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Importance of aerosol phase and water uptake for organic aerosol oxidation mechanisms

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We will present a combined laboratory and modelling study on the atmospheric processing of organic acid aerosols. We investigate the ozonolysis of aerosols under different temperature and humidity conditions. An electrodynamic balance apparatus is used to study single aerosol particles to assess the phase (liquid versus solid) and size of the ageing aerosol (due to water uptake and/or evaporation of ozonolysis products). An aerosol ensemble flow tube combined with high resolution mass spectrometry is utilized to determine the changing chemical composition of the aerosol. Thermodynamic modelling is applied to generalize the laboratory observations.

Three single-compound model aerosols were investigated and compared: maleic acid, oleic acid and arachidonic acid. All three species contain carboxylic acid functionality. Oleic acid and maleic acid contain one unsaturated double bond, and arachidonic acid contains four unsaturated double bonds. The ozonolysis of maleic acid aerosol particles are found to be dependent on the phase of the aerosol, which itself is dependent on the environmental relative humidity. The ozonolysis of deliquesced particles shows distinctly different behaviour from the ozonolysis of effloresced particles. Differences include: changes in aerosol hygroscopicity, volatility and product composition (1). Furthermore the chemistry of a dry ozonolysed particle changes once it is deliquesced post ozonolysis. The products of the ozonolysis of pure oleic acid and arachidonic acid aerosols, which are slightly hygroscopic, are affected by relative humidity even though they are liquid under all conditions.

This study highlights the necessity of investigating organic aerosol reactivity under different relative humidities, because the uptake of water is a key mechanistic step for the chemical and physical processing of the aerosol. The detailed chemical mechanisms responsible for the ozonolysis of aerosol composed of small highly oxidised species (e.g. maleic acid) differs greatly from the chemistry of aerosols composed of larger and less oxidised species (e.g. oleic acid). A discussion on the importance of these chemical differences for global aerosol populations, formation of gaseous oxygenated volatile organic compounds, and the cloud nucleation behaviour of the aerosols will be provided.

(1) Pope et al. (2010) Ozonolysis of maleic acid aerosols: Effect upon aerosol hygroscopicity, phase and mass. Environ. Sci. Technol. 44 (17), 6656.