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Flow tube perspective of organic aerosol aging: condensed phase chemistry and photochemistry

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Understanding the chemical and microphysical evolution of atmospheric aerosol particles during their life-time in the atmosphere is of significant interest for more quantitative assessments of aerosol climate interactions. Especially the organic fractions poses significant challenges. While a more and more consistent picture of chemical evolution has been emerging with respect to some parameters, such as C:O ratio, density and hygroscopicity from observations in smog chamber and field experiments, our predictive capability for more detailed chemical and microphysical properties is still limited. This calls for detailed thermodynamic and kinetic data of important processes. Aerosol particles evolve in complex cycles including gas phase oxidation, phase partitioning and particle phase chemical processes. While a relatively detailed picture of gas phase oxidation of volatile organic compounds is available (though still with a number of important remaining issues) to feed this information into aerosol evolution, phase transfer and condensed phase chemical processes are much less well understood. Therefore, this contribution highlights a few condensed phase oxidation and photooxidation processes from the literature and own studies to emphasize their importance for chemical evolution, their relation to gas phase HOx, to microphysical properties such as hygroscopicity, phase state and morphology. Examples include oleic acid ozonolysis and the formation of hydrogen peroxide under humid conditions, photosensitized oxidation of dicarboxylic acids and phenolic substrates in presence of oxidants such as ozone or nitrogen oxides. Further, first results of new investigations into nanometer resolution X-ray microspectroscopy that allow concomitant measurement of morphology, carbon functional group composition and degree of hydration under controlled conditions of temperature and humidity will be shown. Apart from resolving microstructure and phase state, this allows following slow diffusion controlled oxidation into viscous particles.