Modelling of Tropospheric Multiphase Processes: Tools and Chemical Mechanisms (AFO2000 Join Project: MODMEP)

Guest Contribution

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Summary

Multiphase processes are of increasing importance for the understanding of tropospheric interactions. The complexities of the cloud processes involved have discouraged investigators from simultaneously treating all aspects of multiphase chemistry and microphysics with equal rigor. The description of cloud processes in the currently available box models and Eulerian grid models (cloud or mesoscale models) focuses either on detailed microphysics or complex multiphase chemistry. The chemical conversions in the liquid phase are described only in a few aggregated drop classes (e. g., Chaumerliac et al., 2000), or strongly simplified chemical mechanisms are used (e. g., Wurzler et. al., 2000). The objective of the present project is the development of a cloud module which combines a complex multiphase chemistry with detailed microphysics. The description of both components is given for a fine-resolved drop spectrum. The influence of simplifications within single components and the kind of their coupling on the simulation results will be investigated for different tropospheric situations. In the framework of the joint project, techniques will be provided and tested which allow the description of complex multiphase chemistry and of detailed microphysics in multidimensional chemistry-transport models.

An efficient numerical solution of the entire complex model strongly requires the use of new numerical methods. A major task is the development of implicit time integration methods which integrate all involved processes in a coupled manner. This includes a. o. the implementation and the test of solution methods for large, sparse, linear equation systems. An efficient solution of such systems is only possible utilizing the special structure of the system. The extension of the chemical mechanism will focus on a better description of the oxidation of dissolved organic compounds within aqueous particles. For higher organics which may partition from the gas phase, phase transfer will be described by gas phase diffusion, mass accommodation and Henry solubility. Process parameters from the FEBUKO field experiments will be taken over into the complex chemistry model. Another objective consists in the development and the application of an automated method of analysis and reduction for multiphase reaction mechanisms. This leads to the derivation of reduced mechanisms for specifiable application purposes. The developments are performed in close cooperation with the joint cloud experiment project FEBUKO (Herrmann et al.).

Objectives

Aerosol particles affect cloud microphysics (e.g. number and size of drops, precipitation formation) with their size distribution, their number and chemical composition. In addition, clouds act as chemical reaction chambers, where gases and particles are scavenged. Chemical reactions modify the concentrations of trace gases and oxidants in either phase. The phase interchange depends strongly on the phase surface area. The objective of the present joint project is the development of tools and methods for cloud models which combine a complex multiphase chemistry with detailed microphysics. The description of both components should be given for a high resolution drop spectrum. This decomposition of the droplet spectrum into classes is based on their droplet size and the amount of scavenged material inside the drops, respectively. Numerical approaches for treating such size-resolved multiphase processes have been investigated in Wolke et al. (2001a, b). An efficient numerical solution of the entire complex model strongly requires the development of new numerical methods which utilize the special structure of the system. At the end of the project, the developed multiphase reaction mechanisms and modules of different complexity can be used directly in box models or as a component in multidimensional chemistry-transport codes (cloud resolving or mesoscale models). Furthermore, estimations are provided concerning the expected numerical expense as well as the inaccuracies caused by simplifications of model components or by decoupling. Recommendations for the required model setup are given.

Methods

Coupling of detailed microphysics with multiphase chemistry in several drop classes. The fine resolution of the drop spectrum used in the microphysical cloud model will be extended to the treatment of detailed multiphase chemistry. It will be investigated how a reduction of the number of drop classes by aggregation affects the simulation results. In a first version, operator splitting will be used as coupling scheme. The aqueous phase concentrations are coupled both via the gas phase and via the liquid water transfer (coalescence, drop disintegration). The numerical integration of the multiphase system is done by an implicit integrator making use of the sparsity of the system. The microphysical parameters needed by the multiphase chemistry are supplied by the microphysical model. The sensitivity of the entire model will be analyzed.

Adapted multiphase mechanisms and tools for their implementation into complex models. The existing aqueous phase mechanisms CAPRAM 2.3 and 2.4 (Herrmann et al., 2000) already containing a detailed C_1 and C_2 chemistry will be developed towards a better process description for higher organic compounds which are identified in cloud-water and aerosols in field experiments. For higher organics not only uptake from the gas phase but additionally dissolution out of the cloud droplet precursor CCN has to be implemented as source processes. The extension of the chemical mechanism will focus on a better description of the oxidation of dissolved organic compounds within aqueous particles. For higher organics which may partition from the gas phase, phase transfer will be described by gas phase diffusion, mass accommodation and Henry solubility. Process parameters from the FEBUKO field experiments will directly be taken over into the complex chemistry model. Another objective consists in the development and the application of an automated method of analysis and reduction for multiphase reaction mechanisms. This leads to the derivation of reduced mechanisms for specifiable application purposes.

Methods for coupled time integration. Despite the strong coupling over different processes, the governing equations for dynamic, chemical and cloud parameters are commonly treated separately in chemistry-transport models. It is investigated whether and which processes can be integrated decoupled with which loss of exactness and which gain of efficiency. A major task is the development of implicit-explicit time integration methods which integrate all

involved processes in a coupled manner. This includes a. o. the development and the test of solution methods for large, sparse, linear equation systems. An efficient solution of such systems is only possible utilizing the special structure (Wolke and Knoth, 2002).

Spatial description of clouds in 3D models. Clouds are dynamic objects with high spatial and temporal variability, whose formation depends on the existence of aerosols, their physical and chemical properties and the appearance of local supersaturation. At the boundaries of the clouds a phase transition process between "dry air" and "air with cloud drops" occurs. Eulerian grid models in general do not spatially resolve the cloud boundaries. Due to the gradual representation of the cloud in a grid box, an artificial, numerically caused "smearing" of the cloud properties over the grid box appears in the area of the cloud boundaries. This has an effect both on the computation of cloud-dynamical and cloud-microphysical parameters as well as on the simulation of cloud-chemical properties which are coupled with the gas phase concentrations of the related grid cells. The main goal is the development of a method for the additional prediction of the cloud boundary inside a model grid cell (Margolin et al., 1997). Hereby, by means of the "Volume of Fluid" method, the separate computation of microphysical and chemical processes in the clouded as in the cloud-free area of a grid cell is possible.

First Results

The phase interchange depends strongly on the phase surface area. To emphasize the influence of the resolution of the droplet spectrum, the simulations are performed for different numbers of droplet classes. A log-normal size distribution of the number of droplets is assumed. The size intervals are disposed equidistant in a logarithmic scale in the range from 1 to 64 µm. For the calculation of the mass transfer coefficient the mean droplet radius of the corresponding size fraction is used. For the whole spectrum a mean radius of 10 µm is assumed. For the simulation the CAPRAM 2.4 mechanism (Ervens et al., 2002) is used.

The influence of a size-resolution of the cloud droplet spectrum onto gas phase as well as aqueous phase concentrations is illustrated in Figure 1. The switch from one fraction with a mean radius of 10 µm to a size-resolved spectrum leads to observable changes in most concentration levels. Species, for which the transport processes, i. e. gas phase diffusion and/or mass accommodation, or fast reactions within the aqueous phase determine the efficiency of phase transfer, do not reach the thermodynamic equilibrium given by the Henry´s law constant. The radicals OH and NO_3 are consumed by many reactions in the aqueous phase where the surface area controls the uptake. Their concentration in the aqueous phase would be overestimated with the exclusive approach by Henry´s law. NO in the gas phase shows the contrary behavior. One of its most important sink is the reaction with HO_2 which is transported effectively into the droplets. Because the concentration of HO_2 in the gas phase is decreasing with increasing phase surface area, more NO is remaining in the gas phase at least considering only one size class. Possibly, a similar reason leads to the change in the H_2SO_4 concentration because it is formed by SO_2 and OH.

Conclusions

The goal of the joint project is the development of suitable tools and methods for the treatment of complex cloud-chemical processes under consideration of several drop classes for parcel and multidimensional chemistry transport models (cloud resolving or mesoscale models). At the end of the project multiphase reaction mechanisms and modules of different complexity are available for the coupled simulation of multiphase-chemical and microphysical processes.

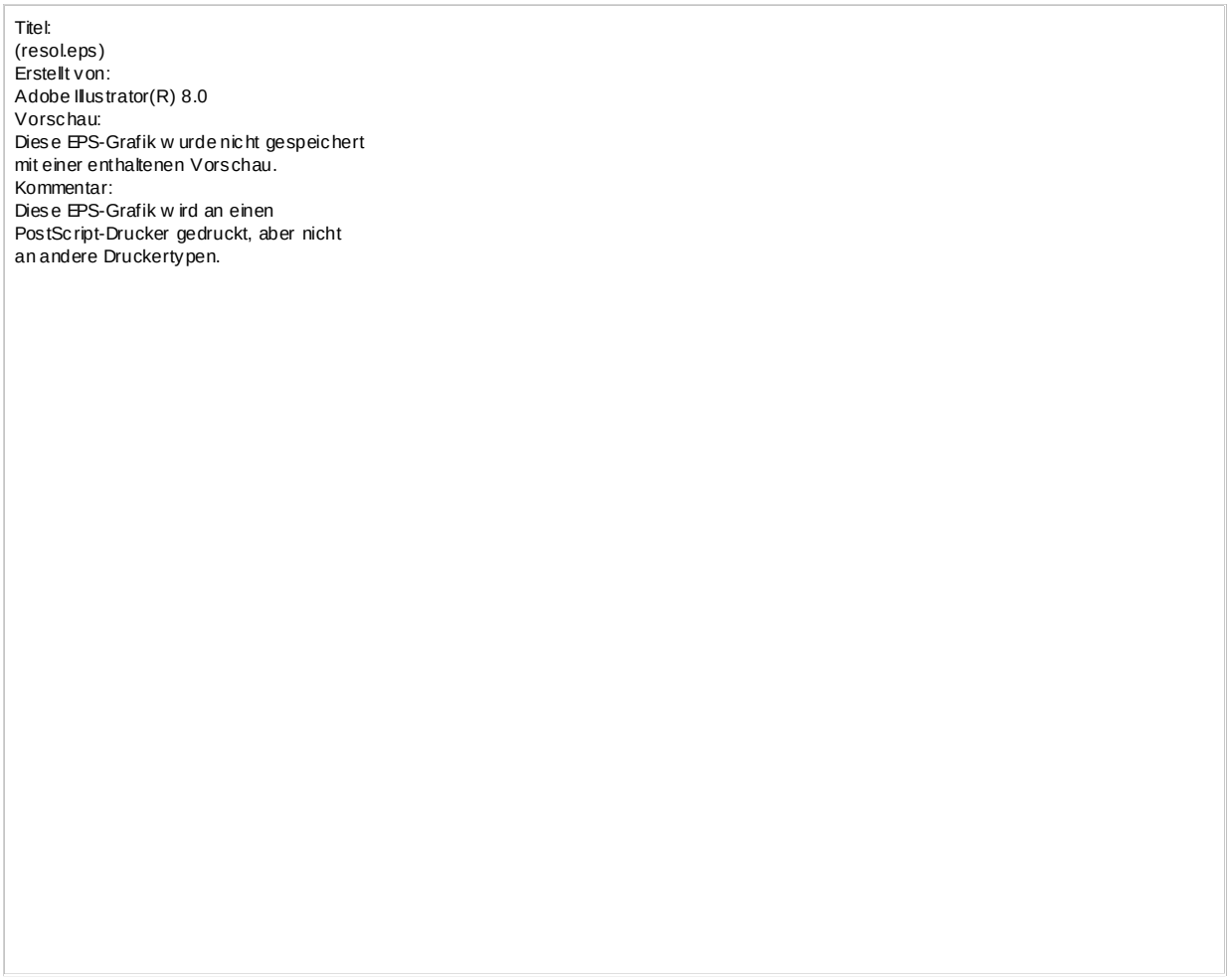

Figure 1: Concentrations of several gas phase and aqueous phase species for a simulation with different numbers of drop classes. The aqueous phase concentrations are averaged over all classes..

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