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Chemical Aging of Tropospheric Aerosols Evaluated by Single Particle Mass Spectrometry in the Field

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There are few techniques applicable to field observations which allow the real-time observation of processes of chemical aging of atmospheric aerosols. Mostly the information has to be inferred from Lagrangian experiments or analysis in relation to backward airmass trajectories. Perhaps the most direct information upon the aging of aerosols can be gained from single particle studies, provided sufficient particles can be examined to give a statistically significant picture of the chemical change. One technique capable of characterising individual airborne particles at a relatively high frequency is Aerosol Time-of-Flight Mass Spectrometry. This allows the continuous collection of data upon particle composition and the construction of time series. In this research, deployment of a TSI Model 3800 Aerosol Time-of-Flight Mass Spectrometer has been used to characterise the aerodynamic diameter and chemical composition of individual airborne particles. The instrument has been deployed in a number of field campaigns including use at urban, rural and remote sites. During a campaign, it is typical for the instrument to collect aerodynamic size and positive and negative ion mass spectra on in excess of 100,000 particles. These data are reduced by using a clustering algorithm (typically the ART2a neural network program or K-means clustering) to yield a manageable number of particle categories with common features in terms of their mass spectra and particle size distribution. A considerable volume of data have been collected at rural sites and show considerable internal mixing of particles from which the processes of aging can be inferred. Examples will be presented of the elucidation of aging processes through the use of mass spectrometer data. These include:

• the aging of marine aerosol as demonstrated by the incorporation of nitrate and depletion of chloride;
• the cycling of nitrate between aerosol and gas phase on a diurnal basis within a polluted urban airshed;
• the diurnal cycling of semi-volatile secondary organic matter between the gas phase and aerosol in a southern European city;
• fog processing of carbonyl compounds with sulphur dioxide;
• the generation of aerosol of mixed composition within a polluted urban atmosphere.

The strengths and weaknesses of the ATOFMS for real-time studies of atmospheric chemistry will be discussed.