Ultrafine Aerosol Dynamics Module for Modal Aerosol Dynamic Model for Europe

A contribution to subproject GLOREAM

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Summary

The treatment of nucleation is an extremely important part of any atmospheric aerosol model. Nevertheless, formulations of binary nucleation rates of water and sulphuric acid have not been in good agreement with measured nucleation rates. Additionally, the understanding of the importance of nucleation on a regional scale is still limited.

New nucleation mechanisms, e.g. ternary nucleation of sulphuric acid-ammonia-water, have been recently suggested as a possible explanation of the observed atmospheric nucleation events. These mechanisms should be parameterised and tested in different aerosol dynamical models to investigate their applicability and to compare them with atmospheric measurements. Therefore, we have continued the development of the Modal Aerosol Dynamic model for Europe (MADE) including an improved representation of the nucleation and the freshly nucleated particles.

Preliminary tests in a zero and one-dimensional version of MADE showed that several changes to the numerical solution method and the modal structure were necessary. The results are compared with results of a sectional model.

Introduction

Atmospheric particulate matter scatters sunlight, serves as cloud condensation nuclei for the formation of cloud droplets and is strongly linked to numerous air pollution phenomena. Furthermore, atmospheric particulate matter is connected to global climate change, but its total effect is not yet well understood. Not surprisingly, formation of new atmospheric particles has received growing interest recently.

On a regional scale the transport and development of aerosol particles can be studied with different physic-chemical models. Chemical transport models (CTM), which include aerosol dynamics, are a valuable tool to analyse the effect of different sources and processes.

Formation of new aerosol particles directly from gas phase, i.e. nucleation, is an important source of particles in the atmosphere. A range of nucleation pathways in the atmosphere (e.g. H_2SO_4-H_2O systems) have been proposed in the past. However, these formation pathways are often unable to produce the observed nucleation rates. Lately, some new and improved nucleation schemes have become available, e.g. ternary nucleation of the H_2SO_4-NH_4-H_2O system (Korhonen *et al.*, 1999) and theory of thermodynamically stable clusters (Kulmala *et al.*, 2000).

The simulation of atmospheric particle nucleation requires taking in account the dynamics of nanometer-scale aerosol particles. Several CTM parameterise these dynamics and assume that particles behave in a classical manner within the time frames associated with the simulations. The results can be misleading especially on small to medium time scales.

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Objectives

The overall objective is to investigate and evaluate the importance of atmospheric particle nucleation on the regional scale. Therefore, detailed subroutines to handle relevant aerosol dynamics are developed applying new nucleation parameterisations. To determine the applicability of the new nucleation schemes on the regional scale the MADE/EURAD model system is used. The model it is required to represent the aerosol dynamics of freshly formed nanometer scale particles. Additionally, parameterisations and numerical formulations are required to have minimal effect to calculation time. The development of suitable parameterisations is therefore also an objective of the project. In the future, the model will be applied to conduct a comparison with measurements of aerosol particle concentrations and size distributions for the measurement site Hyytiälä in central Finland (Kulmala and Hämeri, 2000).

Activities

The MADE (Ackermann *et al.*, 1998) aerosol dynamical model assumes that the total aerosol particle size distribution can be described by a predetermined number of aerosol populations (modes). In the model the modes are assumed to have lognormal size distribution. This choice of the modal structure allows significantly faster calculations than other representations of the size distribution and provides the additional advantage of analytical solutions for most of the required operations (Binkowski, 2000).

During the new development of the ultrafine aerosol dynamics module for MADE special emphasis was put on a correct behaviour of the model at the nucleation event and the subsequent particle growth. The model results will be compared with results of other models and atmospheric measurements.

Results

Zero-dimensional box simulations give a good representation on the development of the aerosol particle size distribution in a single transport time step. In Figure 1 a schematic overview of some of the possible main problems regarding the nucleation treatment within a modal model formulation. MADE usually consists of two dynamic modes (Aitken- and accumulation mode) representing sub micron particles and one mode for coarse particles.

During a strong nucleation event a comparatively large amount of particles is formed in the nanometer size range. If these particles are simply added to the Aitken mode the distribution usually cannot describe both aged particles and the newly formed particles very well (curve A in Figure 1). In general, this leads to abnormal large geometric standard deviations and a poor representation of the physical situation.

Another problem arises with the treatment of condensing material on the freshly nucleated particles (point B in Figure 1). Condensation is an important process for the nucleated particles, because by this process particles can grow to larger sizes. In the current model formulation the Kelvin effect is neglected for submicron particles. However, for freshly nucleated nanoparticles this effect has to be taken into account. Therefore, the condensation formulation had to be updated in the model. The condensation integrals are now solved numerically using Gauss-Hermite integration.

Since nucleation and related processes can change the aerosol size distribution rather fast, the solution methods of the differential equations solution also required consideration. The relatively large mass-transfer rates during a nucleation event require small time steps, since otherwise the solution can be inaccurately (point c in Figure 1). In the current model formulation this is taken into account by a variable time step scheme.

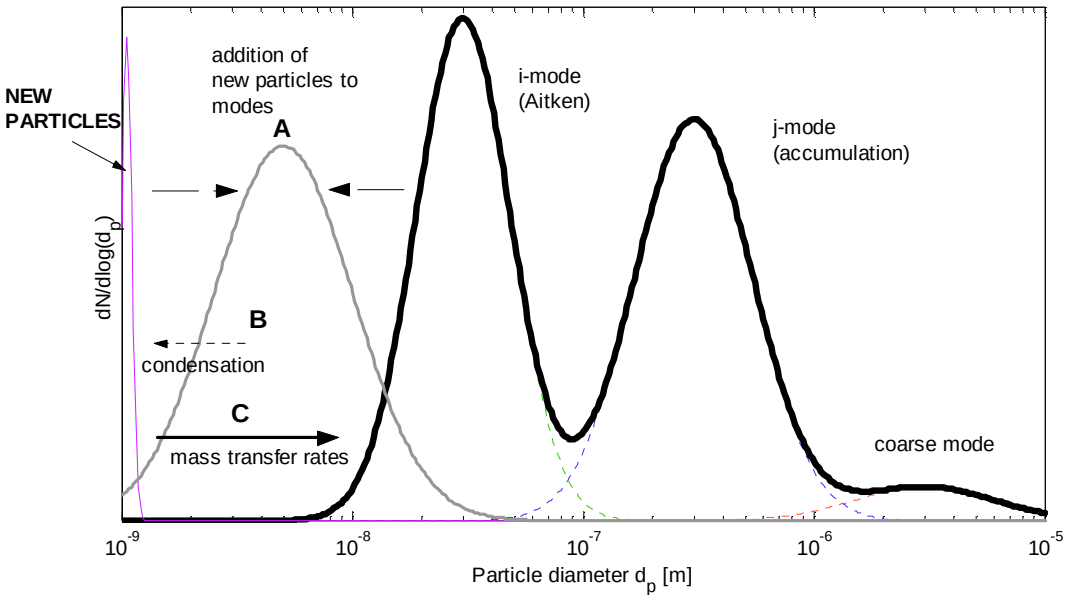

Figure 1. Schematic overview of problems associated with nucleation in a modal aerosol model.

Conclusions

Several key conclusions can be drawn from the preliminary tests and development in 0-d and 1-d versions of MADE. Additionally, the most important changes for the aerosol routines can be summarised as:

- Variable standard deviations are used for all fine particle modes (except coarse mode).
- Addition of a mode to account for newly nucleated particles. It is assumed that freshly nucleated particles form a narrow lognormal mode ($\sigma \approx 1.05$) with a median diameter of 1 nm. This particle mode would then work as a ”transport mode” to the other modes due to condensation and coagulation processes.
- The numerical solution time-step should be variable to take care of rapid changes due to nucleation events.
- Some changes of the equilibrium approach have been included to parameterise the Kelvin effect

Our results indicate, that a more detailed representation of freshly nucleated particles requires significant changes to the common model description of particulate matter within regional scale air quality models. These aspects should be taken into account while simulating nucleation phenomena.

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