribution

Attempts have been made to estimate some of the uncertainties mentioned above. For the present calculations, a log-normal size distribution was assumed with a median particle radius of 0.05 m and a geometric standard deviation of 2.0. To examine the dependence of the physical characteristics of the aerosol on indirect forcing, calculations were made of the globally averaged indirect radiative forcing as a function of the width of the particle size distributions (r) for a fixed geometry mean diameter by volume (DGV) 0.42m. The results are shown in Fig.6. Unlike in the case of the direct forcing, the variation of the indirect radiative forcing with the width of particle size distribution is non-linear. For example a 10 % decrease of width from 2.0 to 1.8 would lead to 10 % decrease in the forcing while a similar amount in increase of the width from 2.0 to 2.2 would lead only to 2 % increase in the forcing.

The average error in the estimate of the cloud droplet concentration obtained using the parameterization between N_{tot} and aerosol concentration A (Eqn. 4) is $\pm$ 45 cm^{-3} (Martin *et al.* 1994). Similarly for the parameterization of Leaitch *et al.* (1992), the standard errors are given in the parantheses of Eqn.5. The changes of the radiative forcing for these standard errors have been determined and the results are shown in Table 4. For the data of Martin *et al.* (1994), the percentage changes in the annually averaged global forcing is 46 % (53 %) for increase (decrease) of N_{tot} by one standard error. For Leaitch *et al.* (1992) case these values are 27 % and 46 % respectively. The sensitivity is higher for decrease than increase in the cloud droplet concentration. If the estimated uncertainty due to errors in cloud droplet concentration and changes in width of particle size (σ_g) are taken together, then the estimated radiative forcing varies from -0.471 to 1.673 Wm^{-2}.

5. Conclusions

The indirect radiative forcing (through the enhancement of cloud albedo) due to anthropogenic sulphate aerosols was calculated using a three-dimensional radiative transfer model.

TABLE 4

Percentage changes in the annually averaged radiative forcing (Wm^{-2}) for increase/decrease of 1 Standard error (SE) in Eqns(3) & (4)

S.No.	Region	Martin *et al.* data Eqn(3)		Leaitch *et al.* data Eqn(4)	
		+SE	-SE	+SE	-SE
1.	Northern hemisphere	31	52	36	56
2.	Southern hemisphere	60	55	18	37
3.	Global	46	53	27	46

In this study only sulphate aerosols are considered as anthropogenic aerosols. We can expect the similar kind of effect (although with smaller magnitude) due to other anthropogenic aerosols, like soot particles due to biomass burning (Penner *et al.* 1992 & 1994). A better understanding of the processes governing aerosol growth and droplet nucleation is needed. More measurements of sulphate and other anthropogenic aerosol mass and cloud droplet number concentration and more comprehensive modeling approaches are further required. Further calculations of these effects are needed with observed cloud fields (obvious choice is to make use of the satellite (derived cloud fields) instead of the model derived cloud fields. It is, however, essential that the climate models should incorporate schemes for the prediction of sources, sinks and advection of aerosols, so that the aerosol-cloud-climate interaction and associated feedback mechanisms can be treated more accurately.

Acknowledgements

The author is thankful to Prof. R.N. Keshavamurty for his guidance and keen interest in this study and to Prof. M.E. Schlesinger, University of Illinois, USA for providing the Atmospheric General circulation Model data and radiative transfer model and to Dr. J. Langner, Swedish Meteorological Office, Stockholm, Sweden for supplying the sulphate aerosol concentration data. The author is also thankful to the referee for his valuable comments. He is also thankful to Smt. N.S. Satbhai for typing the paper.

APPENDIX - A

Log-normal distribution

The sizes of the aerosol particle are represented by a log-normal function,

$$\frac{dN}{d(ln\ r)} = C \exp(-p)\ N1$$

where $C = 1.0/[(2\pi)^{1/2} \ln\sigma_g]$

and $P = 1/2 [\ln (r/r_n) / \ln \sigma_g]^2$

N l is the total number of particles per cubic meter in the accumulation mode, r_n is the geometric mean radius and σ_g is the standard deviation.

For the log-normal distribution the distribution, is normal with respect to ln(r) so that 95% of the particles fall within a size range defined by exp [ln (CMD) ±2 ln σ_g].CMD is the count median diameter. This range is asymmetrical and goes from CMD/σg^2 to CMDσg^2.

To convert the count mean diameter (CMD) to the mass mean diameter (MMD) or geometric mean diameter by volume (DGV) of the distribution, the following expression is used.

$$MMD = DGV = CMD \exp (3.5 \ln^2 \sigma_g)$$

Similarly to convert the count median diameter (CMD) to the particle with average mass (that size particle whose mass multiplied by the total number of particles gives the total mass) the following expression is used.

$$dm = CMD \exp (1.5 \ln^2 \sigma_g)$$

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