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Glass transition measurements in mixed organic and organic/inorganic aerosol particles

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The recent proposal of a semi-solid or glassy state of secondary organic aerosol (SOA) particles has sparked intense research in that area. In particular, potential effects of a glassy aerosol state such as incomplete gas-to-particle partitioning of semi-volatile organics, inhibited chemical reactions and water uptake, and the potential to act as heterogeneous ice nuclei have been identified so far. Many of these studies use well-studied proxies for oxidized organics such as sugars or other polyols. There are, however, few measurements on compounds that do exist in atmospheric aerosol particles.

Here, we have performed studies on the phase state of organics that actually occur in natural SOA particles arising from the oxidation of alpha-pinene emitted in boreal forests. We have investigated the two marker compounds pinonic acid and 3-methylbutane-1,2,3-tricarboxylic acid (3-MBTCA) and their mixtures. 3-MBCTA was synthesized from methyl isobutyrate and dimethyl maleate in two steps.

In order to transfer these substances into a glassy state we have developed a novel aerosol spray drying technique. Dilute solutions of the relevant organics are atomized into aerosol particles which are dried subsequently by diffusion drying. The dried aerosol particles are then recollected in an impactor and studied by means of differential scanning calorimetry (DSC), which provides unambiguous information on the aerosols' phase state, i.e. whether the particles are crystalline or glassy. In the latter case DSC is used to determine the glass transition temperature Tg of the investigated samples. Using the above setup we were able to determine Tg of various mixtures of organic aerosol compounds as a function of their dry mass fraction, thus allowing to infer a relation between Tg and the O:C ratio of the aerosols. Moreover, we also studied the glass transition behavior of mixed organic/inorganic aerosol particles, including the effects of liquid-liquid phase separation upon drying.