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Computation of Phase Equilibria, State Diagrams and Gas/Particle Partitioning of Mixed Organic-Inorganic Aerosols

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The chemical composition of organic-inorganic aerosols is linked to several processes and specific topics in the field of atmospheric aerosol science. Photochemical oxidation of organics in the gas phase lowers the volatility of semi-volatile compounds and contributes to the particulate matter by gas/particle partitioning. Heterogeneous chemistry and changes in the ambient relative humidity influence the aerosol composition as well. Molecular interactions between condensed phase species show typically non-ideal thermodynamic behavior. Liquid-liquid phase separations into a mainly polar, aqueous and a less polar, organic phase may considerably influence the gas/particle partitioning of semi-volatile organics and inorganics (Erdakos and Pankow, 2004; Chang and Pankow, 2006). Moreover, the phases present in the aerosol particles feed back on the heterogeneous, multi-phase chemistry, influence the scattering and absorption of radiation and affect the CCN ability of the particles. Non-ideal thermodynamic behavior in mixtures is usually described by an expression for the excess Gibbs energy, enabling the calculation of activity coefficients. We use the group-contribution model AIOMFAC (Zuend et al., 2008) to calculate activity coefficients, chemical potentials and the total Gibbs energy of mixed organic-inorganic systems. This thermodynamic model was combined with a robust global optimization module to compute potential liquid-liquid (LLE) and vapor-liquid-liquid equilibria (VLLE) as a function of particle composition at room temperature. And related to that, the gas/particle partitioning of semi-volatile components. Furthermore, we compute the thermodynamic stability (spinodal limits) of single-phase solutions, which provides information on the process type and kinetics of a phase separation.

References

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