

## ***Interactive comment on “Temperature dependence of secondary organic aerosol yield from the ozonolysis of $\beta$ -pinene” by C. Stenby et al.***

**Anonymous Referee #2**

Received and published: 11 November 2006

General Comments:

This manuscript reports on investigations of the temperature effects on the secondary organic aerosol (SOA) formation from ozonolysis of beta-pinene in the flow tube reactor. The authors found that SOA yields do not steadily increase as temperature decreases, indicating that SOA yields cannot be explained by a solely conventional partitioning theory and need an additional pathway through the reactions in the particle phase. The importance of heterogeneous reactions as an additional mechanism for the SOA formation has been also suggested in the recent investigations by many other researchers (1-7). Thus, the reevaluation of temperature effect on SOA yields is a timely important subject in the atmospheric chemistry community. Overall, the manuscript is scientifically well written and appropriate for ACP. I recommend the manuscript to be

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published after meeting the following comments.

1. How can the SOA composition within a short time scale in the flow reactor be related to the SOA composition formed for several hours in larger chambers or in the ambient air?

2. All the experiments reported in this study have been operated for the dry air which is not relevant to the realistic atmosphere. What is the tendency of temperature dependence of SOA formation in different humidity close to ambient conditions?

3. Instead of fitting the parameters to experimental data, the authors should provide the appropriate criterion to evaluate temperature dependence of partitioning and heterogeneous reactions on SOA yields. For example, when partitioning is dominant in SOA formation, the temperature effect on SOA yields may be explained mainly by vapor pressure of organic products through the Clausius Clapeyron equation. Then, the deviation from the partitioning aspect can be related to the particle phase reactions. What are the suggested criteria by the authors to the community?

4. In comparison, the authors should provide the implication of this study to other SOA formation from various terpenes or aromatics. For example, the study by Takekawa et al. reported that temperature effects are much sensitive in SOA from photo-irradiation of alpha-pinene than SOA in aromatics. Do the previous studies imply that heterogeneous reactions in the aromatic SOA are more significant and also assist the current study of temperature effects on SOA formation?

Specific comments:

Line 9, p 10279: Why is the flow rate of the sheath air changed?

Pages 10284 and 10280: The two statements for the OH scavenger effects were contradictory. It needs to be clarified.

Lines 16-18, p 10279: It is hard to understand how the sheath air is re-established for equilibrium. If the sheath air does not have the same gas concentration with the reactor,

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the organic compounds can be off-gassed from SOA and sequentially the SOA yields are affected by the sheath air.

p 10280: Current calculations of the consumption of [beta-pinene] based on the assumptions of ozone is not a solid method for the yield quantifications. In particular, beta-pinene is also consumed by OH radicals which are produced from ozonolysis of beta-pinene. The beta-pinene reaction with an OH radical reaction is much faster than those with ozone. If included, the consumption of beta-pinene by an OH radical reduces the SOA yields.

3rd paragraph on page 10286 and Table 3: The authors suggested that the changes in stoichiometric coefficients (alpha values) for the gas phase products are necessary to fit the data. This indicates the alteration of the gas phase reactions and products. Initially, authors emphasize the additional particle phase reactions and their influence on temperature effects on SOA yields. Such particle phase reactions should be independent from the gas phase reactions and not modify stoichiometric coefficients. If occurs, the partitioning behavior or partitioning coefficients would be more likely affected by the particle phase reactions. The fitting parameters in Table 3 should be reconsidered based on the property of the particle phase reactions.

Lines 11-12, p10283. Please provide simple explanation for why the number concentration generally decreases with decreasing temperature.

Lines 9-23, p 10285. The humidity conditions in the previous studies were various. Some studies were performed in the dry air like authors' study, and some employed ambient humidity conditions. Authors should consider potential difference in SOA composition in different humidity.

It is suggested that heterogeneous reactions may influence SOA yield with respect to temperature. How do rate constants for these types of reactions vary with temperature?

In conclusion section, if authors can also include implication of this study to the recent

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investigation of heterogeneous reactions in SOA (please see the references:(1-7)), it will be very useful for the community.

Technical comments:

Please, include humidity in the experimental section.

Line 20, p 10280: It is assumed that OH formation is independent of temperature. Please provide a reference for this statement.

List of citations.

(1) Kalberer, M.; Paulsen, D.; Sax, M.; Steinbacher, M.; Dommen, J.; Prevot, A.S.H.; Fisseha, R.; Weingartner, E.; Frankevich, V.; Zenobi, R.; Baltensperger, U., Identification of Polymers as Major Components of Atmospheric Organic Aerosols. *Science* 2004, 303, 1659-1662.

(2) Jang, M.; Czoschke, N.M.; Lee, S.; Kamens, R.M., Heterogeneous Atmospheric Aerosol Production by Acid- Catalyzed Particle-Phase Reactions. *Science* 2002, 298, 814-817.

(3) Iinuma, Y.; Böge, O.; Gnauk, T.; Herrmann, H., Aerosol-chamber study of the  $\alpha$ -pinene/O<sub>3</sub> reaction: influence of particle acidity on aerosol yields and products. *Atmos. Environ.* 2004, 39, 761-773.

(4) Jang, M.; Czoschke, N.M.; Northcross, A.L.; Cao, G.; Shaof, D., Secondary Organic Aerosol Formation from Partitioning and Heterogeneous Reactions: Model Study. *Environ. Sci. Technol.* 2006.

(5) Johnson, D.; Jenkin, M.E.; Wirtz, K.; Martin-Reviejo, M., Simulating the Formation of Secondary Organic Aerosol from the Photooxidation of Aromatic Hydrocarbons. *Environmental Chemistry* 2005, 2, 35-48.

(6) Gao, S.; Keywood, M.; Ng, N.L.; Surratt, J.; Varutbangkul, V.; Bahreini, R.; Flagan, R.C.; Seinfeld, J.H., Low-Molecular-Weight and Oligomeric Components in Secondary

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Organic Aerosol from the Ozonolysis of Cycloalkenes and  $\alpha$ -Pinene. *Journal of Physical Chemistry A* 2004, 108, 10147-10164.

(7) Kalberer, M.; Sax, M.; Samburova, V., Molecular Size Evolution of Oligomers in Organic Aerosols Collected in Urban Atmospheres and Generated in a Smog Chamber. *Environmental Science Technology* 2006, 40, 5917-5922.

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Interactive comment on *Atmos. Chem. Phys. Discuss.*, 6, 10275, 2006.

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