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MICROPHYSICAL AND COMPOSITIONAL INFLUENCES ON SHORTWAVE RADIATIVE FORCING OF CLIMATE BY SULFATE AEROSOLS

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ABSTRACT

Anthropogenic sulfate aerosols scatter shortwave (solar) radiation incident upon the atmosphere, thereby exerting a cooling influence on climate relative to pre-industrial times. Previous estimates of this forcing place its global and annual average value at about -1 W m^{-2}, uncertain to a factor of somewhat more than 2, comparable in magnitude to greenhouse gas forcing over the same period but opposite in sign and much more uncertain. Key sources of uncertainty are atmospheric chemistry factors (yield, residence time), and microphysical factors (scattering efficiency, upscatter fraction, and the dependence of these quantities on particle size and relative humidity, RH). This paper examines these microphysical influences to identify properties required to obtain more a accurate description of this forcing. The mass scattering efficiency exhibits a maximum at a particle diameter (~0.5 μm) roughly equal to the wavelength of maximum power in the solar spectrum and roughly equal to diameter typical of anthropogenic sulfate aerosols. Particle size, and hence mass scattering efficiency, increase with increasing on RH because of accretion of water by deliquescent salt aerosols. For example the scattering efficiency of aqueous $(NH_4)_2SO_4$ (dry radius 0.2 μm) increases from 8 to 80 m^2/g (SO_4^{2-}) as RH increases from 39 to 97%. The sensitivity of forcing to particle dry mass and relative humidity suggest the need to explicitly represent these properties if the sulfate aerosol forcing is to be accurately described in climate models.

EXTENDED ABSTRACT

Anthropogenic sulfate aerosols scatter shortwave (solar) radiation incident upon the atmosphere and thereby exert a cooling influence on climate relative to pre-industrial times. An initial approach to estimating the magnitude of the direct forcing is given by a box-model description of the phenomenon (Charlson et al., 1992; Penner et al., 1994). Although such a model is incapable of describing the geographical distribution of the forcing and of accounting for correlations among controlling variables, it is a useful first approximation, especially as the forcing is linear in aerosol loading for an optically thin aerosol in which single scattering dominates. This model yields for the area-average shortwave forcing $\overline{\Delta F_{\mathrm{R}}}$ resulting from an increase in sulfate aerosol concentration:

$$\overline{\Delta F_{\mathrm{R}}} = -\frac{1}{2} F_{\mathrm{T}}(1-A_{\mathrm{c}})T^2(1-R_{\mathrm{s}})^2\bar{\beta}\alpha^{\mathrm{RH}_r}_{\mathrm{SO}_4^{2-}} f(\mathrm{RH}) Q_{\mathrm{SO}_2} Y_{\mathrm{SO}_4^{2-}} \tau_{\mathrm{SO}_4^{2-}} \Big/ A. \qquad (1)$$

The negative sign denotes that the forcing represents a cooling tendency; the factor of 1/2 is due to only half the planet being illuminated at a given time. The symbols in the equation are defined as follows:

F_{T} is the solar constant i.e., mean solar radiative flux at distance of the Earth to the sun;

A_{c} is the fractional cloud cover in the area of concern; the factor $(1 - A_{\mathrm{c}})$ is introduced because the albedo enhancement is applicable only for cloud free regions;

T is the fraction of incident or scattered light transmitted through the atmosphere above the aerosol layer;

R_{s} is the albedo of the underlying surface; the factor $(1-R_{\mathrm{s}})^2$ takes into account multiple reflection between the surface and the aerosol layer;

$\bar{\beta}$ is the fraction of the radiation scattered upward by the aerosol, averaged over the sunward hemisphere;

$\alpha^{\mathrm{RH}_r}_{\mathrm{SO}_4^{2-}}$ is the light-scattering mass efficiency of sulfate aerosol, i.e., scattering coefficient per sulfate mass, at a reference low relative humidity ($\mathrm{RH}_{\mathrm{r}} = 30\%$);

$f(\mathrm{RH})$ is the relative increase in scattering cross-section at ambient RH;

Q_{SO_2} is the source strength of anthropogenic SO_2;

$Y_{SO_4^{2-}}$ is the fractional yield of emitted SO_2 that reacts to produce sulfate aerosol;

$\tau_{SO_4^{2-}}$ is the mean residence time of sulfate aerosol in the atmosphere; and

A is the area of the geographical region to which the calculation is applied, e.g., the entire Earth or the Northern Hemisphere.

Previous estimates of this forcing, summarized in Table 1, place its global and annual average value at about -1 W m^{-2}, uncertain to a factor of somewhat more than 2 (Charlson et al., 1992). The overall multiplicative uncertainty is evaluated as $f_t = \exp[\Sigma(\log f_i)^2]^{1/2}$, where the f_i are the multiplicative uncertainties estimated for the individual factors in Eq (1). The estimated forcing is comparable in magnitude to radiative forcing by anthropogenic greenhouse gases (~2.5 W m^{-2}; IPCC, 1994) but opposite in sign and much more uncertain. Similar global average results are obtained in calculations based on geographical distributions of these aerosols calculated with chemical transport models which explicitly show the geographical distribution of this forcing, concentrated in the vicinity of industrial regions of the Northern Hemisphere (Charlson et al., 1991; Kiehl and Briegleb, 1993). Key sources of uncertainty are atmospheric chemistry factors (yield, residence time), and microphysical factors (scattering efficiency and upscatter fraction and the dependence of these quantities on particle size and RH). This paper examines these microphysical influences to identify properties that must be known to obtain a more accurate description of this forcing.

Figure 1 shows the dependence of mass scattering efficiency on particle radius, showing the well known maximum at a particle diameter roughly equal to the wavelength. This particle size corresponds more or less to that commonly observed in anthropogenic sulfate aerosols. Particle size is thus a key determinant of light scattering efficiency. It is also well known that particle size, and hence mass scattering efficiency, depend on RH because of accretion of water by deliquescent salt aerosols. Figure 2 shows this dependence for several sulfate species. Figure 2a, showing the dependence exhibited with increasing RH, displays the abrupt increase in size associated with deliquescence, occurring at a RH corresponding to the vapor pressure of water above a saturated solution. As indicated in Figure 2b, as RH decreases, an aerosol particle can

remain as a supersaturated solution to relative humidities well below the deliquescence point. Such behavior is commonly observed in tropospheric aerosols (Rood et al., 1989).
Figure 3 shows the effect of these phenomena in combination, illustrating the dependence of scattering efficiency, defined as having units m^2 per gram of sulfate, upon particle dry radius and RH. The substantial growth with RH should be noted as well as the shift of the maximum to lower dry radius as RH increases. For example, for 83% RH the efficiency reaches a maximum of ~20 $m^2\ g^{-1}$, compared to the value $f(\mathrm{RH}) \times \alpha(\mathrm{RH_r}) = 8.5\ m^2\ g^{-1}$ employed in the evaluation given in Table 1. For a sulfate concentration of 4 $\mu g\ m^{-3}$, representative of the remote, anthropogenically influenced troposphere (mixing ratio 1 $\mathrm{nmol/mol_{air}}$, i.e., 1 ppb), uniformly distributed through 1 km (column burden 4 $mg\ m^{-2}$), the corresponding aerosol optical depth is 0.08.

Figure 4 examines the dependence of upscatter fraction β on particle radius r as a function of the cosine of the solar zenith angle, μ_0. For the sun at the horizon ($\mu_0 = 0$) β is 0.5, independent of r. For the sun higher in the sky, forward scattering by larger particles leads to a decrease in β. Figure 5 couples this information to evaluate the 24-hour average forcing at a representative midlatitude location, normalized to a unit mass of sulfate aerosol ($W\ m^{-2}$ per gram sulfate m^{-2}, or $W\ g^{-1}$), for $(NH_4)_2SO_4$ aerosol at 83% RH. A rather broad maximum (negative) forcing is exhibited for $r \approx 0.2\ \mu m$. For sulfate column burden 4 $mg\ m^{-2}$, the corresponding 24-hour average forcing is ~1.6 $W\ m^{-2}$.

These calculations illustrate the sensitivity of calculated shortwave radiative forcing by sulfate aerosols to composition and microphysical properties. Accurate description of this forcing and its spatial and temporal dependence in models of climate change will require accurate representation of these properties.

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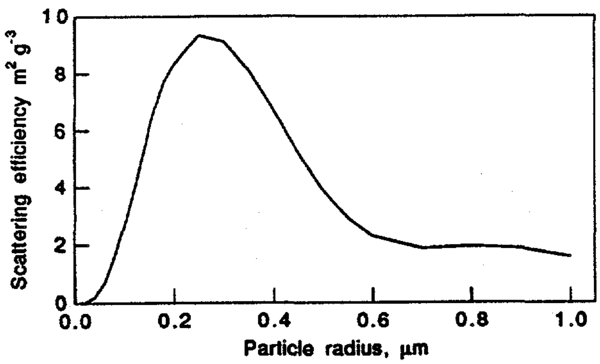

Figure1. Aerosol mass scattering efficiency, i.e., the scattering cross section divided by the mass of the particle, as function of particle radius. Calculation was made for narrow size distribution centered about the radius given on the abscissa. Index of refraction, 1.52-0.008*i*; wavelength, 0.55 µm; density 1 g cm^{-3}.

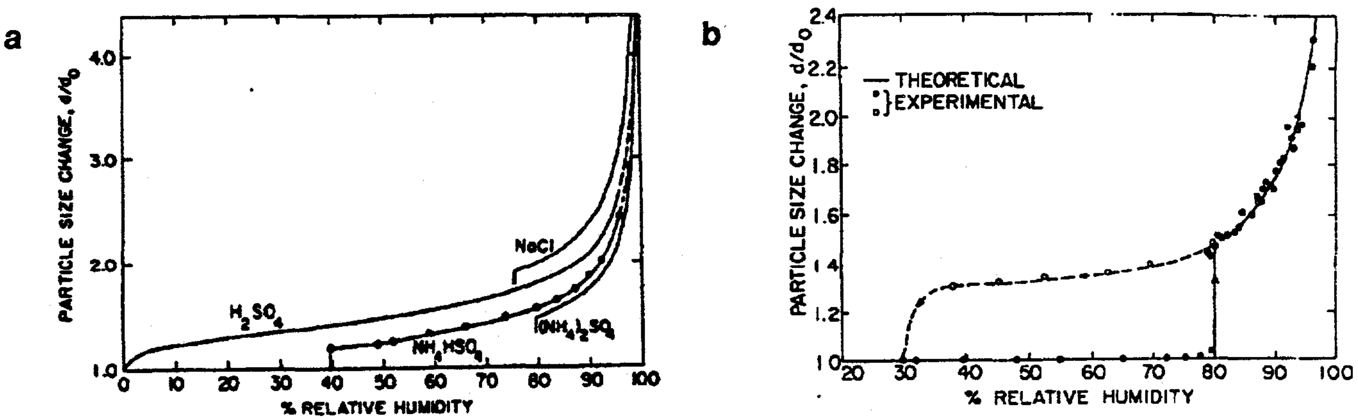

Figure 2. Dependence of aerosol particle radius on relative humidity. a) Theoretical curves showing deliquescence points for several salts. b) Theory and observations for ammonium sulfate, showing supersaturation regime exhibited on decreasing RH below deliquescence point. (Tang and Munkelwitz, 1977; Tang, 1980).

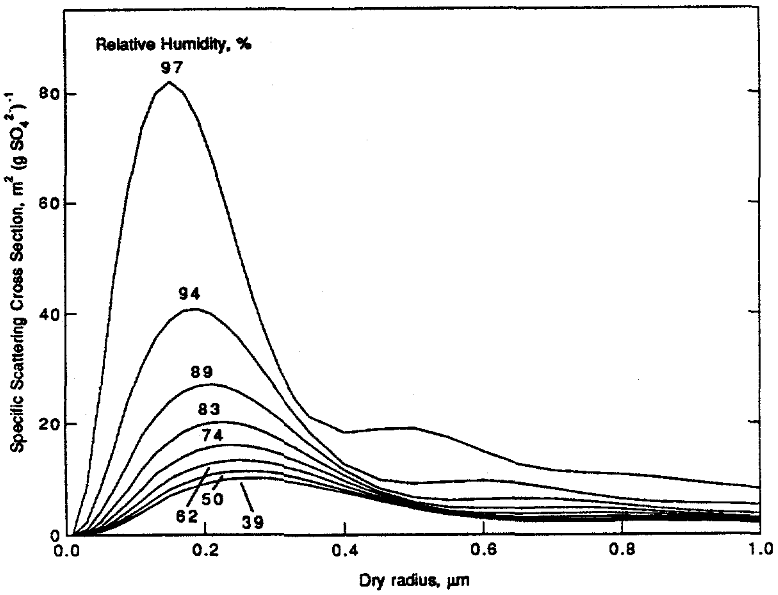

Figure 3. Dependence of mass scattering efficiency of aqueous $(NH_4)_2SO_4^{2-}$ aerosol on particle dry "radius" (evaluated as the radius of the sphere of equal volume) for indicated values of relative humidity. Calculated from density, vapor pressure, and index of refraction data of Tang and Munkelwitz (1994). In order to suppress high frequency resonances a gamma size distribution with a very narrow effective width was employed for each of the particle sizes; wavelength, 0.65 µm.

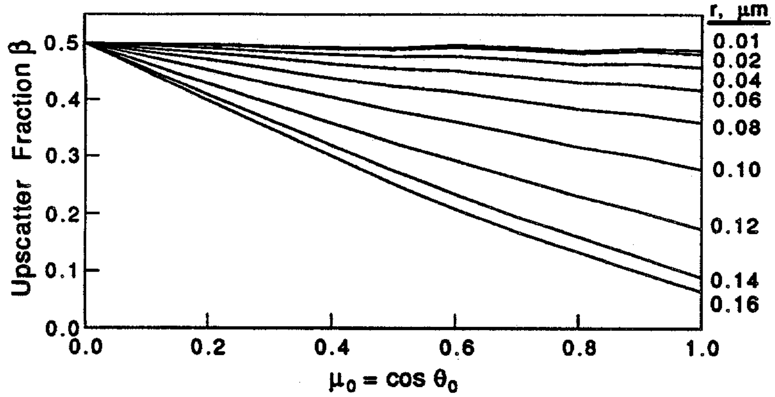

Figure 4. Fraction of incident radiation scattered into the upward hemisphere as a function of cosine of solar zenith angle for indicated values of particle radius. Evaluation is for wavelength 0.55 μm.

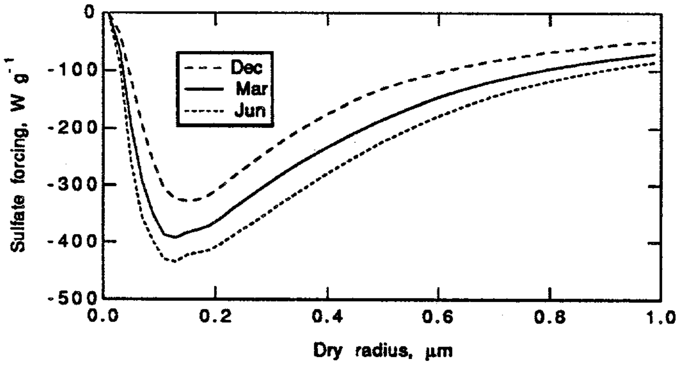

Figure 5. Dependence of normalized shortwave forcing (W m^{-2} per g m^{-2}) of sulfate aerosol on particle size. Normalized forcing represents the 24-hour mean for the 15th of the indicated month for a cloud-free sky, for relative humidity 83%. Latitude 36.7°N and surface albedo 0.11. Single scattering without aerosol absorption is assumed.

TABLE 1. Evaluation of global mean direct radiative forcing due to anthropogenic sulfate aerosol. After Charlson et al.(1992) and Penner et al. (1994).

Quantity	Value	Units	Uncertainty Factor
F_T	1370	W m^{-2}	—
1-A_c	0.4	—	1.1
T	0.76	—	1.15
1-R_s	0.85	—	1.1
$\overline{\beta}$	0.29	—	1.3
$\alpha^{RH_r}_{SO_4^{2-}}$	5	$m^2 (g\ SO_4^{2-})^{-1}$	1.5
f(RH)	1.7	—	1.2
Q_{SO_2}	80	Tg S yr^{-1}	1.15
$Y_{SO_4^{2-}}$	0.4	—	1.5
$\tau_{SO_4^{2-}}$	0.02	yr	1.5
A	5×10^{14}	m^2	—
$\overline{\Delta F_R}$	-1.1	W m^{-2}	2.4