In this document, we present, in the order the comments were raised by the reviewers, the changes made to the manuscript and/or our replies to the reviewers. We believe that we have satisfactorily and completely responded to each and all reviewer issues and that the paper is now in a significantly improved form. We hope that you agree and that the paper is now in publishable form. Thanks to the reviewers for their helpful comments.

Reply to **Anonymous Referee #1** on "Determination of alpha-pinene-derived organic nitrate yields: particle phase partitioning and hydrolysis" by J. D. Rindelaub et al.

Reviewer comments in **bold**, our replies after:

One potential inconsistency in the results cited by the authors concerns the lack of detection of the expected nitrate hydrolysis product, pinanediol. However, Bleier et al. (J. Phys. Chem. A, 117, 4223-4232, 2013) recently reported that the hydrolysis of alpha-pinene oxide leads to the ring-opened product, trans-sobrerol, rather than the ring-retaining product, pinanediol. Since both the anticipated dominant organic nitrate product in this work and alpha pinene oxide should hydrolyze through a common tertiary carbocation intermediate, it seems reasonable to expect that trans-sobrerol would also be a hydrolysis product in the present system. Since trans-sobrerol is commercially available (sold as trans-p-menth-6-ene-2,8-diol), the authors could quite easily confirm or refute the presence of trans-sobrerol as a product, which could help to further elucidate their proposed mechanism for organic nitrate hydrolysis.

On line 363, page 14 of the revised manuscript, we have added the following text: "One such isomer is trans-sobrerol, which has been identified as a ring-opening hydrolysis product of  $\alpha$ -pinene oxide by Bleier and Elrod (2013) and is also a likely product of the chemistry described in this study (the organic nitrate analog of trans-sobrerol is shown in Scheme 3). While trans-sobrerol was not observed in any filter samples, it was observed as a very minor product in denuder extracts. This is likely due to the partitioning of trans-sobrerol back into the gas phase after particle phase production. Bleier and Elrod (2013) report that trans-sobrerol can have further reactivity under acidic conditions, thus, particle phase chemical processing is likely a major reason why trans-sobrerol was not detected in filter extracts." Thank you for the suggestion. We added the Bleier and Elrod reference to the reference list.

On p. 17, line 379, we have modified the previous text to read as follows: "For instance, both pinanediol and *trans*-sobrerol could undergo further reactions, such as oligomerization, sulfate esterification, substitution or elimination mechanisms to produce new products. Example reactions for pinanediol are shown in Scheme 4. Of the products shown in Scheme 4, both the organosulfate product (Surratt et al., 2008; Eddingsaas et al., 2012) and keto-enol tautomerization product (Iinuma et al., 2013) have been previously observed. The proposed dimer resulting from Fischer esterification has not been identified to date."

On p. 17, line 392, we modified the previous text to now read: "The lack of detected particle phase pinanediol and *trans*-sobrerol in this study highlights the need for further study of the condensed phase chemistry of monoterpene oxidation products."

p. 3306, line 21: Since the Shepson group has extensive experience detecting and quantifying organic nitrates via GC-MS methods, there must be a compelling reason that the authors chose to use FT-IR methods instead. It would be helpful to readers to understand why the FT-IR detection method was selected.

Since all organic nitrates have a unique asymmetric stretch at  $\sim 1640~cm^{-1}$  (Nielsen et al., 1995) and do not have significant variation in molar extinction coefficients (Carrington, 1960), quantifying the total organic nitrate species using a commercially available isopropyl nitrate standard was deemed the most logical approach. Additionally, complete detection of organic nitrate species may not be possible using a GC-based method as highly oxidized compounds, such as nitrated organosulfates, will not elute through the column to the detector. On page 6, line 128, we now state: "As the full complement of  $\alpha$ -pinene-derived nitrate standards are currently unavailable, quantification of organic nitrate products using a GC-based method was not possible for this study. Additionally, FT-IR presents the advantage of detecting all organic compounds with the RONO<sub>2</sub> functionality and does not require that they quantitatively pass through a column."

p. 3306, line 28: Since tetrachloroethylene has quite a congested IR spectrum in the region of interest for organic nitrates, I'm assuming that solvent properties dictated this choice. Again, it would be useful to explicitly explain this choice.

On pg. 6, line 135, we have added the following statement: "Tetrachloroethylene was chosen as a solvent because it has minimal absorption features in the infrared region. However, as observed in the IR spectra (Fig. 1), bands from weaker absorption features are more prominent in liquid phase spectra. The non-organic nitrate bands in Fig. 1 are believed to be of solvent origin as both the blank extract and  $C_2Cl_4$  solvent had identical FT-IR spectra."

## p. 3307, line 18: How sensitive are the derived organic nitrate product yields to the assumptions concerning OH rate constants and NOx recycling efficiency?

On page 7, line 160 of the revision, we now state: "Due to the relatively low value estimated for the effective OH rate constant with monoterpene-derived nitrates,  $1.0 \times 10^{-12} \, \mathrm{cm^3}$  molecule-1 s-1, variations in the above assumptions do not have a large impact on the calculated yields, i.e. shifting the effective OH rate constant by an order of magnitude corresponds to less than a 2% difference in final yield values."

p. 3317, line 18: Since the partitioning efficiency is apparently very high at RH > 20% for the alpha-pinene derived organic nitrates, it leads one to consider whether isoprene-derived organic nitrates might also be able to partition significantly (even though one would expect that isoprene-derived organic nitrate partitioning coefficients are probably significantly smaller than the alpha-derived organic nitrates). Given the relative dearth of elevated RH chamber experiments, do the authors think that this kind of partitioning is possible for

## isoprene-derived organic nitrates but has simply has not been observed because of the experimental conditions used in previous studies?

The partitioning of smaller organic nitrate compounds, such as those derived from isoprene, is beyond the scope of this current study. However, our data do suggest that if tertiary or secondary hydroxy nitrate organics do partition into the aerosol phase, hydrolysis chemistry is likely to ensue. As pointed out by the reviewer, a shortage of high humidity chamber studies leaves many open questions about SOA partitioning and particle phase chemistry, specifically for isoprene-derived products. We or others will likely pursue this question in the future!

Reply to **Anonymous Referee #2** on "Determination of alpha-pinene-derived organic nitrate yields: particle phase partitioning and hydrolysis" by J. D. Rindelaub et al.

Reviewer comments in **bold**, **our replies after**:

This reviewer found some of the distinctions made by the authors difficult to follow from the data they show. For example, Figures 3 and 4 show organic nitrate yields as a function of relative humidity. The data for the acidic and unseeded case at low RH (about < 15% RH) are very scattered, covering factors of 3-5. Are there some experimental reasons for this large amount of scatter?

DuPont, the chemical provider of our chamber's Teflon, estimates a water transmission rate of 2 g  $m^{-2}$  day<sup>-1</sup> for PFA Teflon used for our chamber walls

(http://www2.dupont.com/Teflon\_Industrial/en\_US/assets/downloads/DuPont\_Teflon\_PFA\_Film\_Properties\_Bulletin\_K26944.pdf). So for the 28.27  $m^2$  area of exposed chamber Teflon and an average of 2.5 hr for experimental and sampling time, it is possible for 5.9 g of  $H_2O$  to enter the chamber. This corresponds to an estimated increase of 4.0% RH over the experimental and sampling timescale. As seen in Figs. 3-4, the organic nitrate yields are highly dependent on chamber RH, especially at low RH. Thus, the higher degree of scatter at low RH levels is likely due to this effect.

Additionally, variations in sampling time and seed aerosol acidity, arising from differing levels of  $HNO_3$  present in aerosols, may have influence on the organic nitrate yields over the very sensitive low RH conditions.

On page 8, line 188 of the revision, we added the following sentence; "Since organic nitrate yields were extremely sensitive to humidity over the low RH range, the higher degree of scatter under dry conditions in Figs. 3 and 4 may be caused by water vapor permeating into the chamber from the laboratory, potentially significantly changing the RH through the course of the experiment, in this very RH-sensitive regime. The water vapor transmission rate for the PFA Teflon used in this experiment is estimated at 2 g  $m^{-2}$  day<sup>-1</sup> (DuPont<sup>TM</sup>)."

It is not clear from the figure caption whether each point represents one experiment or an average of several (i.e., does "multiple experiments" mean each point is one experiment or each is an average of a number of experiments?).

The revised manuscript now states more clearly in the Fig. 3 caption: "The total organic yield as a function of chamber relative humidity for both the acidic seed aerosol ( $\circ$ ) and neutral seed aerosol ( $\circ$ ) experiments. Each data point represents the organic nitrate yield from a single experiment.", and in the Fig. 4 caption: "The total organic nitrate yield as a function of chamber relative humidity for the unseeded aerosol experiments. Each data point represents the organic nitrate yield from a single experiment."

Assuming each point is one experiment, would it not make more sense, given the scatter, to average these lower RH experiments and show error bars in both the X- and Y- axes, with the data from individual runs in a table to show the ranges?

We find that there are not enough data points for data-averaging to be an effective method for creating a more visually appealing figure. Tabulated results, however, may be useful for the reader and have been added to the manuscript as Tables 1 (seed aerosol experiments) and Table 2 (unseeded experiments).

The revision now states on pg. 9 that we are also providing the data in Table form. These data also help express the answer to the previous question, and make clear the scope and scale of the effort for this paper.

On page 9, line 198 of the revision, we added the following sentence: "Table 1 shows results from both the acidic and neutral seed experiments where the  $A_i$  and  $F_i$  parameters correspond to the concentration, in ppb, of  $RONO_2$  in the gas phase and particle phase, respectively. Table 2 shows the analogous results for the unseeded experiments."

With this much scatter, it is not at all clear how they could carry out a linear regression on the low humidity data (what range did they use?) to get a yield from the Y-intercept, particularly to give an error of only 6% on the yield of 23% they derive.

We have now discussed that there is one point in Figure 3 for the acidic aerosol case that is a clear statistical outlier, with the following addition on page 9, line 195: "The low humidity data from 0 to 20% RH was used for linear regression analysis because, as shown in Fig. 3, the observed yield is very humidity-dependent over this range, and the use of a larger range would underestimate the yield. The acidic seed data point at  $\sim$ 20% RH was omitted from the least squares fit after analysis of its Cook's distance (Cook, 1977)."

On page 9, line 199, the revised manuscript now reads: "Combining the standard errors of the y-intercepts, we report a  $26\pm7\%$  total organic nitrate yield from this oxidation pathway of  $\alpha$ -pinene."

The scatter at low RH for the unseeded aerosol (Fig. 4) is about the same as for the acidic aerosol (Fig. 3) but clearly the intercept in Fig. 4 at low RH would be very hard to determine.

The scatter at low RH for the unseeded aerosol (Fig. 4) is too large to enable determination of a reliable intercept in that case. For this reason, estimated y-intercept and organic nitrate yield values were not reported for the unseeded data set. On page 9, line 201 of the revision, we state: "Due to the high degree of scatter in the unseeded data at low RH (Fig. 4) and the lack of a statistically significant slope from a least squares fit, an analogous linear extrapolation from the unseeded plot was not conducted."

Figure 6 shows organic nitrate yields separately for gas and aerosol. It would be helpful to show the paired data, i.e. in one experiment, what are the values for each? I think this is essentially what is shown in Fig 7, so they might just refer to that in discussion of Fig. 6. If this interpretation is correct, it looks like the ratio of gas to particle nitrate varies by three orders of magnitude for each set of experiments at RH below 40%. A question then is what can be derived from the data with this much scatter.

Figure 6 shows a comparison of gas and particle phase yields for the acidic seed aerosol experiments as a function of experimental relative humidity while the plot in Fig.7 shows the partition coefficients as a function of RH. To address your question we added Fig. 6a in which we plot the observed F/A ratio for both seed aerosol experiments as a function of RH. As seen in the new figure, the neutral seed aerosol maintains an F/A ratio greater than 1.0 at high RH while the F/A ratio in the acidic case approaches zero. What is derived and discussed is that in the acidic high RH case aerosol phase organic nitrates are quickly and completely destroyed, which is useful new information.

Figure 6 caption: "Fig. 6. A.) The ratio of particle phase organic nitrate yield to gas phase organic nitrate yield ( $F_i/A_i$ ) as a function of experimental relative humidity for both the acidic seed aerosol ( $^{\circ}$ ) and neutral seed aerosol ( $^{\circ}$ ) experiments. B.) The gas ( $^{\circ}$ ) and particle phase ( $^{\circ}$ ) organic nitrate yields as a function of experimental relative humidity for the acidic seed experiments. In both plots, each data point represents a single experiment."

Page 10, line 229 of the revision now reads: "The higher degree of scatter in the organic nitrate yields at low RH compared to high RH may be related to the difficulty in maintaining low RH levels resulting from the Teflon chamber's permeability to room  $H_2O$ . In Fig. 6a, we show the ratio of the measured particle phase organic nitrate concentration to gas phase organic nitrate concentration  $(F_i/A_i)$  as a function of relative humidity for each experiment, for the acidic and neutral seed experiments. Figure 6b shows the gas phase and aerosol phase yields separately for the acidic seed experiments."

We now state, on page 13, line 291 of the revision: "Figure 6 indicates that for the high aerosol loadings in these experiments, much of the organic nitrate mass is in the particle phase. The  $F_i/A_i$  ratio of organic nitrates in the system, where  $F_i$  and  $A_i$  are the particle and gas phase concentrations, respectively, was greater than 1.0 for all seed aerosol systems when averaged over the low humidity range (0-20% RH). At elevated RH, only the acidic seed aerosol case revealed  $F_i/A_i$ 

ratios below 1.0, as shown in Fig. 6a. This is consistent with more facile consumption of the organic nitrate in the particle phase under acidic conditions, as shown in Figure 6b."

Much of the last part of the paper is devoted to a discussion of mechanisms and possible products. However, no product data is given (could they not obtain information on specific products from their GC-NICI-MS measurements?), except to say the one product they looked for that might be expected from the nitrate product reaction (pinanediol) was not observed.

Specific products were not able to be identified by GC-NICI-MS because the technique specializes in detection of electronegative compounds such as alkyl halides and nitrates, and in the latter case we do not have available standards relevant to the chemistry of this study.

However, we did identify *trans*-sobrerol in the gas phase and now state, on page 6, line 122: "Filter and denuder extracts were analyzed using GC-(EI)-MS with focus on the detection of expected products with commercially available standards."

## Given the lack of product data, the extensive discussion of potential mechanims and products and five reaction schemes seems inappropriate.

We feel that the use of reaction schemes in this study is necessary to communicate with some mechanistic detail our understanding of the chemistry behind the major conclusions of this manuscript. With the exception of the dimer product shown in Figure 4, the compounds identified in these reaction schemes are not potential products, rather, products that are known to originate from  $\alpha$ -pinene oxidation chemistry, as reported in previous literature. The exception shown in Scheme 4 (dimer product) is a proposed product derived from a very well-known chemical mechanism, Fischer esterification. Since many organic products have yet to be identified in the particle phase, elucidation of condensed phase chemical mechanisms is essential in the effort to further understand aerosol phase chemical processing, and some mechanistic discussion seems warranted. On page 17, line 386 of the revision, we made clear that this mechanism is a speculative example.

The first reaction scheme explains the initial gas phase oxidation mechanism of  $\alpha$ -pinene. It is essential in understanding the gas phase oxidation chemistry and how it relates to NOx, ozone, and regional air quality.

The second scheme is important for the reader to understand hydrolysis chemistry of organic nitrates, which has shown to produce the corresponding alcohol in previous studies (Darer et al. 2010, Hu et al. 2011). Additionally, this scheme also helps clarify our discussion (page 11) of the  $S_N 1$  nature of tertiary organic nitrate hydrolysis.

The third scheme is the rearrangement chemistry proposed by Peeters et al. (2001). It allows the reader to understand the complexity of the organic nitrate products and why analysis (e.g. chromatography) of the reaction mixture was a challenge. Additionally, it shows the chemical rearrangement that may be responsible for the absence of the expected hydrolysis product, pinanediol, as discussed on page 16 of the revision.

The fourth scheme is shown so the reader can understand the further chemical pathways that may be responsible for absence of the hydrolysis product. The organosulfate product on the right of this scheme has been observed in previous studies (Surratt et al., 2008; Eddingsaas et al., 2012) and the keto-enol tautomer of the middle product has also been identified previously (Iinuma et al., 2013). The product on the left has not been identified to date, however, the chemistry in this scheme occurs via Fischer esterification, which is expected to occur at low pH.

The fifth scheme shows that the ketone product presented, which has been identified previously from  $\alpha$ -pinene oxidation experiments (Iinuma et al., 2013), may be a direct product of organic nitrate hydrolysis in addition to a product of pinanediol chemistry. It also indicates that nucleophilic substitution is not the only type of chemistry that is likely to occur in the particle phase.

We have provided two changes; line 383 on pg. 17 now reads: "Example reactions for pinanediol are shown in Scheme 4. Of the products shown in Scheme 4, both the organosulfate product (Surratt et al., 2008; Eddingsaas et al., 2012) and keto-enol tautomerization product (Iinuma et al., 2013) have been previously observed. The proposed dimer resulting from Fischer esterification has not been identified to date." And the new caption for Scheme 5 now reads: "The acid-catalyzed E1 elimination mechanism for a proposed  $\alpha$ -pinene-derived organic nitrate in the condensed phase. This mechanism is in direct competition with the  $S_N1$  mechanism. The product shown, pinocamphone, has been previously identified in a laboratory-based study (Iinuma et al., 2013)."

This reviewer found the discussion of partitioning confusing. The calculated time for uptake into the particles is 194 s, which is relatively short on the time scale of their experiments so that equilibrium between the gas and particle phase should be achieved. However, they state that the plot of Fi/Ai against aerosol mass that should be linear in this case is "highly scattered and does not show such a relationship... cannot be explained by a simple gasparticle equilibrium model". If the plot is that scattered (it is not shown), can they be confident that it really rules out the equilibrium model? And if it is not, what are the alternatives and what would the data be expected to show in that case? On page 13, line 288, the sentence beginning "Then, to maintain equilibrium, uptake of gas phase organic nitrates to the particle phase will occur...." implies that there is equilibrium partitioning occurring, while it was argued a few lines above that it does not.

As seen in the revised Fig. 6a, the F/A ratio is highly dependent on chamber humidity. Thus, the high amount of scatter from the F/A vs. M plot is likely derived from  $RONO_2$  particle phase elimination, providing further evidence that the partitioning in our system has a dependence on particle phase hydrolysis.

Due to the consumption of particle phase  $RONO_2$ , the partitioning model described by Pankow (1994) cannot accurately describe the observed partitioning of our system. The  $RONO_2$  products partition into the aerosol phase and are quickly consumed via hydrolysis before a gas/particle equilibrium can be reached. As this consumption of  $RONO_2$  occurs, more  $RONO_2$  products continually partition into the aerosol to undergo the same fate, leading to an "apparent non-

equilibrium partition system" (see: pg. 14, line 318) where the time scale for hydrolysis is shorter than the time scale for evaporation from the particles.

We now state on pg. 14, line 316, "Instead, it is likely organic nitrate products are partitioning into the aerosol phase and either consumed or converted to an organic nitrate product with a different volatility before a gas-particle equilibrium can be reached, leading to an apparent non-equilibrium partitioning system."

And on pg. 15, line 327, "The decrease in apparent particle phase organic nitrate yields can be explained by hydrolysis in the aerosol phase, where the consumption of organic nitrates perturbs the equilibrium of the system, causing further partitioning of gas phase organic nitrates into the particle phase, followed by subsequent hydrolysis, lowering the apparent yields for both phases, as shown in Fig. 6b."

Similarly, in "Conclusion", it says "The concentration of organic nitrates present after equilibration with particles ..." and also that at high relative humidity, "liquid-like particles where equilibrium partitioning readily occurs.." This reviewer is confused about whether the authors believe equilibrium partitioning occurs, and if so, what data show this, also why the statement is made that the data are not consistent with this.

See our reply to the question above. It will be difficult to assess whether equilibrium conditions truly exist as this will depend on knowledge of the time scale for uptake and release to/from the particles, requiring knowledge of the mass accommodation coefficients for all the nitrates, and how these timescales compare with those for reactive loss in the particles and diffusion within the particles. So, we do not have enough quantitative information to be able to provide a quantitative answer to this question. We have, however, modified the text on page 19, line 416 to read: "The concentration of organic nitrates present after interaction with particles was found to be highly dependent on relative humidity and seed aerosol acidity for both the gas and particle phase products." And, on line 418: "High relative humidity may create well-mixed, liquid-like particles where partitioning readily occurs, followed by the hydrolysis of organic nitrate species, continued partitioning and elimination of such species, and an observed apparent non-equilibrium gasparticle partitioning system."

A related statement in the Conclusion is that "hydrolysis alone does not account for all the variability in the Kp values"; it is not clear to this reviewer what this statement is based on.

This statement is simply based on literature observation of multiple condensed phase processes for similar compounds. On page 19, line 429 of the revision we rephrase to: "However, hydrolysis alone may not account for all the variability in the observed Kp values due to low correlation with chamber RH (Fig. 7), and so other condensed phase reactions that have impact on the  $RONO_2$  functional group may also be important."

Experimentally, there are several questions: 1. Page 5, line 107: The chamber was flushed with air to clean it. Is this enough? Matsunaga and Ziemann (Aer. Sci. Tech- nol.) showed that Teflon takes up organics, particularly polar species that would be formed in these reactions,

and releases them back into the gas phase. Could this be responsible for the non-linear plot of aerosol mass vs time in Fig. 2 where there seems to be more mass than expected initially?

The chamber was flushed extensively with high purity air prior to experiments and, which the original manuscript fails to mention, was done under irradiation. While the uptake of organic compounds to Teflon is of concern, the flushing procedure is believed to release any adsorbed organics from the walls prior to the experiments. This is supported by various chamber blanks taken before injection of any experimental compound, such as those taken by GC-FID for detection of organics and SMPS for aerosol mass concentration measurements. We now state on pg. 5, line 110: "The chamber was flushed continuously under irradiation with at least five chamber volumes of hydrocarbon-free air prior to each experiment."

2. The infrared spectrum in Fig. 1 shows a number of strong peaks that do not appear to be the CCl4 solvent, but surprisingly, they appear in the filter blank as well. Can the authors comment on these? Also, some are saturating at an absorbance of 2.5 and even the nitrate peak at 1640 cm-1 is quite strong, with an absorbance of about 1.3. The latter corresponds to only 5% of the analytical light beam getting through the sample and under these conditions, signals are often non-linear with concentration. This has probably been considered by the authors but a comment on this would help the reader. Would use of a cell of length less than 1 cm be appropriate?

The FTIR features in the solvent spectrum are believed to originate from the concentration-related prominence of weak absorption bands of the tetrachloroethylene ( $C_2Cl_4$ ) solvent typically not observed in FTIR-ATR or gas phase FTIR spectra. Support for this assumption lays in the extreme similarity between the FTIR spectra of filter blanks and the pure solvent. It is also possible these absorption bands could be related to impurities within the solvent.

A length of 1 cm was used due to concern arising from the low organic nitrate concentrations in extracts that occurred in several experiments, e.g. the high RH aerosol phase samples.

We have expanded our description on page 6, line 135 of the revision, to say: "Tetrachloroethylene was chosen as a solvent because it has minimal absorption features in the infrared region. However, as observed in the IR spectra (Fig. 1), bands from weaker absorption features are more prominent in liquid phase spectra. The non-organic nitrate bands in Fig. 1 are believed to be of solvent origin as both the blank extract and  $C_2Cl_4$  solvent had identical FT-IR spectra. The instrument used was calibrated over the range of absorbances observed in this study. Calibration curves were extremely linear, with  $R^2$  values >0.99, to absorbance values above those shown for the organic nitrate stretch in Fig. 1. "

3. Page 7, line 151: Because the particles grew rapidly beyond the SMPS limit, they were "unable to calculate aerosol yields or partition coefficients for the unseeded experiments". Could the sampling be done at shorter times before they have grown this large, or could the initial precursor concentrations be lowered?

Experimental and sampling times were based on the amount of  $\alpha$ -pinene consumption. Experiments were designed to conclude when approximately one-half the precursor BVOC was

consumed in an effort to focus on  $1^{st}$  generation products. The SOA in these experiments was typically well out of the measureable range by the time experiments ended.

The initial precursor concentrations could not be lower due to concern with detecting low product concentrations via FTIR.

4. Does the denuder strip only gas phase organic nitrates or can it also evaporate these compounds from the particles as they pass through?

Calibration of the denuder prior to experimental use indicated that the denuder could absorb organics with greater than 98% efficiency. Measurements downstream of the denuder did not reveal any evaporative losses.

5. What were the number and mass concentrations of the initial seeds?

We have added the following lines to the revision, on line 88, pg. 4 of the Experimental section: "Typical seed aerosol mass concentrations ranged from 50 to 150  $\mu$ g/m³ and number concentrations ranged from approx. 5.0 x 10<sup>4</sup> to 1.5 x 10<sup>5</sup> cm-³. Large initial seed aerosol concentrations were effective in keeping the SOA within the measureable range of the SMPS over the course of the experimental timescale. "

6. Page 10, line 235: "The gas phase yields were not dependent on seed aerosol composition". It appeared to this reviewer that only the acidic seed data are shown (Fig. 6), perhaps it should be shown for all cases?

In the revised manuscript, Fig. 6 shows the F/A ratio for both the neutral and acidic seed aerosol cases plotted as a function of chamber RH.

Technical/minor comments: 1. Page 4, line 81: The Chen et al. (1998) reference is not in the reference list.

This was repaired, thank you.

2. Page 4, line 92: Should "glass tee" (as in golf tee) be "glass T"?

Yes, it should read "glass tee".

3. Page 6, line 124: Need space between the number and unit in "1 cm".

This was repaired.

4. Figure 2: Rather than saying "a representative experiment", more details should be given in caption. For example, text says "a seed aerosol" experiment but in neither the text nor caption which seed was used is given.

The Fig. 2 caption was changed to: "The  $\alpha$ -pinene and aerosol mass concentrations as a function of time for a neutral seed experiment. The open circles ( $\circ$ ) represent  $\alpha$ -pinene (ppb) while the closed

circles ( $\ensuremath{\mathbb{Z}}$ ) represent the aerosol mass concentration ( $\mu g/m^3$ ). Initial  $\alpha$ -pinene and seed aerosol concentrations were 960 ppb and 110  $\mu g/m^3$ , respectively."