Aerosol Chemical Composition and Hygroscopic Growth - first results from field studies

Hanna Herich[1], Lukas Kammermann[2], Daniel J. Cziczo[1], Martin Gysel[2], Ernest Weingartner[2], Urs Baltensperger[2] and Ulrike Lohmann[1]

Summary:

Real-time measurements of the chemical composition of atmospheric aerosols as a function of hygroscopicity have been made using a Hygroscopicity Tandem Differential Mobility Analyzer (HTDMA) and an Aerosol Time-of-Flight Mass Spectrometer (ATOFMS). These instruments were connected in series and thus single particle mass spectra from aerosols leaving the HTDMA could be obtained. Two sets of experiments have been performed: One at the ETH in the urban Zurich environment and the other at the remote high alpine station Jungfraujoch (JFJ). Both studies were conducted during February and March 2007. At Zurich 3500 mass spectra were obtained during two days of measurements. At the JFJ, due to low particle concentrations, 54 spectra were recorded in nine days. In both locations a bimodal growth factor (GF) distribution with a minor peak at GF~1.0 and major peak at GF~1.4 was observed.
Preliminary results show a large contribution of organic matter and combustion particles in the hydrophobic growth mode for both locations. Sulfate was found to be a constituent in more than 80% of the Zurich particles and more than 90% of the JFJ particles independent of their growth factor. The occurrence of black carbon was ~50% in hydrophobic Zurich aerosols and ~20% in hygroscopic ones. At the JFJ elemental carbon was found in the majority of all particles. The results show that aerosols containing elemental and organic carbon and organics referring to the fragment peak -42 are the major source for particles found in the hydrophobic mode and sulfate uptake begins to occur after the production process. Thereby the embedding of sulfate in the aerosol particles seems to be much more complex than so far assumed.

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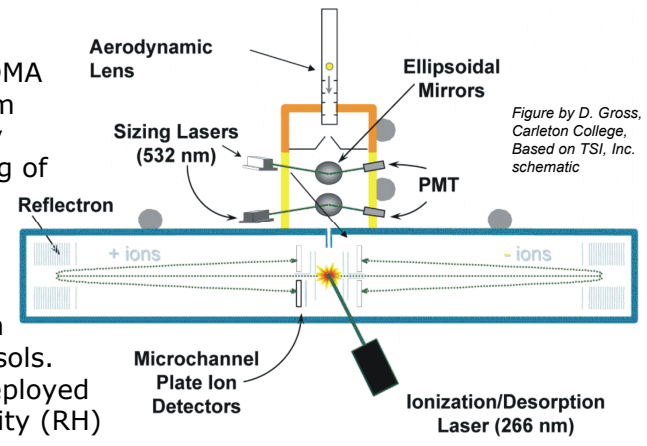
Method

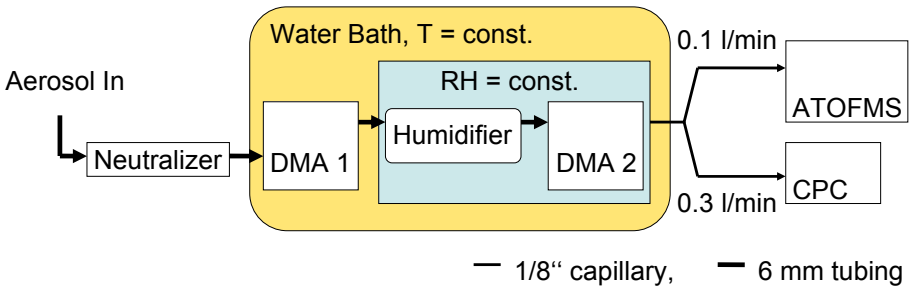
A custom built HTDMA (Humidified Tandem Differential Mobility Analyzer) consisting of two DMAs and a humidification region was used to determine the hygroscopic growth factor (GF) of aerosols. The HTDMA was deployed at a relative humidity (RH) of 82% and particles with a dry diameter of 260 nm were selected.



Aerosol Time-of-Flight Mass Spectrometer (ATOFMS), TSI Model 3800

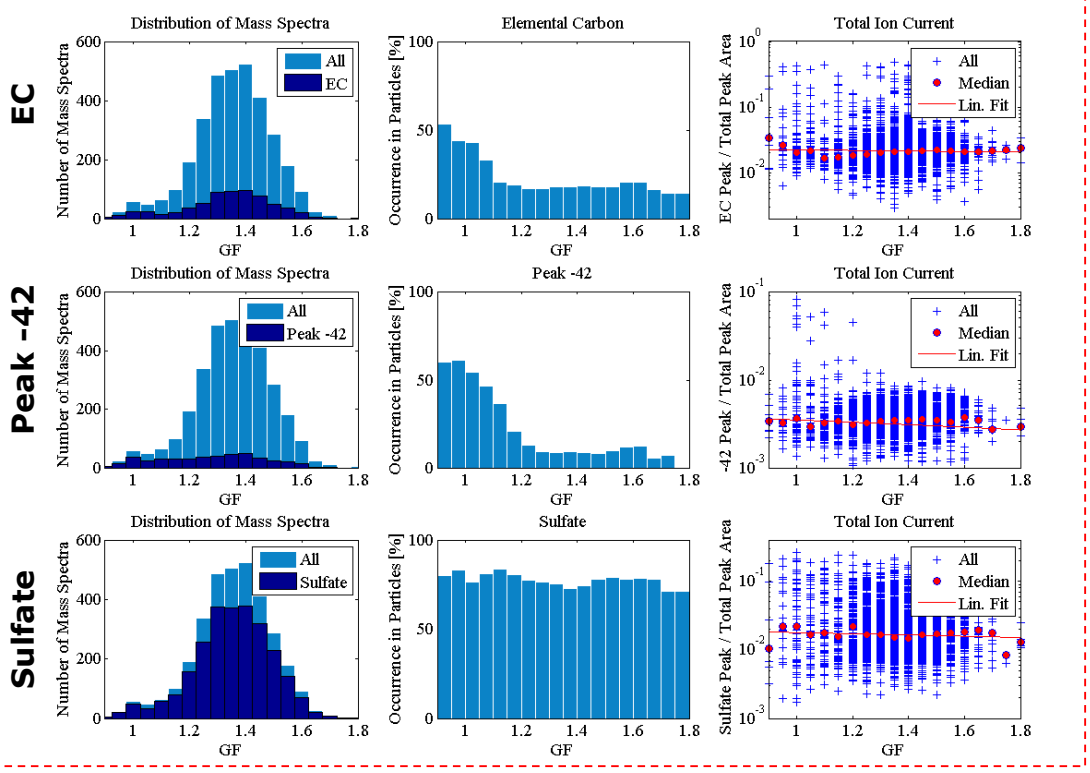
These setup conditions were limited due to the lower size cut of the ATOFMS and arching at very high voltages in the HTDMA. The ATOFMS, a single particle mass spectrometer (Model 3800, TSI, Shoreview, MN), determined size and composition of the aerosols with a certain GF simultaneously and in real time.

Experimental Setup:



Results

Zurich



JFJ

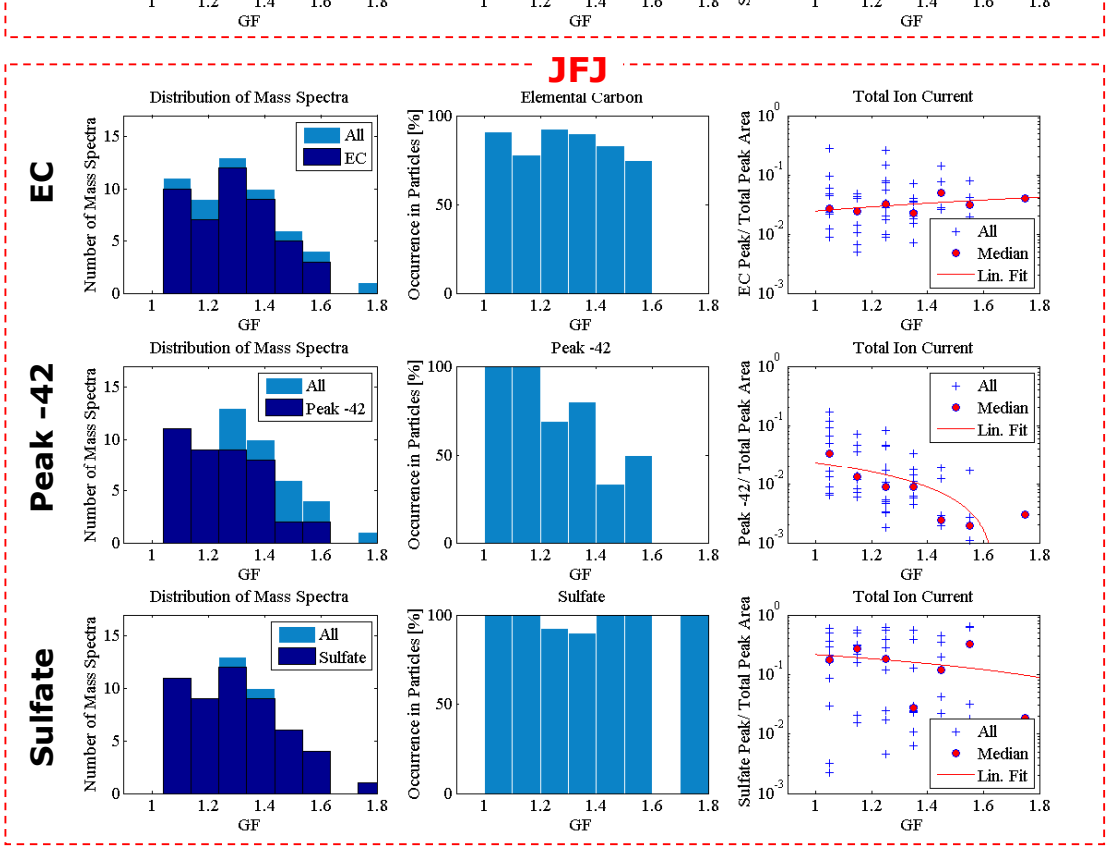


Figure: Occurrence of chemical components in dependence of the GF for the Zurich and the JFJ studies. Left column: Number distribution of all taken mass spectra and number of those containing the compound. Middle: Number distribution of mass spectra containing the certain compound normalized. Right: Peak heights of certain compounds in reference to the total peak area for each mass spectra. Peak height median for each GF and linear fit.

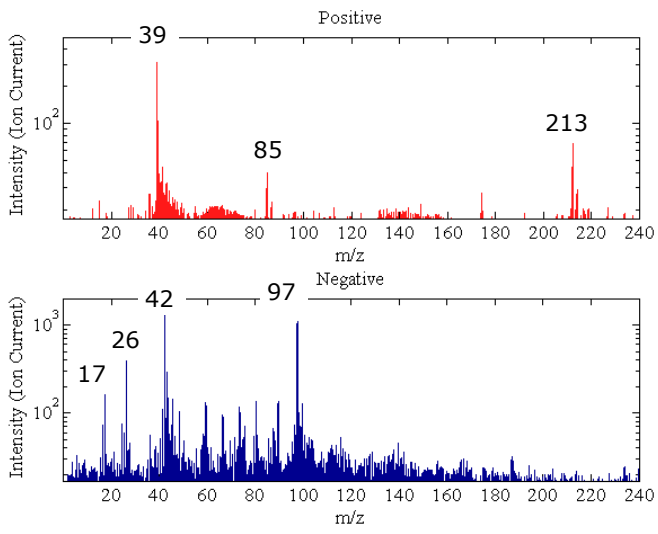
Discussion and Conclusions

During the Zurich experiment more than 3500 mass spectra were obtained. The selected size range of 260nm indicates that the aerosol is aged and not notably influenced by fresh emissions. This corresponds with the observed stable concentrations unaffected by rush hours.
Several chemical compounds were analyzed from the mass spectra and the results for elemental carbon (EC), the peak -42 (C_2H_2O or CNO) and sulfate are presented here. While a clear relation between occurrence and hygroscopic growth can be found for EC and the peak -42, sulfate is present in almost all particles.
Due to matrix effects the ATOFMS mass spectra peak heights provide only a qualitative signal. To get brief information about quantity, peak height ratios of single peaks to the overall mass spectra area were analyzed. In agreement to the first results the highest peak ratios for EC and especially peak -42 were found in the hydrophobic regime. For sulfate the peak height ratios were relatively stable. Although the JFJ dataset is much smaller similar trends in occurrence and peak ratios can be observed.
The results show that aerosols containing elemental carbon and organics referring to the fragment peak -42 are the major source for particles found in the hydrophobic mode and sulfate uptake begins to occur after the production process. Thereby the embedding of sulfate in the aerosol particles seems to be much more complex than so far assumed.

Mass Spectra

Figure:

Example of a single particle bipolar ATOFMS Mass Spectra. This particle spectra was measured at the JFJ and the particle GF was 1.05.



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