We thank the reviewer for their valuable comments. We have addressed each comment below in bold italic text, and indicate where necessary the corresponding changes we have made to the manuscript.

Anonymous Referee #2

General Comments

In this manuscript the authors report results of a study of the formation of aerosol from the OH oxidation of isoprene. Experiments were conducted in an environmental chamber under high and low NOx conditions, and gas and aerosol composition and particle volatility were analyzed using a FIGAERO-CIMS instrument with iodide ionization. A major focus was an evaluation of the effects of NOx on organic nitrate and SOA formation, gas-particle partitioning, and particle volatility. Information was also obtained on the possible role of oligomer formation on SOA composition and yield. The lab results were used to help interpret measurements made with the same instrument at the 2013 SOAS campaign, providing additional insight into the role of isoprene in aerosol C1 chemistry. The study was well done, including measurements, data analysis, and interpretation. The manuscript is also well written. Overall the manuscript is of high quality and represents an important contribution to atmospheric chemistry. I recommend it be published after the following comments are addressed.

Specific Comments

1. Line 160–162: Were the seed particles dried before entering the chamber. If not, might they not stay as deliquesced particles since the 50% RH is above the efflorescence point?

Yes, they passed through a diffusion drier before entering the chamber. The relative humidity of the drier output is routinely verified to be <30%, below the efflorescence point of ammonium sulfate particles [Cziczo and Abbatt, 2000]. We also verified that the seed particles were effloresced by comparing the size distribution of the particles exiting the 50% RH chamber.

2. Lines 212–215: If wall loss of NO is significant would you expect wall loss of organic vapors to also be significant?

This sentence was moved earlier in the paper into the "experimental overview" section from the "Gas-particle Partitioning: Saturation Vapor Concentrations and Oligomeric Content" section into the paragraph where particle wall loss is discussed:

Operating the chamber in continuous flow mode possibly reduces the net flux of organic compounds to the walls, at least for low volatility to semi volatile compounds, as some degree of equilibration can occur [Liu et al., 2016; Shilling et al., 2008].

Nonetheless, our vapor concentration data may be biased low due to some amount of loss to the walls.

Furthermore, we do not have evidence that a significant fraction of NO is lost to the walls, though it was not measured and Teflon is known to be somewhat permeable. We removed the last part of the sentence in question to prevent confusion that wall deposition is a main fate. NO is likely reacting fast enough with peroxy radicals that wall loss is not a significant fate.

3. Lines 226–227: It would be useful to explain how you get the factor of 1.5 used to correct AMS data for particle wall loss.

See our response to reviewer #1's comment above.

4. I am aware of the mechanism for forming IEPOX under low NOx conditions, in which OH is lost from an –OOH group as the epoxide is formed. Is there an analogous mechanism for forming IEPOX under high NOx conditions in which NO2 is lost from a Ǎ˘n–ONO2 group? If so it might be worth mentioning.

Good point, IEPOX can be formed from the reaction of isoprene hydroxy nitrate with OH [Jacobs et al., 2014; Lee et al., 2014; St Clair et al., 2016], albeit at fairly low overall yields. See response to comment #6, we added a sentence mentioning this possibility.

5. Lines 336–338: This sentence is confusing to me. Are you saying that HO2 reacts with an alkene C=C double bond? I don't think this happens.

Agreed. This sentence was changed to this:

Two possible sources of this compound are the reaction of the ISOPOOH derived peroxy radical ($C_5H_{11}O_6$) with RO_2, or a dihydroxy alkene undergoing reaction with OH and O_2 to form a dihydroxy peroxy radical, followed by reaction with HO_2.

6. Lines 344–348 and elsewhere: What about the possibility that HNO3 serves as an acid catalyst under high NOx conditions?

This is an interesting suggestion. Our two major tracers of IEPOX multiphase chemistry, $C_5H_{12}O_4$ and $C_5H_{10}O_3$, both decrease in concentration in the particle phase with increasing NO_x, arguing against HNO_3 acidifying the aerosol sufficiently to enhance "traditional" IEPOX SOA formation, but it may be driving some formation of oligomers from IEPOX. This idea would require more detailed experiments to assess. We have added the following sentences:

These two tracers are also observed in the NO_x addition experiments performed in continuous-flow mode, though at lower concentrations. In these experiments there was not enough NO to completely suppress ISOPOOH formation and therefore IEPOX

formation from ISOPOOH + OH. Moreover, IEPOX can also form, albeit at lower yields, from OH reactions with the first-generation isoprene hydroxy nitrate [Jacobs et al., 2014; Lee et al., 2014; St Clair et al., 2016]. However, the formation of IEPOX SOA tracers is more puzzling given the very low reactive uptake of IEPOX expected on solid inorganic seed coated with isoprene SOA [Gaston et al., 2014; Riedel et al., 2015]. Perhaps nitric acid catalyzed IEPOX multiphase chemistry is contributing to the formation of these tracers at high NO_x. That said, observation of these tracers, also in the absence of NO_x addition, indicates that this explanation not sufficient.

7. Lines 394–395: I'm not sure what "included" means here. Do you mean the estimates have been corrected for these effects? Could make this clearer.

See 8. below. This paragraph was slightly rearranged and sentences reworded to make the error corrections more clear.

8. Lines 393–394. What are the estimated uncertainties in the measured values of Fp?

The measured F_p values contain errors primarily from losses of gaseous vapors to the chamber walls as well as losses in the sampling inlet. Because the sampling inlet was short (1 m) and the flow volume was high (12 lpm) for the data presented in this plot, we expect inlet losses to be at most 25-44% assuming a diffusivity of 0.05-0.1 $cm^2 s^{-1}$ and irreversible loss at the wall. We made this more explicit in the text:

The largest source of error in the measured F_p, beyond thermal decomposition during desorption, is diffusion-controlled vapor losses in the inlet which were corrected for by assuming a diffusivity of 0.05-0.1 $cm^2 s^{-1}$, although the variability is not discernable on the log scale. Inlet losses are 25-44% for the range of diffusivities, a small effect on the comparison of measured and predicted F_p as we show below.

9. Lines 481–502: With regards to the possible presence of accretion reaction products: Do you observe any masses with identical profiles to C5H12O6 or C5H10O6 that would be indicative of co-products of the thermal decomposition reactions?

On lines 445-460 we discuss the reasons these specific compounds do not show evidence of being thermal decomposition products. We added "as opposed to thermal decomposition" to the end of the sentence on line 459-460to make it clearer. We also added a paragraph to the end of section 3.3 and a corresponding figure (now Fig. 6) indicating evidence of accretion products measured by the FIGAERO-CIMS, as well as adding the sum of signal from compounds having 6 or more carbons to the bottom left panel of Figure 7 (previously Fig. 6).

10. Lines 521–525: Is it likely that SOA can have high viscosity at the temperatures used for thermal desorption?

How isoprene SOA viscosity varies as a function of temperature remains poorly studied, though our expectation is that viscosity lowers significantly as the aerosol is heated to 200 ^{0}C. It would be interesting to examine the thermal desorption behavior of SOA as a function of known viscosity but this has not been done. It is possible that there are two phases present, one with low viscosity that persists to temperature higher than 100 ^{0}C. However, we have no evidence for two organic phases.

Technical Comments

1. Line 477: Should be "predicted".

Fixed

2. Line 490: Should be FIGAERO-CIMS

Fixed

3. Throughout the paper it seems like you switch arbitrarily from FIGAERO to FIGAERO-CIMS. I suggest you choose one and stick with it.

We changed all "FIGAERO" references to "FIGAERO-CIMS" except where specifically talking about the unit itself.

4. Line 618: Is the volatility of IEPOX really known to 5 significant figures?

Good catch. We have reduced the number of significant figures to 1, more in line with previously stated c*'s throughout the text, and reduced all other group-contribution method obtained c*'s to 1 significant figure as well.

Note: there was an error in the pie chart (figure 4) where we were previously not accounting for collection volume for the "other" portion of the pie. The updated figure does not change our conclusions.

References

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