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Coexistence of three liquid phases in atmospheric aerosol particles

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Aerosol particles are ubiquitous in the atmosphere and play an important role for air quality and Earth's climate. Primary organic aerosol (POA), secondary organic aerosol (SOA), and secondary inorganic aerosol (SIA) constitute a significant mass fraction of these particles. POA, SOA, and SIA can become internally mixed within the same particle though different processes such as coagulation, gas–particle partitioning. To predict the role of these internally mixed particles in climate and air quality information on their phase behaviour is needed, i.e. information on the number and type of phases present within these particles. As an example, a particle with a single homogeneous liquid phase can have different radiative properties, reaction rates, uptake kinetics, and potential to change cloud microphysical properties by activating into a cloud droplet, compared to a particle with multiple liquid or solid phases.

In the current study we used Nile red, a solvatochromic dye, and fluorescence microscopy in order to determine the phase behaviour of POA+SOA+SIA particles. Squalane was used as a proxy of POA, ammonium sulfate was used as SIA and 1 of 23 different oxidized organic molecules were used as proxies of SOA. We demonstrate that three liquid phases often coexist within individual particles. We find that the phase behaviour strongly depends on the oxygen-to-carbon ratio of the SOA proxies. Experiments with SOA generated by dark ozonolysis of α-pinene in an environmental chamber are consistent with these observations. We also used thermodynamic and kinetic modelling to investigate the atmospheric implications of our experimental results.