Physico-chemical Characterisation of Air, Particles, and Cloud Water in Cloud Experiments (FKZ 07ATF01 - 395)

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MOTIVATION

Aim of the project is the clarification of multi-phase processes under participation of aerosols centered of chemical and physical processes involving organics in the aerosol and cloud droplets. For the understanding of real atmosphere-chemical systems is to be clarified apart from the spectrum of possible products (above all organic substances) also the question about the place of the conversion (gaseous phase, particle surface or within aqueous particles (activated particles and cloud drops)) and the mutual exchange of the formed species between the individual phases. In addition the supply of the won extensive data record is to serve the advancement of tropospheric-chemical multi-phase models (e.g. CAPRAM) and chemistry transport models.

RESULTS

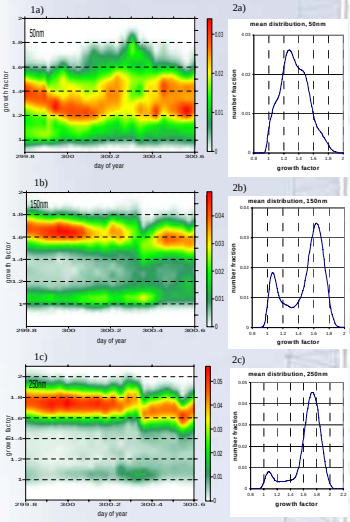

Fig. 2

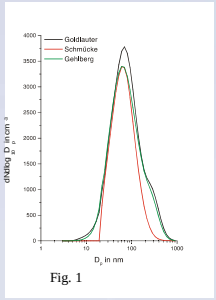

Fig. 1

Within the first measurement campaign in October 2001 17 cloud events occurred which were comprised into 9 events.
The results of the longest event during 0october 26th/27th 2001 are presented.

Fig. 1 shows averages of the particle size distribution at the three measuring sites over the whole cloud period on 26th/27th October 2001. Compared to the valley sites, the scavenging of particles > 160 nm on summit can be seen. This can be traced back to the fact that the cloud on top of the mountain activates such particles by condensation of cloudwater onto the particle surface. Thereby the particles grow up to a size that is undetectable by the instruments used for.

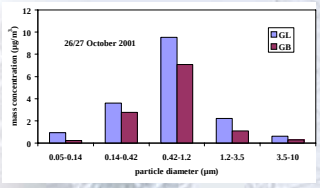

Fig. 3

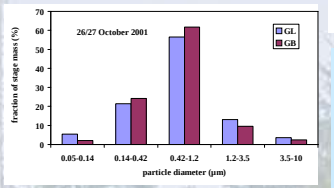

Fig. 4

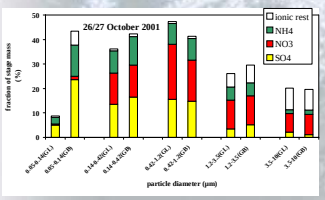

Fig. 5

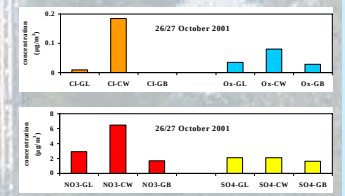

Fig. 6

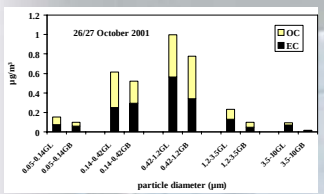

Fig. 7

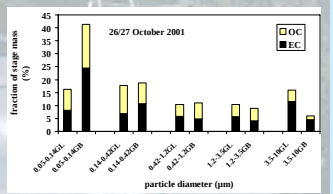

Fig. 8

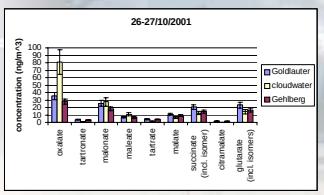

Fig. 9

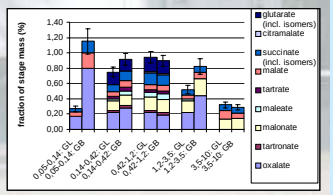

Fig. 10

EXPERIMENTALS

The particle collection was performed at the two valley stations in Goldlauter (GL - luff-side) and Gehlberg (GB - lee-side) and meteorology and gaseous phase characterisation at the three stations with identical equipment:
Meteorology: temperature, humidity, wind direction and velocity, j_{NO2}, (liquid water content).
Gaseous phase characterisation: ozone -, nitrogen oxide -, carbon monoxide-, and sulfur dioxide monitors as well as carbonyl sampling on DNPH cartridges.
Physical characterisation: Number/size distribution measurement with TDMPS and APS as well as additionally in Goldlauter measurements of humidity growth with HTDMA.
Particle collectors: ever 2 five-stage BERNER impactors (0.05/0.14/0.42/1.2/3.5/10 µm), Digitel-$PM_{2.5}$-HV collector and Sierra Andersen -PM_{10}-HV-collector.
Cloud water (CW) was collected with 4 cloudwater collectors (CASCC2) of California Institute of Technology at the mountain site Scmücke.

Analytical methods of chemical characterisation of particles:
- mass: micro balance (UMT2, Mettler/Toledo)
- inorganic mean components: ion chromatography (Metrohm)
- mono- and dicarbonic acid: capillary electrophoresis (TSP)
- OC/EC: thermogravimetric method (C-mat 5500, Ströhlein)
- polar VOC: GC-MS (Thermofinnigan)
- non-polar VOC: GC-MS (GC 6890, MSD 5973N, Agilent)

Analytical methods for cloud water:
- anionic mean components: CE
- mono- and dicarbonic acid: CE
- non-polar VOC: GC-MS

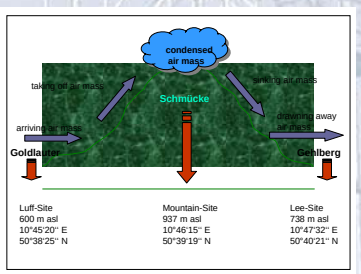

Fig. 2: 1a) to c) show time series of hygroscopic growth curves for 50, 150 and 250 nm particles at 90% r.h. measured with a HTDMA. The mean growth spectra for this time period are displayed in Fig. 2a) to c).The spectra for 150 and 250nm particles reveal a bimodal behaviour with a growth factor (GF) around 1.1 for the hydrophobic mode and a GF around 1.7 for the more hygroscopic particles. A bimodal growth characteristic for 50nm particels was not evident for this cloud event. The peak of the growth curve for 50nm particles was found around a GF of 1.3.

The total mass concentration of particles is classified as low. While the transport over the ridge of the Thüringer Wald a dilution occurs mainly by deposition.

The mass (Fig. 3), ionic, OC/EC (Fig. 7), and organic acids (Fig. 9) concentrations of the collected particles show generally lower concentrations after the cloud than in front of it. The particle size distribution of total mass is presented in Fig. 4. The stages 2 and 3 (0.14/0.42/1.2 µm) show an increase and the stages 1, 4, and 5 (0.05-0.14, 1.2/3.5/10 µm) a decrease of the particle mass.

On stage 1 (0.05-0.14 µm) an increase of ions (Fig. 5) OC/EC (Fig. 8), and organic acids (Fig. 10) in terms of fraction of stage mass (%) was observed. The reason for these changes is not yet clear.

In the cloudwater the sulfate concentration agrees reasonable with the particle concentration, on the other hand nitrate, chloride, and oxalate in the cloud droplets are clearly higher concentrated (Figs. 6 and 9).

Significant lower concentrations for the alkanes and sugars were found after the cloud passage (Fig. 11 and 13). The sugars showed the highest concentrations from the observed organic compounds. The concentrations of dicarboxylic acids and terpene acids were nearly the same on both sites (Fig. 12). Trimethylpentadecanone, jasmonic acid and it methyl esters are ubiquitous in plants. They have hormone properties. These compounds are more concentrated after the cloud, except for 6,10,14-trimethylpentadekanone (Fig. 14).

Summarizing physical and chemical characterisation of particles before and after a cloud passage including the characterisation of cloud droplets as well as of the gaseous phase the FEBUKO data set represents a new quality and an important data source for the corresponding modelling project MODMEP.

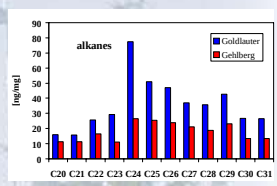

Fig. 11

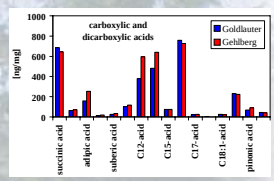

Fig. 12

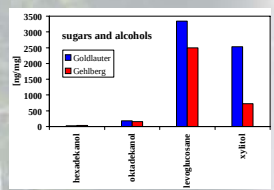

Fig. 13

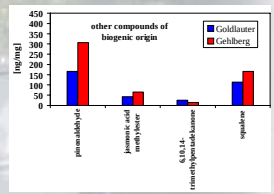

Fig. 14

Acknowledgement

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