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# ***Interactive comment on “Biogenic SOA formation through gas-phase oxidation and gas-to-particle partitioning – comparison between process models of varying complexity” by E. Hermansson et al.***

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The paper is very interesting because it illustrates in a well-structured manner uncertainties of descriptions for the gas-to-particle partitioning of compounds forming in the atmospheric oxidation of biogenic volatile organic compounds. By comparison of different models, including near-explicit descriptions and VBS schemes currently used in regional and global atmospheric models, the study highlights the importance of the various physical and chemical processes affecting the long-term atmospheric simula-

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tion of particle mass concentrations. The paper is well written for the most part and the conclusions of the work are easily accessible. My main reservations are expressed in the following. After addressing these concerns and the minor comments, I think the paper is well worth publishing in ACP.

1.) A recent study by Ehn et al. (2014) found large yields of highly oxygenated products in the first step(s) of the oxidation of  $\alpha$ -pinene under atmospherically relevant conditions in chamber experiments. These highly oxygenated products with both monomer or dimer patterns were irreversibly taken up by the seed particles, even at very low particle loadings. This immediately points to limitations of the concept of equilibrium gas-to-particle partitioning in general, and of the VBS scheme ( $\log_{10}$  of  $C^*=0$  is not defined) specifically. However, the implementation of gas-phase reactions shown in the extended materials of the Ehn et al. (2014) study (Extended Data, Figure 7) into the Master Chemical Mechanism appears to be straight forward, and should be included in the current model comparison.

2.) A discussion of the assumptions behind the use of an air mass trajectory that happens to pass (close-by) all the three sites (at respective ground level) needs to be provided. Important details about the procedure to derive this specific trajectory with the HYSPLIT model are missing. In the boundary layer, the concept of a *single* trajectory being representative for the path of an air parcel does not hold since the air mass parcel will quickly lose its identity by turbulent mixing (Stohl et al., 1998). The accuracy of trajectory calculations will also be affected by the orography between the station locations. Terrain-following trajectories usually neglect vertical motions of synoptic origin, which could quickly modulate the air mass in the boundary layer.

3.) The description of fragmentation in 2DVBS does not state whether fragmentation takes place only for the first oxidation step or also for the subsequent oxidation steps. Did the authors take any precautions to avoid excessive fragmentation of smaller molecules, forming upon first functionalization (that will have high O:C due to short chain length), in the subsequent oxidation steps?

4.) A general problem of using the volatility ( $C^*$ ) axis is that since  $C^*$  depends strongly on temperature (Clausius Clapeyron equation, Eq. (A-2)), a temperature change by 10 degrees or more will cause a change of the bin assignment. This is less critical when relatively low values of  $\Delta H_{vap}$  is used, as in the 1DVBS approach (with  $\Delta H_{vap} = 30$  KJ/mol). It is however critical for the  $\Delta H_{vap}$  dependence on  $C^*$  (which refers to Donahue et al., 2006) used in the 2DVBS model that results in higher values (65-129 KJ/mol). Diurnal variation of ambient air temperature by 10 degrees is quite common in summer. At a temperature of 281 K the bin assignment in the 2DVBS (with reference temperature of 298 K) will change more or less by one decade (depending on the volatility bin). Did the authors consider the bin shifting within (and beyond) the VBS scale for changing temperature?

5.) It is clear from the title of the paper and section 2.4 (Processes not investigated in this study), that condensed phase reactions (accretion reactions, oxidative reactions, and aqueous phase reactions) were not topic of this paper. However, there is some evidence that SOA is not fully describable through equilibrium partitioning theory on short timescales (as in chamber experiments) likely as result of condensed phase reactions (Kroll et al., 2007), which could explain that the volatility of model SOA tends to be substantially higher than that of ambient organic aerosols (Cappa and Jimenez, 2011). The authors should add a discussion of the uncertainties of the modeled SOA in this respect.

#### Technical corrections

Abstract p 11002, line11: Replace “allows us to, under atmospheric relevant conditions compare” by “allows us to compare, under atmospheric relevant conditions,”.

P.11012, lines 4-8; Figure 2: Since an aerosol dynamics model was used (with only condensation) it would be more informative, especially with respect to CCN growth, to show how the modeled number size distribution changes as function of time (instead of volume concentration which merely is mass concentration divided by density). In

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Figure 2 initial distribution (at Abisko) together with size distributions at Pallas and Värriö should be plotted for ADCHEM, MALTE-BOX, and ADCHEM DensMalte.

P.11020, line 20-21: This is an important conclusion that needs to be added to the Abstract.

P.11021, Equation (A-2): Equation (A-2) contains a typo. It has to be “ $\Delta H_{vap}/R$ ” and not “ $\Delta H_{vap}/T$ ”.

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