# Direct Shortwave Forcing of Climate by Anthropogenic Sulfate Aerosol: Sensitivity to Particle Size, Composition, and Relative Humidity

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Recent estimates of global- or hemispheric-average forcing of climate by anthropogenic sulfate aerosol due to scattering of shortwave radiation ("direct" effect) are uncertain by somewhat more than a factor of 2. The principal sources of this uncertainty are atmospheric chemistry properties (yield, residence time), and microphysical properties [scattering efficiency, upscatter fraction, and the dependence of these properties on particle size, composition, and relative humidity (RH)]. Among anthropogenic aerosols, much attention has been focused in sulfates because of their importance and relative ease in quantitative evaluation.

Charlson et al. (1992) and Penner et al. (1994) have estimated the magnitude of the direct forcing using a box model as a first approximation. This approximation is reasonable on a global average given that the forcing is linear in aerosol loading in the optically thin limit. The box model readily allows for the examination of uncertainties but cannot account for geographic distribution for which a chemical transport model is required (Charlson et al. 1991). The box model also does not take into account the wavelength dependence on $\alpha*$ [in the present study, the scattering efficiency is evaluated relative to the mass of sulfate in the aerosol and is expressed as $\alpha* = \alpha x\, F(RH)$] and Rayleigh scattering (Kiehl and Briegleb 1993). The area-averaged shortwave forcing by this model for a specific source strength of $SO_2$ (precursor of sulfate formed by atmospheric oxidation of $SO_2$) is

$$\Delta F_{SO_4^{2-}} = -0.5\, F_T (1 - A_c) T^2 (1 - R_s)^2 \overline{\beta} \alpha^{RH_r}_{SO_4^{2-}} \mathcal{F}(RH) \beta_{SO_4^{2-}} \quad (1)$$

The minus sign indicates that this is representative of a cooling effect and the 0.5 is due to the fact that only half the planet is illuminated at any given period of time. The symbols in the above equation are defined as follows:

$F_T$ = the solar constant, the solar radiative flux at a mean distance from the earth to the sun

$A_c$ = the fractional cloud cover in the area of concern. The factor $(1-A_c)$ is introduced because the albedo enhancement is applicable only for cloud free regions.

$T$ = the fraction of incident or scattered light transmitted through the atmosphere above the aerosol layer

$R_s$ = the albedo of the underlying surface. The factor $(1\text{-}R_s)^2$ takes into account multiple reflection between the surface and the aerosol layer.

$\overline{\beta}$ = the fraction of the radiation scattered upward by the aerosol, averaged over the sunward hemisphere.

$\alpha^{RH_r}_{SO_4^{2-}}$ = the light-scattering efficiency of sulfate aerosol, i.e., scattering coefficient per sulfate mass, at a reference low relative humidity ($RH_r$ = 30%)

$\mathcal{F}(RH)$ = the relative increase in scattering cross-section at an ambient RH

$\beta_{SO_4^{2-}}$ = the average $SO_2$ loading (sulfate column mass burden) that is a product of the source strength of anthropogenic $SO_2$, the fractional yield of emitted $SO_2$ that reacts to produce sulfate aerosol, and the mean residence time of $SO_2$ aerosol in the atmosphere divided by the area of the geographical region to which the calculation is applied.

This paper examines the sensitivity of forcing to these microphysical properties for the purpose of obtaining a better understanding of the properties required to reduce the uncertainty in the forcing. To focus on the radiative terms in Equation 1 it is useful to define a normalized forcing for a cloud-free region as

$$\Delta G_{SO_4^{2-}} = \frac{\Delta F_{SO_4^{2-}}}{\beta_{SO_4^{2-}}} \qquad (2)$$

which has units of $Wm^{-2}$ per $g(SO_4^{2-})m^{-2}$, or $Wg(SO_4^{2-})^{-1}$. The relation ($\Delta G$) between aerosol loading and forcing developed here is suitable for comparing modeled and measured aerosol forcing at specific locations and for use in climate models incorporating aerosol forcing provided aerosol microphysical properties are known or assumed.

The scattering efficiency of an ammonium sulfate aerosol at low RH (40%) varies by about a factor of 2 for particle radius 0.1 to 1 μm, with a maximum of $\approx 8 m^2 g^{-1}$ (sulfate) for particle radius 0.25 μm (see Figure 1). The maximum particle mass scattering efficiency increases with

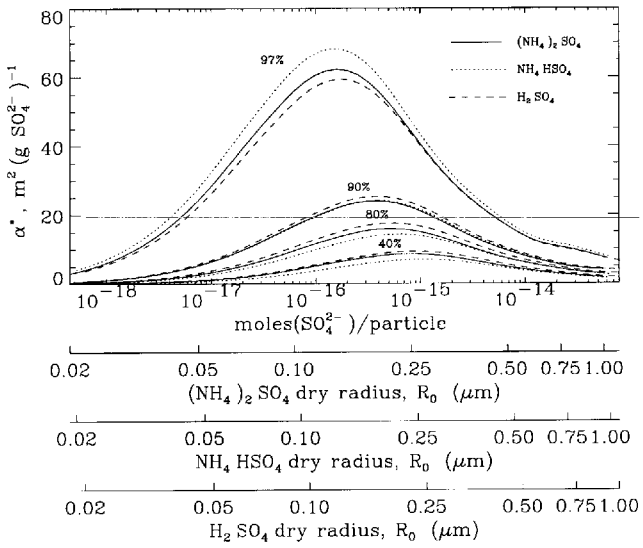


**Figure 1**. Dependence of scattering efficiency, integrated over the solar spectrum, for aqueous $(NH_4)_2SO_4$, $NH_4HSO_4$, and $H_2SO_4$ aerosol on moles per particle of substance for indicated values of relative humidity. Particle dry "radius" (evaluated as the radius of the sphere of equal volume) is also shown as secondary axes.

increasing RH to as great as $70 m^2 g^{-1}$ (Sulfate) at RH=97% because of accretion of water with increasing RH. The upscatter fraction likewise depends markedly on particle radius, with a hemispheric average in the range of 17% to 50%; upscatter fraction also depends strongly on solar zenith angle (SZA), ranging from a small fraction (10% to 30% depending on radius) near zenith to 50% at the limb. Based on the global and annual values given by Charlson et al. (1992), $\Delta G = -715 Wg^{-1}$ for a cloud free planet. Charlson et al. overestimate the normalized forcing by a factor of two (see Figure 2). This factor comes from the fact that a constant $\alpha x F(RH) = 8.5 m^2 g^{-1}$ over the solar spectrum is used. This average value is chosen based on the assumption that Rayleigh scattering is constant throughout the solar spectrum. However, Rayleigh scattering reduces $\Delta G$ more at shorter wavelengths. Thus, the average value should actually be reduced by a factor of about two, placing it in better agreement with the present study. Differences between the results from Kiehl and Briegleb (1993) and this study are primarily due to the fact that although they use the European Centre for Medium-Range Weather Forecasts RH to computer $\alpha*$, they obtain the optical properties of anthropogenic aerosol from the dry particle size distribution that ignores the dependence of RH.

The role of RH is of major importance, especially because of its influence on the scattering efficiency. Seasonal and latitudinal variations in $<\beta>$ can influence $\Delta G$ by as much as a factor of two. However, influences by different particle compositions such as ammonium sulfate,

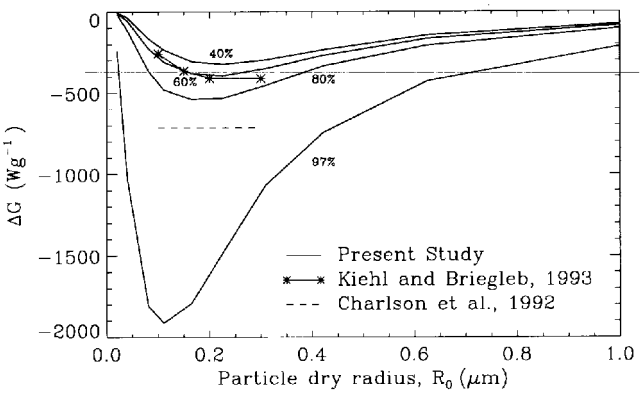


**Figure 2**. Normalized ammonium sulfate aerosol forcing for a hypothetical clear-sky planet as a function of particle dry radius for several RH.

ammonium bisulfate, and sulfuric acid appear to play a more secondary role on the changes in the magnitude of $\Delta G$.

The normalized forcing, $\Delta G$, is a useful quantity for the purpose of focusing solely on the radiative terms that are needed to compute the radiative forcing. $\Delta G$ computed as a function of particle size makes it possible to apply real particle size distributions for the purpose of determining the forcing as a function of variables such as RH. By isolating the influences of the microphysical properties of the aerosol particles from factors such as cloud cover and column mass burden, $\Delta G$ can be used in realistic climate models that use modeled or observed RH.

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