**Phase changes during hygroscopic cycles in polyethylene glycol 400/ammonium sulfate system**

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**Introduction**

Atmospheric aerosols are complex mixtures of inorganic and organic components, whereof organics typically account for 20-50% of the total fine aerosol mass at continental mid-latitudes. The presence of organics impacts the heterogeneous chemistry, CCN activity, hygroscopicity, and the interaction with light of aerosol particles (Kanakidou *et al.*, 2005).

The phases of mixed organic/inorganic aerosol particles are influenced by physical and chemical properties of both components, but also by temperature and relative humidity. An understanding of the phases is required to quantify the impact of aerosols on climate, visibility and atmospheric chemistry (Parsons *et al.*, 2004).

**Experimental procedure**

Single droplets of polyethylene glycol 400/ammonium sulfate were produced from bulk solutions by a droplet generator with a modified ink jet print cartridge and deposited on a hydrophobically coated slide in a heating/cooling stage. The relative humidity over the particle was varied by passing a continuous flow of dry and humidified nitrogen at different ratios, through the cell. The particle’s phases were monitored with a microscope and their composition determined by Raman spectroscopy.

**Results and Conclusions**

With a 50:50 wt% mixture of polyethylene glycol 400/ammonium sulfate we have performed several humidity cycles at 293 K for different particle sizes. For all the particles we have observed a liquid-liquid phase separation when the ammonium sulfate deliquesced. This separation of phases was also observed in bulk measurements performed on the same system (Marcolli & Krieger, 2006). The presence of two liquid phases in a ~70 µm particle is shown in Figure 1. Based on Raman spectra collected from the particle, the ammonium sulfate was found to be in the inner sphere, meanwhile the polyethylene glycol was present in the outer sphere.

By increasing the relative humidity in the heating/cooling stage, the two liquid phases merge into one liquid phase, corresponding to a polyethylene glycol/ammonium sulfate aqueous solution. A decrease in relative humidity leads to liquid-liquid phase separation, which starts at about 90% RH and lasts until ammonium sulfate effloresces.

Such phase separation might be very common in tropospheric aerosol particles and in consequence may be an important factor affecting the climate by influencing heterogeneous and multiphase chemistry, gas/particle partitioning and the aerosol’s hygroscopicity.

![Figure 1](image-url) (a) Liquid-liquid phase separation for the polyethylene glycol 400/ammonium sulfate system (50:50 wt %); 1) aqueous ammonium sulfate solution; 2) mainly PEG 400. (b) Effloresced particle.

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**References**