

Interactive comment on “The role of highly oxidized multifunctional organic molecules for the growth of new particles over the boreal forest region” by Emilie Öström et al.

Anonymous Referee #1

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Öström et al. present a modeling study of growth dynamics and chemistry during new particle formation events at the Pallas boreal forest site in Northern Finland. They use an updated version of ADCHEM as a one-dimensional column trajectory model to simulate aerosol dynamics and chemistry of selected new particle formation events observed at the Pallas station between 2005 and 2010. The modeled particle number size distributions are compared to the measured size distributions, and the modeled contributions to particle growth of different chemical compounds and compound classes, in particular highly oxidized multifunctional organic compounds (HOMs), are discussed.

The manuscript is written in a clear and structured way, and covers a very interesting and timely topic, i.e. the role of HOMs in new particle formation and growth. This mod-

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eling study is instructive and a useful contribution to the field. However, the title and the aim defined at the end of the Introduction section raise very high expectations, which are not entirely fulfilled in the presented manuscript. Therefore, I recommend considering the manuscript for publication after revisions taking into account the following comments:

- 1) The abstract is somewhat lengthy and should be shortened to focus on the key aspects of the manuscript. For example, the very first part of the abstract is quite general and could be more concise.
- 2) On p.3 line 17, "to constrain the contribution of HOMs to the activation and growth of new particles over the boreal forest region" is given as the aim of the study. This is a bold aim, and I don't find any results or conclusions that would constrain the contribution of HOMs to new particle growth over the boreal forest. Please revise the definition of the study aim(s). Also, can you really make a clear statement about the role of HOMs for the growth of new particles over the boreal forest region? I recommend rephrasing the manuscript title to better reflect the nature of the study - a comparison of modeled and measured particle number size distributions and model simulations of the chemical composition of new particles during growth.
- 3) p.5 line 5: What is the reasoning behind the treatment of all monoterpenes other than alpha-pinene, beta-pinene, and limonene as carene? Is this a common procedure?
- 4) p.6 lines 5/6: Clouds and in-cloud aerosol processing were not considered in the study. I think this is fine as a first approach, but given that 34 % of the modeled times may have been influenced by low-level clouds, the potential of aqueous-phase chemistry should be added to the Discussion section, both for the oxidation of organic compounds and H₂SO₄ formation (presented as gas phase reaction of SO₂ and OH on p.4 line 27).
- 5) p.6 line 32: How exactly did you calculate the upper limit molar yield of HOM formation from OH oxidation? In line 29, you give molar yields of HOM formation from

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OH oxidation of alpha-pinene, limonene, and beta-pinene as 13 %, 27 %, and 17 % of the molar yield of HOM formation from alpha-pinene ozonolysis. Then, I would assume that the molar yield of HOM formation from OH oxidation is highest for limonene, which is not the case according to the numbers given in line 32 (beta-pinene 2.5 % vs. limonene 1.5 %)?

6) p.8 lines 10-14: How sensitive are the results about the mass fractions of compound types contributing to particle growth given in Figures 8 and 9 to neglecting interactions between the organic and inorganic compounds, and to setting all organic compound activity coefficients equal to unity?

7) p.12 lines 1-7: Two main conclusions of the study are that the modeled particle size distributions show good agreement with the observations during initial growth up to 20 nm diameter, but underestimate particle growth in the diameter range from 20 - 80 nm. One plausible explanation given in the manuscript is that particle-phase oligomerization involving semi-volatile organic compounds has not been considered in the model. Such particle-phase reactions might increase particle growth and possibly reduce the very high O:C ratio of nearly 1 for the modeled SOA. How would this affect the modeled chemical composition and SOA volatility distribution presented in Figures 8 - 10? This is a very important finding, which should lead to follow-up studies. In my opinion, the statements that the model was able to "capture" the main features of the observed growth (p.1 line 24; p.2 line 1; p.9 line 26; p.12 line 1) should be carefully rephrased. Which physical and chemical features were explained by the model?

Technical comments:

p.3 line 1; p.11 line 32: The presence of gas-phase HOMs has been shown in more than the two studies given as a reference. Please add "e.g." before the given references.

p.3 line 14: Here, the reference should be given as (Roldin et al., 2011a), and modified throughout the manuscript accordingly.

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p.5 line 24: Add superscripts in "H₂SO₄".

p.6 line 4: Give the value of C (= 0.39?) used in your study.

p.6 line 30: Add superscript in "O₃".

p.10 line 14: Change to "... was observed with a nitrate chemical...".

p.17 line 19: Change to "Silver Spring".

p.30 Figure 10 and supplementary Figure S7: Explain the meaning of "HOM C10-NO" shown as red bars.

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