Responses to Referee # 2

We thank the reviewer for carefully reviewing our manuscript; the comments and suggestions are very helpful and greatly appreciated. All the comments have been addressed. We believe that revisions based on these comment have substantially improved our manuscript. In the following please find our responses to the comments one by one and the corresponding changes made to the manuscript. The original comments are shown in italics. The revised parts of the manuscript are highlighted.

Before we start our responses to the reviewer, we would like to briefly clarify the motivation of this manuscript. Our primary goal of this study is to investigate how and to what extent small ambient inorganic trace gases, here NO_x and SO_2, affect the SOA formation in the ambient anthropogenic-biogenic interactions. We aimed to study the more complex situation found in the ambient atmosphere instead of pure VOC reaction systems. Our main goal is not so much to provide a single absolute value of SOA yield to modelers nor to simply "improve" values of the SOA yields from previous studies because all chamber studies have operational limitations compared to the atmosphere (e.g. wall effects). Often different studies have distinct physical and chemical regimes due to the different operational limitations and diverse experimental conditions e.g. chamber size, radical generation, and photolysis rates. We as well as the community are in the course of addressing the influence of wall loss of vapors. We believe that, rather denying the findings from previous studies, our study provided additional knowledge and insights to existing understanding of SOA formation in certain conditions of the real atmosphere. This is based on the fact that our experiments were conducted under conditions relatively close to the ambient anthropogenic-biogenic interactions, including ambient RH, concentrations of SO_2, NO_x, and VOC close to ambient levels, natural sunlight, and low surface-to-volume ratio of our large chamber.

Anonymous Referee #2

This chamber study investigated the effects of SO2 and NOx (NO) on SOA formation from photooxidation of a-pinene and limonene. It was found that SO2 enhanced SOA yield while NOx suppressed SOA yield. The suppression effect of NOx was attributed to the suppressed new particle formation and thus a lack of particle surface area for organics to condense on. The authors concluded that SO2 oxidation produced high number of particles and compensated for the suppression of SOA yield by NOx. SOA composition measured by AMS was also presented and discussed.

This is an interesting study. The gas- and particle-phase measurements are comprehensive and include several important species that have not been typically characterized in previous studies (e.g., OH, HO2 and RO2). The experiments appeared to be carefully conducted. However, I have major concerns regarding data interpretation and some conclusions in the manuscript.

One of the central themes of the manuscript is that the suppression effect of NOx on SOA formation

36   *can be compensated by the presence of SO2. This conclusion is not accurate based on all the data*

37   *presented in this manuscript. For a-pinene, it appears that under high SO2 conditions, the SOA yields*

38   *under low vs. high NOx conditions are comparable. However, this is not the case for limonene, where*

39   *there is still a large difference in SOA yields between low vs. high NOx conditions in the presence of*

40   *high SO2. The manuscript needs to be thoroughly revised to accurately reflect what the data are*

41   *showing. If one set of data is showing one thing and another set of data is showing the opposite, the*

42   *authors need to discuss both datasets equally and cannot conclude that SO2 effect can compensate*

43   *NOx effect.*

44   **Response:**

45   A key result in our study is that for both α-pinene and limonene, the difference in SOA yield between

46   high NO_x and low NO_x was much reduced in the presence of SO_2, although for limonene the SOA

47   yield at high NO_x and high SO_2 was still lower than the yield at low NO_x. This result indicates that the

48   suppression of SOA yield by NO_x was compensated to a large extent by SO_2. This conclusion holds

49   regardless the difference in the detailed results between α-pinene and limonene system.

50   In the revised manuscript, we have revised the conclusion to better represent the results from both

51   α-pinene and limonene cases. The following sentence has been revised in the conclusion part:

52   "SO_2 oxidation produced high number concentration of particles and compensated for the suppression

53   of SOA yield by NO_x to a large extent."

54   The abstract has been revised accordingly as follows.

55   "However, in the presence of SO_2 which induced high number concentration of particles after

56   oxidation to H_2SO_4, the suppression of the mass yield of SOA by NO_x was completely or partly

57   compensated."

58   *The authors concluded that the suppression effect of NOx on SOA yields is mainly due to suppression*

59   *of nucleation (absence of particle surface area as condensation sink) rather than decrease of*

60   *condensable materials. If particle surface area plays a role, this will point to the importance of loss*

61   *process of oxidation products via chemical reactions and/or chamber wall loss. However, the effect of*

62   *loss of organic vapors on chamber walls is not considered in this study. Nevertheless, previous studies*

63   *on a-pinene oxidation suggested that SOA yield is independent of particle surface area. In this regard,*

64   *the interpretation that the suppression effect of NOx arises from a lack of particle surface area*

65   *appears to be at odds with previous studies. All in all, it is not clear how the absence of particle*

66   *surface area can explain the suppressed SOA yields under high NOx condition in this study.*

67   **Response:**

68   In general, condensable SOA materials are chemically produced in the gas-phase and nucleation and

69   condensation on particle surface constitute sinks for the SOA materials besides other sinks e.g. wall

70   loss and flush out. In the case of sufficient nucleation or seeded experiments, condensation on

71   particles is the dominant sink. In absence of nucleation and surface, the other sinks such as wall loss

72 dominate the losses of SOA materials. Therefore, no SOA would be formed without nucleation. This

73 *a priori* tells nothing about the importance of wall losses when SOA is formed.

74 The loss of organic vapors to chamber walls can be important for SOA yield, although in our

75 manuscript we had not corrected for it because it is a challenge to quantify it. In the revised

76 manuscript, we have added a section to estimate the influence of vapor wall loss on SOA yield (Sect.

77 "2.3 Wall loss of organic vapors") and provided more discussion on vapor wall loss. We found that

78 the influence of vapor wall loss on SOA yield is likely to be significant when surface area

79 concentrations of SOA formed were low in the high NO_x and low SO_2 conditions. Yet, the influence

80 of vapor wall loss is likely to be not significant at the higher surface area concentrations of SOA

81 formed in the low NO_x conditions or high NO_x and high SO_2 conditions.

82 We respectfully disagree with the reviewer's statement that "previous studies on α-pinene oxidation

83 suggested that SOA yield is independent of particle surface area" without considering each specific

84 study. Although some studies showed that SOA yield from α-pinene oxidation is independent of seed

85 particle surface area (McVay et al., 2016; Nah et al., 2016), a number of studies showed that SOA

86 yield from α-pinene ozonolysis or photooxidation depends on particle surface area. For example, our

87 previous studies clearly showed that SOA yields from α-pinene photooxidation depend on surface

88 area (Sarrafzadeh et al., 2016; Ehn et al., 2014), and Eddingsaas et al. (2012) also showed that in

89 "high NO" conditions, SOA yield from α-pinene photooxidation is much higher with neutral seed

90 than that without seed. The discrepancy in the dependence of SOA yield on particle surface areas in

91 the literature can be attributed to reaction conditions, surface area range and chamber setup. For

92 example, if the reaction produces enough new particles by itself and results in fast particle growth and

93 larger aerosol surface area as the dominant condensational sink for vapors compared to the loss on

94 chamber walls, SOA yield would be less affected by the seed surface area. On the contrary, if VOC

95 oxidation does not induce nucleation by itself, all vapors would be lost onto the chamber walls and

96 SOA yield would be essentially zero.

97 In this study, at high NO_x and low SO_2, the particle number and surface area concentrations were low

98 (peak surface area concentration of $6.8×10^{-7}$ cm^2 cm^{-3} and particle-to-chamber surface ratio of

99 $7.7×10^{-5}$ for α-pinene), much lower than the aerosol surface area range in the studies by Nah et al.

100 (2016) and McVay et al. (2016) (~10^{-5} cm^2 cm^{-3} and particle-to-chamber surface ratio of >$4×10^{-5}$). In

101 addition, not only seed particle surface area but also total particle surface area formed during reaction

102 provide condensational sinks to compete with vapor wall loss. At such low particle surface area

103 concentrations, the condensation of vapors on particles had a much longer time scale than that of the

104 wall loss and a large fraction of vapors condensed on chamber walls. Therefore, SOA yield was

105 significantly suppressed due to lack of particle surface area.

106 In the revised manuscript, we have discussed the findings in the literature.

107 "Artificially added seed aerosol has been shown to enhance SOA formation from α-pinene and

108   β-pinene oxidation (Ehn et al., 2014; Sarrafzadeh et al., 2016; Eddingsaas et al., 2012a). In some other

109   studies, it was found that the SOA yield from α-pinene oxidation is independent of initial seed surface

110   area (McVay et al., 2016; Nah et al., 2016). The difference in the literature may be due to the range of

111   total surface area of particles, reaction conditions and chamber setup. For example, the peak

112   particle-to-chamber surface ratio for α-pinene photooxidation in this study was 7.7×10^{-5} at high NO_x

113   and low SO_2, much lower than the aerosol surface area range in the studies by Nah et al. (2016) and

114   McVay et al. (2016). A lower particle-to-chamber surface ratio can lead to a larger fraction of

115   organics lost on chamber walls. Hence, providing additional particle surface by adding seed particles

116   can increase the condensation of organics on particles and thus increase SOA yield. However, once

117   the surface area is high enough to inhibit condensation of vapors on chamber walls, further

118   enhancement of particle surface will not significantly enhance the yield (Sarrafzadeh et al., 2016)."

119   *The authors explained the effect of SO2 as 1) inducing new particle formation and providing surface*

120   *area for vapor condensation, 2) acid-catalyzed particle-phase reactions. I have the same question*

121   *regarding the first explanation, i.e., what is the role of vapor wall loss (if any), and how does one*

122   *reconcile this explanation with findings from previous studies? Also, what is the effect of SO2 on*

123   *gas-phase chemistry and SOA yield? This is not considered.*

124   **Response:**

125   The role of vapor wall loss can be referred to our response to the comment above. Wall loss of vapors

126   leads to an underestimate of SOA yield. Condensation of vapors onto aerosol particle surface

127   competes with the loss of vapors on chamber walls. Therefore the surface area provided by nucleation

128   and growth of particles in the presence of SO_2 enhanced the SOA yield in this study.

129   Only few studies have investigated the effect of SO_2 on the SOA yield from α-pinene oxidation. More

130   studies investigated the effect of seed aerosol and particle acidity. Kleindienst et al. (2006) attributed

131   the increase of SOA yield in the presence of SO_2 to the formation of H_2SO_4 acidic aerosol. While

132   particle acidity may contribute to the increased SOA yield, especially at high NO_x, the effect of

133   facilitating nucleation and further providing surface area seems to be more important in our study.

134   The importance of the SO_2 via nucleating and providing particle surface depends on the particle

135   surface area in the absence of SO_2 because the competition for the condensation of vapors between

136   particles and wall depends on particle surface area. When VOC oxidation does not form enough new

137   particles and particle surface by itself, the role of SO_2 via nucleating and providing particle surface in

138   enhancing SOA yield is more important. In the revised manuscript, we have added more discussion on

139   this aspect.

140   "Particle acidity may also play a role in affecting the SOA yield in the experiments with high SO_2.

141   Particle acidity was found to enhance the SOA yield from α-pinene photooxidation at high NO_x

142   (Offenberg et al., 2009) and "high NO" conditions (Eddingsaas et al., 2012a). Yet, in low NO_x

143   condition, particle acidity was reported to have no significant effect on the SOA yield from α-pinene

144 photooxidation (Eddingsaas et al., 2012a; Han et al., 2016). According to these findings, at low NO_x
145 the enhancement of SOA yield in this study is attributed to the effect of facilitating nucleation and
146 providing more particle surface by SO_2 photooxidation. At high NO_x, the effect in enhancing new
147 particle formation by SO_2 photooxidation seems to be more important, although the effect of particle
148 acidity resulted from SO_2 photooxidation may also play a role."
149 The effect of SO_2 on gas phase chemistry is not significant in this study because the reaction rate of
150 SO_2 with OH (~2×10^{-12} molecules^{-1} cm^3 s^{-1}) and with RO_2 (<10^{-14} molecule^{-1} cm^3 s^{-1}) are very low
151 (Lightfoot et al., 1992; Berndt et al., 2015). From the AMS data of SOA formed at high SO_2, we
152 found no significant organic fragments containing sulfur. Also the fragment $CH_3SO_2^+$ suggested by
153 Farmer et al. (2010) was not detected in our data. Therefore, we conclude that in our study, the effect
154 of SO_2 on gas phase chemistry of organics and thus further on SOA yield via affecting gas phase
155 chemistry is not important.
156 In the revised manuscript, we have added the following discussion about this point.
157 "SO_2 has been proposed to also affect gas phase chemistry of organics by changing the HO_2/OH or
158 forming SO_3 (Friedman et al., 2016). In this study, the effect of SO_2 on gas phase chemistry of
159 organics was not significant because of the much lower reactivity of SO_2 with OH compared with
160 α-pinene and limonene (Atkinson et al., 2004, 2006; Atkinson and Arey, 2003) and the low OH
161 concentrations (2-3 orders of magnitude lower than those in the study by Friedman et al. (2016)).
162 Moreover, reactions of RO_2 with SO_2 was also not important because the reaction rate constant is very
163 low (<10^{-14} molecule^{-1} cm^3 s^{-1}) (Lightfoot et al., 1992; Berndt et al., 2015). In addition, from the AMS
164 data of SOA formed at high SO_2 no significant organic fragments containing sulfur were found. Also
165 the fragment $CH_3SO_2^+$ from organic sulfate suggested by Farmer et al. (2010) was not detected in our
166 data. The absence of organic sulfate tracers is likely due to the lack of aqueous phase in aerosol
167 particles in this study. Therefore, the influence of SO_2 on gas phase chemistry of organics and further
168 on SOA yield via affecting gas phase chemistry is not important in this study."
169 *It appears that the SOA yields in high SO2 experiments might be overestimated by double counting*
170 *the density of ammonium sulfate/ammonium bisulfate in the SOA mass calculation. This is not entirely*
171 *clear.*
172 **Response:**
173 The density of SOA in high SO_2 experiments was not double counted. In the revised manuscript, we
174 have clarified this point. Please also refer to our response to the similar comment below ("detailed
175 comment" 3).
176 "…and their respective density (1.32 g cm^{-3} for organic aerosol from one of our previous studies
177 (Flores et al., 2014) and the literature (Ng et al., 2007) and ~1.77 g cm^{-3} for ammonium
178 sulfate/ammonium bisulfate)…"
179 We would like to note that the SOA yield in this study was derived by adjusting the density of SOA to

180 1 g cm^{-3}.

181 *Finally, the authors need to conduct a more careful and accurate comparison with previous studies. It*

182 *was noted that in high SO2 conditions, their findings that SOA yields are comparable under high NOx*

183 *and low NOx conditions are in line with Sarrafzadeh et al. and Eddingsaas et al. I do not think that*

184 *the data in Eddingsaas et al. showed this. SOA yields are also a function of deltaMo (as well as*

185 *various experimental conditions and parameters) and this could play a role, see detailed comment*

186 *below. Also, the a-pinene yields in this study under comparable NOx/SO2/OH exposure are much*

187 *lower than Eddingsaas et al.. This is not mentioned and discussed in the manuscript.*

188 **Response:**

189 In the revised manuscript, we have added more discussion to compare this study with previous studies

190 and added a table summarizing previous studies.

191 Please refer to our detailed responses to the specific comments below ("detailed comments" 11d, e).

192 *Major revisions are needed before the manuscript can be published. Specific comments are listed*

193 *below.*

194 *Detailed comments*

195 *1. Line 18-20. This statement is not true for limonene data presented in this study.*

196 **Response:**

197 In the revised manuscript, we have modified this statement. Please refer to our response to a similar

198 comment above (Pg. 1, lines 35-43).

199 *2. Line 79 -81. This sentence seems to imply that previous studies that used higher NOx and SO2*

200 *concentrations are not atmospherically relevant. I think these sentences should be revised and*

201 *clarified to more accurately reflect the experimental design and results from previous studies. For*

202 *instance, the use of high levels of NOx (e.g., from HONO or CH3ONO) in some studies is to push the*

203 *RO2 radical fate to the extreme (i.e., RO2+NO or RO2+NO2) to investigate SOA yields and*

204 *composition under such conditions. Thus, the use of high levels of NOx do not necessarily mean that*

205 *the results are not applicable to ambient conditions.*

206 **Response:**

207 We understand that in some studies high concentration of NO_x is intentionally used to push the RO_2

208 radical fate to an extreme in order to study the SOA yields and composition. Nevertheless,

209 experiments under ambient levels of high NO_x concentrations are more transferable to the ambient

210 anthropogenic-biogenic interaction than the experiments conducted at extremely high NO_x

211 concentrations considering the shortened lifetime of RO_2 and the potential secondary processes as

212 well as the effect of NO_x on OH concentration at extremely high NO_x. In the revised manuscript, we

213 have modified this sentence.

214 "For example, many studies used very high NO_x and SO_2 concentrations (up to several hundreds of

215 ppb). High NO_x can make the RO_2 radical fate dominated by one single pathway (i.e., RO_2+NO or

216 RO$_2$+NO$_2$) to investigate SOA yields and composition under such conditions. Yet, the effects of NO$_x$

217 and SO$_2$ at concentration ranges for ambient anthropogenic-biogenic interactions (sub ppb to several

218 tens of ppb for NO$_2$ and SO$_2$) have seldom been directly addressed."

219 *3. Line 132. Is the organic aerosol density 1.32 g/cm3 from Eddingsaas et al. (2012a)? If this is the*

220 *case, note that this density used in Eddingsaas et al. is directly taken from the results in Ng et al.*

221 *(2007), and that this density was obtained in the presence of seeds already. Therefore, it appears that*

222 *there might be a double counting of the density of ammonium sulfate/ammonium bisulfate in the data*

223 *presented here?*

224 **Response:**

225 Organic aerosol density was based on our previous study (Flores et al., 2014) as well as the study of

226 Eddingsaas et al. (2012) and thus Ng et al. (2007).

227 Although the density of organic aerosol in the study of Ng et al. (2007) was obtained in presence of

228 seed, the contribution of seed aerosol (ammonium sulfate) to particle volume and thus density has

229 been taken into account (Bahreini et al., 2005) . The value reported by Ng et al. (2007) is the density

230 of organic aerosol instead of the density of mixed aerosol. There is no double counting of the density

231 of ammonium sulfate/ammonium bisulfate in our study.

232 4. *Line 162. How were OH and O3 formed in the experiments (under each combination of NOx/SO2*

233 *condition). Please provide more info. Also, please provide typical time profiles of VOC (either*

234 *a-pinene of limonene), O3, OH, NO, NO2, SO2 for each combination of NOx/SO2 condition. These*

235 *are important to help the readers obtain a better idea of the reaction pathways/regimes under each*

236 *condition.*

237 **Response:**

238 OH was formed via HONO photolysis, which was produced from a photolytic process on the Teflon

239 chamber wall as we described in the manuscript. The details can be found in a previous study on our

240 chamber (Rohrer et al., 2005). In the revised manuscript, we have added one more sentence to better

241 clarify this point.

242 "OH was formed via HONO photolysis, which was produced from a photolytic process on the Teflon

243 chamber wall (Rohrer et al., 2005)."

244 In addition, in all VOC photooxidation of our study, OH was partly contributed by the recycling

245 reaction of HO$_2$ with NO. The reaction of HO$_2$ and RO$_2$ with NO also produces NO$_2$, whose

246 photolysis further forms O$_3$. While the detailed mechanism of O$_3$ formation is beyond the scope of

247 this study, we have provided a brief description in the revised manuscript.

248 "O$_3$ was formed in photochemical reactions since NO$_x$, even in trace amount (<~1 ppbV), was present

249 in this study."

250 "In the photooxidation of VOC, OH and O$_3$ often co-exist and both contribute to VOC oxidation

251 because O$_3$ formation in chamber studies is often unavoidable during photochemical reactions of

252 <mark>VOC even in the presence of trace amount of NO_x."</mark>

253 In the revised manuscript, we have provided time profiles of VOC, O_3, OH, NO, and NO_2 in low NO_x

254 and high NO_x conditions for α-pinene and limonene (Fig. S5). Time profiles of these species at high

255 SO_2 were similar to those at low SO_2 because SO_2 had little effect on gas phase chemistry due to its

256 low reactivity of SO_2 with OH and RO_2, and thus are not further shown. SO_2 time series are shown

257 separately (Fig. S2).

258 *5. Line 163. Was all the VOC reacted in each experiment?*

259 **Response:**

260 All the VOC precursor was consumed in the experiments of this study except for one experiment

261 where small amount of VOC (~10%) was still left at the end of the reaction due to the cloudy

262 condition and thus lower photolysis rates.

263 *6. Lin 163. There is no "typical" experiment in this study, as each experiment was conducted under a*

264 *different NOx/SO2 condition. Please clearly state that this is only for low NOx condition. Also, what*

265 *about high NOx condition? Was it exclusively OH reaction? Please also specify clearly.*

266 **Response:**

267 The relative dominance of OH oxidation over ozonolysis (as shown in Fig. 6) is similar in both the

268 low NO_x and high NO_x conditions. At high NO_x, OH was often higher and meanwhile more O_3 was

269 also produced.

270 In the revised manuscript, we have modified this sentence as follows.

271 "<mark>For all the experiment in this study, the VOC loss was dominated by OH oxidation over ozonolysis</mark>

272 <mark>(see Fig. S6 as an example). The relative importance of the reaction of OH and O_3 with monoterpenes</mark>

273 <mark>was similar in the low NO_x and high NO_x experiments. At high NO_x, OH was often higher while more</mark>

274 <mark>O_3 was also produced</mark>."

275 *7. Line 173 – 177. Here, under low NOx condition, RO2+NO dominates throughout the entire*

276 *experiment (RO2+HO2 only contributes to 40% at most).*

277 *a. These sentences clearly demonstrate the shortcomings of classifying the experiments as low NOx vs.*

278 *high NOx as discussed in Wennberg et al. (IGAC news, 2103). I suggest the authors to characterize*

279 *reactions conditions by explicitly stating the RO2 fates, rather than as low vs. high NOx.*

280 **Response:**

281 We had clearly defined our low NO_x and high NO_x conditions using the RO_2 fate in our study.

282 Therefore, we respectfully do not think that using the terms "low NO_x" and "high NO_x" caused

283 ambiguity as long as we define them clearly.

284 In the revised manuscript, we have added the following sentence to emphasize the RO_2 fates under

285 different NO_x conditions.

286 "<mark>Note that the RO_2 fate in the low and high NO_x conditions quantified here are further used in the</mark>

287 <mark>discussion below since the information of RO_2 fate is important for data interpretation of experiments</mark>

288 conducted at different NO$_x$ levels (Wennberg, 2013)."

289 *b. It is stated that under low NOx conditions, in the beginning of the experiment, a trace amount of*

290 *NO is formed from photolysis of HONO from the chamber wall. Is this just in the beginning of the*

291 *experiment, or there is a continuous NO source from HONO photolysis throughout the entire*

292 *experiment? Please specify.*

293 **Response:**

294 There is a continuous NO source from HONO photolysis. In the revised manuscript, we have clarified

295 this as follows.

296 "The trace amount of NO (up to ~0.2 ppbV) was from the photolysis of HONO, which was

297 continuously produced from a photolytic process on chamber walls throughout an experiment (Rohrer

298 et al., 2005)."

299 *8. Line 189. The authors attributed the lower particle number concentration and nucleation rate at*

300 *high NOx to the decreasing RO2+RO2 reaction in the presence of NOx. However, in line 182, the*

301 *authors noted that RO2+RO2 reaction is negligible in this study to start with. Please reconcile these*

302 *seemingly contradictory statements. Also, can be suppressed nucleation under high NOx due to the*

303 *higher volatility of organic nitrates as compared to peroxides (from RO2+HO2)?*

304 **Response:**

305 The compounds responsible for nucleation only account for a very small fraction of RO$_2$ reaction

306 products. Although the contribution of RO$_2$+RO$_2$ reaction to the total RO$_2$ loss is negligible, it can

307 contribute a lot to the compounds responsible for nucleation because RO$_2$+RO$_2$ reactions form dimers,

308 which have high molecular weight and extremely low volatility (Ehn et al., 2014; Kirkby et al., 2016).

309 Generally, organic nitrates are not expected to be the main compounds responsible for nucleation

310 since their volatility is not low enough to nucleate, nor primary organic peroxides (from

311 RO$_2$(C$_{10}$)+HO$_2$). Therefore, although under high NO$_x$ more organic nitrates were found, organic

312 nitrates are unlikely to be the reason for the suppressed nucleation.

313 *9. Line 205-206. There is nucleation (from organics) in the presence of NOx as shown in Fig. 4. In*

314 *this sense, "absence of nucleation" here is a bit confusing. Perhaps would be clearer to say "absence*

315 *of seed particles".*

316 **Response:**

317 In the revised manuscript, we have modified "absence of nucleation" as follows.

318 "Because NO$_x$ suppressed new particle formation, the suppression of the SOA yield could be

319 attributed to the lack of new particles as seed and thus the lack of condensational sink, or to the

320 decrease of condensable organic materials."

321 *10. Line 211 and Figure 3. The author concluded that the suppression effect of NOx on SOA yields*

322 *was mainly due to suppression of nucleation, i.e., to the absence of particle surface as condensation*

323 *sink. Many critical aspects are not discussed, making this conclusion not well-justified and*

324   *well-supported.*

325   *a. If the absence of seed particle surface area is the reason for the low yield under high NOx*

326   *condition (at low SO2), this will point to the importance of loss of semivolatile species via chemical*

327   *reactions or chamber wall loss (Kroll et al., ES&T, 2007). However, the effect of vapor wall loss in*

328   *not considered in this study. Zhang et al. (PNAS, 2014) first systematically investigated the effects of*

329   *particle surface area and vapor wall loss on SOA yields. For a-pinene photooxidation and ozonolysis*

330   *specifically, it was found that SOA yields are largely independent of seed surface area (McVay et al.,*

331   *2016, Nah et al., ACP, 2016; Nah et al., ACP, 2017). Therefore, taken all these together, it is not*

332   *clear how the absence of particle surface area can explain the suppressed SOA yields under high NOx*

333   *condition in this study.*

334   **Response:**

335   Please refer to our response to a similar comment above (Pg. 2, lines 58-66).

336   *b. The authors dismissed the "decrease of condensable organic materials" in high NOx conditions as*

337   *an explanation for the observed decrease in yield. Why? If more volatile organic nitrates are formed*

338   *in high NOx conditions, why can't this be an explanation for the suppressed SOA yield? For limonene*

339   *data (Line 213-218), the authors appeared to embrace the role of volatility of oxidation products.*

340   **Response:**

341   In our study, when new particle formation was already enhanced by added SO_2, the SOA yield at high

342   NO_x was comparable to that at low NO_x for α-pinene and the difference in SOA yield between high

343   NO_x and low NO_x was much smaller (Fig. 3a). If the organic materials such as organic nitrate formed

344   in high NO_x conditions were more volatile, the SOA yield in high NO_x should be low regardless of

345   SO_2 concentration unless in addition to that, SO_2 enhanced the SOA yield at high NO_x via the

346   influence other than surface area effect, e.g., acidity effect. Organic nitrates formed at high NO_x was

347   proposed to be more volatile (Presto et al., 2005; Kroll et al., 2006). However, many organic nitrate

348   formed in our study is highly oxidized organic molecule (HOMs) containing multi-functional groups

349   besides nitrate ($C_{7-10}H_{9-15}NO_{8-15}$). The compounds are expected to have low volatility and they were

350   found to have an uptake coefficient on particles of ~1 (Pullinen et al. in preparation). A recent study

351   also implied that organic nitrate may have low volatility (Hakkinen et al., 2012). Therefore, the

352   suppressing effect of NO_x on SOA yield was more likely due to suppressed nucleation, i.e., to lack of

353   particle surface as condensational sinks.

354   In the revised manuscript, we have elaborated our discussion.

355   "This finding ==can be attributed to two possible explanations. Firstly==, NO_x did not significantly

356   suppress the formation of ==low volatile== condensable organic materials, although NO_x obviously

357   suppressed the formation of products for nucleation. ==Secondly, NO_x did suppress the formation of==

358   ==low-volatility condensable organic materials via forming potentially more volatile compounds and in==

359   ==addition to that, the suppressed formation of condensable organic materials was compensated by the==

10

360 presence of SO_2, resulting in comparable SOA yield. Organic nitrates are a group of compounds
361 formed at high NO_x, which have been proposed to be more volatile (Presto et al., 2005; Kroll et al.,
362 2006). However, many organic nitrates formed by photooxidation in this study were highly oxidized
363 organic molecules (HOMs) containing multi-functional groups besides nitrate group
364 ($C_{7-10}H_{9-15}NO_{8-15}$). These compounds are expected to have low volatility and they are found to have an
365 uptake coefficient on particles of ~1 (Pullinen et al., in preparation). Therefore, the suppressing
366 effect of NO_x on SOA yield was mostly likely due to suppressed nucleation, i.e., the lack of particle
367 surface as condensational sink."

368 For limonene data, please refer to our response to the comment below ("detailed comments" 10c).

369 *c. Line 217. How does the different range in VOC/NOx for a-pinene and limonene experiments*
370 *explain the differences in yields in high SO2 conditions? Please elaborate and explain clearly.*

371 **Response:**

372 The cause of the difference between the α-pinene and limonene cases is unknown for the moment and
373 it would be pure speculation when discussing reasons for this difference. Therefore, in the revised
374 manuscript, we only state that the reason is unknown so far and as possible explanation, we note that
375 the average volatility of limonene oxidation products may be higher at higher NO_x.

376 In the revised manuscript, we have revised this sentence as follows.

377 "The cause of this difference is currently unknown. Our data of SOA yield suggest that the products
378 formed from limonene oxidation at high NO_x seemed to have higher average volatility than that at low
379 NO_x."

380 *11. Line 225-237. Comparisons with previous studies. Many critical details are not considered and*
381 *discussed. I think the authors jumped to the conclusion on whether their study agree/disagree with*
382 *previous studies too quickly.*

383 *a. Line 225. This sentence is only true for a-pinene data in this study, but not for limonene. Please*
384 *state clearly.*

385 **Response:**

386 In the revised manuscript, we have revised this sentence as follows.

387 "Our finding that the difference in SOA yield between high NO_x and low NO_x conditions was highly
388 reduced at high SO_2 is also in line with the findings of some previous studies using seed aerosols
389 (Sarrafzadeh et al., 2016; Eddingsaas et al., 2012a)."

390 b. *Seed particles were generated via SO2 oxidation in this study (for high SO2 experiments).*
391 *Previous studies directly injected seeds into the chamber. In comparing SOA yields, the author should*
392 *also consider the role of gas-phase chemistry and particle phase chemistry. For instance, what about*
393 *the reaction of SO2 and criegee intermediates? What about the effect of particle acidity on*
394 *particle-phase reactions (in this study vs. previous studies)? Please discuss.*

395 **Response:**

396 In our study, the reaction of SO_2 with Criegee intermediates was not important to the formation of

397 oxidized organics and SOA formation for the following reasons. 1) The reaction of VOC with O_3 only

398 contributed to a small fraction of VOC loss in this study and thus formation of Criegee intermediates

399 was not significant. 2) At the water vapor concentration of this study, water may compete for Criegee

400 intermediates with SO_2 to a large extent.

401 Particle acidity may affect the SOA yield via acid-catalyzed reactions, as we had discussed in our

402 manuscript. In the revised manuscript, we have elaborated this discussion by comparing with previous

403 studies as follows.

404 "Particle acidity may also play a role in affecting the SOA yield in the experiments with high SO_2.

405 Particle acidity was found to enhance the SOA yield from α-pinene photooxidation at high NO_x

406 (Offenberg et al., 2009) and "high NO" conditions (Eddingsaas et al., 2012a). Yet, in low NO_x

407 condition, particle acidity was reported to have no significant effect on the SOA yield from α-pinene

408 photooxidation (Eddingsaas et al., 2012a; Han et al., 2016). According to these findings, at low NO_x

409 the enhancement of SOA yield in this study is attributed to the effect of facilitating nucleation and

410 providing more particle surface by SO_2 photooxidation. At high NO_x, the effect in enhancing new

411 particle formation by SO_2 photooxidation seems to be more important, although the effect of particle

412 acidity resulted from SO_2 photooxidation may also play a role."

413 *c. The experiments in this study were conducted in the presence of humidity but previous studies were*

414 *mostly conducted under dry conditions. RH can affect gas-phase and particle-phase chemistry, and*

415 *subsequently SOA yields.*

416 **Response:**

417 We agree with the reviewer that RH can affect gas-phase and particle-phase chemistry and thus may

418 also affect SOA yield. Because humidity is ubiquitous in the real atmosphere, we conducted our

419 experiments in the presence of humidity in order to better represent ambient conditions. In the revised

420 manuscript, we have emphasized this point.

421 "RH of this study is different from many previous studies, which often used very low RH (<10%)."

422 *d. The authors noted that the finding that SOA yields at high NOx is comparable to that at low NOx in*

423 *high SO2 conditions is in line with findings in Sarrafzadeh et al. (2016) and Eddingsaas et al. (2012a).*

424 *I do not think that the data in Eddingsaas et al. showed that "in presence of seed aerosol, the*

425 *difference in the SOA yield between low and high NOx is much reduced". SOA yield is also a function*

426 *of deltaMo. Considering the data in Table 1 of Eddingsaas et al., the difference in yields between low*

427 *and high NO experiments for nucleation is 19%, and for seeded experiments are 15% and 10%.*

428 *However, the difference in deltaMo for the nucleation experiments is also the largest and this will*

429 *play a role in the yield difference.*

430 **Response:**

431 As the reviewer noticed, the data in Eddingsaas et al. (2012) showed that in the absence of seed

432   aerosol SOA yield at low NO_x is 2.5 times higher than that at high NO_x, while in the presence of seed

433   aerosol SOA yield at low NO_x is only 1 and 0.6 times higher than that at high NO_x for neutral seed

434   and acid seed, respectively. Therefore, the data in Eddingsaas et al. (2012) did show that "in presence

435   of seed aerosol, the difference in the SOA yield between low and high NO_x is much reduced".

436   We noticed that in the data given by Eddingsaas et al. (2012) the difference in $deltaM_o$ for nucleation

437   experiments is the largest, which plays a role in SOA yield. However, the large difference in $deltaM_o$

438   between high NO_x and low NO_x cases is because the $deltaM_o$ at high NO_x is the lowest in absence of

439   seed, much lower than $deltaM_o$ in presence of seed when other conditions are largely the same. The

440   higher $deltaM_o$ and the smaller difference in $deltaM_o$ between low NO_x and high NO_x in presence of

441   seed also originated from the seed aerosol since other conditions were kept constant. This result

442   agrees with our finding that "in presence of seed aerosol, the difference in the SOA yield between low

443   and high NO_x is much reduced".

444   *e. The SOA yields in this study are much lower than previous studies, why? Consid-ering the low NOx*

445   *low SO2 experiment, with OH dose of 1e11 molecules cm-3 s, the yield in this study is 7%. However,*

446   *the corresponding yield in Eddingsaas et al. is > 30% (Figure 3 of Eddingsaas et al.).*

447   **Response:**

448   The difference in SOA yield between this study and the study by Eddingsaas et al. (2012) can be

449   explained by several reasons. Firstly, SOA yield in this study was calculated using a density of 1 g

450   cm^{-3} while in Eddingsaas et al. (2012) SOA yield was calculated using a density of 1.32 g cm^{-3}.

451   Secondly, reaction conditions such as VOC concentrations, NO_x concentrations, and OH source and

452   concentrations of our study at low NO_x are different from those in   Eddingsaas et al. (2012). For

453   example, in our study at low NO_x, RO_2+NO account for a large fraction of RO_2 loss while in

454   Eddingsaas et al. (2012) RO_2+HO_2 is the dominant pathway of RO_2 loss. These differences in reaction

455   conditions may affect SOA yield. Thirdly, the organic aerosol concentration of this study is much

456   lower than that in Eddingsaas et al. (2012). Fourthly, the total particle surface area may be different

457   from Eddingsaas et al. (2012) (the data are not available to compare with), which can also affect the

458   measured SOA yield. Also note that the exceptionally high SOA yield at lower α-pinene

459   concentration is an "outlier" to other data in Eddingsaas et al. (2012) and could not be explained by

460   the authors.

461   In the revised manuscript, we have added the following discussion regarding the comparison of SOA

462   yield from α-pinene photooxidation with the literature.

463   "The reaction conditions of this study often differ from those described in the literature (see Table

464   S2).

465   The difference in these conditions can result in both different apparent dependence on specific

466   parameters and the varied SOA yield. For example, SOA yield from α-pinene photooxidation at low

467   NO_x in this study appeared to be much lower than that in Eddingsaas et al. (2012a). The difference

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

485 In the revised manuscript, we have also added a figure plotting SOA yield as a function of organic

486 aerosol mass loading (Fig. S8).

487 *12. Line 238 onwards, effect of SO2.*

488 *a. One of the proposed reasons to explain the effect of SO2 is that it induces nucleation and provides*

489 *more particle surface area for condensation. Again, if this is the case, it will point to the importance*

490 *of loss of organic vapors to chamber walls, though previous studies suggested that this process does*

491 *not effect SOA yields from a-pinene oxidations to a large extent. With this, it is not clear if this is*

492 *indeed a reason for the observed SO2 effect. Please explain.*

493 **Response:**

494 Please refer to our response to the similar comments above (Pg. 2, lines 58-66; Pg. 4, lines 119-123)

495 and corresponding revisions to the manuscript.

496 *b. Line 258. Is "counterbalance" the appropriate word? If the suppression effect of NOx is*

497 *counterbalanced by the enhancement effect SO2, in going from "low NOx low SO2" to "high NOx*

498 *high SO2" one shall not observe change in SOA yields? Also, note that the limonene data showed*

499 *very different trends comparing to the a-pinene data. This needs to be accurately and clearly stated.*

500 **Response:**

501 We have changed "counterbalance" to "compensated". The revised sentence is as follows.

502 "<mark>The presence of high SO_2 enhanced the SOA mass yield at high NO_x conditions, which was even</mark>

503 <mark>comparable with the SOA yield at low NO_x for α-pinene oxidation</mark>. This finding indicates that the

504   suppressing effect of NO_x on SOA mass formation was ==compensated to large extent== by the presence

505   of SO_2."

506   As for the difference in the limonene and α-pinene data, please refer to our response to the similar

507   comments above (Pg. 1, lines 35-43) and our corresponding revisions to the manuscript.

508   *13. Line 315-318. This explanation is a stretch and not well-justified. There is extensive fragmentation*

509   *in the AMS and so the H/C ratios of oxidation product molecules do not necessarily translate to the*

510   *H/C ratios measured. As shown in Chhabra et al., not all experiments conducted under low NOx*

511   *condition have higher H/C ratios.*

512   **Response:**

513   In our opinion, the reaction pathway of RO_2 at least provides a likely explanation for the difference in

514   H/C at different NO_x. Although there is extensive fragmentation in the AMS, H/C measured generally

515   reflects the H/C ratios of the overall compounds. In addition, as we discussed, Chhabra et al. (2011)'s

516   data also show that for α-pinene photooxidation, SOA formed at high NO_x generally has lower H/C,

517   consistent with our study. Admittedly, the RO_2 reaction pathway is not the only factor affecting H/C

518   and O/C. Other factors such as VOC identity, oxidants, and reaction mechanisms, including various

519   functionalization, fragmentation, and oligomerization in both the gas phase and particle phase also

520   play important roles in the chemical composition and thus H/C and O/C. We did not intend to apply

521   our explanation here to all other reaction systems.

522   *14. Line 262 onwards. Did the ratio of nitrate mass concentration to organics mass change over*

523   *time?*

524   **Response:**

525   The ratio of nitrate mass concentration to organics gradually decreased in the beginning of the

526   reactions (2-3 h) and then leveled off. Also, note that in the very beginning of a reaction, the data have

527   large uncertainties due to the low concentration of nitrate and organics. In our manuscript, the average

528   ratios were used to compare different experiments. In the revised manuscript, we have clearly

529   described this in the captions of Fig. 4.

530   "==The average ratios of nitrate to organics during the reaction are shown and error bars indicate the==

531   ==standard deviations.=="

532   Minor comments

533   *1. Line 72. Why "in contrast"?*

534   **Response:**

535   The finding here is different from those in the studies discussed before. In the revised manuscript, we

536   have modified this sentence as follows.

537   "In constrast, Eddingsaas et al. (2012a) found that particle yield increases with aerosol acidity ==only== in

538   "high NO" condition (NO_x 800 ppb, α-pinene: 20-52 ppb), but is independent of the presence of seed

539   aerosol or aerosol acidity in both ==high NO_2" condition (NO_x 800 ppb)"== and low NO_x (NO_x lower than

540   the detection limit of the NO_x analyzer).”

541   *2. Line 84 citation. There are more studies on OH oxidations of a-pinene and they should also be*

542   *cited here (for example, some of the studies cited in page 2).*

543   **Response:**

544   In the revised manuscript, we have added more studies on OH oxidation of α-pinene. However, we

545   would like to note here that many studies on α-pinene photooxidation did not quantitatively

546   distinguish the contributions of oxidation by OH and by O_3.

547   *3. Line 125. “mass” should be “volume”? SMPS measures volume concentration.*

548   **Response:**

549   We have changed the “mass” to “volume” in the revised manuscript.

550   *4. Line 126. Delete “with”.*

551   **Response:**

552   Done.

553   *5. Line 256. Sentence not clear.*

554   **Response:**

555   This sentence has been revised as follows.

556   “The presence of high SO_2 enhanced the SOA mass yield at high NO_x conditions, which was even

557   comparable with the SOA yield at low NO_x for α-pinene oxidation. This finding indicates that the

558   suppressing effect of NO_x on SOA mass formation was compensated to large extent by the presence

559   of SO_2.”

560   *6.nFigure 1 caption should specify the SO2 condition.*

561   **Response:**

562   Done.

563   **References**

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