Responses to Referee # 1

We thank the reviewer for the careful review of our manuscript. The comments and suggestions are greatly appreciated. All the comments have been addressed. In the following, please find our responses to the comments one by one and the corresponding revisions made to the manuscript. The original comments are shown in italics. The revised parts of the manuscript are highlighted.

Anonymous Referee #1

Received and published: 26 April 2017

This manuscript describes the competing roles of NOx and SO2 on SOA formation of a-pinene and limonene. The ability of SO2 to enhance seed aerosol surface area appears to be a dominant factor, and that enhancing seed aerosol reduces the NOx suppression of SOA yields, at least in some monoterpenes. The authors use their AMS data to determine the role of organic nitrates in SOA, and find that organic nitrates account for a substantial fraction of the SOA mass. Overall, this is an interesting piece of work, and warrants publication in the ACP following some revision.

Major Comments

The nature of the experimental design was not so much to look at the impact of SO2 – but to look at the role of a sulfate seed aerosol. From the manuscript, my interpretation is that the SO2 additions were used to nucleate (inorganic) seed aerosol. Was there any SO2 left over to impact VOC oxidation? It is not clear to me if the SO2 additions really paralleled the NOx additions, because the experimental design was different. That's not to say that these aren't valuable experiments that add to the literature! I merely question whether this was truly an 'SO2 addition' rather than a 'sulfate aerosol addition' to VOC oxidation experiments.

Response:

We thank the reviewer for the supporting remarks.

In the experiments with SO_2 added, SO_2 concentration decreased slowly and most of SO_2 was still left (typically around 8 ppb) at the end of an experiment because of the low reactivity of SO_2 with OH (~2×10^{-12} molecules^{-1} cm^{3} s^{-1} at 298 K). SO_2 time series in a typical experiment are shown in a newly added figure (Fig. S2). Therefore, the experiments with SO_2 not only included the effect of sulfate formed from SO_2 oxidation as seed but also the potential role of SO_2 on VOC oxidation, although the role on VOC oxidation turned out to be likely not significant. We have added the follow sentence in the revised manuscript to clarify this point.

"SO_2 concentration decayed slowly in the experiments with SO_2 added and most of the SO_2 was still left (typically around 8 ppb from initial 15 ppb) at the end of an experiment due to its low reactivity with OH. Typical SO_2 time series in high SO_2 experiments are shown in Fig S2."

Although the SO_2 addition did not exactly parallel NO_x addition, by adding SO_2 and inducing nucleation first, we can make sure that in high SO_2 conditions enough nucleated particles were represent for the oxidation products to condense on once VOC oxidation started. Otherwise, it would be unclear whether the low SOA yield at high NO_x was due to missing nucleation or lack of condensable products. In this way, we can somewhat differentiate the role of promoting nucleation from the role of affecting the condensable products from VOC oxidation.

In the revised manuscript, we have added the following sentence.

"Adding SO_2 first and initializing nucleation by SO_2 photooxidation ensured that enough nucleating particles were present when VOC oxidation started."

The effect of SO_2 on VOC oxidation is provided in the response to one similar comment below (Pg. 7, lines 205-209).

SOA yield is influenced strongly by OA mass. The authors plot SOA yield versus OH dose, which is certainly a useful figure to see – but it is hard to compare the SOA yields if the SOA mass has not been accounted for. The authors need to also show SOA yield versus OA mass so that the readers can contrast the relationships to other studies. It would be useful to compare the SOA yields to other studies: how do the yield values compare to other measurements of OH oxidation of a-pinene? This will allow readers to place the studies in context.

Response:

We have accepted the reviewer's suggestion. In the revised manuscript, we have added a figure of SOA yield versus OA mass concentration (Fig. S8 and S9) and compared the SOA yield in this study to previous studies. We have also discussed other factors influencing SOA yield.

The results of SO2 and NOx effects on SOA yield are consistent with the Sarrafzadeh and Eddingsaas studies, which found that the presence of seed aerosol suppresses the 'NOx effect' on SOA yield. However, they contradict previous studies (e.g. Ng et al. 2007, Presto et al. 2005). The authors need to do a better job of contrasting their studies – they attribute the difference to a vague collection of parameters (e.g. NO:NO2 ratio, OH concentrations, etc.). It would be extremely helpful if the authors could synthesize the information (i.e. put numbers on those parameters) to help readers understand the differences in experimental conditions across the studies. A table would be particularly helpful.

Response:

We have accepted the reviewer's suggestions. In the revised manuscript, we have added one table summarizing the reaction conditions of previous studies (Table. S2) and elaborated the discussion related to the difference between our study and previous studies.

"…The reaction conditions of this study often differ from those described in the literature (see Table S2).

The difference in these conditions can result in both different apparent dependence on specific parameters and the varied SOA yield. For example, SOA yield from α-pinene photooxidation at low NO_x in this study appeared to be much lower than that in Eddingsaas et al. (2012a). The difference between the SOA yield in this study and some of previous studies and between the values in the literature can be attributed to several reasons: 1) RO_2 fates may be different. For example, in our study at low NO_x, RO_2+NO account for a large fraction of RO_2 loss while in Eddingsaas et al. (2012a) RO_2+HO_2 is the dominant pathway of RO_2 loss. This difference in RO_2 fates may affect oxidation products distribution. 2) The organic aerosol loading of this study is much lower than that some of previous studies (e.g., Eddingsaas et al. (2012a)) (see Fig. S9). SOA yields in this study were also plotted versus organic aerosol loading to better compare with previous studies (Fig. S8 and S9). 3) The total particle surface area in this study may also differ from previous studies, which may influence the apparent SOA yield due to vapor wall loss (the total particle surface area is often not reported in many previous studies to compare with). 4) RH of this study is different from many previous studies, which often used very low RH (<10%). It is important to emphasize that reaction conditions including the NO_x as well as SO_2 concentration range and RH in this study were chosen to be relevant to the anthropogenic-biogenic interactions in the ambient atmosphere. In addition, difference in the organic aerosol density used in yield calculation should be taken into account. In this study, SOA yield was derived using a density of 1 g cm^{-3} to better compare with many previous studies (e.g., (Henry et al., 2012)), while in some other studies SOA yield was derived using different density (e.g., 1.32 g cm^{-3} in Eddingsaas et al. (2012a))."

Lines 118: the use of the HR-ToF-AMS to derive elemental ratios uses the older Aiken method. However, as the authors note, the newer 2015 approach corrects some underestimation. Because readers may wish to compare results across studies in the future, it is appropriate and prudent to update the results to the newer calculations.

Response:

We have calculated the H/C and O/C data using the newer approach by Canagaratna et al. (2015) and compared them with the data derived from the older method (Aiken et al., 2007) (Fig. S1). The H/C derived using the newer method strongly correlated with that derived using older method and just increased by 27%. Similarly, O/C just increased by 11%. In the revised manuscript, we have discussed this difference.

"The H/C and O/C were also derived using the newer approach by Canagaratna et al. (2015) and compared with the data derived from the Aiken et al. (2007) method. The H/C values derived using the Canagaratna et al. (2015) method strongly correlated with the values derived using Aiken et al.

(2007) method (Fig. S1) and just increased by 27% as suggested by Canagaratna et al. (2015). Similar results were found for O/C and there was just a difference of 11% in O/C. Since only relative difference in elemental composition of SOA is studied here, only the data derived using Aiken et al. (2007) method are shown as the conclusion was not affected by the methods chosen."

Line 128: the authors note that they account for particle wall loss and dilution loss, but not for vapor wall loss. Recent papers have shown this to be a chemically-dependent and substantial effect on SOA yields, and most rigorous SOA yield work now accounts for these effects. How will ignoring vapor wall loss influence the results – and the interpretation thereof?

Response:

The wall loss of vapors causes an under-estimate of the SOA yield. In the revised manuscript, we have estimated the influence of the vapor wall loss on SOA yield using the measured wall loss rate of vapors. And we have added a section to address the influence of vapor wall loss.

"Wall loss of organic vapors

The loss of organic vapors on chamber walls can influence SOA yield (Kroll et al., 2007; Zhang et al., 2014; Ehn et al., 2014; Sarrafzadeh et al., 2016; McVay et al., 2016; Nah et al., 2016; Matsunaga and Ziemann, 2010; Ye et al., 2016; Loza et al., 2010). The wall loss rate of organic vapors in our chamber was estimated by following the decay of organic vapor concentrations after photooxidation was stopped in the experiments with low particle surface area (~5×10^{-8} cm^2 cm^{-3}) and thus low condensational sink on particles. Such method is similar to the method used in previous studies (Ehn et al., 2014; Sarrafzadeh et al., 2016; Krechmer et al., 2016; Zhang et al., 2015). A high-resolution time-of-flight chemical ionization mass spectrometer (HR-ToF-CIMS, Aerodyne Research Inc.) with nitrate ion source ($^{15}NO_3^-$) was used to measure semi/low-volatile organic vapors. The details of the instrument were described in our previous studies (Ehn et al., 2014; Sarrafzadeh et al., 2016). The decay of vapors started from the time when the roof of the chamber was closed. The data were acquired at a time resolution of 4 s. A typical decay of low-volatile organics is shown in Fig. S3 and the first-order wall loss rate was determined to be around 6×10^{-4} s^{-1}.

The SOA yield was not directly corrected for the vapor wall loss, but the influence of vapor wall loss on SOA yield was estimated using the method in the study of Sarrafzadeh et al. (2016) and the details of the method are described therein. Briefly, particle surface and chamber walls competed for the vapor loss (condensation) and the condensation on particles led to particle growth. The fraction of organic vapor loss to particles in the sum of the vapor loss to chamber walls and to particles (F_p) was calculated. The vapor loss to chamber walls was derived using the wall loss rate. The vapor loss to particles was derived using particle surface area concentration, molecular velocity and an accommodation coefficient α_p (Sarrafzadeh et al., 2016). $1/F_p$ (f_{corr}) provides the correction factor to obtain the "real" SOA yield. f_{corr} is a function of particle surface area concentration and accommodation coefficient as shown in Fig. S4. Here a range of 0.1-1 for α_p was used, which is generally in line with the ranges of α_p found by Nah et al. (2016) by fitting a vapor-particle dynamic model to experimental data. At a given α_p, the higher particle surface area, the lower f_{corr} and the lower the influence of vapor wall loss are because most vapors condense on particle surface and vice versa. At a given particle surface area, f_{corr} decreases with α_p because at higher α_p a larger fraction of vapors condenses on particles. An average molecular weight of 200 g/mol was used to estimate the influence of vapor wall loss. For the aerosol surface area range in most of the experiments in this study, the influence of vapor wall loss on SOA yield was relatively small (<~40% for particle surface area larger than 3×10^{-6} cm^2 cm^{-3}, Fig. S4). Yet, for the experiments at high NO_x and low SO_2 for α-pinene and limonene, the influence of vapor wall loss on SOA can be high due to the low particle surface area, especially at lower α_p."

In the Introduction, the authors do a good job of summarizing the reasons why such a study would be interesting. Much of the discussion focuses on the role of NOx on SOA yields – this is reasonable as most of the literature has focused on that problem! However, there is some relatively recent literature regarding the role of SO2 in affecting SOA chemistry and monoterpene OH oxidation that the authors should consider. In particular:

Photooxidation of cyclohexene in the presence of SO2: SOA yield and chemical composition.

Shijie Liu, Long Jia, Yongfu Xu, Narcisse T. Tsona, Shuangshuang Ge, and Lin Du. Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-30, 2017

Synergetic formation of secondary inorganic and organic aerosol: effect of SO2 and NH3 on particle formation and growth. Biwu Chu, Xiao Zhang, Yongchun Liu, Hong He, Yele Sun, Jingkun Jiang, Junhua Li, and Jiming Hao. Atmos. Chem. Phys., 16, 14219-14230, doi:10.5194/acp-16-14219-2016, 2016

Formation of secondary aerosols from gasoline vehicle exhaust when mixing with SO2. T. Liu, X. Wang, Q. Hu, W. Deng, Y. Zhang, X. Ding, X. Fu, F. Bernard, Z. Zhang, S. Lü, Q. He, X. Bi, J. Chen, Y. Sun, J. Yu, P. Peng, G. Sheng, and J. Fu. Atmos. Chem. Phys., 16, 675-689, doi:10.5194/acp-16-675-2016, 2016

Anthropogenic Sulfur Perturbations on Biogenic Oxidation: SO2 Additions Impact Gas- Phase OH Oxidation Products of _- and _-Pinene. Beth Friedman, Patrick Brophy, William H. Brune, and Delphine K. Farmer. Environmental Science & Technology 2016 50 (3), 1269-1279. DOI: 10.1021/acs.est.5b05010

Is there any evidence for organic sulfates in the SOA from the AMS data? This has been a subject of some debate in the literature, and an additional datapoint would be useful. This may also clarify the role of acid catalysis, as I believe that has been linked to the formation of organic sulfates.

Response:

We thank the reviewer for raising these papers. In the revised manuscript, we have enriched the discussion on the role of SO_2 by including some of the papers.

From our AMS data, we did not find evidence of organic sulfate. For SOA formed at high SO_2, we found no significant organic fragments containing sulfur. Also the fragment $CH_3SO_2^+$ from organic sulfate suggested by Farmer et al. (2010) was not detectable in our data. We found that the pattern of sulfate in mass spectra had no significant difference from the pattern of pure ammonium sulfate. However, we would like to note that AMS has very limited capability to differentiate organic sulfate and inorganic sulfate (Farmer et al., 2010).

Moreover, according to the literature, organic sulfate is mainly formed by aqueous reaction of sulfate with organics. In the conditions of our study, there was no aqueous phase as we stated based on the AIM model. Therefore, experimental conditions in our study did not favor the formation organic sulfate.

In the revised manuscript, we have clarified these results.

"In addition, from the AMS data of SOA formed at high SO_2 no significant organic fragments containing sulfur were found. Also the fragment $CH_3SO_2^+$ from organic sulfate suggested by Farmer et al. (2010) was not detected in our data. The absence of organic sulfate tracers is likely due to the lack of aqueous phase in aerosol particles in this study. Therefore, the influence of SO_2 on gas phase chemistry of organics and further on SOA yield via affecting gas phase chemistry is not important in this study."

Minor Comments

Line 136. The authors note an average RH of 28-42% for the experiments. This seems like a relatively large range: will this affect the SOA yields, or interpretation of the data?

Response:

The average RH was in the range of 28-42% taking into account all experiments. Actually, except one experiment, the average RH was in the range of 28-34%. For particle phase reactions, the particle water content absorbed by organic aerosol in the range of 28-42% RH is low and the difference of water content between 28% and 42 % is very minor (typically <~2% of the particle volume based on our hygroscopic growth measurement). The RH variations are not expected to significantly change the particle phase chemistry. Moreover, since water vapor is abundant and in excess in the gas phase, the

RH variations are not expected to significant change gas phase chemistry either. Therefore, we do not expect that would significantly change the SOA yield. In the revised manuscript, we have clarified this point.

"The average RH for the period of monoterpene photooxidation was 28-34% except for one experiment with average RH of 42% RH."

Re: Discussion of SO2 effects. The authors dominantly attribute the enhancement of SOA by SO2 to increased particle surface area, or perhaps to acid catalysis. These seem like extremely likely reasons; however, there is one study that suggests that SO2 will influence gas-phase oxidation products (Friedman et al.), which could also be a confounding factor unless all of the SO2 is in the particle phase before VOC oxidation commences… This would be a useful clarification.

Response:

The influence of SO_2 on gas phase oxidation is likely to be trivial in this study for two reasons. Firstly, the reactivity of SO_2 with OH is very low (2×10^{-12} vs. 5.3×10^{-11} molecules^{-1} cm^{3} s^{-1} for α-pinene with OH) and SO_2 only accounts for a very small fraction of the OH loss (typically ~2% in the beginning of an experiment). Secondly, the OH concentration is 2-3 orders of magnitude lower than those in the PAM chamber used by Friedman et al. (2016). Therefore, either the change in OH/HO_2 ratio or SO_3 concentration, which is attributed to the reason of changed oxidation products by Friedman et al. (2016), is much lower in our experiments. In the revised manuscript, we have added a brief discussion of the effect of SO_2 on gas phase oxidation of monoterpenes in this study as follows.

"SO_2 has been proposed to also affect gas phase chemistry of organics by changing the HO_2/OH or forming SO_3 (Friedman et al., 2016). In this study, the effect of SO_2 on gas phase chemistry of organics was not significant because of the much lower reactivity of SO_2 with OH compared with α-pinene and limonene (Atkinson et al., 2004, 2006; Atkinson and Arey, 2003) and the low OH concentrations (2-3 orders of magnitude lower than those in the study by Friedman et al. (2016)). Moreover, reactions of RO_2 with SO_2 was also not important because the reaction rate constant is very low ($<10^{-14}$ molecule^{-1} cm^{3} s^{-1}) (Lightfoot et al., 1992; Berndt et al., 2015). In addition, from the AMS data of SOA formed at high SO_2 no significant organic fragments containing sulfur were found. Also the fragment $CH_3SO_2^+$ from organic sulfate suggested by Farmer et al. (2010) was not detected in our data. The absence of organic sulfate tracers is likely due to the lack of aqueous phase in aerosol particles in this study. Therefore, the influence of SO_2 on gas phase chemistry of organics and further on SOA yield via affecting gas phase chemistry is not important in this study."

Technical comments.

Line 26: should read "compared to low NOx"

Response: Corrected.

Line 29: should read “SO2 can compensate for such effects”

Response: Corrected.

Introduction: line 34: sentence has repetitive ‘importants’: consider removing at least one (e.g. “SOA is an important class of atmospheric aerosol” seems like an unnecessary statement for the journal’s audience). This adjective is used heavily throughout the introduction (lines 45, 49), and I recommend removing or replacing the adjective to improve readability.

Response: We have accepted the reviewer’s suggestion. In the revised manuscript, we have removed the “as an important class of atmospheric aerosol”, and removed or replaced “important” where it is necessary.

Line 56: hydroperoxides should be plural

Response: Corrected.

Line 57: need comma between ‘NO’ and ‘forming’

Response: Corrected.

Line 87: should read “might have either counteracting or synergistic effects on SOA: : :”

Response: Corrected.

Line 126: remove the with following ‘multiplied by’

Response: Done.

Line 135: should read ‘there was no aqueous..’

Response: Corrected.

Line 221, remove comma between ‘that’ and ‘high’

Response: Done.

Line 360: should read ‘in the ambient atmosphere’

Response: Corrected.

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