Atmos. Chem. Phys., 6, 4231-4251, 2006
www.atmos-chem-phys.net/6/4231/2006/

Columnar modelling of nucleation burst evolution in the convective boundary layer – first results from a feasibility study
Part III: Preliminary results on physicochemical model performance using two "clean air mass" reference scenarios

O. Hellmuth
Leibniz Institute for Tropospheric Research, Modelling Department,
Permoserstrasse 15, 04318 Leipzig, Germany

Abstract. In Paper I of four papers, a revised columnar high-order model to investigate gas-aerosol-turbulence interactions in the convective boundary layer (CBL) was proposed. In Paper II, the model capability to predict first-, second- and third-order moments of meteorological variables in the CBL was demonstrated using available observational data. In the present Paper III, the high-order modelling concept is extended to sulphur and ammonia chemistry as well as to aerosol dynamics. Based on the previous CBL simulation, a feasibility study is performed using two "clean air mass" scenarios with an emission source at the ground but low aerosol background concentration. Such scenarios synoptically correspond to the advection of fresh post-frontal air in an anthropogenically influenced region. The aim is to evaluate the time-height evolution of ultrafine condensation nuclei (UCNs) and to elucidate the interactions between meteorological and physicochemical variables in a CBL column. The scenarios differ in the treatment of new particle formation (NPF), whereas homogeneous nucleation according to the classical nucleation theory (CNT) is considered. The first scenario considers nucleation of a binary system consisting of water vapour and sulphuric acid (H_2SO_4) vapour, the second one nucleation of a ternary system additionally involving ammonia (NH_3). Here, the two synthetic scenarios are discussed in detail, whereas special attention is payed to the role of turbulence in the formation of the typical UCN burst behaviour, that can often be observed in the surface layer. The intercomparison of the two scenarios reveals large differences in the evolution of the UCN number concentration in the surface layer as well as in the time-height cross-sections of first-order moments and double correlation terms. Although in both cases the occurrence of NPF bursts could be simulated, the burst characteristics and genesis of the bursts are completely different. It is demonstrated, that observations from the surface layer alone are not conclusive to elucidate the origin of newly formed particles. This is also true with respect to the interpretation of box modelling studies. The binary and ternary NPF bursts observed in the surface layer differ with respect to burst amplitude and phase. New particles simulated in the binary scenario are formed in the forenoon in the upper part of the growing CBL, followed by turbulence-induced top-down transport. Hence, with respect to the burst observation site in the surface

layer, new particles are formed ex situ. In opposite to this, the ternary case reveals a much more complex pattern. Here, NPF is initiated in the early morning hours in the surface layer, when temperature (*T*) is low and relative humidity (*RH*), sulphur dioxide (SO_2) and NH_3 concentrations are high, hence new particles are formed in situ. Shortly after that, ex situ NPF in the free troposphere sets in, followed by entrainment and top-down diffusion of newly formed particles into the surface layer. Altogether, these processes mainly contribute to the formation of a strong burst in the morning hours in the ternary scenario. While the time-height cross-section of the binary nucleation rate resembles a "blob"-like evolution pattern, the ternary one resembles a "sucking tube"-like pattern. The time-height cross-sections of the flux pattern and double correlations could be plausibly interpreted in terms of CBL turbulence and entrainment/detrainment processes both in the binary and in the ternary case. Although the present approach is a pure conceptual one, it shows the feasibility to simulate gas-aerosol-turbulence interactions in the CBL. Prior to a dedicated verification/validation study, further attempts are necessary to consider a more advanced description of the formation and activation of thermodynamically stable clusters according to modern concepts proposed by Kulmala et al. (2000), Kulmala (2003) and Kulmala et al. (2004a).

Final Revised Paper (PDF, 10897 KB)   Discussion Paper (ACPD)

Citation: Hellmuth, O.: Columnar modelling of nucleation burst evolution in the convective boundary layer – first results from a feasibility study
Part III: Preliminary results on physicochemical model performance using two "clean air mass" reference scenarios, Atmos. Chem. Phys., 6, 4231-4251, 2006.   Bibtex   EndNote   Reference Manager