4.025 Diffusivity measurements of volatile organics in levitated viscous aerosol particles.

Early Career Scientist

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Abstract:

Field measurements indicating that atmospheric secondary aerosol particles can be present in a highly viscous, glassy state [1] have spurred numerous studies addressing kinetic limitations of hygroscopic growth due to slow condensed phase diffusion and measurements of water diffusivities in viscous organics [2, 3]. In contrast, little is known about diffusion limitations of organic molecules in viscous matrices and how these might affect gas-particle phase partitioning of complex mixtures with constituents of different volatility, the questionable assumption often being that viscosity data can be used to infer diffusivities according to Stokes-Einstein.
In this study, we developed a measurement procedure and provide a quantitative estimate for the diffusivity of a volatile organic in a viscous matrix.
Single particles generated from an aqueous solution of sucrose and a small quantity of volatile polyethylene glycol (PEG) are investigated in an electrodynamic balance at controlled humidity conditions over a large temperature range, thereby varying the viscosity of the sucrose matrix. Mie resonance spectroscopy is used to determine the particle size and evaporation rate.
At high temperatures, no diffusion limitations apply and the particle remains well-mixed throughout the evaporation process. In the kinetically limited regime at low temperatures, the surface concentration of PEG is depleted as the diffusion flux emanating from the center of the particle is too low to maintain a homogeneous concentration throughout the particle. Therefore, we observe a deviation of the evaporation rates from the temperature dependence as predicted by the pure component enthalpy of vaporization under well-mixed conditions. The resulting reduced evaporation rates are used to determine the PEG activity at the surface and to quantify the PEG diffusivity.
 [1] Virtanen, A., et al., Nature, 2010
[2] Lienhard, D., et al. ACP, 2015
[3] Davies, J.F. and Wilson, K.R., Anal.Chem.,2016