

Interactive comment on "The role of organic condensation on ultrafine particle growth during nucleation events" by D. Patoulias et al.

Anonymous Referee #1

Received and published: 3 February 2015

General Comments:

The authors present an updated version of the Dynamic Model for Aerosol Nucleation (DMAN) model, DMANx which includes the condensation of organic vapors on particles and the Volatility Basis Set (VBS) framework.

The paper is certainly within the scope of ACP and I would recommend publication, following clarification on the below, only minor issues. The manuscript is reasonably well written, but I would recommend that the authors take another read through the discussion sections to see if they can improve the clarity and readability.

Specific / Minor Comments:

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p30769, line 23: the statement "Sulfuric acid is assumed to be in pseudo-steady state in DMANx" is not particularly useful by itself, you could add that you are talking about the concentration of sulfuric acid and referring to a steady state with its source (oxidation of SO2) and sinks (nucleation and condensation)

p30763, lines 27-29: I would recommend adding two more recent references to this list: Riccobono et al. (2014) for organics and Almeida et al. (2013) for amines.

p30770, line 22: this section is called "Aging of OA" and the process is referred to as such (although elsewhere you also mention "biogenic chemical aging" and "chemical aging of biogenic SOA"), but you're talking about chemical gas-phase aging so it's not really the OA that is aging? I think it's important to distinguish because there could (in reality) be other processing in the particle phase that would need to be referred to as aging of OA. I would recommend deciding on one term, defining it, and using consistently throughout.

p30772, line 22: it would be useful to have further details on what constitutes a "typical spring day" at these locations, given that the meteorological inputs have come from observations – how did you choose what to use?

p30772, lines 23-25: This is slightly confusing and would benefit from clarification: the number size distribution is taken from observations, so do you just assume that each size bin contains half organics and half ammonium sulfate, by mass? Could this have been constrained by these, or any, observations?

p30773, line 7 and Figures 1 & 2 (b), 4 (a), 6 (a): What size are the "initial / new / fresh" particles that you are tracking the composition of. In Section 2.1 you say "it is assumed that the nucleated particles consist of NH4HSO4", so at what size do your new particles "appear"?

p30775, lines 6 – 11: you could move these extra details into Section 2.6 and have a more complete description there; it's slightly confusing to refer to the same rate con-

stant twice but differently i.e., $1 \times 10-11$ (in Section 2.6) and $10 \times 10-12$ (in Section 4.3). You should also mention the fact that you are generating these low-volatility organics assuming a reaction with OH, whereas the results of Ehn et al. (2014) were based on ozonolysis reactions.

p30776, line 13: you could state here that you have set $\sigma = 0$ N/m, since this is how you discuss the surface tension in Section 4.2

p30776, lines 18-19: What are you basing this statement on? Figure S5 just tells you about the size of the particles

p30777, lines 7-8: again, I'm not sure that you have shown that? The Figures just show the mass fraction and the increasing contribution of organics over time, but we don't know anything about the actual mass of the ultrafine particles? You could replace "mass" with "size"

p30777, line 10: a significant reduction relative to what? In this paragraph you could add some citations to literature that discusses these processes as I don't think you can demonstrate this from your own simulations.

p30778, line 9: this seems like a smaller increase in N100, rather than a "similar increase" due to sulfuric acid

p30778, line 13: you could add some discussion here on the reason for the shorter delay before the onset of the N100 increase in the no organics case, compared to Hyytiala?

Technical Comments:

p30763, line 15: consider rephrasing "damage can be bigger" with "damage can be greater"

p30766, line 7: replace "Basic" with "Basis"

p30770, line 14: replace "Basic" with "Basis"

C11834

p30771, line 8: replace "test" with "tested"

p30773, line 19: suggest replacing "in the end of the day" with "at the end of the day"

p30774, line 17: insert "new" or "newly formed" before "particles"

p30775, line 11: the Murphy et al. (2012) reference is missing from the reference list, should it be Murphy et al. (2009) instead?

p30776, line 7-9: suggest rephrasing here (for ease of reading) to something like: "During the day the organic mass fraction increases, reaching a maximum of 45% of the nucleated particle mass and consisting of 30% low-volatility and 70% semi-volatile organics."

p30777, line 3: replace "and" with a comma?

p30778, line 6: are you referring to the daily mean number concentration?

p30778, line 18: suggest replacing "in the end of the day" with "at the end of the day"

p30779, line 25: you need to say what this increase is relative to (i.e., a simulation without organics), otherwise it could be interpreted as an increase over time

p30798, Figure 7: It would be useful to have a legend to indicate what the different colored lines mean, rather than having to read this in the caption. Caption: remove space between "B" and "lack" in second sentence.

Comments on Supplementary Material:

p39, Figure S4: Based on the text I think this caption should say Finokalia, rather than Hyytiala?

Figure S5 - S8: For clarity, I would add to these captions that these simulations do not include the chemical aging of biogenic oxidation products.

References:

Almeida, J. et al., Molecular understanding of sulphuric acid-amine particle nucleation in the atmosphere, Nature, 502, 359-363, doi: 10.1038/nature12663, 2013.

Riccobono, F. et al., Oxidation Products of Biogenic Emissions Contribute to Nucleation of Atmospheric Particles, Science, 344, 717-721, doi: 10.1126/science.1243527, 2014.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 30761, 2014.

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