Response to Referee #1

General Comments:

1. 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.

We do appreciate the positive assessment. We have made several changes in the revised text in an effort to further improve it and avoid any misunderstandings. These changes are described below.

Specific / Minor Comments:

2. 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).

We have rephrased this sentence to clarify that the sulfuric acid concentration is calculated by assuming that its production rate (oxidation of sulfur dioxide) is equal to its consumption rate (condensation and nucleation).

3. 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.

We have added the recommended references.

4. 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.

This is a valid concern. We now use the term “chemical aging of SOA precursors” throughout the revised manuscript.

5. 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?

Using the available measurements at Hyytiala in April 2007 we have selected the days with nucleation events and estimated the observed average diurnal profile of the various meteorological variables. We did the same for Finokalia for the period of May 2008. The corresponding average profiles were assigned to the typical day. We have now added this information in the revised text to explain better the approach that we used to construct the “typical” nucleation day for each location and season.

6. 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?

That is correct. For the initial distribution that was used as input in the model we assume that each size bin contains half organics and half ammonium sulfate. Our results are not especially sensitive to this assumed initial composition; the initial particle size distribution is a lot more important. We do clarify this point in the revised manuscript.

7. 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”?

The newly formed particles have a diameter of 1 nm and consist of ammonium bisulfate. We have added this information to the text.

8. 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 constant twice but differently i.e., 1 x 10-11 (in Section 2.6) and 10 x 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.

We have moved these details to Section 2.6. We have changed the rate constant into $1\text{x}10^{-11}$ cm^{3} $molec^{-1}$ s^{-1} and clarified the differences with the Ehn et al. (2014) mechanism.

9. 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.

We have added the suggested information.

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

We have changed the sentence "The mass concentration of new particles increases due to the condensation of organics" to "The size of the new particles increases due to the condensation of organics."

11. 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".

To be more precise, we have replaced the word "mass" with "size" in this sentence.

12. 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.

The "significant reduction" refers to the number concentration of small particles and is relative to the case without condensation of organic vapors. We have now added this in the text as well as some references for the discussed coagulation probability (Kuang et al., 2009; Westervelt et al., 2013; 2014) as suggested by the reviewer.

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

We have replaced the phrase "a similar increase" with "a smaller increase".

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

This is due both to the importance for sulfuric acid for the growth of particles to sizes larger than 100 nm in Finokalia but also to the faster photochemistry (much higher OH levels). We have added the corresponding discussion in the text.

Technical Comments:

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

Rephrased.

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

Done.

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

Done.

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

Done.

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

Done.

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

Done.

21. p30774, line 17: the Murphy et al. (2012) reference is missing from the reference list, should it be Murphy et al. (2009) instead?

We have corrected the typo replacing "Murphy et al. (2012)" with "Murphy et al. (2009)".

22. 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.”

Done.

23. p30777, line 3: replace “and” with a comma?

Done.

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

Yes, we refer to the daily mean number concentration. We have added this information.

25. p30778, line 18: suggest replacing “in the end of the day” with “at the end of the day”

Done.

26. 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.

We have rephrased this sentence to clarify that the increase is relative to a simulation in which the condensation of organics is neglected.

27. 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.

We have added the legend and corrected the typo.

Comments on Supplementary Material:

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

We thank the reviewer for this correction. We have replaced “Hyytiala” with “Finokalia”.

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

Done.

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.

Westervelt, D. M., Pierce, J. R., and Adams, P. J.: Analysis of feedbacks between nucleation rate, survival probability and cloud condensation nuclei formation, Atmos. Chem. Phys., 14, 5577-5597, doi:10.5194/acp-14-5577-2014, 2014.

Westervelt, D. M., Pierce, J. R., Riipinen, I., Trivitayanurak, W., Hamed, A., Kulmala, M., Laaksonen, A., Decesari, S., and Adams, P. J.: Formation and growth of nucleated particles into cloud condensation nuclei: model-measurement comparison, Atmos. Chem. Phys., 13, 7645–7663, 10.5194/acp-13-7645-2013, 2013.

Kuang, C., McMurry, P. H., and McCormick, A. V.: Determination of cloud condensation nuclei production from measured new particle formation events, Geophys. Res. Lett., 36, L09822, doi:10.1029/2009GL037584, 2009.

Response to Referee #2

1. This manuscript introduces a new version of a previously-published aerosol dynamic model and applies it to two environments in order to get new insight into the role of organic vapor condensation in the growth of small particles formed by atmospheric nucleation. The paper is original, scientifically sound and relatively well written. There are a few issues (see below) that required some rewriting of the text. After these minor modifications, I recommend accepting this paper for publication in ACP.

We do appreciate the positive assessment of our work.

2. The description and discussion of chemical aging reactions needs to be improved in the manuscript to avoid confusion. The authors use the terms "OA aging" or "SOA aging" (in one place even "biogenic aging" which is definitely incorrect) for reactions that essentially convert gaseous semi-volatile organic vapors to less volatile ones. In that respect, it is not really OA or SOA that ages but their precursors. I understand that the commonly used terminology is not well established, yet there is a clear danger that the readers not familiar with this topic misunderstand "OA aging" to mean e.g. heterogeneous reactions taking place in the particulate phase. I encourage the authors to reconsider the used terminology (e.g. "aging of SOA precursors" or something like that) are rewrite parts of the text accordingly. It might also worth considering combining sections 2.5 and 2.6 to explain the series of processes leading to SOA formation in one package (what happens in gas phase, how this leads to partitioning and SOA formation).

Following the reviewer's suggestion we define and use the term "chemical aging of SOA precursors" consistently throughout the revised manuscript. Furthermore, we combined sections 2.5 and 2.6 in one section and also added more text to describe in more detail the processes leading to SOA formation in this model application.

3. Another issue that requires some rewriting is related to the representativeness of the results. The authors state that they simulate a "typical" day with nucleation in two locations (page 30772). What is meant by "typical" here? Based on the figures, the authors have selected one day from these two locations and compare their simulations results to those two days in their analysis. This sounds like two case studies, provided that the model input corresponds to the conditions met during those two days. When comparing observed and simulated nuclei growth rates (section 4.1), the authors talk about typical growth in Hyytiala (why not to compare to the growth rate on the simulated day?), while they do not define at all what is meant by the observed growth rate of 5 nm/h in Finokalia. Is this 5 nm/h the average growth rated observed during some campaign(s) in Finokalia, or growth rate that was observed in the simulated day? In summary: are these real cases studies or some mixture between real cases and "average behavior" observed at the two sites?

We have re-written this part to avoid any misunderstandings. We have used the following process to obtain the characteristics of a typical nucleation event day in Hyytiala in April 2007 and in Finokalia in May 2008. We have first found the days with observed particle formation and growth. We have then averaged the measurements during these days generating in this way the meteorological and chemical characteristics of an "average" nucleation day for the specific periods in the two locations. For the parameters for which measurements were not available, but were needed for the model input, (e.g. OH concentration) we followed the same process using the predicted values from the 3-D chemical transport model PMCAMx. Therefore we do not choose specific days to simulate but rather try to simulate a "representative" nucleation day. In this way, we compare our results (e.g., for the growth rate) to the average growth rates and their corresponding ranges observed during this period. We do explain this process in detail in the revised manuscript.

4. Related to the previous comment, the authors provide several conclusions that appear general even though in reality they are based on the simulated two cases. For example, by reading the abstract one easily gets the impression that the given numbers (45% contribution of nuclei growth and 13% and 25% increases in CCN concentrations) are generally valid for those two locations. This should be corrected to avoid confusion or misunderstandings.

This is a valid concern and it is not our intention to over-generalize our conclusions. These are clearly applicable to nucleation days in the two locations during the simulated periods. We have qualified the corresponding conclusions in the abstract and the conclusions section to avoid misinterpretation of our findings.