Atmospheric
Chemistry
and Physics
Discussions

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 #2

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The following is a review of "The role of highly oxidized multifunctional organic molecules for the growth of new particles over the boreal forest region" by Öström et al. This manuscript describes a modeling study of new particle formation and growth in which the performance of the model was assessed by comparing predicted particle number size distributions with those measured at the Pallas Station in the boreal forest in Northern Finland. It has long been recognized that improved model representations of the growth of nanometer-sized particles formed from nucleation are needed in order to adequately assess the potential role of new particle formation on atmospheric chemistry and climate. This manuscript can play an important role in addressing this need. However, I have some concerns about this manuscript that should be addressed

in order for me to consider this suitable for publication in Atmospheric Chemistry and Physics. These are listed below, in no particular order.

1. As the title suggests, this study focuses on the question of whether the partitioning of highly oxidized multifunctional organic molecules (HOMs) could account for all of the observed growth in the atmosphere above the boreal forest. The implementation of the 1D model (ADCHEM) used in this study was impressive in its ability to account for unique emissions along the air parcel that encounters the measurement site. However the model, as described on page 8, makes very simple assumptions regarding gas-particle partitioning of HOMs. Essentially, all HOMs are represented as undergoing non-reactive, reversible partitioning. Particle phase chemistry is assumed to be non-existent and HOMs that partition into the particle phase form ideal solutions. This simplification is recognized numerous times by the authors, but is so far from representing aerosol chemistry that I fear that the main conclusions from the study have been pre-determined by their modeling approach. I feel a more accurate title would refer specifically to reversible partitioning of low-volatility HOMs.

2. Model predictions of chemical composition are extremely important in this study, however no field measurements of particle chemistry are provided and I am certain that such data exist for the Pallas Station. For example, for many years FMI has participated in the Pallas Cloud Experiment (PaCE), where aerosol properties were measured along with cloud properties. Without some chemical composition data to compare modeling results to, it seems likely that the model could be getting the right answer for the wrong reasons. The only two reference to prior Pallas measurements was applied to a few sentences that described their DMPS and state that new particle formation occurs there, however there are numerous published studies to which the authors may refer (http://en.ilmatieteenlaitos.fi/pallas-publications). The only comparisons to measurements of particle composition refer to measurements at the CLOUD chamber, which mostly focus on processes for particle sizes smaller than those measured at Pallas and for which the authors make no attempt to define relevance by comparing the

chemical and environmental conditions of the two studies. I feel that the authors must provide some discussion of that which is known about Pallas aerosol, preferably co-incident measurements but even measurements performed during other time periods would still provide some insight. Even studies that have taken place at Hyytiälä, some of which have focused on characterizing the composition of nanometer-sized particles (ref: the published work of Johnston and Smith groups) would be useful.

3. There is much discussion in the introduction of the importance of aerosol formation and growth to CCN populations; however, a key component of this is understanding vertical transport of newly formed particles to parts of the atmosphere where they have the ability to affect cloud properties. While I understand that this manuscript is focusing on growth, ADCHEM was used in this study as a 1D vertical resolving model and can provide important insights into the potential role that these formation events may play on climate. I am sure that readers would be very interested to know if the events that were observed on the ground might potentially represent populations throughout the boundary layer. If the authors could comment on this in the manuscript, they would actually address the very same crucial research needs that they claim motivate their study.

4. If the SIMPOL model calculates that HOM vapor pressures are very low, then HOMs will partition to particles irrespective of diffusion limitations that may exist in particles. Therefore, one would not expect any changes in size-resolved chemistry in particles when comparing fast and slow particle phase mass transport in particles, and in fact this is borne out in Figure 9. What is the reader to learn from this exercise? Wouldn't it have been better to use the somewhat higher vapor pressures calculated using COSMO, especially if the authors felt that it better represented processes in the small diameter range?

5. Overall I felt that the Results and Discussion section of this manuscript provided very little analysis of the data. Rather, the discussion minimally knits together references to each of the 8 figures and sometimes just stated rather obvious attributes of

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the figures while providing very little insight. For example, in reference to Figure 5 the authors state that the model run that did not include auto-oxidation mechanisms did not grow particles large enough to replicate the measured size distribution. This is clear from the figure. But there is no discussion of what the model DID predict, and this might be important when one considers environments in which auto-oxidation is curtailed by RO2-RO2 or RO2-NO chemistry. Another example: what is the importance of the distributions in Figure 10? They look nearly identical, except for small differences in the lowest volatility bins (which the authors attribute to dimerization). Why doesn't the volatility distribution evolve? Why compare the volatility distribution to Trostl et al.? Where the precursors and conditions (temperature, radiation, etc.) in that experiment identical to those at Pallas? Would it not be accurate to compare these distributions to those obtained at some of the SMEAR stations? Additionally, the authors state that lack of particle-phase chemistry of SVOC is likely to blame for many of the observed discrepancies between measured and modeled size distributions. As I say in item (1), it is of little surprise that ignoring LVOC and SVOC chemistry will lead to such problems. Studies like this modeling one can actually provide some insights into the types of processes that may actually explain observed growth rates. Even if the authors chose not to perform additional calculations, it would be useful for them to provide an assessment of the potential mechanisms that could help to address these discrepancies. Based on the list of co-authors, I believe that additional insights that would boost the quality of the analysis in this paper are indeed possible.

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