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Columnar modelling of nucleation burst evolution in the convective boundary layer – first results from a feasibility study
Part IV: A compilation of previous observations for valuation of simulation results from a columnar modelling study

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Abstract. In the preceding Papers I, II and III a revised columnar high-order modelling approach to model gas-aerosol-turbulence interactions in the convective boundary layer (CBL) was proposed, and simulation results of two synthetic nucleation scenarios (binary vs. ternary) on new particle formation (NPF) in the anthropogenically influenced CBL were presented and discussed. The purpose of the present finishing Paper IV is twofold: Firstly, an attempt is made to compile previous observational findings on NPF bursts in the CBL, obtained from a number of field experiments. Secondly, the scenario simulations discussed in Paper III will be evaluated with respect to the role of CBL turbulence in NPF burst evolution. It was demonstrated, that completely different nucleation mechanisms can lead to the occurrence of NPF bursts in the surface layer, but the corresponding evolution patterns strongly differ with respect to the origin, amplitude and phase of the NPF burst as well as with respect to the time-height evolution of turbulent vertical fluxes and double correlation terms of physicochemical and aerosoldynamical variables. The large differences between the binary and ternary case scenario indicate, that ammonia (NH_3) can not be considered as a time-independent tuning parameter in nucleation modelling. Its contribution to the evolution of the NPF burst pattern is much more complicated and reflects the influence of CBL turbulence as well as the strong non-linearity of the ternary nucleation rate. The impact of water (H_2O) vapour on the nucleation rate is quite varying depending on the considered nucleation mechanism. According to the classical theory of binary nucleation involving H_2O and sulphuric acid (H_2SO_4), H_2O vapour favours NPF, according to the classical theory of ternary nuncleation involving H_2O, H_2SO_4 and NH_3 and according to organic nucleation via chemical reactions involving stabilised Criegee intermediates (SCIs), H_2O vapour disfavours nucleation, and according to the parameterisation of the collision-controlled binary nucleation rate proposed by Weber et al. (1996), H_2O vapour does not explicitly affect the particle formation. Since the H_2SO_4 concentration is overpredicted in the simulations presented in Paper III, the nucleation rates are too high compared to previous estimations. Therefore, the results are not directly comparable to measurements. Especially NPF events, where organics are suspected to play a key role, such as those observed at the boreal forest station in Hyytiälä (Southern

Finland) or at Hohenpeissenberg (mountain site in Southern Germany), can not be explained by employing simple sulphur/ammonia chemistry. However, some valuable hints regarding the role of CBL turbulence in NPF can be obtained. In the literature a number of observations on the link between turbulence and NPF can be found, whose burst patterns support a strong contribution of CBL turbulence to the NPF burst evolution simulated here. Observations, that do not correspond to the scenarios are discussed with respect to possible reasons for the differences between model and observation. The model simulations support some state-of-the-art hypotheses on the contribution of CBL turbulence to NPF. Considering the application of box models, the present study shows, that CBL turbulence, not explicitly considered in such models, can strongly affect the spatio-temporal NPF burst evolution. The columnar high-order model presented here is a helpful tool to elucidate gas-aerosol-turbulence interactions, especially the genesis of NPF bursts in the CBL. An advanced description of the cluster formation and condensation growth is required as well as a comprehensive verification/validation study using observed high-order moments. Further scenario simulations remain to be performed.

Final Revised Paper (PDF, 395 KB) Discussion Paper (ACPD)

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