Physico-chemical characterisation of air, particles and cloud water in cloud experiments

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Introduction

Within the joint research project FEBUKO *(field investigations of budgets and conversions of particle phase organics in tropospheric cloud processes)* two series of ground-based cloud experiments were performed in autumn 2001 and 2002. The aim of the project is to obtain a better insight into aerosol-cloud interactions and chemical conversions especially of organic compounds in tropospheric multiphase systems.

To this end, a detailed physical and chemical characterisation of air masses (particle phase, gas phase and aqueous phase) before, while and after passing an orographic cloud took place at three stations in a forestal region in Germany (Thuringian forest).

Furthermore, the results of the field studies will serve as an initialisation parameter set for complex multiphase models developed in a sister project (MODMEP) and a comparison data set for the calculated cloud water and processed aerosol composition.

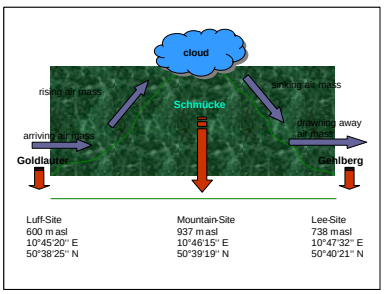

Figure 1: Schematic view of the sampling sites during the cloud experiments

Experimental

Air mass characterisation took place at the two valley sites Goldlauter (upwind) and Gehlberg (downwind) and at the top of the mountain Schmücke (937 m asl) (Figure 1).

Physical characterisation of particles:
- Twin Differential Mobility Particle Sizer (TDMPS) at the valley stations
- Hygroscopicity Tandem Differential Mobility Analyser (HTDMA) at the upwind site
- Differential Mobility Particle Sizer (DMPS) connected to a counterflow virtual impactor (CVI) and an interstitial inlet at the mountain site
- PVM (Particle Volume Monitor) for liquid water content of cloud

Sampling of atmospheric particles at both valley stations:
- Digitel DHA-80 filter sampler for $PM_{2.5}$
- Sierra-Andersen sampler for PM_{10}
- 5-stage BERNER impactors for size resolved particle sampling

Sampling of cloud water at the mountain site:
- 4 bulk cloud water collectors (CASCC2, loan from the Colorado State University, Fort Collins, USA)

Chemical characterisation of particles and cloudwater was performed using different instruments:
- mass: micro balance (UMT2, Mettler/Toledo)
- inorganic cations: ion chromatography (Metrohm)
- inorganic anions: capillary electrophoresis (thermo separation products)
- dicarboxylic acids: capillary electrophoresis (thermos separation products)
- OC/EC: thermogravimetric method (C-mat 5500, Ströhlein)
- polar VOC: GC-MS (Thermofinnigan)
- non-polar VOC: GC-MS (GC 6890, MSD 5973N, Agilent)

Results

Table 1: Top events of the measurement campaigns

Event #	Date / Time (UTC)	Sampling time (h)
1	26.10.02 8:30 – 16:30	8
2	02.10.02 8:00 – 12:45	4,75
3	26./27.10.02 22:00 –13:00	15
4	02.10.02 8:00 – 12:45	4,75

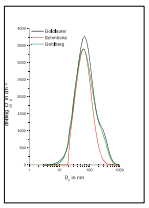

Figure 2: Number size distributions for event #1

- Within the two measurement campaigns 2001 and 2002 27 cloud events occurred which were comprised into 14 periods. However, only 4 of them came with the right conditions for an orographic cloud (Table 1).

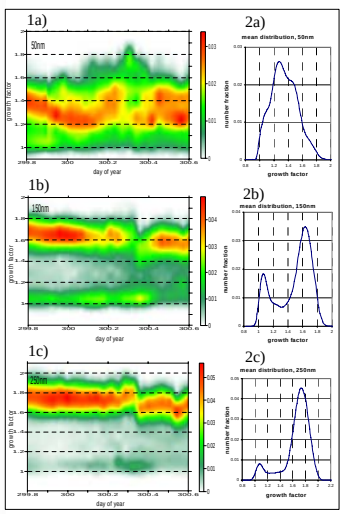

Figure 3: Hygroscopic growth factors for 26th/27th of october 2001

- Figure 2 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 cloud water onto the particle surface. Thereby the particles grow up to a size that is undetectable by the instruments used for.

- Figure 3: 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 particles was not evident for this cloud event. The peak of the growth curve for 50nm particles was found around a GF of 1.3.

- Total particle mass concentration (PM_{10}) decreases in the case of the transport from upwind side to downwind station with nearly all events (Figure 4), which is probably caused by deposition and a dilution by entrainment.

- A detailed chemical composition of size-resolved particles at the upwind site is given in Figure 5. Besides the sum parameters water soluble organic carbon (WSOC), water insoluble organic carbon (WISOC) and elemental carbon (EC), a speciation of inorganic ions, dicarboxylic acids (C2 – C5 and hydroxylated acids) and metals was performed. The water content was calculated using growth factors for every single impactor stage.

- Figure 6 shows a comparison of ion concentrations before, while and after the cloud passage of the air masses. For chloride and nitrate the cloud concentrations are substantially higher than the concentrations at the valley stations. This could be due to higher gas phase concentrations of these compounds outside the cloud or losses during impactor sampling. Sulphate and oxalate show good agreement of concentrations at all three stations.

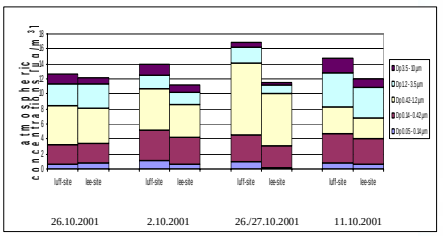

Figure 4: Concentrations of total particle mass for events #1 - 4

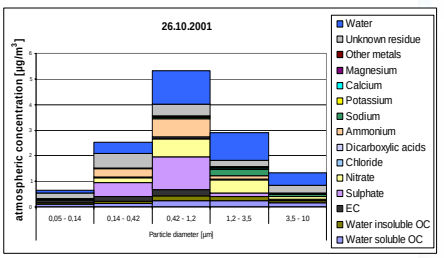

Figure 5: Chemical composition of particles for event #1

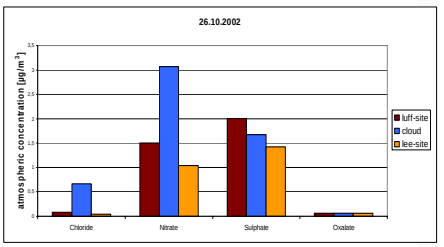

Figure 6: Comparison of concentrations upwind, in-cloud and downwind for event #1

Modelling and Outlook

As a joint effort of the cluster projects FEBUKO and MODMEP and the activities of the IFT chemistry and modelling departments the combined aerosol/cloud model SPEKMEP has been developed. It combines complex size-resolved microphysics of aerosols and clouds with complex state-of-the-art chemical mechanisms provided by the CAPRAM mechanism line, see http://www.tropos.de/CHEMIE/multimod/CAPRAM/capram.html. The model runs are initialised by the aerosol and gas phase chemical composition as experimentally determined at the Goldlauter station luff of the Schmücke hill capped orographic cloud. As a first model output, Figure 7 shows a graphical presentation of the liquid water content developing when an air parcel treated in a size-resolved manner moves from Goldlauter up to the Schmücke. Currently, the results for chemical cloudwater constituents from SPEKMEP are compared with the actual experimental cloud water measurements and process analysis and model characterisation is performed.

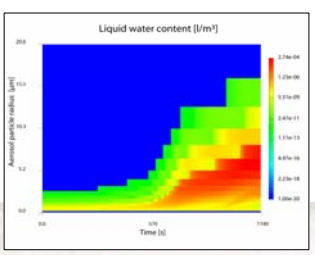

Figure 7: LWC developing from valley station to mountain site

Acknowledgment

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