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Atmospheric
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Discussions

Interactive comment on "Simulating ultrafine particle formation in Europe using a regional CTM: contribution of primary emissions versus secondary formation to aerosol number concentrations" *by* C. Fountoukis et al.

C. Fountoukis et al.

spyros@chemeng.upatras.gr

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This paper discusses a modeling study of aerosol particle formation and evolution with a sectional 3-D model. The paper is generally well written and has important new information. Below are some requested edits and calculations.

(1) *Introduction. Mention that secondary particles also form by in cloud processing of aerosols followed by cloud drop evaporation, releasing a modified aerosol.*

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This is now mentioned in the revised manuscript.

(2) *P. 13588. "…assuming a density of 1.4 g/cm3." Is this a constant density assumed for all particles of all sizes and for all times? What about when soil dust particles are present in larger size bins?*

No, the density is not constant. The aerosol density is calculated and updated continuously in each size bin based on the particle composition. The value of 1.4 g/cm^3 mentioned in the original text was indicative to estimate the corresponding size range. To avoid confusion we have deleted this statement.

(3) *P. 13588. "For the integration of the differential equations during nucleation…" Does this mean that nucleation is operator split from condensation? If so, please state explicitly.*

Nucleation and condensation rates are simulated together using the pseudo-steady-state approximation (PSSA) for sulfuric acid vapor (Pierce and Adams, 2009). We have re-phrased this sentence to avoid any misunderstandings.

(4) *P. 13588. "Condensation of ammonia…" Ammonia is more likely to dissolve in solution than condense. Is its dissolution treated? What about crystallization to ammonium nitrate or ammonium sulfate?*

Ammonia dissolution as well as formation of ammonium salts is treated by making use of the ISORROPIA (Nenes et al., 1998) aerosol thermodynamic model.

(5) *P. 13588. "We limit the ammonia condensation flux in each time step to prevent condensing more than what is available…" Shouldn't the flux also be constrained by the pH of solution in drops of each size, which itself is a function of all other acids and* *the condense or dissolve (e.g., Jacobson, Aerosol Science, and Technology 39, 92-103, 2005). Even if this is not included, it would be useful to clarify what the ammonium concentration in solution is a function of.*

This is actually constrained by the ISORROPIA model. We have now deleted this sentence to avoid any misunderstandings.

(6) *P. 13592. "The first two days of each simulation were excluded..." Did you check whether results from the first two days are really inaccurate compared with other days or were they just automatically excluded? I would suggest to check whether there really was much error.*

We did check the results from the first two days and no major problems were evident compared to the other days. Despite that, we decided to be cautious and exclude these first 2 days to minimize any influence of the rather arbitrary initial conditions on our results.

(7) *P. 13597. What was the operator-splitting time step used and what was the order of calculation?*

Based on results of Jung et al. (2006) the operator-splitting time step used here is 300 s (which corresponds to the master time step of PMCAMx-UF). The calculations follow in general the order of gas-phase chemistry – nucleation – coagulation – condensation. More specifically, in the operator-splitting scheme, the condensation of ammonia is simulated for $\Delta t/2$ (where Δt is the operator time step) and then particles are allowed to coagulate for $\Delta t/2$. In the next step, the gas-phase chemistry and nucleation equations are integrated. Finally the overall step ends with calculation of coagulation for $\Delta t/2$ and condensation for $\Delta t/2$.

(8) *P. 13597. Since the authors are operator splitting nucleation and growth, I think it is important to do the following two sensitivity tests: 1) Solve condensation before rather*

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than after nucleation each time step to see whether the depletion of sulfuric acid (and ammonia) during condensation reduces the nucleation rate. 2) Reduce the operator-split time step between nucleation and condensation to see whether increased competition reduces the nucleation rate.

This is not applicable to the current structure of the model (see also reply to previous comment for the order of calculation). As stated in the text, the two species (i.e. sulfuric acid and ammonia) are treated independently in the aerosol microphysics scheme. For sulfuric acid vapor, nucleation and condensation rates are calculated simultaneously. We have included some text explaining this and also stating that a different model structure (e.g. Jacobson, 2002) could also be used.

(9) *Table 2. Use the same number of significant digits for the model and observations.*

Done.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 13581, 2012.