Atmos. Chem. Phys. Discuss., 15, C1697–C1700, 2015
www.atmos-chem-phys-discuss.net/15/C1697/2015/

Atmospheric Chemistry and Physics
Discussions
Open Access

Interactive comment on "Contribution from biogenic organic compounds to particle growth during the 2010 BEACHON-ROCS campaign in a Colorado temperate needle leaf forest" *by* L. Zhou et al.

Anonymous Referee #1

Received and published: 20 April 2015

Zhou et al. studied the process of new aerosol particle formation in and above the temperate forest in Colorado (US) using the 1D-chemical transport model SOSAA. The major focus was set on the biogenic contribution to early particle growth and the contribution of methylbutenol (MBO) oxidation products with OH. The study is well done, the references list conclusive and the plots clearly presented. Thanks very much. There a a couple of minor questions and aspects that should be discussed before the final acceptance, which the paper deserves. Those are listed in the following. Some aspects appear at various places in the document, so that only the first point

is or in case of Figures and Tables the text location and the reference are being named.

minor comments:

Abstract, p.3,l.3: Molecular clusters do not form by nucleation, but by molecular interactions. This is named nucleation once it overcomes the nucleation barrier (critical size). Reformulate!

p.3,l.3: growth by condensation is one part of the whole process. As the Kelvin effect hinders condensation at size ranges especially below 5-10 nm, which is critical for the nucleation to occur, other processes contribute as well that cannot be named condensation: (i) coagulation and coalescence, (ii) dissolution in particle mass or water (Raoult effect) and (iii) reactive attachment (e.g. polymerization). Please reformulate "by the uptake of vapours".

p.3, l.10-11: "suggesting missing atmospheric sulfuric acid sources" is probably a too strong statement as this may be caused by false assumption of sinks too. Common models assume a negligible saturation vapour pressure of sulfuric acid, which is according to literature not exactly true. As vapour pressures are functions of temperature this is expected to vary notably throughout the day. Please add the potentially different sink terms as well. The calculated OH concentration is one of the most critical in this respect, as OH reacts with nearly any species available except a handfull of substances. There might be an additional OH production via the ozonolysis of alkenes and the hydroperoxide channel. This impacts on the simulation approach via a specific assumption during night and early morning as investigated by the co-athors earlier on: E.g. all the monoterpenes are treated in a certain mixture out of three types, α-pinene, β-pinene and limonene which even provides a huge amount of species and reaction speeds. However, the real mixture is more complex with some notable differences in OH production and the individual terpenoid contributions may vary notably throughout the day affecting OH-cycling.

p.3, l.11ff: The impact of MBO+OH is a very nice result! But it's hard to understand the specific processes. MBO has got a molar mass of 86 g/mole with a single hydroxyl group that doubles during the reaction with OH. However no organic compound with 5 carbon atoms being a dialcohole will presume a saturation vapour pressure or even partitioning coefficient for the early stage of particle formation. So understanding seems only possible if treating MBO as a marker for BVOCs (oxidized VOCs)+OH. Thus, MBO is the wrong candidate at the right place at the time of interest with a similar bevaiour. This results in multifunctional organic peroxy radicals leading to some kind of polymerization. There are multiple of articles on this point. Due the authors have any suggestion about the involved processes? If so, please name it to focus future investigations!

Model validation for meteorology and chemistry, p.13/14: The model SOSAA seems to underestimate the mixing layer height notably. This is not a local phenomenon but applies at different sites in a similar way. Could the authors briefly indicate about the magnitude of the impact of this on the calculated results e.g. by taking measurements instead of calculations with interpolation in between the observations? This would cause a different dilution and different deposition rates.

p.15: Different timescales for mean daily pattern of compounds seems very critical. If possible the same time frame should be applied for all important gases as small changes sometimes have notable consequences, hiding important features. The uncertainty range is as large as always. How well the model performs if the upper concentrations of OH and VOCs are assumed (read-in)? Is that capable in explaining

the deviations between model and observations?

p.16 and Fig.6: As OH sometimes compensates missing production and sink terms due to the multitude of connections at reasonable photolysis rates this may explain the morning and early midday behaviour of OH. What happened during the afternoon, i.e. cloudy sky, differences between the different days of averaging? This is indicated in Fig.7 and the photolysis rate of NO_2 as well. Please provide more info on this.

p.32, Table 1: Please note, there is NO organic condensation but partitioning in the atmosphere as there is always organic material present casing subsidence below the saturation levels. Please modify the expression "organic condensing vapor" and replace it by "organic vapor type assumed" or something similar.

p.40: How well the usually taken 3-component assumption (α-pinene, β-pinene and limonene) matches with the plots shown? Is the simplified assumption made elsewhere justified or not? Please provide a brief statement. Regarding the plot only parts of it are informative. Could you provide more information shortly: Which kind of species are summarized in here, i.e. the stable ones or stable and radical products? Otherwise skip that plot.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 9033, 2015.