

Refractive Indices of Three Hygroscopic Aerosols and their Dependence on Relative Humidity

The following briefly describes laboratory measurements of refractive indices that are available for common hygroscopic aerosol species, i.e., ammonium sulfate $(\text{NH}_4)_2\text{SO}_4$, sea salt, and ammonium nitrate NH_4NO_3 . These are used to develop a FORTRAN code to interpolate and extrapolate real and imaginary refractive indices as functions of wavelength from 0.2 to 2000 μm ($50,000$ to 5 cm^{-1}) for specified relative humidity between 0.0 to 1.0.

Laboratory measurements of real and imaginary refractive indices are needed over the full range of solar and thermal wavelengths in order to compute the radiative forcing due to atmospheric aerosols. Such measurements are available for dry ammonium sulfate [Toon and Pollack, 1976] over the spectral range 0.3 – 40 μm , and for dry sea salt [Shettle and Fenn, 1979; Nilsson, 1979; both based on Volz, 1972 measurements] over 0.2 – 40 μm . Partial spectrum measurements from 0.7 to 2.6 μm of the imaginary refractive index of ammonium sulfate and ammonium nitrate are also available [Gosse *et al.*, 1997] for solutions with dry mass fractions of 0.10, 0.25, and 0.40.

Ammonium sulfate, sea salt, and ammonium nitrate are typical hygroscopic aerosol species in that they readily absorb water vapor from the atmosphere. Typically, the aerosol remains in its initial dry crystalline state until it encounters its critical relative humidity of deliquescence (RHD) whereupon it rapidly absorbs sufficient water to fully dissolve, becoming a solution whose concentration stays in equilibrium with subsequent changes in relative humidity. If the relative humidity falls below the critical value for crystallization (RHC), the aerosol droplet rapidly loses its remaining water, returning to its original dry crystalline state.

The refractive index of hygroscopic aerosols changes with the additional amount of water that is absorbed in response to changing relative humidity. This change in refractive index, including also the change in specific density, size, and mass fraction, have been measured very accurately as functions of relative humidity [Tang and Munkelwitz, 1991; 1994; 1996]. The measurements are made at the HeNe laser wavelength of 0.633 μm , and parametric formulas derived for the changes in real part of the refractive index, specific density, size, and water activity as functions of mass fraction. We use these formulas to interpolate spectrally between the refractive indices of dry ammonium sulfate, sea salt, and ammonium nitrate and those of water.

The real and imaginary refractive indices of water are smoothly interpolated utilizing tabulated data from Hale and Querry [1973] for 0.2 to 0.7 μm , Palmer and Williams [1974] for 0.7 to 2.0 μm , and Downing and Williams [1975] for 2.0 to 1000 μm . The value of 1.330 for the real refractive index at 0.633 μm as measured by Tang and Munkelwitz [1991] is used to anchor the smooth interpolation between the Hale and Querry [1973] and Palmer and Williams [1974] data. For the imaginary refractive index of water between 0.7 to 2.7 μm , we use the more recent measurements of Kou *et al.* [1993].

Kou *et al.* [1993] suggest that linear interpolation is appropriate for changes in refractive index due to changing mass fraction for ammonium sulfate and ammonium nitrate. A more complete description of the methodology and parameterization of relative humidity effects on radiative transfer for hygroscopic aerosols in GCM applications is given in Lacis *et al.* 2002.

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