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# Liquid-liquid phase separation in atmospheric aerosol particles: dependence on organic functionalities and mixture complexity

M. Song (1), C. Marcolli (1), U.K. Krieger (1), A. Zuend (2), and T. Peter (1)

(1) ETH Zürich, Institute for Atmospheric and Climate Sciences, Zürich, Switzerland, (2) California Institute of Technology, Department of Chemical Engineering, California, USA

In the troposphere, aerosol particles undergo phase transitions such as deliquescence and efflorescence during humidity cycles (Marcolli and Krieger, 2006). In addition, interactions between organic and inorganic compounds lead to liquid-liquid phase separation (LLPS) (Ciobanu et al., 2009). Recent studies on a limited number of model systems have shown that oxygen-to-carbon ratios (O:C) of the organic aerosol fraction might be a good predictor for LLPS in mixed organic/ammonium sulfate (AS) particles (Bertram et al., 2011; Song et al., 2011). However, in order to corroborate this hypothesis experiments with an organic fraction that consists of a higher number of components with different O:C ratios and functional groups are needed.

In order to determine the influence of O:C ratio, the specific organic functionalities and the mixture complexity on LLPS, we subjected organic/AS particles deposited on a hydrophobically coated substrate to relative humidity (RH) cycles and observed phase changes using optical microscopy and micro-Raman spectroscopy. To determine the influence of mixture complexity, we mixed together up to 10 organic compounds. We also prepared mixtures that were rich in different types of functional groups like polyols, aromatics and dicarboxylic acids which were identified from field measurements. We screened for a miscibility gap by varying the organic-to-inorganic ratio from 2:1 to 1:6.

AS in the investigated single particles effloresced at 27 - 50 %RH and deliquesced at 72 - 79 %RH during humidity cycles. The occurrence of LLPS is determined to a high degree by the O:C of the organics: there was no LLPS for mixtures with O:C > 0.8 and there was always LLPS for mixtures with O:C < 0.57. In the range in between, we observed a dependence on the specific functional groups: a high share of aromatic functionalities shifts the range of O:C for which LLPS occurs to lower values. A correlation was also found for the onset RH of LLPS as a function of O:C. We did not find any dependence of LLPS on the complexity of the mixture. Overall, the RH range of coexistence of two liquid phases depends in first place on the O:C ratio of the particles and in second place also on the specific organic functionalities.

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