

Response to Anonymous Referee #2

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

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.

Reply> We have added more explanations on the features of the chamber. The chamber is aimed for exploratory research, especially, to investigate the influence of the acidity of pre-existing particles on SOA formation at different temperatures. Using the present compact chamber, multiple experiments under different temperature, seed particle, relative humidity, oxidant, and radiation conditions can be executed in relatively short time. The major limitations of the present chamber are wall-loss of particles and oxidized vapors are high because of the high surface-to-volume ratio of the chamber. We have carefully revised the entire manuscript, while considering both the merits and limitations of the present chamber.

A brief explanation of the capabilities of the compact chamber after the discussion of its limitation in Sect. 4.1 has been included into the revised manuscript as follows: “We also note that the chamber is aimed for exploratory research, where multiple experiments under different temperature, seed particle, relative humidity, oxidant, and radiation conditions can be executed within relatively short periods.” (Page 10 lines 338–341).

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.

Reply> We have carefully estimated the uncertainties of the values reported in this study. Regarding the comparison with SOA yields of previous studies, we recognize that our values are largely underestimated when compared to those from the large chambers, mainly due to the wall-loss of oxidized vapors (Sect. 4.1). We focus on the influence of the acidity of seed particles on the SOA yields and chemical compositions, in addition to the influence of the chamber temperature (Sects. 4.2, 4.3). With regard to

the VBS distributions, we have referred to a recent work by Morino et al. (2020), where the most potential VBS distributions of the SOA from α -pinene ozonolysis among the reported ones were suggested based on model simulations of SOA concentrations from SOA formation experiments and VFRs (volume fraction remaining) from heating experiments. From the analysis of SOA compositions (Sec. 4.3), we attempted to find an indicator of the acidity of pre-existing particles in ambient aerosol, which we have highlighted in the revised manuscript.

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.

Reply> From the systematic experiments regarding the temperature and acidity dependence of SOA yields and chemical compositions from α -pinene ozonolysis, we claim the following findings: (1) The SOA yields increased with an increase in the acidity of seed particles (solid/near solid state) in the 278–298 K temperature range at low SOA mass loadings. If the SOA mass loading is too high compared to the amounts of pre-existing particles, the enhancement of the SOA yield under acidic conditions would be limited. (2) Whereas the abundance of some chemical compounds such as organosulfates and oligomers increased with an increase of the acidity of seed particles, the acid-catalyzed decomposition of some chemical compounds was also observed, which suggests that little or no enhancement of SOA could occur under acidic conditions in field observations when the acid-catalyzed decomposition is dominant. (3) The organosulfates and the oligomers that increased with an increase in the acidity of the seed particles could be indicators of the acidity of pre-existing particles in the field. The findings are highlighted in the revised manuscript.

Specific comments

Try to include all major results in the abstract.

Reply> We have revised the abstract carefully as follows:

“Secondary organic aerosols (SOAs) affect human health and climate change prediction; however, the factors (e.g., temperature, acidity of pre-existing particles, and oxidants) influencing their formation are not sufficiently resolved. Using a compact chamber, the temperature and acidity dependence of SOA yields and chemical components in SOA from α -pinene ozonolysis were systematically investigated under 278 K, 288 K, and 298 K temperatures using neutral $((\text{NH}_4)_2\text{SO}_4)$ /acidic $(\text{H}_2\text{SO}_4 + ((\text{NH}_4)_2\text{SO}_4))$ seed aerosols. SOA components with m/z less than 400 were analyzed using negative electrospray ionization liquid-chromatography time-of-flight mass spectrometry. Based on the slightly negative temperature dependence of the SOA yields, the enthalpies of vaporization under neutral and acidic seed conditions were estimated to be 25 and 44 kJ mol^{-1} , respectively. In addition, SOA yields increased with an increase in the acidity of seed particles (solid/near solid state) at low SOA mass loadings, when compared with the seed particle amounts. Acidity dependence analysis of the chemical formula, molecular mass, and O:C ratio of the detected compounds indicated the enhanced formation of multiple oligomers in the wide

molecular mass range with a wide range of O:C ratios under acidic seed conditions. The abundance of some chemical compounds increased with an increase in the acidity of seed particles (e.g., m/z 197, 311, 313, 339, 355 and 383), while decreases in the abundance of some chemical compounds were observed (e.g., m/z 171, 185, 215, 343, and 357). The acidity dependence could be explained by acid-catalyzed heterogeneous reactions or acid-catalyzed decomposition of hydroperoxides. In addition, organosulfate (OS) formation was observed under acidic seed conditions. Six out of the eleven detected OS were potentially formed via the aldehyde + HSO₄⁻ pathway.”

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

Reply> We have revised the abstract. We have added more explanations on the characteristics of the chamber.

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

Reply> The word “photooxidation” has been changed to “oxidation” ([page 1 line 26](#)).

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

Reply> The phrase “saturate concentrations” has been changed to “saturation concentrations” ([Page 1 line 34](#)).

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).

Reply> The LVOCs here indicate all low volatility organic compounds, including ELVOCs, with saturation concentrations less than 10^{-0.5} µg m⁻³. Regrettably, we noticed that the original description was not clear, and the reference was not appropriate. Hence, the original expression “The importance of the formation of low volatility organic compounds (LVOCs) with saturate concentrations of less than 10^{-0.5} µg m⁻³ has been highlighted in the SOA formation mechanisms of recent studies (Ehn et al., 2014; Shrivastava et al., 2017).” has been modified to

“The importance of the formation of low (and/or extremely low) volatility organic compounds (LVOCs) with saturation concentrations less than 10^{-0.5} µg m⁻³ through heterogeneous/multiphase accretion processes has been highlighted in SOA formation mechanisms in recent studies (Ziemann and Atkinson, 2012; Shrivastava et al., 2017).” ([Page 1 lines 34–36](#)).

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

Reply> The sentence “However, contrary results have been reported for the influence of the acidity of pre-existing particles on SOA yields from chamber experiments. For example, whereas Offenberg et al. (2009) reported a positive relationship between the acidity of seed particles and the ratio of SOA concentrations from the photooxidation of α -pinene with NO_x at an elevated acidity relative to neutral seed conditions, Eddingsaas et al. (2012) reported greater SOA yields under acidic than neutral seed conditions from photooxidation of α -pinene under high-NO_x conditions, and no influence of seed particle acidity under low-NO_x conditions.” has been modified to:

“However, the influence of the acidity of pre-existing particles on SOA yields from chamber experiments is poorly understood. Previous studies have reported complex results. For example, Eddingsaas et al. (2012) reported greater SOA yields under acidic than under neutral seed conditions from photooxidation of α -pinene under high-NO_x conditions, and no influence of seed particle acidity on SOA yields under low-NO_x conditions.” ([Page 2 line 50–53](#)).

Page 2 line 54: Do you mean contradictory results?

Reply> The phrase “contrary” has been changed to “inconsistent” ([Page 2 line 54](#)).

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

Reply> The citation has been added ([Page 3 line 97](#)).

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

Reply> The phrase “in atmospheric pressure” has been changed to “at atmospheric pressure” ([Page 3 line 119](#)).

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.

Reply> The humidification method, which was explained in the original manuscript as: “The RH of the air was adjusted by passing the G3 pure air through MiliQ water (resistivity of 18.2 M Ω ·cm) before entering the Teflon bag.”, has now been updated to “The RH of the air was adjusted by passing the G3 pure air through MiliQ water (resistivity of 18.2 M Ω ·cm, total organic carbon content \leq 5 ppb) before it entered the Teflon bag.”. ([Page 4 line 121–123](#)) No evident contamination from the humidification process of G3 air was observed according to results of blank chamber experiments (see Text S2).

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?

Reply> Ozone produced by irradiation of pure O₂ with vacuum ultraviolet light from a low-pressure mercury lamp ozone generator (Model 600, Jelight Company Inc., USA) was introduced into the chamber at a flow rate of 200 standard mL min⁻¹, for 1.5 min. After the ozone generator was turned off, the introduction of pure O₂ continued for another one minute to purge all generated ozone into the Teflon bag. The mixing in the chamber was achieved through the introduction of G3 pure dry air at a flow rate of 20 standard L min⁻¹ for 1 min after the introduction of ozone. These have been explained in the manuscript as follows: “After obtaining the initial concentrations of α-pinene and seed particles, excess ozone produced by irradiation of pure O₂ with vacuum ultraviolet light from a low-pressure mercury lamp ozone generator (Model 600, Jelight Company Inc., USA) was introduced into the chamber at a flow rate of 200 standard mL min⁻¹ for 1.5 min, to initiate the ozonolysis reactions. After the ozone generator was turned off, the introduction of pure O₂ continued for another minute to purge all generated ozone into the Teflon bag. Subsequently, the G3 pure air was introduced for one minute to facilitate the mixing of the chamber air.” (Page 4 line 147–151)

The mixing by introducing air at a flow rate of 20 SLM was probably completed within 55 s from the time profile of α-pinene after its introduction in the experiments where ozone was introduced into the chamber before α-pinene. This has been explained in the manuscript as follows: “In the experiments in which O₃ was first introduced, the introduction of G3 pure air was sustained for one more minute after the injection of α-pinene to purge all α-pinene into the Teflon bag and to facilitate the mixing of the chamber air. In the latter case, the maximum α-pinene concentrations appeared within 55 s of its introduction, which indicated that the mixing by introducing air with a flow rate of 20 SLM was probably completed within 55 s.” (Page 4 line 153–157).

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.

Reply> The phrase “aerosols” has been modified to “aerosol particles” or “particles” throughout the manuscript when necessary.

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

Reply> At preliminary runs, we observed that the particle number and mass concentrations became sufficiently low after the clean process mentioned in the text. Indeed, the particle number and mass concentrations were reduced to less than 20 cm⁻³ and 0.02 μg m⁻³ (density = 1 g cm⁻³), respectively. This, in addition to a detailed explanation of the clean process, have been included in the manuscript by modifying the original expression in *Page 4 lines 139-140* “The Teflon bag was flushed with G3 pure air at least three times between two successive experimental runs, which took approximately 40 min.” to “Before each experimental run, the Teflon bag was cleaned by filling it with pure G3 air and then evacuating all the air from the bag at least three times, which took approximately 40 min. The very low chamber background particle concentrations indicate that the bag was sufficiently cleaned (Text S2).” (Page 5 lines 170–172).

Page 5 line 161: It should read: “saturation concentrations”.

Reply> The phrase “saturated concentrations” has been changed to “saturation concentrations” (Page 5 line 198).

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.*

Reply> The phrase “chemical formulae” has been changed to “molecular formulae” (Page 5 line 199). The compound names and molecular structures of the tentatively identified major products are now presented in Table S4. The calculation of C* was explained in Sect. 3.2 of the preprint manuscript, which corresponds to Sect. 3.3 in the revised manuscript (Page 7 lines 263–275).

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.

Reply> Although coagulation does not affect the total volume/mass of the aerosol particles, it influences the size distributions of the particles and the apparent wall-loss of sub-100 nm particles.

We think that electrostatic particle losses were low 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.

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?

Reply> Simulations of wall-loss performed with seed particles have been carried out and presented in Text S3, and are 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).

Comparisons of the reported size-resolved wall-loss rates with the results of Hu et al. (2014) and Wang et al. (2018a) have been included in the revised manuscript as follows: “The size-distributions of the measured particle wall-loss rates presented shapes similar to that of a 0.83 m³ Teflon chamber (Hu et al., 2014), whereas in the latter, the lowest wall-loss rates appeared in the smaller size end (~70–110 nm) and were greater (~0.2 hr⁻¹) than those in the present study. The large apparent wall-loss rates of sub-100 nm particles were also similar to those of a 1.5 m³ Teflon reactor (Wang et al., 2018a).” (Page 6 lines 210–213)

The capacity of the current chamber has been described in Sect. 4.1 in the revised manuscript as follows: “We also note that the chamber is aimed for exploratory research, where multiple experiments under different temperature, seed particle, relative humidity, oxidant, and radiation conditions can be executed within relatively short periods.” (Page 10 lines 338–340)

All seed particles used for the wall-loss rate measurements were in solid (neutral seed conditions) or near solid states (acidic seed conditions) in the studied 26–55 % RH range because they were dried into effloresced states before being introduced into the chamber (Tang and Munkelwitz, 1977).

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

Reply> Simulation results in Text S3 assuming pure sedimentation deposition indicates that it does not contribute much to the overall particle loss. Instead, model simulations indicate that turbulent deposition were likely the major reason for the deposition of super-200 nm particles. Therefore, the original expression “Large wall-loss was observed for particles with mobility diameters of less than 100 nm, caused mainly by coagulation (Wang et al., 2018a). Large wall-loss for particles with mobility diameters of larger than 200 nm was also observed. This is a shortcoming of this chamber and could be explained by the shorter sedimentation time in the compact space compared with large chambers.” has been changed to: “Large wall-loss was observed for particles with mobility diameters less than 100 nm and larger than 200 nm. The size-distributions of the measured particle wall-loss rates presented shapes similar to that of a 0.83 m³ Teflon chamber (Hu et al., 2014), whereas in the latter, the lowest wall-loss rates appeared in the smaller size end (~70–110 nm) and were greater (~0.2 hr⁻¹) than those in the present study. The large apparent wall-loss rates of sub-100 nm particles were also similar to those of a 1.5 m³ Teflon reactor (Wang et al., 2018a). 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 209–216)

In addition, according to the measurement data (see new Fig. S4), the wall-loss rates of super 200 nm particles tend to be greater for seed particles with smaller mean diameters, leading to an increase in bulk wall-loss rates. We cannot clearly explain it, but we decided to wait for 30 min after the introduction of seed particles before initiating the reaction so that the size distributions of the seed particles could shift to the larger size end in our study. This was also explained in the text (currently Page 6 lines 216–220).

Page 5 line 176: It should read: “larger particles”.

Reply> The word “large” has been changed to “larger” (page 6 line 220).

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.

Reply> The SOA mass was derived from its volume concentration, which was derived by the subtraction the volume of seed particles with the correction of the wall loss of particles (Text S4). This method would introduce larger uncertainty to the determined SOA mass if the increase in the volume concentration of particles as a result of SOA formation was low compared with the volume concentration of seed particles. As can be observed from Tables S1 and S2, the relative uncertainty tends to be greater when the SOA mass concentration or yield is smaller. This point is now explained in the revised manuscript as follows: “A detailed explanation of the derivation of m_{SOA} is presented in Text S4. Note that when the mass loadings of SOA are low, the obtained m_{SOA} and related yields retain greater uncertainties because the subtracted volume concentrations of seed particles from the measured volume concentrations are large (Mei et al., 2013).”. (page 6 line 235–238)

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)?

Reply> The uncertainties of the wall-loss correction method can be estimated as the uncertainty (2σ) of the residual of the first-order bulk-volume wall-loss constant fitting. The derived relative uncertainties (2σ divided by the arithmetic mean of the measured bulk volume of seed particles) were in the 2.7–13 % range (Text S4). The loss of semi volatile gases was only qualitatively discussed in Sect. 4.1.

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

Reply> The word “and” was added to the sentence (Page 7 line 264).

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.

Reply> The evolutions of the measured particle number-size distributions and total particle number concentrations are now presented in Fig. S6.

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

Reply> The uncertainties of the SOA mass and yield have been included in the manuscript, as well as the associated SI (Text S4, Tables S1 and S2, Fig. 3, and Page 8 line 287).

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

Reply> The original expression “As the SOA concentration was kept constant after 50 min, ...” has been modified to “As the corrected SOA particle concentration was constant after 50 min, ...” (Page 8 line 288).

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

Reply> The original expression "... concentration of SOA at 90 min could be underestimated ..." has been modified to "... concentration of SOA particles at 90 minutes would be underestimated ..." (Page 8 lines 289–290).

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)."

Reply> The original sentence "In this study, seven experiments with varied initial α -pinene concentrations (54–323 ppbv) at 298 K under neutral seed conditions were conducted under the RH conditions of approximately 26–27 % (Table S1)." has been modified to "In this study, seven experiments with different initial α -pinene concentrations (54–323 ppbv) at 298 K and neutral seeds were conducted under a RH condition of approximately 26–27 % (Table S1)." (Page 9 line 319–320).

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.

Reply> SMPS measurements with only ozone and OH scavenger in the chamber filled with G3 pure air (Text S2) indicated that the aerosol mass before the addition of seed particles and α -pinene was $0.037 \mu\text{g cm}^{-3}$, which was very low compared to the large mass of seed particles ($33.6\text{--}120 \mu\text{g cm}^{-3}$). (Unity density was assumed here.)

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.

Reply> The uncertainties of the final SOA mass concentration and yield have been included as error bars in Fig. 2. Note that although it is possible, the uncertainties of the real time SOA mass concentration and yield haven't been presented to maintain the clarity of the figure. For lower mass loadings, the relative uncertainties of both SOA loadings and yields tended to be larger because of the uncertainty that originated from subtraction of the volume concentration of seed particles was high (Mei et al., 2013). This point has been included in Sect. 3.1. The caption of Fig. 2 has been modified to clarify that the lines represent parameterization results by Pathak et al. (2007a). Only the results of Pathak et al. (2007a) were compared with the results of the present study because Pathak et al. (2007a) summarized the previous studies. Data recently reported by Saathoff et al. (2009), Wang et al. (2011), Wang et al. (2014), Nah et al. (2016), Ye et al. (2018), Kenseth et al. (2020), and Czoschke and Jang (2006) have been added in Fig. 2 in the revised manuscript, as follows.

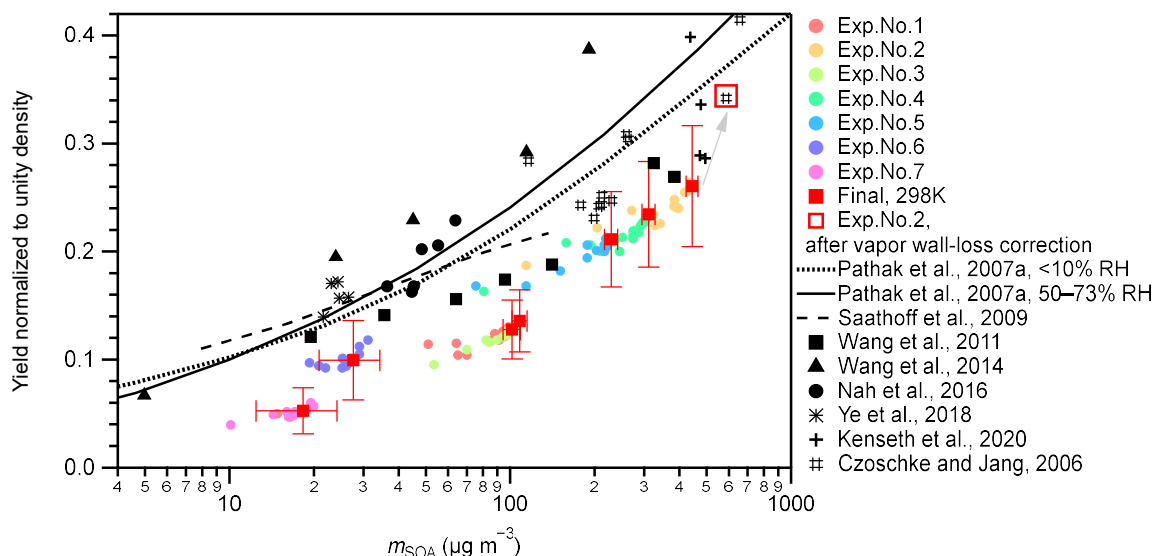


Figure 2: Yield comparison. SOA mass yields measured at 298 K under neutral seed conditions in the present study were compared to those of previous studies. Colored markers represent the results of this study. Colored circular markers represent the real-time SOA yields, i.e., the SOA yields along with the α -pinene ozonolysis reactions from 0 to 90 min. Different experimental runs are differentiated by colors. Red solid square markers represent the final SOA yields of the seven experiments. Horizontal error bars indicate the uncertainties of the final SOA concentrations; vertical error bars indicate the uncertainties of the final SOA yields. However, the systematic errors from vapor wall-loss are not included. The open square indicates the result of Exp. No. 2 after gas-phase wall-loss correction (Text S5). Black markers and curves represent results of previous studies. Black markers represent experimental results. The solid and dotted black curves represent the parameterized results from the four-product volatility basis-set fittings of previous α -pinene ozonolysis experiments under low NO_x and dark conditions summarized in Pathak et al. (2007a). The solid curve represents results under a 50–73 % RH range and the dotted curve represents results under RH<10 %. The dashed curve represents the results calculated using Eqs. 1, 6, 7, 10, and 11 at 303 K in Saathoff et al., 2009. The experiments of Saathoff et al. (2009) were carried out at 303 K, 48–37 and 0.02 % RH, without or with OH scavenger (cyclohexane or 2-butanol); experiments of Wang et al. (2011) were carried out at 295 K, < 1 % RH, without OH scavenger; experiments of Wang et al. (2014) were carried out at 295 K, < 5 % RH, without OH scavenger; experiments of Nah et al. (2016) were carried out at 298 K, < 5 % RH, with cyclohexane as OH scavenger; experiments of Ye et al. (2018) were carried out at 296 K, 12–14 and 48–49 % RH, with cyclohexane as OH scavenger; experiments of Kenseth et al. (2020) were carried out at 295 K, <5 % RH, without OH scavenger; and experiments of Czoschke and Jang (2006) were carried out at 294–300 K, 14–67 %, without OH scavenger. Note that all data presented in this figure are normalized to unity density (1 g cm⁻³).

(Page 10 lines 299–317)

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

Reply> Yes. The surface to volume ratio of the current compact 0.7 m³ chamber is 7.1 m⁻¹, while those of most previous studies were less than 3 m⁻¹. The whole sentence “Possible reasons may include no consideration of the wall-loss of oxidized organic vapors because the surface to volume ratio of the chamber used in this study could be much greater than those of previous studies (the volume of the chamber used in this study is 0.7 m³, whereas those of previous studies are 10–200 m³ (Pathak et al., 2007a and references therein))” has been modified to:

“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).” (Page 9 lines 325–327)

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.

Reply> We did not make any correction when we discussed the temperature dependence of SOA yields, i.e., when we derived the ΔH_{vap} values. We have estimated the wall-loss of the organic vapors for the experimental runs of which the volatility distributions were obtained from the chemical analysis (Text S5). We found that the ratios of the corrected SOA masses to the uncorrected ones had no substantial temperature-dependence and we have added the following sentences in the revised manuscript: “Furthermore, the vapor wall-loss correction factors of the SOA mass presented no obvious temperature-dependence (Text S5). Therefore, the temperature dependence of SOA yields will be discussed assuming that the underestimation of the SOA yield due to the wall-loss of oxidized organic vapors does not affect the temperature dependence.” (Page 10 lines 340–342).

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

Reply> We think it is difficult to estimate the uncertainties of the VBS parameters arithmetically. Instead, sensitivity analyses of the four-product VBS fitting curves achieved 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 for neutral and acidic seed conditions, respectively. This is now presented 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).

Page 8 line ~~266-267~~ 272: Do you mean: “...lowering of their vapor pressures.”?

Reply> The expression “...lowering their volatilities” means “...lowering of their saturation vapor pressures.” (Page 10 lines 354).

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

Reply> The expression “...lower than that of Saha et al. (2016) and much lower than that of Epstein et al. (2010).” has been modified to “...lower than those of Saha et al. (2016) and much lower than those of Epstein et al. (2010).” (Page 10 line 359–360).

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

Reply> The results of the present study indicated that when SOA concentrations were high, the acidity dependence could not be observed; however, it could be observed when SOA concentration was low. This is reasonable because the acidity effect decreases after the seed particles are covered with viscous SOA components (Shiraiwa et al., 2013b). The structure and part of the whole paragraph has been modified to as follows:

“Figure S9 presents the comparisons of the SOA yields under neutral and acidic seed conditions at different temperatures. It indicates that the SOA yields were enhanced under acidic seed conditions when the SOA loadings were low. When the SOA loadings were high, the enhancement disappeared. This is consistent with the results of Gao et al. (2004), which reported obvious initial α -pinene concentration dependence of the enhancement of SOA yields under acidic conditions when compared with neutral seed conditions. For the initial α -pinene concentrations of 12, 25, 48, 52, 96, 120, and 135 ppbv, the relative enhancements of SOA yields were 37, 34, 26, 24, 15, 10, and 8 %, respectively (Gao et al., 2004). This is probably because the SOA components can be of high viscosity under conditions where RH is smaller than around 50 %, and if high SOA mass loadings coated the seed particles, the acid-catalyzed heterogeneous SOA formation reactions could be impeded (Shiraiwa et al., 2013b; Zhou et al., 2013). In this study, the initial concentrations of α -pinene were 54–323 ppbv at 298 K, suggesting that the enhancement could be less than 24 %. When the SOA volume loading was $50 \mu\text{m}^3 \text{cm}^{-3}$, the fitted SOA yields under acidic conditions were enhanced by 11, 17, and 25 % when compared to the neutral seed conditions under 298, 288, and 278 K, respectively. This is consistent with the findings of Gao et al. (2004) and is also comparable to the results of Iinuma et al. (2005). In Iinuma et al. (2005), the experiment with 2-Butanol as an OH radical scavenger under room temperature (294–298 K) reported an enhancement of 19 %, with a final SOA volume concentration of approximately $50 \mu\text{m}^3 \text{cm}^{-3}$. Furthermore, the degree of acidity of the seed aerosols could have also influenced the enhancement (Gao et al., 2004; Czoschke and Jang, 2006). Further comprehensive studies are warranted (including the consideration of the particle viscosity and phase separation) on the influence of seed aerosol acidity on α -pinene ozonolysis SOA formation.” (Pages 10–11 lines 369–385).

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

Reply> Please refer to the responses to the previous comment (Pages 10–11 lines 369–385).

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

Reply> We suggest that the acidity dependence is more significant at low SOA concentrations. This should be confirmed by a more complex and larger chamber, in which the influence of wall-loss is relatively low.

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

Reply> The uncertainty of SOA mass loadings and yields have been included in Fig. 3. Comparisons to literature data at 298 K under neutral seed conditions had been included in Fig. 2, and associated discussions had been integrated in the second paragraph of Sect. 4.1 in the manuscript. Both Fig. 2 and the associated discussions have been updated in the current manuscript.

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

Reply> We think it is difficult to estimate the uncertainties of the VBS parameters arithmetically. Instead, sensitivity analyses of the four-product VBS fitting curves were achieved by changing ΔH_{vap} while fixing the stoichiometric yields α_i , revealing 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). Conversely, the α_i values were compared with the observations in Fig. 4b and in the text (Page 12 lines 428–432).

Comparisons of ΔH_{vap} with literature values was presented in Table 2 and discussed in the second paragraph of Sect. 4.2 in the manuscript.

According to Morino et al. (2020), both the root-mean-square errors between the observed and simulated SOA concentrations in the formation experiments and between the observed and simulated volume fraction remainings in the heating experiments were minimized in the case of the C^* distribution reported by Sato et al. (2018). As mentioned in the text, the C^* distribution patterns in this study were confirmed to be similar to those reported by Sato et al. (2018). We have noted this in the text as follows: “According to Morino et al. (2020), both the root-mean-square errors between the observed and simulated SOA concentrations for the formation experiments and between the observed and simulated volume fraction remainings for the heating experiments were minimized in the case of the C^* distribution reported by Sato et al. (2018).” (Page 12 lines 425–427).

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

Reply> The expression “...measured intensities of aerosol phase compounds...” has been modified to “...measured intensities of particle phase compounds...” (Page 12 line 405).

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

Reply> The expression “The intensity of both the aerosol and gas phases were...” has been modified to “The intensities of both the particle and gas phases were...” (Page 12 line 411).

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.

Reply> The expression “As the α -pinene ozonolysis rate constant does not vary much under the temperature range of 278–298 K (Akimoto, 2016) and...” has been modified to “As the α -pinene ozonolysis rate constant at the temperature range of 278–298 K does not vary considerably (within 15 %; IUPAC Task

Group on Atmospheric Chemical Kinetic Data Evaluation, (<http://iupac.pole-ether.fr>, last access: 1 February 2021)),...” (Page 12 lines 414–416).

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..

Reply> The small variation of the rate constant and the currently identified mechanisms (The Master Chemical Mechanism, <http://mcm.leeds.ac.uk/MCMv3.3.1/home.htm>) of α -pinene ozonolysis reactions indicate that the change of product branching ratios could be insignificant in the studied temperature range. In addition, the temperature-dependence of gas-phase wall-loss was also considered to be low (Text S5). The original expression “As the α -pinene ozonolysis rate constant does not vary much under the temperature range of 278–298 K (Akimoto, 2016) and α -pinene was completely consumed at the reaction time of 90 min, the total amount of formed SVOCs should be similar at the three temperatures.” has been modified to: “As the α -pinene ozonolysis rate constant at the temperature range of 278–298 K does not vary considerably (within 15 %; IUPAC Task Group on Atmospheric Chemical Kinetic Data Evaluation, (<http://iupac.pole-ether.fr>, last access: 1 February 2021)), α -pinene was completely consumed at the reaction time of 90 min, and the temperature-dependence of gas-phase wall-loss was considered insignificant (Text S5), the total amounts of SVOCs formed should be similar at the three temperatures.” (Page 12 lines 414–418) .

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

Reply> The phrase “aerosol” in both the y-axis label and the caption have been modified to “particle”.

It is difficult to estimate the uncertainty of the volatility distribution because the mass of compounds identified by LC-MS analysis could explain at most 30 % of the total SOA mass (Nozière et al., 2015). However, we note that the calculated distribution includes the uncertainties that result from compound specific sensitivities because the sensitivity of ESI mass spectrometry is compound specific in Sect. 3.3: “As has been noted previously, the sensitivity of ESI mass spectrometry is compound specific, thus the calculated distribution includes the uncertainties that result from compound specific sensitivities.” (Page 7 lines 275–276). In addition, the volatility distribution pattern obtained in this study is like those of Sato et al. (2018), in which the distribution obtained with the same method as in this study has been compared with other methods.

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

Reply> The expression “...were concentratedly distributed in the O:C ratio range...” has been modified to “...were concentrated in the O:C ratio range...” (Page 14 line 465).

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.

Reply> The abundances (i.e., total area of extracted ion chromatogram) of all the 362 chemicals that were determined in this study have been included in Table S3. The compound names and molecular structures of the tentatively determined major products are now presented in Table S4.

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

Reply> The chromatograms were extracted from the LC-TOF-MS data file for each OS assuming a relative m/z uncertainty of 20 ppm. The peaks that appeared at different retention times in the chromatogram should indicate isomers of the same OS formula. For OS with low signal intensities (i.e., m/z 247, 249, 251, 253, 267, 269, and 283), the variation of the baseline could also be observed from the figure. Note that the baseline wasn't subtracted from the EIC.

The Fig. S5 in the previous manuscript has now been numbered Fig. S11.

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

Reply> The phrase "might be of a high uncertainty" has been changed to "might be underestimated" (Page 16 line 518).

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

Reply> As has been explained previously, it is difficult to estimate the uncertainty of the volatility distributions. Both trends are significant. Panel (a) indicates the volatility distribution of SOA compounds that presented lower intensity under acidic seed conditions. Panel (b) indicates the volatility distribution of SOA compounds that presented higher intensity under acidic seed conditions. The mechanisms of such differences are interpreted in the manuscript.

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.

Reply> Molecular formulae have been included in the text. (Page 16 lines 534–536)

It is difficult to estimate the uncertainties of ethyl-d5-sulfate equivalent (EDSeq.) yields of OS compounds because we have only one dataset and the sensitivity of the ESI mass spectrometry is compound specific. This has been explained in the revised manuscript as follows:

"3.2 Derivation of the ethyl-d5-sulfate equivalent (EDSeq.) yield of OS

Before the extraction of filter samples, 20 μL sodium ethyl-d5 sulfate methanol solution ($50 \mu\text{g mL}^{-1}$) was added to each sample filter as an internal standard for the quantification of OS. The ethyl-d5-sulfate equivalent (EDSeq.) masses of OSs were determined by comparing the total chromatographic peak areas

of OSs to that of the EDS standard with known mass. The EDSeq. masses of OSs were divided by the corresponding air volumes collected to obtain the EDSeq. concentrations of OSs. The EDSeq. molecular yield of OS is defined as the ratio between the estimated EDSeq. concentration of OS (m_{OS} , $\mu\text{g cm}^{-3}$) and the reacted mass concentration of α -pinene (Δ_{VOC} , $\mu\text{g cm}^{-3}$). Note that the sensitivity of ESI mass spectrometry is compound specific; therefore, the calculated EDSeq. yield includes the uncertainties that result from compound specific sensitivities." (Page 7 lines 258–261).

The potential reaction mechanisms have been included in the supplementary file as Scheme S1, which is referred as "The former is referred hereafter as the alcohol pathway, and the latter the aldehyde pathway (aldehyde + HSO_4^- ; Surratt et al., 2007) (Scheme S1)." (Page 17 lines 555–556).

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."

Reply> The original sentence "The possible formation mechanisms of the OS compounds were proposed based on previous literatures combined with the experimental settings of this study." has been revised to "The potential formation mechanisms of the OS compounds were proposed based on literature data in combination with the results of the experiments of this study." (Page 17 line 537–538).

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,..."

Reply> The original expression "While all eleven OS compounds detected in this study were very likely from α -pinene ozonolysis reaction because the presence of excess OH scavenger in this study,..." has been modified to "While all eleven OS compounds detected in this study are formed from α -pinene ozonolysis due to the presence of excess OH radical scavenger,..." (Page 17 line 548–549).

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

Reply> The expression "...and one (m/z 265) was from the sulfation of the aldehyde compound, specifically pinonaldehyde (Liggio and Li, 2006; Surratt et al., 2007)." has been changed to "...and one (m/z 265) was said to be from the sulfation of pinonaldehyde (Liggio and Li, 2006; Surratt et al., 2007)." (Page 17 line 554–555).

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

Reply> The original expression "...could only be observed at relatively low temperatures." has been changed to "...could only be observed at the lower temperatures." (Page 17 line 570).

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.”

Reply> The original sentence “When the structure of the OS can be determined and the precursor compound can be expected, the formation mechanism of the OS in α -pinene ozonolysis with acidic seeds will be confirmed.” has been modified to “When the structures of the OSs can be determined and the precursor compounds can be assumed, the formation mechanisms of OSs in α -pinene ozonolysis with acidic seeds may be confirmed.” (Page 17 lines 577–578).

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 C7H11O6S and C10H15O7S.

Reply> The yield is related to the mass concentration of consumed α -pinene. It is the ethyl-d5-sulfate equivalent (EDSeq.) yield, the derivation of which has been included in the manuscript as **Sect. 3.2 Derivation of the ethyl-d5-sulfate equivalent (EDSeq.) yield of OS**. The original “Sect. 3.2 Volatility distribution analysis” has been moved to Sect. 3.3. The mean EDSeq. OS yields have been included in Fig. 8 and the interpretation of Fig. 8 has been modified to as follows:

“As shown in Fig. 8, the ethyl-d5-sulfate equivalent (EDSeq.) yields of m/z 247, 249, and 265 decreased with the increase of temperature. In fact, the OS at m/z 247 and 249 could only be observed at the lower temperatures. Conversely, the EDSeq. yields of other OS compounds increased with the temperature, or at least the yields at 278 K were the lowest among the three temperature conditions. The temperature dependence of OS EDSeq. yields seems not to be directly related to the formation mechanisms of either the alcohol pathway or the aldehyde pathway. Nevertheless, the mean EDSeq. yields of the eleven OS compounds, which were dominated by m/z 223 and m/z 279, increased with the increase of reaction temperature. As eleven OS (including three unreported) were observed in the α -pinene ozonolysis reactions with an OH scavenger and acidic seed particles, the identification of the structures of the OSs using high-resolution ion mobility mass spectrometry is planned as the next step. When the structures of the OSs can be determined and the precursor compounds can be assumed, the formation mechanisms of OSs in α -pinene ozonolysis with acidic seeds may be confirmed.” (Page 17 lines 569–578).

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

Reply> Modifications have been made accordingly.

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

Reply> The expression “...under acidic/neutral seed conditions.” has been modified to “...with acidic and neutral seed aerosol particles.” (Page 20 line 597).

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

Reply> The expression "Among the 362 identified compounds,..." has been modified to "Among the 362 compounds identified,..." (Page 20 line 605).

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.

Reply> We have revised the summary and conclusions, taking the responses to the general comments into consideration.

"Using the compact chamber system, SOA formation from α -pinene ozonolysis was studied with diethyl ether as an OH radical scavenger at temperatures of 278, 288, and 298 K, with acidic and neutral seed aerosol particles. The SOA yields and compounds with a molecular mass of less than 400 Da determined using a LC-TOF-MS were analyzed from the perspectives of temperature and seed particle acidity dependence.

The SOA yield increased slightly with the decrease of chamber temperature. The enthalpies of vaporization under neutral and acidic seed conditions was estimated to be 25 and 44 kJ mol⁻¹, respectively. The acidity dependence of the SOA yields at low SOA loadings were comparable to those reported by Gao et al. (2004) and Iinuma et al. (2005). However, the enhancement of the SOA yields under acidic conditions would be limited if the SOA mass loadings are much greater than the amounts of pre-existing particles.

Among the 362 compounds identified, the volatility of 331 was distributed in the VBS bins between -8 and 3. The temperature dependence of the volatility distribution of those identified compounds (particle phase + gas phase) could be consistently explained by the enthalpy of vaporization derived in this study.

The compounds whose intensities under acidic seed conditions were less than 0.9 times those of neutral seed conditions were dominated by monomers, whereas the compounds whose intensities under acidic seed conditions were more than 1.1 times those under neutral seed conditions were dominated by oligomers. The O:C ratios of the former were concentrated in the range of 0.4–0.75. The O:C ratios of the latter were broadly distributed. The compounds with O:C ratios less than 0.4 were all oligomers, which accounted for 61 % of the oligomers with high relative intensity under acidic conditions, whereas those with O:C ratios of greater than 0.75 were highly oxidized molecules, and only contributed to 1 % of the oligomers. In addition, the mean molecular mass of the former compounds (204 ± 4 g mol⁻¹) were evidently lower than those of the latter (284 ± 14 g mol⁻¹). The differences indicated that the formation of many oligomers, especially with low O:C ratios, was enhanced under acidic seed conditions. The acidity-dependence of certain major compounds could be explained by acid-catalyzed heterogeneous reactions (e.g., m/z 171, 185, 343, and 357) or acid-catalyzed decomposition reactions (e.g., m/z 215 and 197), which suggests that little or no enhancement of SOA under acidic conditions in field observations could occur when acid-catalyzed decomposition is dominant.

For the first time, organosulfate compounds were studied for α -pinene ozonolysis reaction in the presence of an OH scavenger and acidic seed particles. Eleven OS compounds were determined from LC-TOF-MS analysis. All of them on average presented higher yields under acidic than under neutral seed conditions.

Six of the OS compounds were potentially formed via the aldehyde + HSO₄⁻ pathway, which should be confirmed in future studies through high resolution mass spectrometry analyses.

Finally, the organosulfates and the oligomers that increased with an increase in acidity of the seed particles could be indicators of the acidity of pre-existing particles in the field, and the new findings obtained from this study should be confirmed using more complex and larger chambers.”

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.

Reply> The letter c at the head of the column with ozone concentrations in Table S1 is not necessary and has been omitted. In Table S2, the letter c is used to indicate that α-pinene was introduced earlier than ozone. The uncertainties of both SOA mass loadings and yields have been added, and the numbers of significant digits have been unified in both Tables S1 and S2.

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

Reply> Tentative names and molecular structures for some major products are now presented in Table S4. References for the C* calculation have been added as a footnote in Table S3.

Figure S3: Add a legend.

Reply> A legend has been added to the figure. The previous Fig. S3 is now numbered as Fig. S5.

Figure S4: Are the differences between the yield parameterizations significant?

Reply> The yield parameterizations represent the measured mean SOA mass loadings and yields. Based on these parameterization results, the difference was significant when the SOA loadings were low (< approximately 100 μg m⁻³) but not significant when the SOA loadings were high.

Is it correct that the temperature dependence of the yields is more significant than the acidity dependence?

Reply> It is difficult to tell which is significant based on the current data. Our aim was to provide information on the influence of particle acidity on SOA yield to SOA models such as CMAQ (Carlton et al., 2010; Pye et al., 2017). Although the temperature dependence of SOA formation in CMAQ is represented by ΔH_{vap}, the acidity dependence of monoterpene oxidations hasn't been considered. Our idea is to include the influence of particle acidity on the rate constants of the conversions from SVOC to NVOC in CMAQ model. This will be reported in a separate work.

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

Reply> The chromatograms were extracted from the LC-TOF-MS data file for each OS assuming a relative m/z uncertainty of 20 ppm. The peaks appeared at different retention times in the chromatogram should indicate isomers with the same OS formula. For OSs with low signal intensities (i.e., m/z 247, 249, 251, 253, 267, 269, and 283), the variation in the baseline could also be observed from the figure. Note that the baseline wasn't subtracted from the EIC.

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