

Review of the manuscript acp-2020-798 with title: “Temperature and acidity dependence of secondary organic aerosol formation from α -pinene ozonolysis with a compact chamber system” by Deng et al.

General comments:

This manuscript describes a new atmospheric simulation chamber and its application to study the secondary organic aerosol from the oxidation of alpha-pinene by ozone at different temperatures and for seed aerosol particles of different acidity. Therefore, it fits well in the scope of the journal of atmospheric chemistry and physics and focusses on an interesting scientific topic which has been subject of many previous studies. The scientific results are presented in a clear and well-structured way and are based on a reasonable scientific approach and valid methods. However, this manuscript represents only a limited contribution to scientific progress in this field.

The description of the new simulation chamber is quite limited and rewards a more detailed discussion of its capabilities and limitations compared to other simulation chambers as well as how suitable it is for this kind of studies. Especially, the impact of the chamber limitations on the uncertainties for the major scientific results should be addressed and quantified.

The scientific results lack a reasonable representation of their uncertainties as well as a suitable comparison with data from the large number of previous studies on this subject. This is especially the case for the yields and VBS distributions. Some of the most interesting findings in this study are the analysis and identification of SOA compounds for which also potential formation mechanisms are discussed. This discussion may be restructured and focused on new findings.

Most of the scientific results presented don't go much beyond current knowledge and some of them can be expected to have higher uncertainties than those of previous studies. Therefore, the manuscript contains only a limited contribution to the scientific understanding of formation and composition of secondary organic aerosol in our atmosphere. Hence, this manuscript should only be accepted for publication after major improvements and focusing on new scientific findings.

Specific comments

Try to include all major results in the abstract.

Page 1 line 10: Actually, you did not describe the development of a chamber in this study. Please reformulate.

Page 1 line 25: SOA is not only formed by photo oxidation. Please reformulate.

Page 1 line 34: Please use “saturation concentrations”.

Page 1 line 35: Notify the importance of ELVOCs and that ELVOCs and LVOCs are also formed in the gas phase (see e.g. Tröstl et al., Nature, 2016).

Page 2 line 50-54: Reformulate to shorter sentences.

Page 2 line 54: Do you mean contradictory results?

Page 3 line 89-93: You may cite Brüggemann et al., EST, 54, 3767, 2020 here as well.

Page 3 line 112: It should read: “at atmospheric pressure”.

Page 3 line 114-115: Explain how the pure air is humidified (bubbling through water?) and how this affects the purity of the G3 pure air regarding gases (e.g. VOC) and small particles.

Page 3 line 131: Explain how ozone was added to the chamber and if you can estimate a mixing time for the reactants. How is mixing achieved in your chamber?

Page 4 line 134: Here it should read: "aerosol particles". Generally, distinguish between aerosols = airborne particles and gases, aerosol particles = airborne particles, and gases throughout the manuscript.

Page 4 line 139-140: Explain the criteria for achieving a sufficient cleanliness of your chamber by flushing.

Page 5 line 161: It should read: "saturation concentrations".

Page 5 line 161: It should read: "molecular formulae" while chemical formulae would already contain some structural information. It would be very useful if you would add the names of those compounds identified in table S3. Explain how the C* values have been calculated.

Page 5 line 169-170: Coagulation is not a wall loss. Please rephrase. Specify if you have any indications of electrostatic particle losses in your chamber especially for new Teflon foil.

Page 5 line 169-172: Please fit the particle losses in your chamber e.g. using the formulation given by Lai and Nazaroff, J. Aerosol Sci., 31, 436, 2000. Discuss how the wall loss parameters of your chamber compare to others and how suitable your chamber is for different kind of applications. Did you also measure the wall losses for solid particles?

Page 5 line 173: How can the seed particle size affect wall losses like sedimentation for super 200 nm particles?

Page 5 line 176: It should read: "larger particles".

Page 5 line 182: Please explain how you separated the SOA mass from the seed particle mass and what uncertainty this means for the determination of the SOA mass.

Page 5 line 182-186: Please discuss the uncertainties caused by the different approximations applied in this section. Did you take direct losses of semi volatile gases to the wall into account (Zhang et al., PNAS, 2014)?

Page 6 line 204: It should read "...was calculated and then ascribed...".

Page 6 line 219: I think it would be helpful to add the evolution of measured particle number size distribution e.g. as a contour plot in Figure 1 as well as the evolution of the particle number.

Page 6 line 224: Add the uncertainty of the SOA yield. It should read

Page 6 line 224-225: It should read: "As the corrected SOA particle concentration was constant after 50 min,...".

Page 6 line 226: It should read: "...concentration of SOA particle mass at 90 minutes would be underestimated...".

Page 8 line 243: It should read: "...seven experiments with different initial α -pinene concentrations (54–323 ppbv) at 298 K and neutral seeds were conducted at a relative humidity (RH) of approximately 26–27 % (Table S1).".

Page 8 line 244/Table S1: Indicate how much aerosol volume or mass was formed after adding ozone to your chamber but before adding pinene.

Page 8 line 248: Add the error bars for your data in Figure 2 and enlarge the symbols. Discuss if the uncertainties are larger for the lower mass loadings. Note that you have only 5 minutes time resolution of the SMPS data. Please change the caption indicating that the lines represent a parameterization by Pathak et al. 2007a and not measured yields. Discuss how representative a comparison to only the data of Pathak et al. is considering the large amount of data available in the literature as well as more recent studies.

Page 8 line 250: Do you mean: "...used in this study was significantly larger than for most previous studies..."?

Page 8 line 258-261: Given the methods you applied to correct for potential artifacts what uncertainty remains for SOA yields for different conditions and what are e.g. resulting concentration limit for your chamber.

Page 8 line 266-267: Give the uncertainties for these parameters.

Page 8 line 266-267: Do you mean: "...lowering of their vapor pressures."?

Page 8 line 277: It should read: "...lower than those of Saha et al...".

Page 9 line 285: Indicate if the dependence of SOA yields on the acidity of seed particles is significant or not.

Page 9 line 299: Indicate if the SOA yields were significantly enhanced comparing acidic to neutral seeds or not.

Page 9 line 306-308: Which measurement accuracy would be needed to achieve significant results?

Page 9 line 310: Add uncertainties to your data points in Figure 3 and compare to literature data.

Page 10 line 316: Add the uncertainties to the VBS parameters in Table 1 and compare to literature values.

Page 10 line 325: It should read: "...measured intensities of particle phase compounds...".

Page 10 line 331: It should read: "The intensities of both the particle and gas phases were...".

Page 10 line 334: Give how much the rate coefficients varies with temperature. Please cite original references like Tillmann et al., PCCP, 11, 2323, 2009.

Page 10 line 335-336: How valid is this assumption as the total amount of SVOCs may be influenced by temperature dependent wall losses, changing product branching ratios, etc..

Page 11 Figure 4: Replace Aerosol by Particle on the y-axis label and caption. Indicate the uncertainty of your data.

Page 12 line 380: It should read: "...were concentrated in the O:C ratio range...".

Page 13: I think it would be useful to add a table of major products, their abundances and their tentative chemical identification (compound, structure) either separately or in table S3.

Page 13 line 429: Please explain the unspecific chromatograms in Figure S5.

Page 14 line 433: Can you give an estimate of the uncertainties for the OS intensities?

Page 14 Figure 7: Add uncertainties and discuss which differences or trends are significant.

Page 14 line 448-449: For the mechanistic discussion it would be more useful to give here and in the following either the compound name or molecular formulae for each compound identified. State the uncertainties of the molecular yields. Consider adding potential reactions mechanisms e.g. in the supplement.

Page 14 line 450-451: Consider revising: "The possible formation mechanisms of the OS compounds were proposed based on literature data combined with the experimental results of this study."

Page 15 line 461: It should read: "While all eleven OS compounds detected in this study are formed from α -pinene ozonolysis due to the presence of an excess of the OH radical scavenger,..."

Page 15 line 467: It should read: "...and one (m/z 265) was said to be from the sulfacation of pinonaldehyde..."

Page 15 line 485: "...could only be observed at the lower temperatures."

Page 15 line 488-489: Consider revising: "When the structure of the OS can be determined and the precursor compound can be assumed, the formation mechanism of the OS in α -pinene ozonolysis with acidic seeds may be confirmed."

Page 16 Figure 8: Indicate to what the yield is related to. You may add total OS yield vs. temperature which will be positive and dominated by $C_7H_{11}O_6S$ and $C_{10}H_{15}O_7S$.

Page 17 Table 3: Try to enlarge the structures as much as possible as well as the letters indicating the various references.

Page 18 line 508: Consider revising: "...with acidic and neutral seed aerosol particles."

Page 18 line 513: Consider revising: "Among the 362 compounds identified,..."

Page 18: In section 5 you should go beyond a summary and add conclusions for our understanding of atmospheric SOA and OS. You may also discuss the relevance of your findings vs. existing literature data.

Tables S1 and S2: Why is there a letter c at the head of the column with ozone concentrations? Add uncertainties for your data. Give only significant digits.

Table S3: Please add compound names where possible and mention that C^* values were calculated according to Li et al., 2016.

Figure S3: Add a legend.

Figure S4: Are the differences between the yield parameterizations significant? Is it correct that the temperature dependence of the yields is more significant than the acidity dependence?

Figure S5: How do you explain the rather unspecific chromatograms e.g. for m/z 249, 253, 267, 283 ?

References:

Brüggemann, M., Xu, R., Tilgner, A., Kai Kwong, C., Mutzel, A., Poon, H.Y., Otto, T., Schaefer, T., Poulain, L., Chan, M.N., and Herrmann, H., Organosulfates in Ambient Aerosol: State of Knowledge and Future Research Directions on Formation, Abundance, Fate, and Importance, *Environmental Science & Technology* 54 (7), 3767-3782 (2020). DOI: 10.1021/acs.est.9b06751

Lai A.C.K., and Nazaroff, W.W. , MODELING INDOOR PARTICLE DEPOSITION FROM TURBULENT FLOW ONTO SMOOTH SURFACES, *J. Aerosol Sci.*, 31, 463-467 (2000) doi.org/10.1016/S0021-8502(99)00536-4

Tillmann, R., Saathoff, H., Brauers, T., Kiendler-Scharr, A., Mentel, T.F., Temperature dependence of the rate coefficient for the α -pinene reaction with ozone in the range between 243 K and 303 K, *Phys. Chem. Chem. Phys.*, 11, 2323-2328 (2009) DOI: 10.1039/B813407C

Tröstl, J., Chuang, W., Gordon, H. *et al.* The role of low-volatility organic compounds in initial particle growth in the atmosphere. *Nature* 533, 527–531 (2016). doi.org/10.1038/nature18271