APPENDIX B

A MIE SCATTERING MODEL AND AEROSOL OPTICAL PROPERTIES

In Chapter II and Appendix A, we stated that the radiative effect of the tropospheric sulfate aerosols has been included in the 24–layer ST–GCM. In Chapter III, we used a 1–D radiative–transfer model to estimate the radiative forcing of volcanic aerosol with stratospheric temperature adjustment. The optical properties (specific extinction, single–scattering albedo and asymmetry factor) of these aerosols were prescribed and set to be invariant in time and space. They were pre–computed by using a Mie scattering model we built. In this appendix, we describe the structure of the Mie scattering model and its application in calculating the optical properties of sulfate aerosols. It should be pointed out that the optical properties of the Pinatubo volcanic aerosol were reconstructed separately by using Stenchikov et al. (1998)'s method as described in Chapter III.

1. Optical Properties of Individual Aerosol Particles

The commonly used parameters for characterizing the scattering properties of an individual aerosol particle based on the Mie theory are (Twomey 1977),

extinction efficiency, the ratio between extinction cross section (σ_e) and the cross section area of the particle (πr^2),

$$Q_e = \frac{\sigma_e}{\pi r^2} = \frac{2}{x^2} \sum_{n=1}^{\infty} (2n+1) \mathrm{Re}(a_n + b_n) \quad , \qquad \text{(B.1)}$$

scattering efficiency, the ratio between scattering cross section (σ_s) and the cross section area of the particle (πr^2),

$$Q_s = \frac{\sigma_s}{\pi r^2} = \frac{2}{x^2} \sum_{n=1}^{\infty} (2n+1) \left\{ |a_n|^2 + |b_n|^2 \right\} \quad , \qquad \text{(B.2)}$$

single scattering albedo, the ratio between scattering efficiency and extinction efficiency,

$$\varpi_0 = Q_s / Q_e \quad , \tag{B.3}$$

and asymmetry factor, the average value of the phase function for the scattered radiation,

$$g = \frac{2}{x^2 Q_s} \sum_{n=1}^{\infty} \left\{ \frac{n(n+2)}{n+1} \mathrm{Re}\left(a_n a_{n+1}^* + b_n b_{n+1}^*\right) + \frac{2n+1}{n(n+1)} \mathrm{Re}\left(a_n b_n^*\right) \right\} \quad , \tag{B.4}$$

where
$$a_n = \frac{S_n'(y) S_n(x) - m S_n(y) S_n'(x)}{S_n'(y)\left[S_n(x) + iC_n(x)\right] - m S_n(y)\left[S_n'(x) + iC_n'(x)\right]} \quad ,$$

$$b_n = \frac{m S_n'(y) S_n(x) - S_n(y) S_n'(x)}{m S_n'(y)\left[S_n(x) + iC_n(x)\right] - S_n(y)\left[S_n'(x) + iC_n'(x)\right]} \quad ,$$

where the prime means derivative;

$i = \sqrt{-1}$;

a_n^* and b_n^* are the complex conjugates of a_n and b_n, respectively;

m is refractive index, $m = m_r - i m_i$;

$x = 2\pi r / \lambda$, r radius of the particle, λ wavelength of incident radiation;

$y = mx$;

$S_n(z) = J_{n+1/2}(z)\sqrt{\pi z / 2}$, $J_{n+1/2}$ the first kind of Bessel function;

and $C_n(z) = -N_{n+1/2}(z)\sqrt{\pi z / 2}$, $N_{n+1/2}(z)$ the second kind of Bessel function.

For a given refractive index, particle radius and the wavelength of incident radiation, the above optical properties of an individual aerosol particle can be determined. As an example, we presented in Fig. B–1 the variations of extinction efficiency against particle radius at wavelength $\lambda = 0.55$ µm with six different refractive indices. For example, Case a represents non–absorbing particles and Case f represents highly absorbing particles. The particle radius varies from 0.01 µm to 9.99 µm with an interval of 0.02 µm. Fig. B–1 reproduces the features of Fig. 9.3a in Twomey (1977). It verifies the Mie model we built.

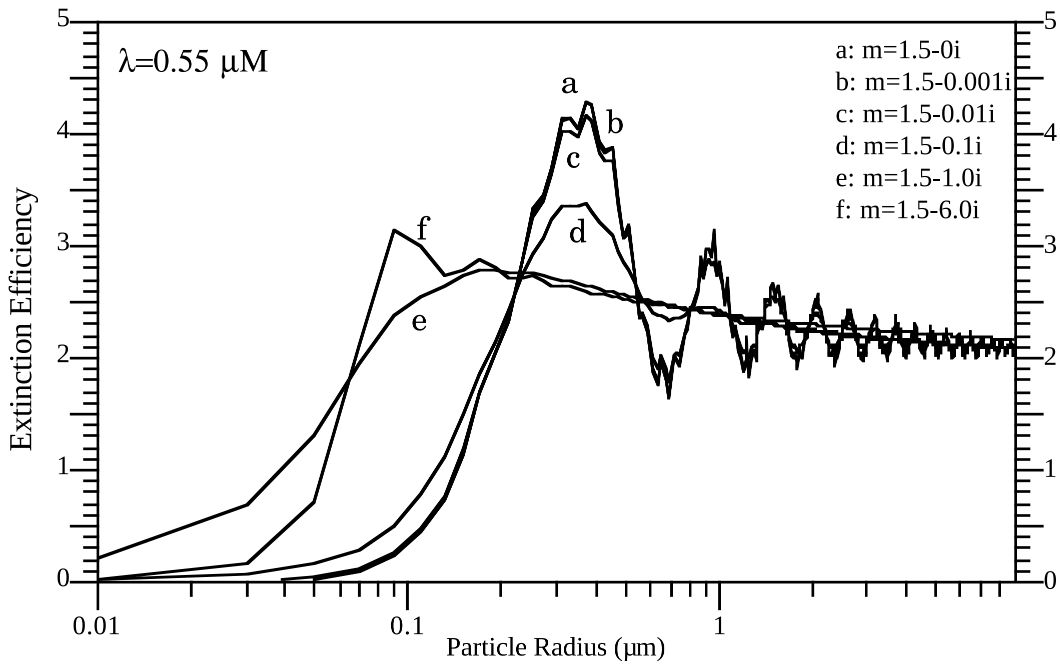

Fig. B–1. *Variations of extinction efficiency against particle radius for individual particles.*

2. Mean Optical Properties

In the real atmosphere, aerosol particles with a variety of radii coexist. Statistically averaged optical properties in a unit volume are computed with certain analytical aerosol–particle size distributions, $n(r)$. At a given wavelength, λ, volumetric mean aerosol optical properties are, Volume extinction coefficient $$E(\lambda) = \int_0^\infty \pi r^2 Q_e(\lambda, r) n(r) dr \quad , \tag{B.5}$$

single scattering albedo $$\varpi(\lambda) = \frac{\int_0^\infty \pi r^2 Q_s(\lambda, r) n(r) dr}{\int_0^\infty \pi r^2 Q_e(\lambda, r) n(r) dr} \quad , \tag{B.6}$$

asymmetry factor $$\overline{g}(\lambda) = \frac{\int_0^\infty \pi r^2 Q_s(\lambda, r) g(\lambda, r) n(r) dr}{\int_0^\infty \pi r^2 Q_s(\lambda, r) n(r) dr} \quad , \tag{B.7}$$

where $n(r)dr$ is the number of particles falling in the range of $\{r, r + dr\}$. The commonly used size distributions for atmospheric aerosol particles are the log–normal distribution and the modified Gamma distribution. For a log–normal size distribution

$$n(r) = \frac{N_0}{r \ln\sigma \sqrt{2\pi}} \exp\left[-\frac{(\ln r - \ln \bar{r})^2}{2(\ln\sigma)^2} \right] \quad , \tag{B.8}$$

where N_0 is the total number of aerosol particles in a unit volume, σ is the geometric standard deviation, and $\bar{r}$ is the geometric mean radius. The distribution is asymmetric. 95% of the particles fall in the range from $\bar{r}/2\sigma$ to $2\bar{r}\sigma$ (For a normal distribution, about 95% of the particles fall between $\bar{r} \pm 2\sigma$). For the modified Gamma size distribution (WMO 1986),

$$n(r) = A r^\alpha \exp\left[-b r^\beta \right] \quad , \tag{B.9}$$

where α, b and β are tunable parameters, which enable the model to produce desirable distributions with different orders of moments. A is a coefficient defined such that the total number of particles is normalized to unity.

3. Optical Properties of Sulfate Aerosol

We calculated the optical properties of sulfate aerosol, which contains 75% sulfuric acid and 25% water (WMO 1986). This type of sulfate aerosol has been widely applied to study volcanic aerosols and tropospheric aerosols (e.g., Kiehl and Briegleb 1993; Stenchikov et al. 1998). It is customary to express the extinction efficiency as extinction per unit mass, i.e. the so–called specific extinction (unit: m^2/g(aerosol)). The specific extinction of sulfate aerosol can be written as,

$$\Psi_e(\lambda) = \frac{E(\lambda)}{\chi_{SO_4} m} = \frac{\int_0^\infty \pi r^2 Q_e(\lambda, r) n(r) dr}{\chi_{SO_4} \rho_s \int_0^\infty \frac{4}{3} \pi r^3 n(r) dr} \quad , \qquad (B.10)$$

where χ_{SO_4} is the fraction of mass that is SO_4^{2-}, ranging from 55% to 65% (Kiehl and Briegleb 1993). We used 60% in our calculations. ρ_s=1.7 g/cm^3 is the density of dry aerosol particles.

To test the influence of particle size distribution on the calculated mean optical properties, we ran two cases, one with a log–normal size distribution and the other with the modified Gamma size distribution. The size distributions and parameters used are given in Fig. B–2. Refractive indices of sulfate aerosol were taken from the WMO report (WMO 1986). They are complex numbers and vary with wavelength. The Mie scattering model was run for the wavelengths from 0.05 μm to 100 μm with a high spectral resolution of 0.005 μm to compute the volumetric mean specific extinction, single–scattering albedo and asymmetry factor. The procedures are summarized in a flowchart (Fig. B–3).

Fig. B–4 shows the volumetric mean aerosol optical properties between 0.05 μm and 10 μm computed by the Mie scattering model of the Climate Research Group (CRG) using the log–normal size distribution and the modified Gamma size distribution in Fig. B–2. For comparison, Kiehl and Briegleb (1993)'s (hereinafter referred to as K&B) calculations for wavelength between 0.3 μm to 4 μm using the same log–normal size distribution but a different Mie model and different refractive indices are also included in Fig. B–4. Comparison between the two CRG's calculations indicates

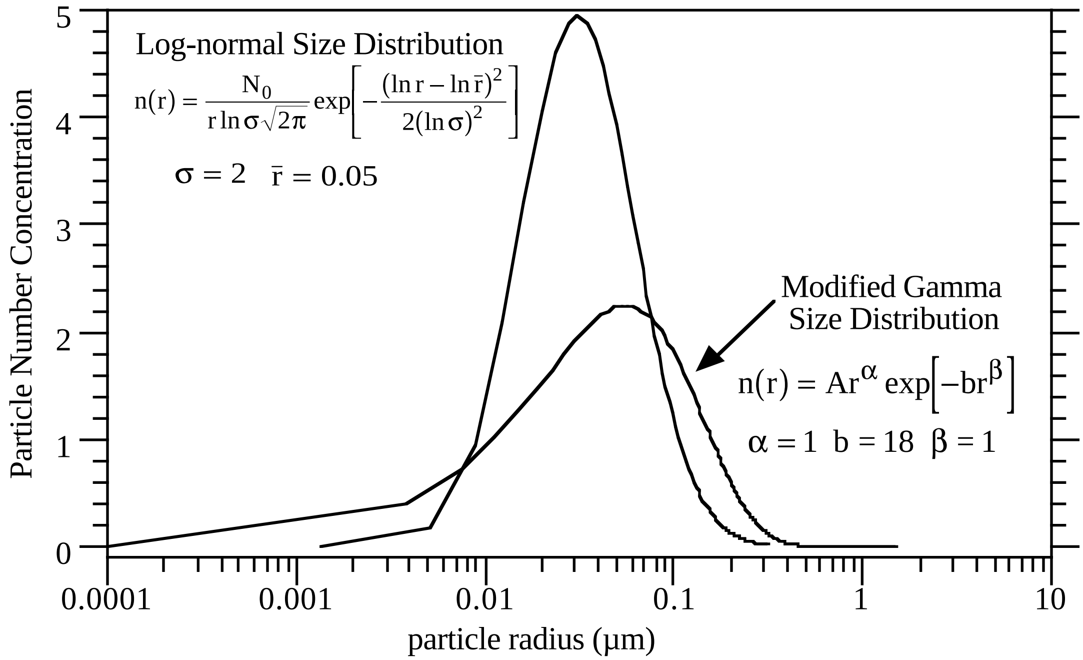

Fig. B–2. *Size distributions of aerosol particles. The vertical axis is in an arbitrary unit.*

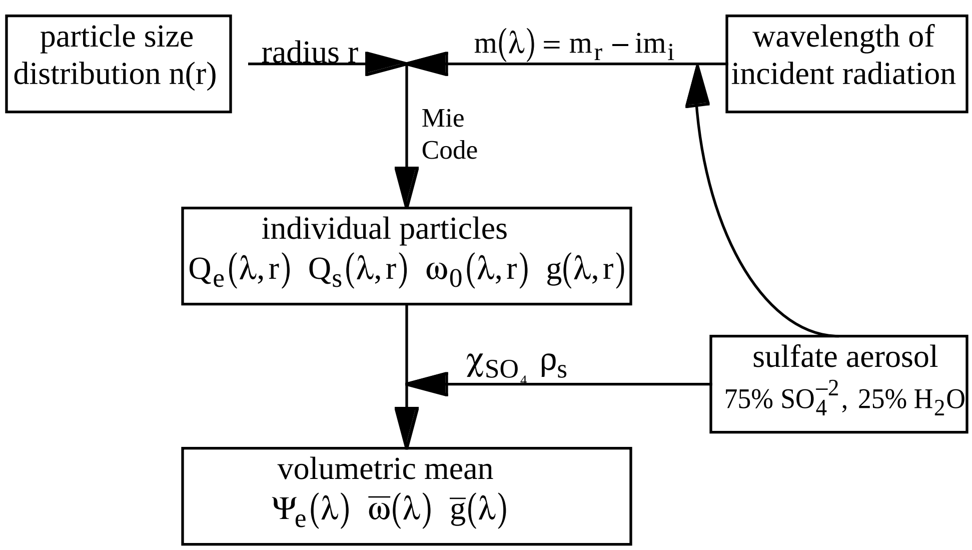

Fig. B–3. *Flowchart: calculation of sulfate aerosol optical properties by a Mie theory model.*

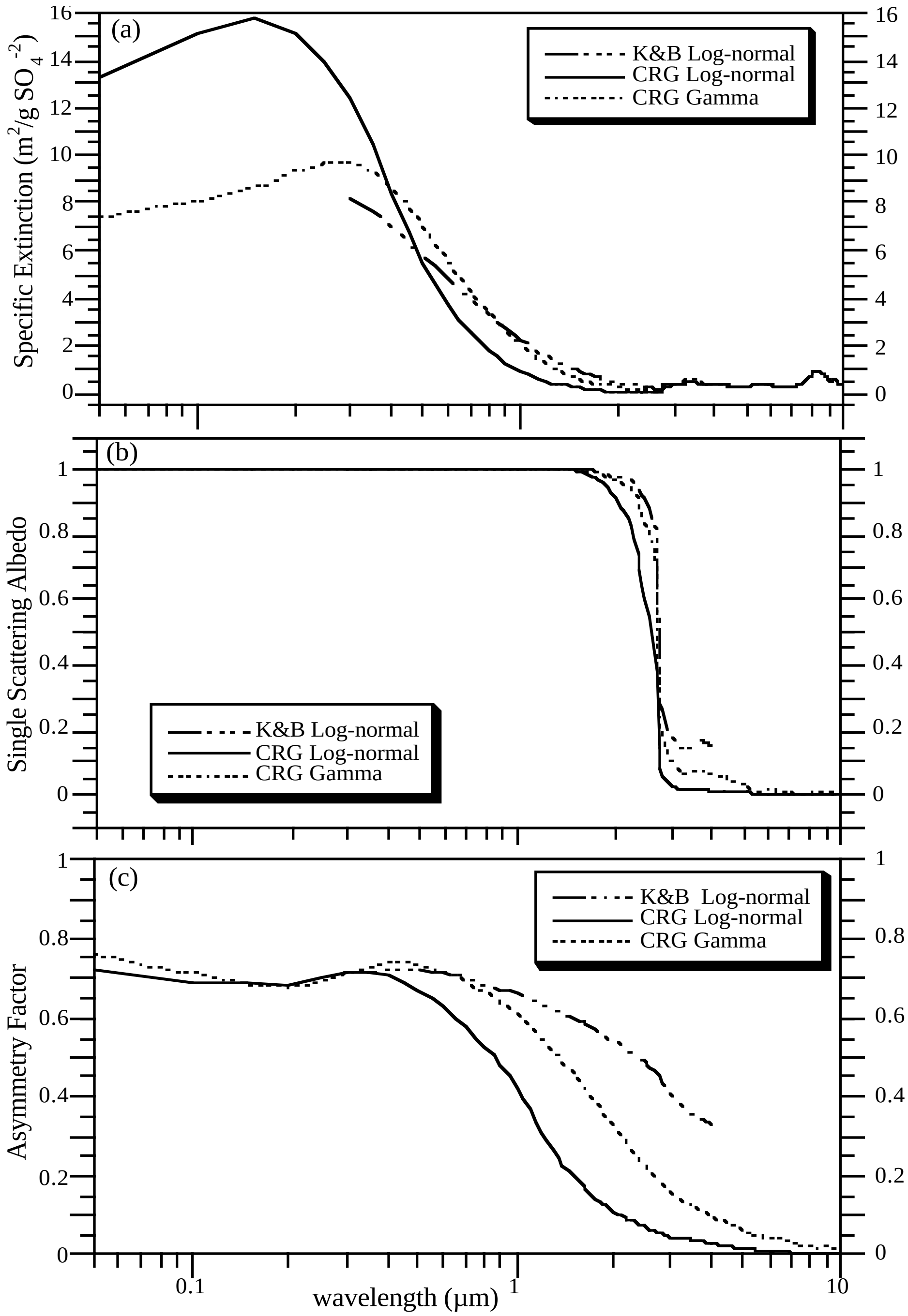

Fig. B–4. *Aerosol optical properties, (a) specific extinction, (b) single scattering albedo and (c) asymmetry factor, computed by the CRG Mie scattering model using the size distributions in Fig. B–2, and computed by Kiehl and Briegleb (1993) (K&B).*

that volumetric mean specific extinction and single scattering albedo are not sensitive to size distribution in the visible, near–infrared and far–infrared regions (0.3~10.0 μm). The two calculations also agree well with K&B's calculation between 0.3 μm and 4.0 μm. In ultra–violet region, the calculated specific extinction by the CRG Mie model with the log–normal size distribution is about 50% larger than that calculated with the modified Gamma size distribution. However, the percentage of solar energy to the total incident solar energy in this region is rather small. For the asymmetry factor, the three calculations are comparable in the visible region, but differ from each other in the near infrared region. Comparison between the two CRG calculations indicates that asymmetry factor is rather sensitive to size distribution. For the same log–normal size distribution, K&B's asymmetry factor is also much larger than the CRG's in the visible and near–infrared regions. The discrepancy may be attributed to the differences of refractive indices and sampling of particle radius in calculating the optics of individual aerosol particles. More sensitivity tests showed that how to sample the particle radius has significant influence on the calculated asymmetry factor. We did not attempt to systematically examine the uncertainties of these parameters and their influence on radiative forcing calculation. However, Boucher et al. (1998) showed that the contribution of the uncertainties of aerosol optical properties to the uncertainty of the calculated aerosol radiative forcing is rather small compared to other factors such as the radiative transfer scheme and the concentration of aerosols measured in the atmosphere.

4. Spectral–Band Averaged Optical Properties of Sulfate Aerosol

The specific extinction, single scattering albedo and asymmetry factor in a 0.05 μm spectral resolution calculated by the CRG Mie model with the modified Gamma size distribution and presented in last Section were then integrated into the 11 broad bands of the solar radiation and the 9 broad bands of longwave radiation for the UIUC 24–layer ST–GCM (Appendix A). The integration was performed by using the Planck function as weight. The calculated broadband specific extinction, single scattering albedo and asymmetry factor for solar radiation are presented in Table A–1 in Appendix A. In Table B–1 we present the broadband specific extinction for longwave radiation. Since no scattering of longwave radiation by sulfate aerosol occurs, the single scattering albedo and asymmetry factor are all equal to zero. The optical properties of this specific type of sulfate aerosol have also been integrated into the broad radiation bands of the UIUC 11–layer AGCM with appropriate modifications. They have been used in the studies of global and regional climate changes induced by the direct solar radiative forcing of anthropogenic sulfate aerosol (Schlesinger et al. 1997a). Finally, it should be pointed out that for tropospheric sulfate aerosol the influence of water vapor on the aerosol particle size must be considered to obtain more accurate radiative forcing. We obtained the dependence of sulfate–aerosol particle size on relative humidity by fitting observations reported by Charlson et al. (1984) (Fig. B–5).

Table B–1. *Specific extinction of sulfate aerosol (75% H_2SO_4 and 25% H_2O) in the 9 broad longwave bands of the UIUC 24–layer ST–GCM.*

wavelength (cm^{-1})	0~ 340	340~ 540	540~ 800	800~ 980	980~ 1100	1100~ 1215	1215~ 1380	1380~ 1900	1900~ 3000
Extinction (m^2/g)	0.0276	0.0729	0.1575	0.2427	0.4886	0.7893	0.6277	0.2930	0.3675

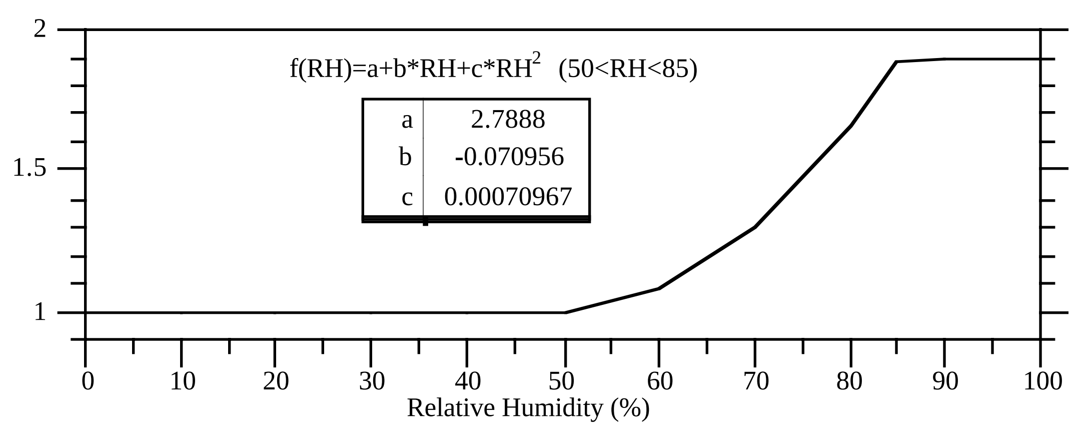

Fig. B–5. *Dependence of sulfate–aerosol particle size on relative humidity.*