PHYSICO-CHEMICAL CHARACTERISATION OF AIR, PARTICLES AND CLOUD WATER IN CLOUD EXPERIMENTS

D. v. PINXTEREN, E. BRÜGGEMANN, D. GALGON, T. GNAUK, K. LEHMANN, A. MASSLING, S. MERTES, K. MÜLLER, A. NOWAK, A. PLEWKA, A. WIEDENSOHLER and H. HERRMANN

Institut für Troposphärenforschung, Permoserstr. 15, 04318 Leipzig, Germany

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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 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 processes. 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 (i) an initialisation parameter set for complex multiphase models developed in a sister project (MODMEP) and (ii) a comparison data set for the calculated cloud water and processed aerosol composition.

EXPERIMENTAL SETUP

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).

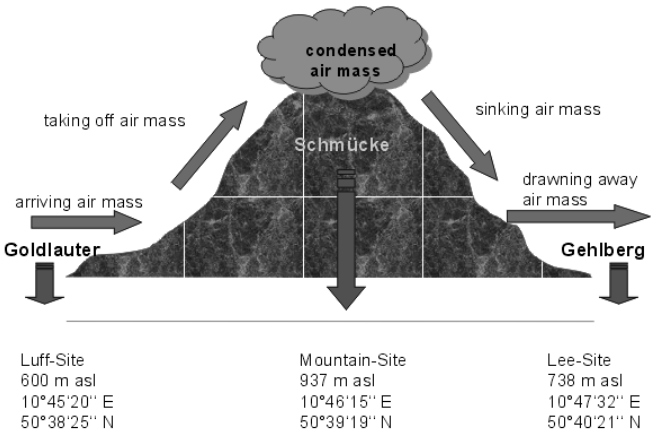

Figure 1: Schematic view of the sampling sites during the cloud experiments

For the physical characterisation of particles at both valley sites number size distributions of atmospheric particles were measured within the measurable range 3-900 nm by means of two TDMPS systems (Twin Differential Mobility Particle Sizer). Parallel to the TDMPS systems in luff a HTDMA (Hygroscopicity Tandem Differential Mobility Analyser) was operated for the determination of the hygroscopic characteristics of sub-micron aerosol particles. On the mountain site the residual and interstitial particle size distributions were measured by two DMPS systems connected to a counterflow virtual impactor (CVI) and an interstitial inlet. The liquid water content was determined by a particle volume monitor (PVM).
For the chemical characterisation of particles different filter samplers were operated (Digitel DHA-80 for $PM_{2.5}$ and Sierra-Andersen sampler for PM_{10}). The quartz filter samples of the large volume collectors served for the identification and quantification of particulate organic compounds as well as some metals. For size-resolved particle sampling BERNER impactors were applied. At each valley station two impactors, equipped with tedlar foil for ion analysis (inorganic main ions and carboxylic acids) and aluminium foil for carbon analysis (TC, EC, OC) were operated.

Cloud water was collected at the mountain site by means of 4 bulk cloud water collectors (CASCC2, loan from the Colorado State University, Fort Collins, USA) for the determination of carboxylic acids, organic single species and selected metals. Characterisation of the gas phase included measurements of O_3, NO_x, CO and SO_2 as well as sampling of volatile carbonyl compounds on DNPH (dinitrophenylhydrazine) cartridges.

FIRST RESULTS

Within the two measurement campaigns in autumn 2001 and 2002 27 cloud events occurred which were comprised into 14 periods. The longest cloud event from 26th/27th October 2001 had a duration of 15 hours. In Figure 2 averages of the particle size distribution at the three measuring sites over this cloud period are shown. Compared to the valley sites, the scavenging of particles > 100 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 water onto the particle surface. Thereby the particles grow up to a size that is undetectable by the instruments used for. The total mass concentration of particles on the impactor foils (summarized five BERNER impactor stages) is classified as low and amounts to between 11 and 17 µg m^{-3} upwind and 6 and 12 µg m^{-3} downwind for the first campaign in 2001. In the case of transport over the ridge of the Thuringian forest a loss occurs mainly by deposition and a dilution by entrainment. The distribution of particle mass shows for particles with diameter from 0.42-1.2 µm the highest mass concentration, which is characteristic for the size range considered. The effect of mass decrease can be observed for the most size ranges of all events. The total soluble mass concentration (sum of mass concentration from chloride, nitrate, sulphate, sodium, ammonium, potassium, magnesium, and calcium over all size ranges) amounts to between 3 and 7 µg m^{-3} in luff and 2 and 5 µg m^{-3} in lee, that are about 40 % in average from the total mass. The decrease of soluble mass after the cloud passage can also be observed for all events. A summary of the chemical characterisation of particles at both sites is shown in Figure 3. Summarizing physical and chemical characterisation of particles before and after a cloud passage including the characterisation of cloud droplets as well as gaseous phase the FEBUKO data set will serve as an important data source for the corresponding modelling project MODMEP.

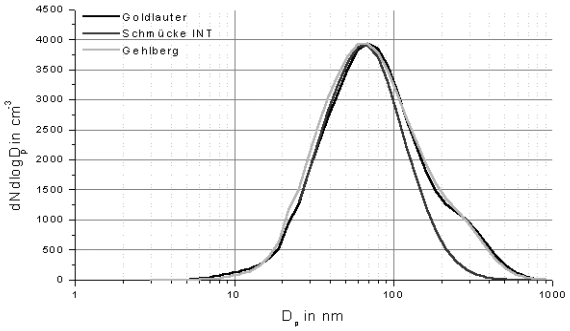

Figure 2: Size distributions during event #1 in FEBUKO 2001

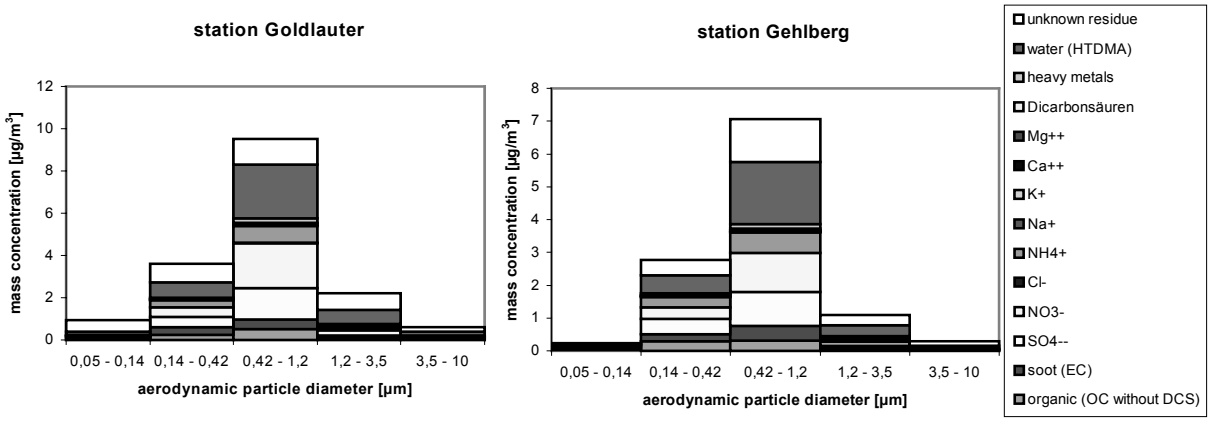

Figure 3: chemical composition of the particles during event #1 in FEBUKO 2001

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