

Response to Anonymous Referee #1

We appreciate the invaluable comments. Our answers to the comments are provided below. The reviewer comments are written in italic.

General remarks:

In this paper, Temperature and acidity dependence of secondary organic aerosol formation from α -pinene ozonolysis with a compact chamber system have been investigated. This is a nice piece of work where the temperature and acidity dependence of SOA yields and chemical components were investigated. In addition, the formation of organosulfates (OS) was observed under acidic seed conditions. The data presented are of significant importance; however some modifications would be good to improve the article. The article should be published after major modifications.

Some characteristics of the new chamber are missing. We have developed a new chamber, it would be good to have more specifications. The temperature effect on the SOA formation has been relatively little studied, mainly due to the low number of temperature controlled chamber. Give more precision on the temperature controlled chamber.

There is no uncertainties, it would be good to add it in the text, in the figure and in the table. For example, no uncertainties has given for the temperature, the yield, the concentration

It would be good also to correct the SOA yield obtained by all wall losses (particle and also oxidized organic vapors) to improve the article.

It would be good to add also some comments and reference (list of some references given at the end of this pdf file) about the relative humidity (RH) because it's not the same in each experiment and the humidity can also influence the SOA yields and chemical compounds. The effect of RH on aerosol formation has yet to be optimized. Some experiments performed in flow reactor have shown that the aerosol yield was dependent on RH and other studies performed in simulation chamber have reported no effect. For example, the study of Saathoff [2009] on the ozonolysis of α -pinene have shown that water had little influence on aerosol yields at room temperature, however, a significant positive effect of water is observed for lower temperatures. Relative humidity is dependent on temperature.

For more explanation, see specific comments.

Reply> We respond to the general comments within the following responses to the specific comments.

Specific comments:

Introduction

Line 41-59: *The role of acidity is really good explain. However, the role of the other parameters as the temperature is very succinct, just one sentence. It would be good to explain more the influence of the other parameters on the SOA yields and chemical composition.*

Reply> Examples of the temperature-dependence of SOA formation have been added to the revised manuscript as follows: “For example, the relative importance of the temperature-dependencies of the volatilities of oxidation products and of the gas-phase and multiphase chemical reactions, which could result in different temperature-dependence of SOA yields (Pathak et al., 2007b; von Hessberg et al., 2009), are still not well constrained.” (Page 2 lines 59–62)

Line 63-81: The dependence of humidity is not explained for the specific case of α -pinene ozonolysis. It would be good to add also some sentences and references (list of some references given at the end of this pdf file) about the relative humidity (RH) because it's not the same in each experiment and the humidity can also influence the SOA yields and chemical compounds. The effect of RH on aerosol formation has yet to be optimized. Some experiments for α -pinene ozonolysis performed in flow reactor have shown that the aerosol yield was dependent on RH and other studies performed in simulation chamber have reported no effect. For example, the study of Saathoff [2009] on the ozonolysis of α -pinene have shown that water had little influence on aerosol yields at room temperature, however, a significant positive effect of water is observed for lower temperatures. Relative humidity is dependent on temperature.

Reply> We agree with the reviewer. The SOA yields and chemical compounds can be influenced by the humidity in the chamber. However, we did not investigate the influence of humidity on SOA yield in the present study. Since very dry conditions are not realistic in ambient air, we carried out the experiments at medium humidity (~26–27 % RH at 298 K, ~32–34 % RH at 288 K, and ~45–55 % RH at 278 K), which did not vary considerably at each temperature. We believe that the differences in RH among our experiments would not influence SOA formation significantly, as explained in the following section. First, nucleation was negligible in all the experiments because of the high concentrations of seed particles applied; therefore, the influence of RH on SOA formation would be reflected in the particle phase (Kristensen et al., 2014). Second, because the seed particles were dried into effloresced states before being introduced into the chamber, all particles in the chamber were in solid (neutral seed conditions) or near solid state (acidic seed conditions) (Tang and Munkelwitz, 1977). Consequently, the influence of water on the particle phase through physical partitioning or chemical reactions should be minor (Faust et al., 2017). We have mentioned this in the revised manuscript as follows:

“In the present study, we did not investigate the influence of humidity on SOA yield. Since very dry conditions are not realistic in ambient air, we carried out the experiments at medium humidity (26–55 % RH, Tables S1 and S2). The differences in RH among experiments in the present study would not influence SOA formation significantly, as explained in the following section. First, nucleation would be negligible in all experiments because of the high concentrations of seed particles applied. Consequently, the influence of RH on SOA formation would be reflected in the particle phase (Kristensen et al., 2014). In addition, because the seed particles were dried into effloresced states before being introduced into the chamber, all particles would be in solid (neutral seed conditions) or near solid states (acidic seed conditions) (Tang and Munkelwitz, 1977). Therefore, the influence of water on the particle phase through physical partitioning or chemical reactions would be minor (Faust et al., 2017).” (Pages 4–5 lines 162–170).

Experimental

Line 109-140: A new chamber is developed. It would be good to have more specifications of this chamber and the analytical tools. Which instrument allow to control and to measure the temperature and the humidity in the chamber? What is the range possible in the chamber? What is the uncertainties and the precision of each measurement? The temperature is homogeneous in all chamber?

Reply> The RH of the chamber air was not measured during but after the experimental run by pumping the remaining chamber air into a separate small TEFLON bag, to which a VAISALA RH&T probe (model, HMP76) equipped with a measurement indicator (model, M170) was attached. In experimental runs with the same temperature setting, similar flow rates and filling times were applied for both humid and dry G3 air, the RH was only measured for 4, 3, and 4 of the experimental runs for temperature settings of 298, 288, and 278 K, respectively. The measured RH ranges are presented in Tables S1, S2, and Table 1. The measurement of the RH has been included in the revised manuscript as follows: “The RH of the chamber air was measured after the experimental run by pumping the remaining chamber air into a separate small TEFLON bag, to which a VAISALA RH&T probe (model, HMP76) equipped with a measurement indicator (model, M170) was attached.”. (Page 4 lines 123–125)

The temperature inside the cabinet was measured using a thermocouple attached to the inside of the cabinet (T3 in Fig. S1). The potential temperature range in the cabinet is 5–40 °C. The evaluation of the thermostat capacity of the cabinet is presented in Text S1 and Fig. S2. Under initial temperature settings of 4.6, 14.6, and 24.7 °C, the temperature inside the cabinet within an hour varied within the 3.9–5.5, 14.0–15.3, and 24.6–25.3 °C ranges, respectively, and the standard deviations are 0.36, 0.27, and 0.18 °C, respectively. The temperature ranges inside the Teflon chamber within one hour were 4.8–5.2, 14.8–15.1, and 24.9–25.2 °C, respectively, and the standard deviations were 0.15, 0.10, and 0.11 °C, respectively. Besides, we waited for 30 min before the start of chemical reactions. Therefore, we think that the temperature was well controlled (uncertainties were < 1 °C) during the SOA formation reactions. This has been added to the revised manuscript, as follows:

“The temperature inside the cabinet was measured using a thermocouple attached to the inside of the cabinet (T3 in Fig. S1). The achievable operating temperature range of the chamber was 5–40 °C. A detailed evaluation of the thermostat capacity of the chamber under dark conditions is presented in Text S1, which indicates that the temperature inside the chamber was well controlled (varied within ± 1 °C).” (Page 3 lines 115–119)

Line 125: Add more precision on the PTRMS analysis: flow of sampling, time resolution, m/z range, E/N

Reply> The following details have been included in the revised manuscript: “The PTR-MS was operated at a flow rate of approximately $250 \text{ cm}^3 \text{ s}^{-1}$ under a field strength (E/N , where E is the electric field strength (V cm^{-1}) and N is the buffer gas number density (molecule cm^{-3}) of the drift tube) of 106 Td. The length of the drift tube was 9.2 cm. The drift voltage was set to 400 V. The temperatures of the inlet and drift tubes were set to 105 °C and the pressure at the drift tube was set to 2.1 mbar. The signal intensities of ions with m/z s of 21, 30, 32, 37, 45, 46, 75, 81, and 137 were recorded approximately every 4.5 sec. The detection sensitivity of α -pinene was $3.3 \pm 0.6 \text{ ncps ppbv}^{-1}$ (ncps means normalized counts per second to 10^6 cps of H_3O^+).” (Page 4 lines 138–143)

Line 136: *Add the uncertainties for each temperature*

Reply> As stated previously, evaluation of the thermostat capacity presented in Text S1 indicates that the temperature inside the chamber was well controlled with uncertainties < 1 °C. This point is presented in the revised manuscript as follows: “A detailed evaluation of the thermostat capacity of the chamber under dark conditions is presented in Text S1, which indicates that the temperature inside the chamber was well controlled (varied within ± 1 °C).” (Page 3 lines 117–119). Hereafter, we don’t show the uncertainties of the temperature in the text.

Line 137: *Modify the sentence “For experiments with the same temperature setting, the RH settings were also similar (Tables S1 and S2). » There is more of 10% of difference for RH for a same temperature, it’s not really similar. Add the range of humidity in the text for each temperature. Furthermore, the humidity is really variable between 26 and 55% for the three temperature.*

Reply> The sentence “For experiments with the same temperature setting, the RH settings were also similar” has been deleted in the revised manuscript. Instead, the RH range is displayed using the following sentence “In the present study, we did not investigate the influence of humidity on SOA yield. Since very dry conditions are not realistic in ambient air, we carried out the experiments at medium humidity (26–55 % RH, Tables S1 and S2).” (Pages 4–5 lines 162–164)

As has been explained earlier, we think the differences in RH among the experiments do not influence SOA formation in our study considerably. We have added the following explanations in the revised manuscript: “The differences in RH among experiments in the present study would not influence SOA formation significantly, as explained in the following section. First, nucleation would be negligible in all experiments because of the high concentrations of seed particles applied. Consequently, the influence of RH on SOA formation would be reflected in the particle phase (Kristensen et al., 2014). In addition, because the seed particles were dried into effloresced states before being introduced into the chamber, all particles would be in solid (neutral seed conditions) or near solid states (acidic seed conditions) (Tang and Munkelwitz, 1977). Therefore, the influence of water on the particle phase through physical partitioning or chemical reactions would be minor (Faust et al., 2017).” (Page 5 lines 164–170).

Line 141-146: *I would like to know if you have perform some blank filter and some blank of chamber. If yes, it would be good to add a sentence as « These control runs led to no detectable SOA production, hence confirming that no “memory effects” of the chamber were contaminating our results ».*

Reply> Information concerning blank filter and blank of chamber has been included in the manuscript and Text S2.

With regard to the chamber blank, a sentence “Particle number concentration and VOC mixing ratio measurements indicate that the chamber background concentrations of particles and VOCs were negligible, and no extra contamination was observed by the humidification process of the G3 air (Text S2).” has been added to the revised manuscript (Page 4 lines 125–127).

About the filter blank, a sentence “A blank filter was also analyzed using a procedure similar to that of the sample filters. The results confirmed no substantial contamination in the filter and the filter analysis procedure (Text S2).” has been added to the revised manuscript (Page 5 lines 176–177).

Line 150: It would be good to have the parameters for the LC-MS (at least in Supporting Information). For example, what is the configuration of the ESI source in this analysis in negative mode.

Reply> The relative configuration parameters have been included in the revised manuscript as follows:

“The key configuration parameter settings were as follows: nebulizer pressure was 0.21 MPa; the voltage in the spray chamber was –3500 V; the drying nitrogen gas temperature was 325°C and flow rate was 5 L min⁻¹; and the fragmentor voltage was 175 V.” (Page 5 lines 185–187).

Line 162: 362 products have been identified. I would like to know if some standard has been analyzed in LCMS to confirm the compounds in this study. If it's not the case, it would be good to say that the identification method was based on retention times and on mass spectra interpretation. Note that in absence of authentic standard, the identification should be regarded as tentative. A recent article Kenseth et al, EST 2020, shows that the use of unrepresentative surrogates can lead to substantial systematic errors in quantitative LC/ESI-MS analyses of SOA.

Reply> The original sentence “We determined the chemical formulae and signal intensities for 362 products (including eleven organosulfates) of different m/z (Table S3)” has been changed to “We tentatively determined the molecular formulae and signal intensities of 362 products (including 11 organosulfates) with different m/z (Table S3) based on retention times and interpretation of mass spectra.” (Page 5 lines 198–200).

Line 164-176: What type of seed (neutral, acidic?) is used to determine the aerosol wall loss in the chamber? In which conditions (RH, T) this experiment of aerosol wall loss has been performed? In SI, it's written that this experiment has been performed under an RH range of 17–23 %, but this range is different during the experiment (26-55%). Can this difference cause different wall losses?

Reply> Fig. S2 (currently Fig. S4) is for checking the size-dependent wall-loss rate. However, the bulk wall-loss rate has been measured under different seed particle and temperature conditions. As was indicated in Sect. 2.3 “The measurements were carried out whenever a new Teflon bag was used or the experimental conditions (i.e., temperature or seed particle acidity) were changed under humid air conditions. The latest measured bulk wall-loss rate (Sect. 3.1) was applied for each SOA formation experiment.” (Currently Page 6 lines 205–207).

There are not only aerosol wall loss but there is also the wall-loss of oxidized organic vapors. I think it would be good to add some sentence on this wall loss even if no experiment would be performed. It's already mentioned in this article in line 250 but it's too late. To improve the article, I would correct the yield by all the wall losses.

Reply> A qualitative discussion of the influence of gas-phase wall-loss on SOA yield has been included in Sect. 4.1. In addition, an associated introduction has been included at the end of Sect. 2.3, as follows: “Wall-loss of gas-phase organic compounds in the Teflon chamber could also cause the underestimation of SOA yields (Zhang et al., 2014; Krechmer et al., 2016). Although not experimentally determined in the present study, the influence of gas-phase wall-loss on SOA yields will be discussed based on the studies of Zhang et al. (2014) and Krechmer et al. (2016) in Sect. 4.1.” (Page 6 lines 221–223).

Line 165: When a new Teflon bag is used? After a blank experiment?

Reply> A new Teflon bag was used whenever the old Teflon bag had a leak problem.

Line 168-176: It would be good to model (i.e. Lai and Nazaroff) of the wall loss performed with seed. This well-known process depends on chamber geometry, static charge build-up on the walls, air flow and particle size. Since aerosol deposition was measured for $(\text{NH}_4)_2\text{SO}_4$ seed with $d = 1.77 \text{ g/cm}^3$, due to the gravitational effect the wall loss rates for SOA (with different density) would be different. For more explanation, see the article of Lai and Nazaroff 2000.

Reply> Simulations of wall-loss performed with seed particles have been performed and presented in Text S3 and referred to in Sect. 2.3 as: “Model simulation (Text S3) and literature survey results revealed that the high wall-loss rates of sub-100 nm particles were mainly caused by particle coagulation (Nah et al., 2017; Wang et al., 2018a) and those of super-200 nm particles were likely the result of turbulent deposition (Lai and Nazaroff, 2000).” (Page 6 lines 213–216).

The models of Lai and Nazaroff (2000), which focused on turbulent deposition, ignoring Brownian diffusion, but considering the gravitational settling deposition, and that of Hinds (1999), which considered only Brownian diffusivity deposition or settling deposition, were applied. The simulation results (Fig. S4) revealed that pure gravitational settling deposition played a minor role in the total observed particle loss. The influence of Brownian diffusivity deposition was also minimal after 30 min of stabilization. Whereas the large wall-loss of super-200 nm particles can be explained by the turbulent deposition, that of sub-100 nm particles cannot be explained by any of the simulated mechanisms. According to the study of Wang et al. (2018a), the changes in particle number-size distributions caused by coagulation could be the major reason for the large apparent wall-loss of sub-100 nm particles. In addition, electrostatic particle losses were small even for a new Teflon bag in our experiment, because the wall-loss of a new bag was often lower than that of an old bag. Besides, humid air was applied during the experiment, which may prevent electrostatic particle losses.

Data analysis

Line 179-201: It would be good to correct also the SOA yield by the wall loss of oxidized organic vapors. Maybe give a range of yield with the particle wall loss correction for the smallest value and the vapor wall loss correction for the largest value.

Reply> The influence of the gas-phase wall-loss on SOA yield is qualitatively discussed in Sect. 4.1. A sentence indicating this point has been included in this section in the revised manuscript, as follows: “The influence of gas-phase wall-loss on SOA yield is discussed qualitatively in Sect. 4.1.” (Page 6 lines 238).

Line 193: « α is assumed to be temperature-independent ». Do you have a reference to assume that α is assumed to be temperature-independent? If yes, it would be good to add it.

Reply> Yes. A reference (Pathak et al., 2007a) has been added (Page 7 line 244).

Result and discussion

Line 230 figure 1: It would be good to homogenize the axe of the figure, for example use the same nomenclature in the figure and also in the text. For example, in this figure it would be good to modify SOA ($\mu\text{g}/\text{m}^3$) by $m\text{SOA}$ ($\mu\text{g}/\text{m}^3$). In figure 1, it would be good to add a banana plot for the aerosol. I think it would be a good idea to correct the data by also the wall loss of oxidized organic vapors.

Reply> The name of the left axis in panel (a) has been changed from “Concentrations of α -pinene, O_3 (ppbv)” to “ α -Pinene, O_3 (ppbv)”, and the name of the right axis has been changed from “SOA ($\mu\text{g m}^{-3}$)” to “ m_{SOA} ($\mu\text{g m}^{-3}$)”. A banana plot for the aerosol has been included as a supplementary figure (Fig. S6). A relevant explanation has been included in the manuscript: “Evolution of the particle number-size distribution is presented in Fig. S6.” (Page 8 lines 291–292).

Qualitative discussions of the influence of the wall loss of oxidized organic vapors based on the results of Fig. 2 have been included in the revised manuscript as follows: “The SOA yields in this study were 25–60 % lower than those of Pathak et al. (2007a). Possible reasons may include lack of consideration of the wall-loss of oxidized organic vapors because the surface to volume ratio of the chamber used in this study (7.1 m^{-1}) was much larger than those of previous studies ($<3 \text{ m}^{-1}$; Pathak et al., 2007a and references therein). According to Zhang et al. (2014), the vapor wall-loss bias factor, R_{wall} (defined as the ratio of the SOA mass when the vapor wall loss is assumed to be zero, to the SOA mass when the optimal vapor wall loss rate is applied), was reported to be ~ 4 at the initial seed surface area of $\sim 2 \times 10^3 \mu\text{m}^2 \text{ cm}^{-3}$ (seed-to-chamber surface area ratio $\sim 1 \times 10^{-3}$) and ~ 2 at the initial seed surface area of $>6 \times 10^3 \mu\text{m}^2 \text{ cm}^{-3}$ (seed-to-chamber surface area ratio $>3 \times 10^{-3}$) during the photooxidation of toluene. As the initial seed surface area in the present study was in the $(1\text{--}3) \times 10^3 \mu\text{m}^2 \text{ cm}^{-3}$ range (seed-to-chamber surface area ratio $(1\text{--}4) \times 10^{-4}$), R_{wall} in the Teflon bag might be at least twice those of the large chambers. This leads to the underestimation of the SOA yield of 50 % when compared with the values obtained from the large chambers. A low limit correction of the gas-phase wall-loss influence for Exp. No. 2 and other experimental runs in which chemical composition analysis had been conducted based on the method of Krechmer et al. (2016) (Text S5, Fig. 2), which confirmed that gas-phase wall-loss is one reason for the lower SOA yields in the present study compared with Pathak et al. (2007a) and other previous studies presented in Fig. 2.” (Pages 9–10 lines 324–337).

Line 235 figure 2: It would be good to compare the yield corrected by all wall loss and not only by the particle wall loss. A comparison is performed with Pathak et al. 2007, it's good to do this comparison but

it's difficult to understand why a comparison is performed with two curves (less of 10% RH and another with a range between 50 and 73%) because in our data the experiment are performed at 26% RH. I would like to know if no literature data exists for this range of humidity (26%). If yes, it would be good to compare the data with this reference. If not, it would be good to say that this data are new and it's difficult to compare with literature because no data have been performed at 298K and 26% RH in presence of seed.

Reply> As mentioned above, we have estimated the amount of the wall-loss of oxidized organic vapors for Exp. No. 2 and the result is shown in Fig. 2. In the figure, we have added some yield data, which were obtained under different RH conditions. As has been explained previously (Pages 4–5 lines 162–170), we think that the differences in RH among the experiments do not influence the SOA yields substantially, and that the present data are categorized in the data with medium RH conditions.

Line 267: *Thanks to add the uncertainties on the ΔH_{vap} obtained in this study*

Reply> Estimating the uncertainties of the VBS parameters arithmetically is challenging. Instead, obtaining sensitivity analyses of the four-product VBS fitting curves by changing ΔH_{vap} while fixing the stoichiometric yields α_i revealed that the effective ΔH_{vap} could be in the 0 to 70 and 0 to 80 kJ mol^{-1} ranges under neutral and acidic seed conditions, respectively. This has been stated in the revised manuscript, as follows: “Notably, sensitivity analyses achieved by fixing the stoichiometric yields α_i while changing ΔH_{vap} and comparing the resulting VBS curves with measured data (Fig. S8) indicated that the effective ΔH_{vap} could be in the 0 to 70 and 0 to 80 kJ mol^{-1} ranges for neutral and acidic seed conditions, respectively.” (Page 10 lines 366–368).

Table 1: *Add the humidity in the table and give the same number of significant number*

Reply> The RH has been included in Table 1. The significant number of α_i has been unified.

Table 2: *It would be good to compare the data on a graphs, the comparison would be more visual. Why the comparison with CMAQv4.7 model in the table? It would be good also to perform it.*

Reply> Information on the CMAQv4.7 SOA module (Carlton et al., 2010) has been included in Table 2. A supplementary figure (Fig. S7) has been illustrated for comparison of ΔH_{vap} in the present study with those in previous studies based on ΔH_{vap} versus C^* plots.

Line 321-324: *« Among the 362 compounds identified through LC-ToF-MS analysis in this study (Table S3), 331 compounds were ascribed to VBS bin ranges of –8–3. The other 31 compounds were ascribed to higher VBS bin ranges of 4–6, which is unrealistic for the compounds in the aerosol phase. Additionally, those latter compounds only accounted for an average of 12 % of the total mass of identified compounds. Thus, only the former 331 compounds are targeted in the following discussions. » I think that the conclusion is very rapid. The VBS is not good consequently you remove 31 compounds. It would be good to add more comments on this compounds even if you decide to follow the discussion with 331 compounds. 31 compounds were ascribed to higher VBS bin ranges of 4–6, which is unrealistic for the compounds in the*

aerosol phase; it's not the first time this observation is performed. This observation strongly supports the resistance to diffusion imposed by the viscosity of the aerosol to the products formed inside the aerosol itself. As previously mentioned in the literature (Shiraiwa et al 2013) this observation indicates that volatility can be strongly minimized by viscosity and that relying only on gas-phase equilibria to represent partitioning can be misleading.

Reply> We agree with the reviewer's suggestion that high-volatility compounds produced inside the aerosol particles might have been wrapped into the particle phase because of the high viscosity of the BSOA. We further note that only less than a half of the compounds in VBS bin ranges greater than four are in the particle phase (Donahue et al., 2006), and that larger compounds might have been fragmented into small molecules during the ESI MS measurements (Fig. S10).

Accordingly, the sentence “, which is unrealistic for the compounds in the aerosol phase. Additionally, those latter compounds only accounted for an average of 12 % of the total mass of identified compounds. Thus, only the former 331 compounds are targeted in the following discussions.” has been modified to: “. Only the former 331 compounds are targeted in the following discussions for two reasons. First, less than a half of the compounds that belong to VBS bins four or greater could exist in the particle phase (Donahue et al., 2006), which would introduce large uncertainties for the estimation of the mass concentrations of the compounds in gas-phase from the particle phase. In addition, LC-ToF-MS analysis of pure compounds indicated that fragmentation of high molecular compounds during the ionization could occur, e.g., pinic acid ($C_9H_{14}O_4$) could be fragmented into $C_8H_{14}O_2$, and the latter was assigned to VBS bin 6 (Fig. S10). Note that due to the potentially high viscosity of the newly formed SOA, high-volatility compounds formed inside the aerosol particle could have been wrapped into the particle phase and detected (Shiraiwa et al., 2013b).” (Pages 11–12 lines 397–404).

Line 332: How is obtained the total amount? By SMPS? It would be good to add a sentence to explain that.

Reply> For each tentatively identified compound, the amount of the aerosol phase is the sum of the area of extracted ion chromatography. The gas phase was derived assuming gas-particle partitioning equilibrium. The total amount in [Line 332](#) (currently [Page 12 line 412](#)) is the sum of the aerosol phase and gaseous phase. The derivation of the quantity of the gas-phase was explained in the two sentences before [Line 332](#): “The corresponding gas phase concentrations of each compound were derived assuming gas-particle partitioning equilibrium (Odum et al., 1996). The ΔH_{vap} values derived in this study (Table 1) were used to calculate the saturation concentration under 278 and 288 K following Eq. (3).” (currently [Page 12 lines 409–411](#)).

Line 350, Figure 4: There is no axes for the yield in the graph B; it would be good to add it.

Reply> The curve does not represent the yields, but the α_i obtained from the four-product VBS fitting normalized based on the total mass fraction of compounds in VBS bins 0 to 3. This is explained in the current version of the text by changing the original expression “In Fig. 4b, the volatility distributions determined from SOA yield curves were also presented.” (original manuscript [Page 10 lines 343–344](#)) to “In Fig. 4b, the volatility distributions determined by normalizing the stoichiometric SOA yields (Table 2)

based on the total mass fractions of compounds in VBS bins 0 to 3 are also presented.” (Page 12 lines 428–429).

Line 448-449: Replace « M/z 223 was the most abundant OS identified at 298 k, followed by m/z 279, 281, 269, 283, 265, 253, 267, 251, 249, and 247. » by « m/z 223 was the most abundant OS identified at 298 K, followed by m/z 279, 281, 269, 283, 265, 253, 267, 251, 249, and 247. »

Reply> Modification has been made accordingly. In addition, the chemical formulas of each m/z are also presented (Page 16 lines 534–536).

Line 500, Table 3: it's difficult to follow the table 3, it would be good to simplify to know what is really proposed by this study. Pleased find an example of table.

<i>m/z</i>	<i>Formula</i>	<i>VBS bin</i>	<i>Tentative structure proposed in this study</i>	<i>Literature</i>
<i>[M-H]-</i>	<i>[M-H]-</i>			

Reply> Modifications have been made to Table 3. (Page 19).

SUPPORTING information

Table S1 Remove the C near O₃ in table S1. I would add if it's possible a column with Δ α -pinene IN THE TABLE S1. It would be good to add the uncertainties in the table. For the yield, it would be good to correct it by all the wall losses. Replace also SOA by m_{SOA}

Reply> In Table S1, the ^c near O₃ in has been removed; a column with Δα-Pinene (μg m⁻³) has been added; uncertainties of the SOA mass and yields have been added; SOA has been replaced with m_{SOA}. However, the correction of gas-phase wall-loss hasn't been reflected in Table S1. It is only qualitatively discussed in Sect. 4.1.

Table S2 I would add if it's possible a column with Δ α -pinene in the table S2. It would be good to add the uncertainties in the table. For the yield, it would be good to correct it by all the wall losses. Replace also SOA by m_{SOA}

Reply> In Table S2, a column with Δα-Pinene (μg m⁻³) has been added; uncertainties of the SOA mass and yields have been added; SOA has been replaced with m_{SOA}. However, the correction of gas-phase wall-loss hasn't been reflected in Table S2. It is only qualitatively discussed in Sect. 4.1.

Figure S2: It would be good to model these experiment of wall loss and add the model in this figure

Reply> The original Figure S2 is currently Figure S4. Model simulation of the particle wall-loss has been included in the figure. A discussion of those simulation results is now presented in Text S3.

References:

Carlton, A. G., Bhawe, P. V., Napelenok, S. L., Edney, E. D., Sarwar, G., Pinder, R. W., Pouliot, G. A., and Houyoux, M.: Model Representation of Secondary Organic Aerosol in CMAQv4.7, *Environmental Science & Technology*, 44, 8553-8560, 10.1021/es100636q, 2010.

Donahue, N. M., Robinson, A. L., Stanier, C. O., and Pandis, S. N.: Coupled partitioning, dilution, and chemical aging of semivolatile organics, *Environmental Science & Technology*, 40, 2635-2643, 10.1021/es052297c, 2006.

Faust, J. A., Wong, J. P. S., Lee, A. K. Y., and Abbatt, J. P. D.: Role of Aerosol Liquid Water in Secondary Organic Aerosol Formation from Volatile Organic Compounds, *Environmental Science & Technology*, 51, 1405-1413, 10.1021/acs.est.6b04700, 2017.

Hinds, W. C.: *Aerosol Technology: Properties, Behavior, and Measurement of Airborne Particles*, Wiley-Interscience, United States, 483 pp., 1999.

Krechmer, J. E., Pagonis, D., Ziemann, P. J., and Jimenez, J. L.: Quantification of Gas-Wall Partitioning in Teflon Environmental Chambers Using Rapid Bursts of Low-Volatility Oxidized Species Generated in Situ, *Environmental Science & Technology*, 50, 5757-5765, 10.1021/acs.est.6b00606, 2016.

Kristensen, K., Cui, T., Zhang, H., Gold, A., Glasius, M., and Surratt, J. D.: Dimers in alpha-pinene secondary organic aerosol: effect of hydroxyl radical, ozone, relative humidity and aerosol acidity, *Atmospheric Chemistry and Physics*, 14, 4201-4218, 10.5194/acp-14-4201-2014, 2014.

Lai, A. C. K., and Nazaroff, W. W.: Modeling indoor particle deposition from turbulent flow onto smooth surfaces, *Journal of Aerosol Science*, 31, 463-476, 2000.

Nah, T., McVay, R. C., Pierce, J. R., Seinfeld, J. H., and Ng, N. L.: Constraining uncertainties in particle-wall deposition correction during SOA formation in chamber experiments, *Atmospheric Chemistry and Physics*, 17, 2297-2310, 10.5194/acp-17-2297-2017, 2017.

Odum, J. R., Hoffmann, T., Bowman, F., Collins, D., Flagan, R. C., and Seinfeld, J. H.: Gas/particle partitioning and secondary organic aerosol yields, *Environmental Science & Technology*, 30, 2580-2585, 10.1021/es950943+, 1996.

Pathak, R. K., Presto, A. A., Lane, T. E., Stanier, C. O., Donahue, N. M., and Pandis, S. N.: Ozonolysis of alpha-pinene: parameterization of secondary organic aerosol mass fraction, *Atmospheric Chemistry and Physics*, 7, 3811-3821, 10.5194/acp-7-3811-2007, 2007a.

Pathak, R. K., Stanier, C. O., Donahue, N. M., and Pandis, S. N.: Ozonolysis of alpha-pinene at atmospherically relevant concentrations: Temperature dependence of aerosol mass fractions (yields), *Journal of Geophysical Research-Atmospheres*, 112, 10.1029/2006jd007436, 2007b.

Shiraiwa, M., Zuend, A., Bertram, A. K., and Seinfeld, J. H.: Gas-particle partitioning of atmospheric aerosols: interplay of physical state, non-ideal mixing and morphology, *Physical Chemistry Chemical Physics*, 15, 11441-11453, 10.1039/c3cp51595h, 2013b.

Tang, I. N., and Munkelwitz, H. R.: Aerosol growth studies—III ammonium bisulfate aerosols in a moist atmosphere, *Journal of Aerosol Science*, 8, 321-330, 10.1016/0021-8502(77)90019-2, 1977.

von Hessberg, C., von Hessberg, P., Poschl, U., Bilde, M., Nielsen, O. J., and Moortgat, G. K.: Temperature and humidity dependence of secondary organic aerosol yield from the ozonolysis of beta-pinene, *Atmospheric Chemistry and Physics*, 9, 3583-3599, 10.5194/acp-9-3583-2009, 2009.

Wang, N. X., Jorga, S. D., Pierce, J. R., Donahue, N. M., and Pandis, S. N.: Particle wall-loss correction methods in smog chamber experiments, *Atmospheric Measurement Techniques*, 11, 6577-6588, 10.5194/amt-11-6577-2018, 2018a.

Zhang, X., Cappa, C. D., Jathar, S. H., McVay, R. C., Ensberg, J. J., Kleeman, M. J., and Seinfeld, J. H.: Influence of vapor wall loss in laboratory chambers on yields of secondary organic aerosol, *Proceedings of the National Academy of Sciences of the United States of America*, 111, 5802-5807, 10.1073/pnas.1404727111, 2014.

References:

Bonn, B., G. Schuster, and G.K. Moortgat, *Influence of water vapor on the process of new particle formation during monoterpene ozonolysis. Journal of Physical Chemistry A*, 2002. **106**(12): p. 2869-2881.

Cocker, D.R., et al., *The effect of water on gas-particle partitioning of secondary organic aerosol. Part I: alpha-pinene/ozone system. Atmospheric Environment*, 2001. **35**(35): p. 6049-6072.

Glasius, M., et al., *Carboxylic acids in secondary aerosols from oxidation of cyclic monoterpenes by ozone. Environmental Science & Technology*, 2000. **34**(6): p. 1001-1010.

Hakola, H., et al., *Product Formation from the Gas-Phase Reactions of Oh Radicals and O₃ with a Series of Monoterpenes. Journal of Atmospheric Chemistry*, 1994. **18**(1): p. 75-102.

Jonsson, A.M., M. Hallquist, and E. Ljungstrom, *Impact of humidity on the ozone initiated oxidation of limonene, Delta(3)-carene, and alpha-pinene. Environmental Science & Technology*, 2006. **40**(1): p. 188-194.

Jonsson, A.M., M. Hallquist, and E. Ljungstrom, *The effect of temperature and water on secondary organic aerosol formation from ozonolysis of limonene, Delta(3)-carene and alpha-pinene. Atmospheric Chemistry and Physics*, 2008. **8**(21): p. 6541-6549.

Jonsson, A.M., M. Hallquist, and H. Saathoff, *Volatility of secondary organic aerosols from the ozone initiated oxidation of alpha-pinene and limonene. Journal of Aerosol Science*, 2007. **38**(8): p. 843-852.

Kenseth Christopher M., Nicholas J. Hafeman, Yuanlong Huang, Nathan F. Dalleska, Brian M. Stoltz, and John H. Seinfeld *Synthesis of Carboxylic Acid and Dimer Ester Surrogates to Constrain the Abundance and Distribution of Molecular Products in α -Pinene and β -Pinene Secondary Organic Aerosol Environmental Science & Technology Article ASAP DOI: 10.1021/acs.est.0c01566*

Lai K., A. C.; Nazaroff, W. W. *Modeling indoor particle deposition from turbulent flow onto smooth surfaces. J. Aerosol Sci.* 2000, 31 (4), 463–476.

Shiraiwa, M.; Zuend, A.; Bertram, A. K.; Seinfeld, J. H., *Gas-particle partitioning of atmospheric aerosols: interplay of physical state, non-ideal mixing and morphology. Physical Chemistry Chemical Physics* **2013**, 15 (27), 11441-11453.

Winterhalter, R., et al., *LC-MS analysis of aerosol particles from the oxidation of -pinene by ozone and OH radicals. Atmospheric Chemistry and Physics Discussions*, 2003. **3**: p. 1-39.

Yu, J.Z., et al., *Gas-phase ozone oxidation of monoterpenes: Gaseous and particulate products. Journal of Atmospheric Chemistry*, 1999. **34**(2): p. 207-258.

Yu, Y., et al., Nitrate ion photochemistry at interfaces: a new mechanism for oxidation of alpha-pinene. *Physical Chemistry Chemical Physics*, 2008. **10**(21): p. 3063-3071.