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Diffusivity Measurements of Volatile Organics in Levitated Viscous Aerosol Particles

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Field measurements indicating that atmospheric secondary aerosol (SOA) particles can be present in a highly viscous, glassy state have spurred numerous studies addressing low water diffusivities in glassy aerosols, focusing on kinetic limitations to hygroscopic growth and the plasticizing effect of water. Less is known about diffusion limitations of organic molecules and oxidants in viscous matrices and how these might affect atmospheric chemistry and gas-particle phase partitioning of complex mixtures with constituents of different volatility. Often viscosity data has been used to infer diffusivity via the Stokes- Einstein relationship even though strong deviations from this relationship have been observed for matrices of high viscosity. In this study, we provide a quantitative estimate for the diffusivity of a volatile organic in a viscous matrix. Evaporation of single particles generated from an aqueous solution of sucrose and a small quantity of volatile tetraethylene glycol (PEG-4) is investigated in an electrodynamic balance at controlled relative humidity (RH) and temperature conditions, thereby varying the viscosity of the sucrose matrix. The evaporative loss of tetraethylene glycol as determined by Mie resonance spectroscopy is used in conjunction with a diffusion model to retrieve translational diffusion coefficients of tetraethylene glycol. The evaporation of PEG-4 shows a pronounced RH and temperature dependence and is severely depressed for RH 30% corresponding to diffusivities $< 10^{-14}$ cm^2/s at temperatures as high as 15 °C, implying that atmospheric volatile organic compounds (VOC) can be subject to severe diffusion limitations in glassy SOA. Comparison of the experimentally derived diffusivities with viscosity estimates for the ternary system reveals a breakdown of the Stokes-Einstein relationship.