The growth of sulfuric acid films on mineral dust for stratospheric science; a laboratory and modelling study

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The presence of an inorganic film on a stratospheric aerosol will alter the atmospheric aerosols ability to scatter and absorb incoming solar radiation. The work presented quantifies the optical change caused by the growth of a thin film of sulfuric acid on an optically trapped mineral aerosol. Mie spectroscopy was applied to determine the film thickness (to one nm) and the refractive index (and thus the optical properties) of the trapped aerosol throughout film development.

Optical trapping techniques created contact free experimental conditions for the trapped particles, whilst the radius and refractive index of the aerosol was obtained through application of Mie theory on backscattered white light spectra from the optically trapped aerosol (Bohren & Huffman 1998). The refractive index describes the scattering and absorbing properties of the aerosol.

Trial experiments to determine the behaviour of sulfuric acid upon trapping were run prior to filmdevelopment. Regardless of the initial acid concentration, all optically trapped droplets equilibrated with surrounding water vapour, at a given relative humidity, to reach the same concentration. Sulfuric acid droplets are dependent on their environment, and thus the same theory can be applied to sulfuric acid films.



Figure 1: The black refractive index dispersion depicts the final concentration of a trapped sulfuric acid droplet upon reaching equilibrium with the relative humidity of the surrounding environment. Comparing the refractive index dispersion to dispersions from literature (Krieger et al. 2000), the sulfuric acid concentration upon trapping is approximately 48 % wt, a much higher concentration to its intial concentration (20 % wt).

The refractive index of the mineral aerosol, silica, was determined before film formation. The refractive

index of 1.3713 ± 0.0182 at wavelength of 589 nm was determined from analysis of eighteen individually trapped silica particles. The refractive index value for silica is lower than published refractive index values due to the silica particles porosity (Zhishang & Ripple 2014).

To replicate coagulation between stratospheric aerosol and sulfuric acid, single mineral aerosol particles were trapped and a film of sulfuric acid formed from an acid mist, which wetted and spread on the silica aerosol. Mie spectra were obtained throughout the condensation of sulfuric acid on the mineral aerosol.



Figure 2: Mie spectra depicting the alteration of Mie resonances during the development of a sulfuric acid film on a silica mineral aerosol.

Analysis of Mie spectra throughout sulfuric acid film growth provided knowledge of the change in refractive index of the mineral particle: upon film formation the resultant refractive index is that of sulfuric acid. The results confirm that sulfuric acid and mineral aerosols readily coalesce, and upon coalescence the refractive index of the initial aerosol alters greatly, thus changing the solar radiation scattering and absorbing potential of the aerosol enormously.

Presentation of the study to the aerosol community is crucial for future stratospheric aerosol research; determination of the refractive index change upon film formation has a direct impact on the modelling of stratospheric aerosols and their impact on controlling global temperatures.

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