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Feedbacks between microphysics and photochemical aging in viscous aerosol

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Fe(III)-citrate complex photochemistry, which plays an important role in aerosol aging, especially in lower troposphere, has been widely recognized in both solution and solid states. It can get excited by light below about 500 nm, inducing the oxidation of carboxylate ligands and the production of peroxides (e.g., OH•, HO_2•), which have a significant impact on the climate, air quality and health. Recently, there is literature reporting that aqueous aerosol particles may attain highly viscous, semi-solid or even glassy physical states under a wide range of atmospheric conditions. However, systematic studies on the effect of high viscosity on photochemical processes are scarce.

In this research, mass and size changes of a single, aqueous Fe(III)-citrate/citric acid particle levitated in an electrodynamic balance (EDB) are tracked during photochemical processing. We observe an overall mass loss during photochemical processing due to evaporation of volatile (e.g., CO_2) and semi-volatile (e.g., ketones) compounds. It is known that relative humidity and temperature strongly effects the viscosity of citric acid. Hence, under light intensities large enough not limiting photochemical processing (at a wavelength of either 375 nm or 473 nm), the quasi-steady state evaporation rate in our experiments depends on relative humidity and temperature. The same holds true for the characteristic time scale for reaching thermodynamic equilibrium after switching off the light source. We are focusing on the high viscosity case (i.e. reduced molecular mobility and low water content), which slows down the transport of products but can also affect chemical reaction rates (e.g., initial absorption process, charge and energy transfer). Data are compared to kinetic modeling and diffusivities for semi-volatile compounds are estimated aiming at a more detailed understanding of the feedbacks between microphysics and photochemical aging.