Atmos. Chem. Phys. Discuss., 4, S471–S476, 2004
www.atmos-chem-phys.org/acpd/4/S471/

Atmospheric
Chemistry
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Interactive comment on "Multicomponent aerosol dynamics model UHMA: model development and validation" *by* H. Korhonen et al.

H. Korhonen et al.

Received and published: 13 April 2004

Response to comments by Reviewer #1:

Page 5, section 2.2.4: As the reviewer points out, detrainment and entrainment of particles can effect the size distribution notably and thus play a role in the onset of new particle formation. Including these processes into the model is relatively simple and we have added them into the list of processes that the model is capable of simulating.

Page 5, section 2.3: We agree with the reviewer that a simple figure presenting the main differences of the size distribution methods used could be helpful to non-expert readers; such a figure was in fact originally planned to be included in the manuscript. Coming up with a simple and non-confusing way to highlight the differences in a figure is, however, very challenging due to e.g. different number of time steps taken between the splitting of particles between the sections. We decided, therefore, to leave the

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figure out and explain the methods in detail in the text. These explanations together with references to original work presenting the methods should enable also the non-expert reader to follow the manuscript.

Page 6, section 3.1: The following comments addressing the points raised by the reviewer have now been added to the manuscript at the end of paragraph 3 and before the last paragraph of the section:

ŞAlthough only one set of initial conditions is presented here, the conclusions of the comparison of the methods are valid for new particle formation simulations in general.Ť

ŞWhen logarithmic spacing for size sections is used, the problems associated with numerical diffusion and additional dents in the size distribution are most pronounced for the nucleation mode particles. A denser size grid in this region would eliminate numerical diffusion to some extent but at the same time increase the computational burden of the code; as explained above, a denser grid would not, however, smooth out the particle distribution when the moving center approach is used. Another solution to the problems encountered in the nucleation regime could be an accurate parameterization which takes into account not only nucleation but also the early stages of particle growth and thus essentially predicts the appearance rate of particles at a larger size (e.g. at 5 nm). In addition, such a parameterization would reduce the number of size sections needed to describe the smallest particles and could therefore decrease the computational cost of the code.Ť

Page 7, section 3.2:

The organic vapours responsible for the growth of newly formed particles in the ambient atmosphere have not been identified and the oxidation path of their precursors is not known. In this study, we have therefore assumed a semisinusoidal production rate peaking at day time for the condensable vapours; an assumption which implicitly indicates that photochemical reactions control their production. Assuming the semisinusoidal behaviour, the organic vapour concentrations were optimized Ű making sure the concentrations agreed with estimates for boreal forest sites Ű to reproduce the growth rates observed. This is now explained in section 3.2.

As stated in the abstract, the model captures the key qualitative behaviour of the observed system well. One reason for the quantitative differences in the observed and simulated particle evolution for nucleation mode particles lies in numerical diffusion as pointed out by Lehtinen and Kulmala (2002). This is now explained in section 3.2.

Taking the possible dilution effect of the pre-existing particle concentration into account in the model simulations would decrease the sink for the nucleating and condensing vapours. Overestimation of the condensation sink during the simulation can in fact be one explanation why the simulated growth rate for nucleation mode particles is lower than the observed one. Capturing the observed process that seems to first affect Aitken mode particles and only some hours later accumulation mode particles is, however, difficult Ű if not impossible Ű in the model based mainly on first principles. This is now explained in section 3.2.

In the last paragraph of section 3.2 it now reads: ŞĚThe dark blue colour indicates that either no particles exist in the size section or, as is the case with the very smallest particles, the existing particles contain no organic matter. The simulation results suggests that the nucleated particles grow by condensation of sulphuric acid until the size of about 2 nm after which an organic vapour described by nano-Kohler mechanism begins to condense onto them. Once the particles have reached a diameter of 3-4 nm, half of their volume may consist of organic matter whereas the organic volume fraction in 10 nm particles may exceed 0.8.Ť

Technical corrections suggested by the reviewer were made.

The reason for using the ammonia concentration 0.1 ppt as a border value lies in the validity range of the ternary parameterization; this is now explained in the text. As the nucleation rates corresponding to ammonia concentrations below this value are very small, this assumption does not introduce errors in the model runs.

Response to comments by Reviewer #2:

Additional remark:

Although we agree with the reviewer that the thermodynamics used and assumptions made in nano-Kohler theory are important considering simulation of the initial steps of the particle growth in the model, they are fairly complex and we feel that a detailed description of them here would unnecessarily burden the reader and distract from the main topic of the paper. The treatment of activity coefficients, surface tensions and other details regarding the condensation of organic vapours in the context of this theory are reported in Kulmala et al. (2004a) which is now out in Journal of Geophysical Research. Calculation of the diffusion coefficient follows Reid et al. (1987) which is now mentioned in the text.

Specific comments:

Page 478-479: The calculation of equilibrium and organic vapour pressure is based on earlier work as explained in the original manuscript; the former follows a parameterization by Napari et al. and the latter is based on Nano-Köhler theory (Kulmala et al., 2004a). To further clarify the treatment of condensation in the model, it now reads explicitly at the end of first paragraph in section 2.2.1 that water and ammonia are the species assumed to be in equilibrium and that the condensation of all other species is treated dynamically. Regarding sulphuric acid condensation, we have added a sentence to the end of section 2.2.1, 4th paragraph stating that sulphuric acid saturation pressure is assumed negligible.

Page 482, lines 13-22: The description of coagulation has been reformulated as suggested by the reviewer.

Page 438, line 1: The reviewer suggests that the cause-effect relation between accumulation mode deposition velocity and deposition mechanism efficiencies should be reversed. We find this comment confusing since the reason for the deposition velocity minimum in the accumulation mode range clearly is that the Brownian mechanism (effective for small particles), and inertial impaction and gravitational settling (effective for large particles) cease to be effective in this size range Ű not vice versa.

Page 438, lines 9-10: The size dependent semiempirical deposition model presented in Rannik et al. (2003) was applied to particles smaller than 10 nm by using the three empirical parameters (which determine the level of collection curve, the diameter of minimum collection efficiency, and the steepness of increase in collection efficiency after the minimum) derived from the measurements of 10-500 nm particles. This is now explained in more detail in section 2.2.4.

Page 486, line 9: The reason for the partitioning of higher volatile organic species into large pre-existing particles rather than small nucleated particles lies in the Kelvin effect.

Page 486, lines 24->: A comparison of surface and volume concentrations given by the three methods supports essentially conclusions made based on Figures 1 and 2. New figures were therefore not added but the results of these comparisons are now explained at the beginning of paragraph 4 in section 3.1.

Page 489, line 1->: The results from the other two methods agree with the ones presented in the manuscript which is now mentioned in the text. The reason for presenting the results from the hybrid structure (i.e. the structure that performed the worst in the comparison in section 3.1.) is purely technical as the results from the other two structures are not easy to compare with DMPS checkerboard plots such as in Fig. 5: The moving center structure frequently produces dents and respective peaks as discussed in the text and thus does not show a uniform growth in the checkerboard plot. The same is true for the retracking method as the retracking once an hour redistributes the particles to the fixed grid. This is now explained in the text but as a comparison of the three methods has already been presented in 3.1., we feel that additional figures here would not provide any new information.

The technical corrections suggested by the referee were made.

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