

Computation of anthropogenic sulphate aerosol forcing using radiative perturbation theory

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ABSTRACT

The radiative forcing of the climate by anthropogenic aerosols has been a matter of some concern for many years now, especially in the northern hemisphere. Recently in these pages, Charlson et al. attempted to quantify this forcing. However, that calculation involved relatively crude optical and radiative transfer models. In this paper, we use a far more detailed sulphate optical model, and employ radiative perturbation theory (a technique ideally suited to answering questions of this sort) to repeat this radiation calculation. We obtain results which are similar to Charlson et al., provided that proper allowance is made for the effects of humidity.

1. Introduction

In a recent article in these pages, Charlson et al. (1991) suggested that one reason that the global warming signal is not as evident in the northern hemisphere as it is in the southern, (see, Jones, 1988) may be the presence of a significant sulphate aerosol layer there, due to the much greater industrial activity in that hemisphere. (Northern hemisphere temperatures dropped considerably between 1940 and 1970 (Jones, 1988) a period of rapid industrialization.) This concept has been addressed by other workers over the years (see for example, Wigley, 1991; Grassl, 1988; Bolin and Charlson, 1976). However, Charlson et al. (1991, 1992) appear to have gone further than most, in attempting to quantify this sulphate aerosol forcing.

The calculation they performed can be divided into 3 parts: an assessment of the hemispheric burden of sulphate aerosol based on flux estimates and residence times, the conversion of this mass burden into an optical model (i.e., extinction, scattering albedo, and asymmetry factor or back-

scattered fraction), and an estimate of the radiative effects of this model. In this paper, we address the 2nd and 3rd of these steps, by performing a full radiative calculation of the change in surface flux, or forcing, due to an appropriate aerosol optical model, based on radiative perturbation theory.

In Section 2, we summarise the radiative model used by Charlson et al. (1991), and extract their result in a form suitable for comparison with ours. In Section 3, we outline the ideas of our radiative perturbation technique, before discussing the sulphate aerosol model. We obtain results (for 6 different solar zenith angles), ranging between 40% and 50% of those of Charlson et al. (1991) for dry aerosol, but which are in substantial agreement once the effects of humidity are carefully included.

2. Charlson's model

Charlson et al.'s (1991) radiative model may be summarised quite simply as follows:

$$\overline{\Delta F_R} = \frac{1}{2} Q_0 \overline{T_i^2} (1 - \overline{A_c}) (1 - \overline{R_s})^2 \overline{\beta \delta}, \quad (1)$$

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where

ΔF_R is the reduction in surface insolation

$\frac{1}{2}$ allows for day-night averaging

$Q_0 = 1370 \text{ W m}^{-2}$ is the solar constant

T_i is the atmospheric transmissivity

$A_c \approx 0.61$ is the cloud cover fraction

$R_s \approx 0.15$ is the surface albedo

β is the backscattered (actually upscattered) fraction (Wiscombe and Grams, 1976)

δ is the anthropogenic sulphate aerosol optical thickness.

In all cases, the over-bar denotes an appropriate averaging, over the hemisphere, or solar elevation and over wavelength. (If required, a further factor to allow for variations in humidity may be included: see Section 4.)

In our work, we have concerned ourselves with clear-sky forcing only, and so we have ignored the effects of cloud cover, as well as day-night averaging, and surface reflection, regarding these as correction factors to be added later. With these factors removed, the expression with which we wish to compare our results is

$$\overline{\Delta F_R} = Q_0 \overline{T_i^2} \bar{\beta} \delta. \quad (1')$$

Charlson et al. (1991) chose a value of 0.76 for the atmospheric transmittance, and 0.29 for the backscattered fraction, so that their result is

$$\begin{aligned} \overline{\Delta F_R} &= 1370 \times 0.76^2 \times 0.29 \delta \\ &= 230 \delta. \end{aligned} \quad (2)$$

3. Radiative perturbation theory

The radiative perturbation technique (Box et al., 1989a, b) was recently borrowed from the neutron transport literature (Bell and Glasstone, 1970; Gerstl and Stacey, 1973), as the transport processes have much in common, especially mathematically. In this approach, one first solves the radiative transfer equation, and the adjoint equation (Box et al., 1988), corresponding to a specified base model of the atmosphere. The goal is to obtain a specified radiative effect, such as the flux at the ground.

Then one asks questions such as "how would this effect change, if the atmospheric model were to be changed?" The answer, at least to first order in perturbation, is that the change in surface flux (the effect of interest in this work) is given by

$$\Delta F = \langle I_0^+, \Delta L I_0 \rangle,$$

where

I_0 is the base model intensity function,

I_0^+ is the base model adjoint intensity (Box et al., 1988),

ΔL is the perturbation to the atmospheric model and angular brackets denote integration over angles and height, plus wavelength in this case.

Recently, radiative perturbation theory has been applied in a large-scale study of the effects of different levels and types of aerosols on atmospheric heating rates and surface fluxes (Trautmann et al., 1992). The results presented here should be seen as an extension of that work. Full details of our atmospheric base model may be found in that paper.

In our extended study, we made use of a set of aerosol models proposed by the WMO (Lenoble, 1985; Shettle and Fenn, 1979). With the exception of the background and volcanic stratosphere models, these models comprise a mixture of components, each with its corresponding size distribution and optical properties (Lenoble, 1985).

In the present study, as in our earlier work, we have taken the WMO continental model as our base model. This model is comprised of three aerosol types, each with its own size distribution and refractive index at each wavelength. The three components, with their % by volume and size distribution parameters (see, eq. (3)) are: water soluble (29%, $r_m = 0.005$, $\sigma = 0.475$), dust-like (70%, $r_m = 0.5$, $\sigma = 0.475$) and soot (1%, $r_m = 0.0118$, $\sigma = 0.301$). Its optical properties are dominated by the large, dust-like mode, especially at longer wavelengths. For the perturbing aerosol, we took optical data for a sulphate aerosol model from Tables 6–10 of d'Almeida et al. (1991). This model is discussed more fully in the next section.

The variation of the optical properties (extinction coefficient, single scattering albedo and asymmetry factor) with wavelength may now be determined using Mie theory (Bohren and Huffman,

1983). (Note that we have, in fact, used the Henyey-Greenstein (1941) phase function in all radiative transfer calculations.) Overall normalization is set by choosing the optical thickness at the reference wavelength of $0.55 \mu\text{m}$. The continental model has an optical thickness of 0.225 (0.2 below 2 km, and 0.025 between 2 and 12 km). In addition, there is a background stratospheric aerosol of optical thickness 0.003. (The effect of the choice of base case model on the final perturbation result is usually negligible.)

Calculations were performed at 23 wavelengths over the spectral region 0.3 to $4.0 \mu\text{m}$, as described in Trautmann et al. (1992), with a preponderance at the shorter wavelength end. We used 24 stream Gaussian quadrature for the angular integrations, and selected 6 solar zenith angles (being alternate downward quadrature streams). Note that a perturbation of the type considered here is linear in the amount of additional aerosol inserted (as measured, for example, by the optical thickness at $0.55 \mu\text{m}$). This is in accord with eq. (2). We may thus define the perturbation factor as the actual perturbation divided by the optical thickness at a chosen wavelength, in our case $0.55 \mu\text{m}$. Note that we have not used a separate symbol here, as it will be the perturbation factor on which we shall focus in this work.

4. The sulphate aerosol model

As indicated above, we took our optical data on sulphate aerosols from the recent compilation of d'Almeida et al. (1991). In this section, we describe the model involved, and give some justification for its use.

The analytic form of this model is lognormal:

$$n(r) \sim \frac{1}{r} \exp \left\{ -\frac{1}{2} \left[\frac{\log(r/r_m)}{\sigma} \right]^2 \right\}, \quad (3)$$

where the normalization may be determined as required. For the sulphate model, d'Almeida et al. (1991) have selected $r_m = 0.0695 \mu\text{m}$, and $\sigma = 0.307$.

This function is plotted (with arbitrary normalization) in Fig. 1, and is seen to be a fairly typical accumulation-mode size distribution, suitable for an aerosol which forms by gas-to-particle conversion (Whitby, 1978; Hoppel, 1982). It has a

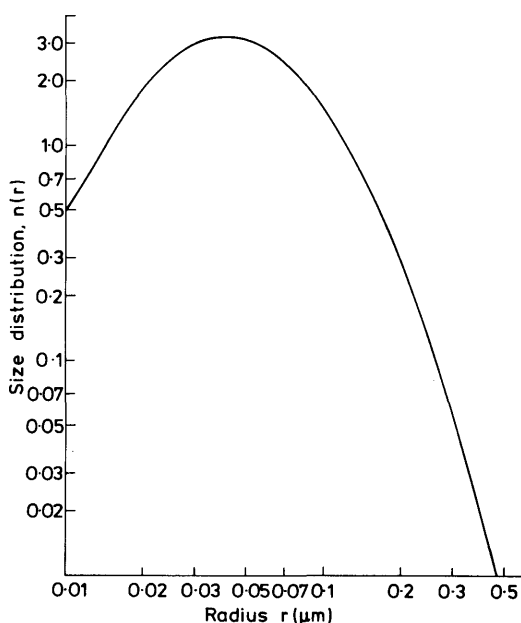


Fig. 1. The sulphate aerosol size distribution for dry particles (arbitrary normalization).

geometric mean radius by volume, $\bar{r}$, which is its third moment, divided by its second moment, of

$$\begin{aligned} \bar{r} &= r_m \exp[2.5(\sigma \ln 10)^2] \\ &= 0.25 \mu\text{m}. \end{aligned}$$

In Fig. 2, we plot the extinction coefficient (normalized to 1.0 at $\lambda = 0.55 \mu\text{m}$), single scattering

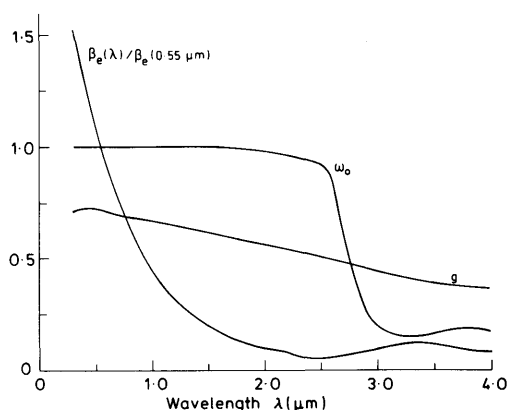


Fig. 2. Normalized extinction coefficient, single scattering albedo, and asymmetry factor, as functions of wavelength, for the (dry) sulphate aerosol model.

Table 1. *Humidity effects on the sulphate aerosol optical model*

Humidity (%)	Growth factor, G	Mean radius $\bar{r}$ (μm)	Specific extinction, ψ ($\text{m}^2 \text{g}^{-1}$)		
			$\lambda = 0.55 \mu\text{m}$	$1.06 \mu\text{m}$	$2.0 \mu\text{m}$
0	1.0	0.25	3.0	1.2	0.3
50	1.2	0.30	4.6	2.0	0.5
70	1.3	0.32	5.6	2.6	0.7
80	1.4	0.35	6.7	3.2	0.9
90	1.65	0.41	10.2	5.3	1.7
95	2.0	0.50	16.2	9.6	3.5
98	2.5	0.62	27.2	18.5	7.7
99	3.0	0.75	40.5	30.6	14.5

albedo, and asymmetry factor against wavelength. Similar plots for the continental aerosol may be found in Trautmann et al. (1992).

Sulphate aerosols are quite hygroscopic, so that their optical properties may be strongly modified in a high humidity atmosphere. Following on the pioneering work of Hanel (1976), d'Almeida et al. (1991) have presented results for the equilibrium size of particles of various compositions, including $(\text{NH}_4)_2\text{SO}_4$, which we take to be typical of the sulphate aerosol. A range of dry sizes is considered, from $0.01 \mu\text{m}$ to $20.0 \mu\text{m}$, plus a range of humidities to 99%. These results show that, with the exception of the very smallest particles (whose optical contribution are negligible), particles of all sizes grow by almost the same factor, which we call the growth factor, G . We can thus allow for the effects of G , i.e., of humidity, on the size distribution by simply replacing r_m by Gr_m . In Table 1 we list G for a series of relative humidities, based on the data in Table 6.3 of d'Almeida et al. (1991).

In addition to the change in volume, there is a change in refractive index, to reflect the aerosol/water mix. Using our assumption of equivalent growth of all particles, the relative proportions of aerosol and water are the same, and we may express the effective refractive index as (cf. eq. (6.3) of d'Almeida et al. (1991))

$$n = n_w + (n_d - n_w)/G^3, \quad (4)$$

where n_d is the refractive index of dry aerosol, and n_w that of water.

We have re-computed the optical properties of the sulphate aerosol (extinction coefficient, asymmetry factor and single scattering albedo) as a function of wavelength, for all relative humidities

indicated in Table 1, using the modified size distribution and refractive index. The simplest measure of the effects of humidity is the change to the extinction at the wavelength of $0.55 \mu\text{m}$. In Table 1 we have included the specific extinction (Kiehl and Briegleb, 1993) at three wavelengths: $0.55 \mu\text{m}$, $1.06 \mu\text{m}$, and $2.0 \mu\text{m}$. Note that the enhancement effects of humidity are not uniform, but are in fact greater at longer wavelengths.

The effects on single scattering albedo are relatively small for all wavelengths to $2.5 \mu\text{m}$, but start to become relatively large beyond $3.0 \mu\text{m}$. The single scattering albedo shows a modest increase with increasing particle size, as would be expected, being around 10% for typical humidities, and wavelengths to around $2.0 \mu\text{m}$. For the larger wavelengths the increase is greater.

5. Results and discussion

The sulphate perturbation for dry sulphate aerosol was integrated over the solar spectrum to give the final result for each solar zenith angle. The results are presented in Table 2. Here, F_0 is the base case surface flux, ΔF_0 is the perturbation factor corresponding to the base case (continental) boundary layer aerosol (cf. Trautmann et al., 1992) while ΔF_1 is the perturbation factor due to our assumed sulphate aerosol. These numbers should be compared with the coefficient of δ in eq. (2), i.e., with 230.

From these results we see that perturbation with the continental (base model) aerosol gives results in very good agreement with Charlson et al. (1991), whereas our results for the sulphate pertur-

Table 2. *Surface flux perturbation factors*

	5.6°	20.2°	34.9°	49.6°	64.3°	79.0°
F_0	1214	1138	979	749	466	165
ΔF_0	250	254	260	266	258	174
ΔF_1 (0%)	86	90	98	111	121	92
ΔF_1 (50%)	136	143	156	176	194	150
ΔF_1 (70%)	168	177	193	219	241	188
ΔF_1 (80%)	205	216	235	267	295	232
ΔF_1 (90%)	320	336	367	418	465	372
ΔF_1 (95%)	536	564	616	705	790	646
ΔF_1 (98%)	959	1009	1105	1268	1434	1204
ΔF_1 (99%)	1516	1597	1750	2012	2289	1962

bation are smaller by a factor of at least 2. These differences must clearly be related to differences in the two aerosol models, which require investigation.

As can be seen in both Figs. 2, 3, the sulphate extinction coefficient shows considerable variation with wavelength, so that the value at $0.55 \mu\text{m}$ is clearly not representative of the whole of the solar spectrum. In the Charlson et al. (1991) calculation, they were required to adopt a single, average value for δ , and we regard this as the major deficiency of their approach. Our decision to identify this average with the value at $\lambda = 0.55 \mu\text{m}$ (in line with the practice for the WMO models) is thus open to question.

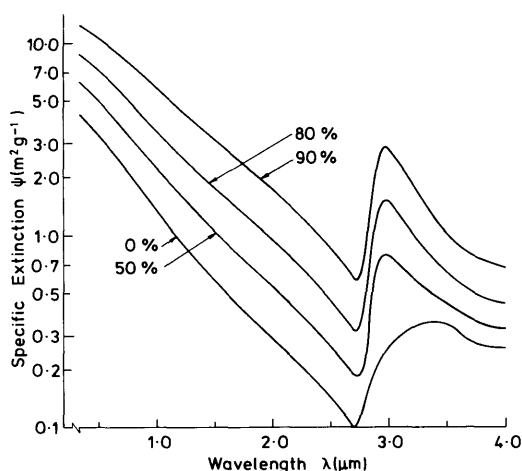


Fig. 3. Specific extinction versus wavelength for the sulphate aerosol model, for relative humidities of 0%, 50%, 80% and 90%.

So far, our results have not included the effects of humidity. As already indicated, we have computed the optical properties of our sulphate aerosol model for a series of relative humidities. Since the primary effect of humidity is on the extinction, and our perturbation technique is linear in this quantity, we have rescaled the results for dry sulphate, at each wavelength, and then simply repeated the final spectral integral. Although this ignores changes to the asymmetry factor and single scattering albedo, we believe that these effects are almost certainly smaller than the remaining uncertainties in these calculations. The full set of results, for all relative humidities, is given in Table 2. Note that for a relative humidity of 70% to 80%, as suggested by Charlson et al. (1991), our results are now in close agreement with theirs. Remember that these results must be multiplied by the sulphate optical depth at $0.55 \mu\text{m}$ for dry aerosol. If this value is obtained from direct measurements, then it must be corrected for humidity, using the results in Table 1.

Due to the large spectral variation in sulphate extinction, it is essential that any optical thickness value be quoted at a specified wavelength. In addition, if this is a measured value, then the relative humidity is also required. These problems may be avoided if the column burden of sulphate (natural, anthropogenic, or total) is available. Charlson et al. (1991) estimate the anthropogenic global average burden to be $3.3 \times 10^{-3} \text{ g m}^{-2}$. For our sulphate model this translates to an optical thickness at $\lambda = 0.55 \mu\text{m}$ of 0.01 for dry aerosol, 0.02 for relative humidities of around 70% to 80%. Since the vast majority of anthropogenic sulphate is found in the northern hemisphere, these results should probably be doubled. If we do this, we obtain a northern hemisphere clear-sky forcing of $8\text{--}10 \text{ W m}^{-2}$. Day-night averaging, plus the effects of surface reflection, reduce this to $2.9\text{--}3.6 \text{ W m}^{-2}$. When averaged over both clear and cloudy regions of the hemisphere, the final result becomes $1.1\text{--}1.4 \text{ W m}^{-2}$.

5. Conclusion

It has been argued by many authors over the years that anthropogenic aerosols are likely to have a cooling effect on the earth's surface temperature, by reflecting sunlight back to space.

In order to quantify that forcing, we need to complete a 3 stage calculation: we need to calculate (estimate) the mass loading of anthropogenic aerosols (Langner et al., 1992); this must be converted to an optical model; and then the radiative effects of this optical model must be evaluated. In this paper we have concentrated on the second and third of these stages, to determine the forcing produced per unit of model aerosol.

We have found that the perturbation factor due to an additional aerosol loading with dry sulphate aerosols, as estimated by Charlson et al. (1991) is too large, by a factor of between 2 and 3. The main reason for this is that the spectral dependence of the aerosol optical properties is ignored in the method used by Charlson et al. (1991). However, when the effects of humidity were taken into account, our final results are very close to those of Charlson et al. (1991).

This is in sharp contrast to the very recent calculations of Kiehl and Briegleb (1993), despite

their use of a fully realistic sulphate optical model, which appears quite similar to ours. However, one key difference is in their handling of the effects of humidity, where they use a single factor to rescale the specific extinction. Our calculation, outlined in Section 4 above, show that this rescaling is itself wavelength-dependent, as can best be seen from the numbers quoted in Table 1. Although we believe that our results are the most accurate to date, it is clear that subtle details of the sulphate aerosol model, including the effects of humidity, can have a significant effect on the final result.

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