2.029 Particle phase state and viscosity in atmospheric secondary organic aerosols.

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Abstract:

Secondary organic aerosols (SOA) are ubiquitous in the atmosphere. SOA can occur in amorphous solid or semi-solid phase states depending on chemical composition, relative humidity (RH), and temperature. The phase state of SOA is important for their effects on climate and air quality, but its global distribution is poorly characterized. Our analysis of SOA phase state builds on the molecular corridor approach, which is a two-dimensional framework of volatility and molar mass of SOA components constrained by boundary lines of low and high molecular O:C ratio. We developed a method to estimate glass transition temperatures based on the molar mass and molecular O:C ratio of SOA components. We predict viscosity from the Tg-scaled Arrhenius plot of fragility as a function of the fragility parameter. We estimated viscosity of α-pinene and isoprene SOA as a function of RH by accounting for hygroscopic growth of SOA and applying the Gordon-Taylor mixing rule, reproducing previously published experimental measurements very well. Viscosity of toluene SOA was predicted using the elemental composition obtained by high-resolution mass spectrometry (HRMS), resulting in a good agreement with the measured viscosity. Further, we used the global chemistry climate model EMAC with the organic aerosol module ORACLE to predict the phase state of atmospheric SOA. For the planetary boundary layer, global simulations indicate that SOA are mostly liquid in tropical and polar air with high relative humidity, semi-solid in the mid-latitudes, and solid over dry lands. We find that in the middle and upper troposphere SOA should be mostly in a glassy solid phase state. Thus, slow diffusion of water, oxidants, and organic molecules could kinetically limit gas-particle interactions of SOA in the free and upper troposphere, promote ice nucleation and facilitate long-range transport of reactive and toxic organic

pollutants embedded in SOA.